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Tuning Trap Level with in the Band Gap of $KY_3F_{10}:Ho^{3+}$ Phosphor Powder

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Abstract: The variable heating rate (VHR) method of thermoluminescence (TL) has been employed to investigate how the trap (defect) level changes its position within the band gap of $KY_3F_{10}:Ho^{3+}$commercial phosphor powder after thermal annealing. Thermal annealing alters the structure of glow curves of some TL materials. The effect of annealing on TL glow curves of $KY_3F_{10}:Ho^{3+}$commercial phosphorwas analyzed and the result was compared with that of un annealed one. The sample powder was annealed at different temperatures of 400ºC, 500ºC and 600ºC. The trap depth of the phosphor powder after being annealed at each temperature was calculated using variable heating rate method (VHR). It is found that the trap level can be tuned within the band gap of $KY_3F_{10}:Ho^{3+}$by annealing it at different temperatures. The higher the annealing temperature, the closer the trap level to the conduction band edge. The annealing process also shifts the peak temperatures of the glow curves to higher temperature region.
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Keywords: Tuning, Trap depth, Thermal annealing.

1 Introduction

Heating rates and thermal annealing are the two examples of the most important experimental parameters that cause a change in the shape of TL glow curves [1]. A decrease in TL intensity was observed with increasing heating rate and in general, this phenomena is also observed in $KY_3F_{10}:Ho^{3+}$phosphor powder in our case and it has been explained that this is due to the thermal quenching effectin which the effectiveness of the luminescence decreases as temperature increases because of an increase in the probability of non-radiative transitions [2]. Another effect of heating rate is the alteration of peak temperature of TL peak which shifts to higher values as heating rate increases.
Annealing is a necessary process for dosimetric purposes to erase all information in TL materials and to get good reproducibility. The annealing temperature critically affects the luminescent behavior of a material. Annealing brings about enhancement or decline in the intensity of TL glow Peaks of many TL materials [3-6]. Particularly in this study of $KY_3F_{10}:Ho^{3+}$phosphor powder, an enhancement in its TL intensity was observed following thermal annealing and the peak temperature of each TL peak shifts to higher temperature region. TL study and the effect of thermal treatment of this commercial phosphor are not yet investigated. The change in the location of trap depth after thermal treatment is of our particular interest because of its great impact on band gap engineering of this phosphor. The emission wavelength from this phosphor can be tuned following change in the position of trap depth within the band gap.
It is important to understand the influence of thermal treatments on the TL properties of a crystal. In this work the effect of thermal annealing on the trap depth and TL peak positions of thermoluminescent$KY_3F_{10}:Ho^{3+}$phosphor powder is presented for the first time using the variable heating rate method (VHR). This study shows that thermal annealing affects the TL peak temperatures and the level of the trap below the edge of the conduction band.

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2 Experimental

Commercially available $KY_3F_{10}:Ho^{3+}$standard phosphor powder from phosphor technology (UK) was annealed at temperatures of 400, 500 and 600ºC for one hour in air. The TL glow curves of this phosphor powder annealed at different temperatures at different heating rates are then recorded and compared with that of un annealed one. A UV source was used for TL excitation prior to heating. TL measurements were taken in a nitrogen atmosphere usinga Riso model TL/OSL-DA-20 luminescence reader. The TL was detected by an Electron Tubes Ltd model 9235QB photomultiplier through a BG-39 band-pass filter (transmission band 340-620 nm FWHM). Samples were heated from 0 to 400ºC at various heating rates and for 10 minutes of UV dose. The effect of thermal annealing on trap level and TL sensitivity of the powder phosphor was then investigated. TL measurements were done immediately after stopping the UV exposure. The level of the trap with in the band gap of the phosphor powder was evaluated for each annealing temperature. The structure of the sample powder was also studied by X-ray diffraction (XRD) using a Bruker D8 advance X-ray diffractometer operating at 40kv and 4mA using cu kα= 0.15406 nm. The Morphology was studied using high resolution Transmission Electron Microscope (TEM).

3 Results and Discussion

TL spectra were taken following furnace annealing at different temperatures for 1 h in air. After thermal annealing, there are changes in the TL glow curves.Figure 1ashows the un annealed glow curve (at different heating rates) of $KY_3F_{10}:Ho^{3+}$phosphor. TL glow curves of this phosphor annealed at different temperatures and at different heating rates after irradiation with the same UV dose (10 minutes) are also shown in Figures 1b, 1c and 1d. It is found that the TL intensity increases with the increase of the annealing temperature of the sample.

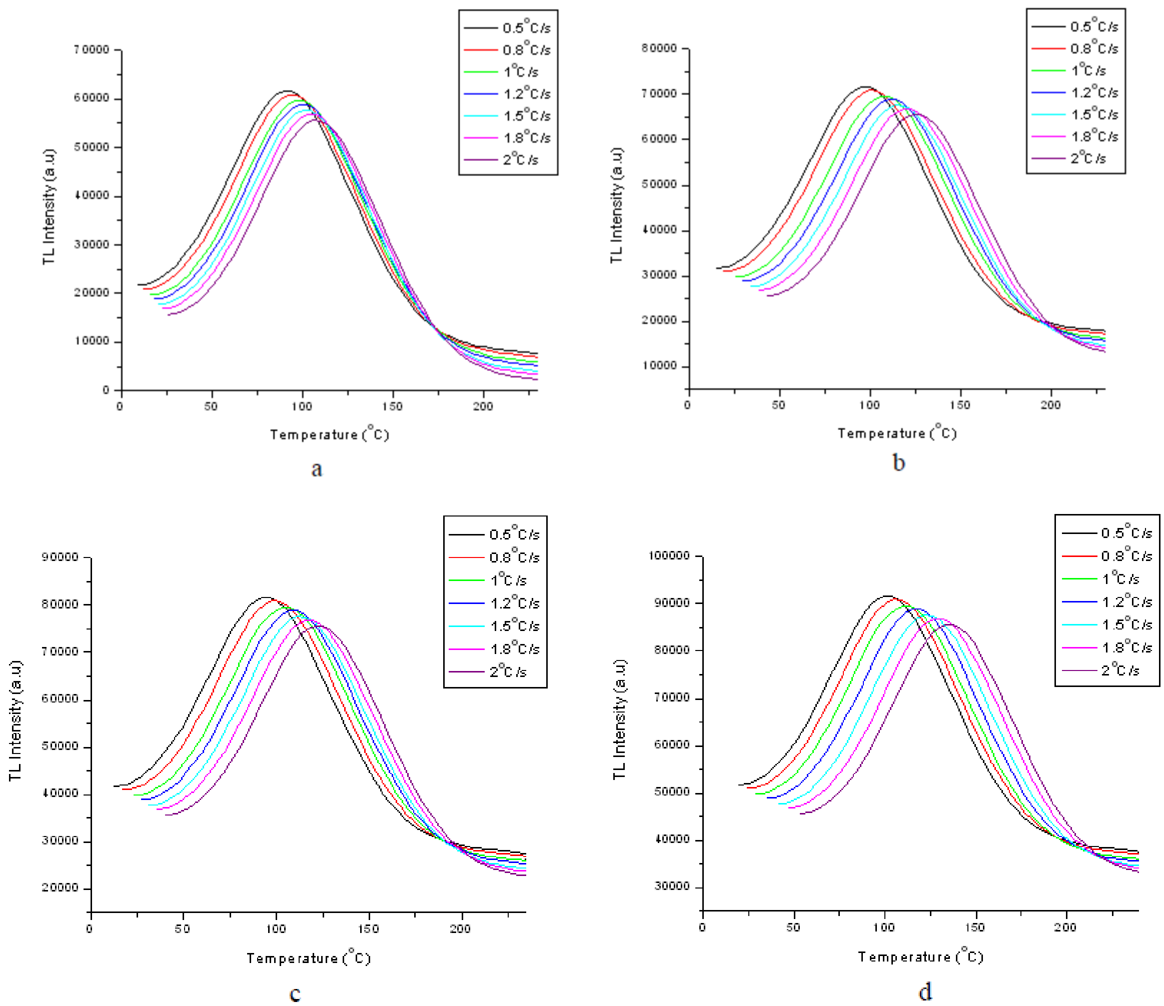

Fig. 1: TL glow curves $KY_3F_{10}:Ho^{3+}$phosphor. (a) Un annealed sample (b) annealed at 400ºC (c) annealed at 500ºC and (d) annealed at 600ºC.

We account this to the fact that higher annealing temperature minimizes the residual TL signal by emptying the high temperature traps and restore the original sensitivity and glow curve structure. The initial non zero value of the TL intensity attributed to the phenomena of fluorescence during UV irradiation. The effect of thermal treatment on the peak temperatures of the glow curves was also analyzed and shown in Figure 2.

This figure clearly shows that the annealing process affects the peak temperatures of glow peak. In all variations, the peak temperatures of all glow peaks of the annealed $KY_3F_{10}:Ho^{3+}$phosphor are higher than that of unannealed one. In addition, the shift in the peak temperature of the glow curves to the higher temperature sides increases with the increasing heating rate except for thermal treatment at 500ºC which shows less shift in peak temperatures as compared to thermal annealing at 400ºC.

Let us now analyze the evolution of the TL maximum with the heating rate in order to determine the level of the trap beneath the edge of the conduction band. For this purpose, we performed a variable heating rate(VHR)analysis that considers how the traps are emptied at different heating rates while all other parameters are held constant. As the heating rate is increased, the position of the TL maximum $T_m$shifts toward higher temperatures (Fig.1). The heating rate βcan be related to the T_m of its corresponding peak using an equation of the following form [7-9]:

$$\frac{E}{kT_m{}^2} = sexp\left(-\frac{E}{kT_m}\right)$$

where$k$ is the Boltzmann constant (in units of eV/K) and E is the activation energy (in units of eV). Change in the linear heating rate βresults in the change of the temperature $T_m$of the maximum TL intensity of the peak: faster heating rates produce a shift in temperature relatively towards higher values of $T_m$as shown in Figure 1. By means of the VHR method, the trap depth (the activation energy) was derived from the variation of $ln\left(\frac{T_M{}^2}{\beta}\right)$ as a function of $\frac{1}{kT_M}$ to see how the annealing process affects the level of the trap within the band gap. When $ln\left(\frac{T_M{}^2}{\beta}\right)$ is plottedagainst$\frac{1}{kT_M}$, the resulting graph consists of astraight line of slope E and intercept (on$ln\left(\frac{T_M{}^2}{\beta}\right)$ axis) $ln\left(\frac{E}{Sk}\right)$. The calculated value of E is 0.92 for un annealed sample, 0.54 for a sample annealed at 400ºC, 0.52 for a sample annealed at 500ºC and 0.44 for a sample annealed at 600ºC.

The comparison of the un annealed and annealed phosphor shows that the annealing process shifts the peak temperatures of the glow curves; the peak temperatures of the glow peaks of the annealed phosphor being higher than that of un annealed phosphor. One of the reasons of the shift in the peak temperatures after annealing can be due to the cluster formation after annealing. The formation of traps clusters instead of randomly distributed defects causes variation in the kinetics of trapping and recombination process [1]. Another reason of the alteration of the peak temperatures after annealing can be the effect of removal and creation of defects as the result of annealing [10].

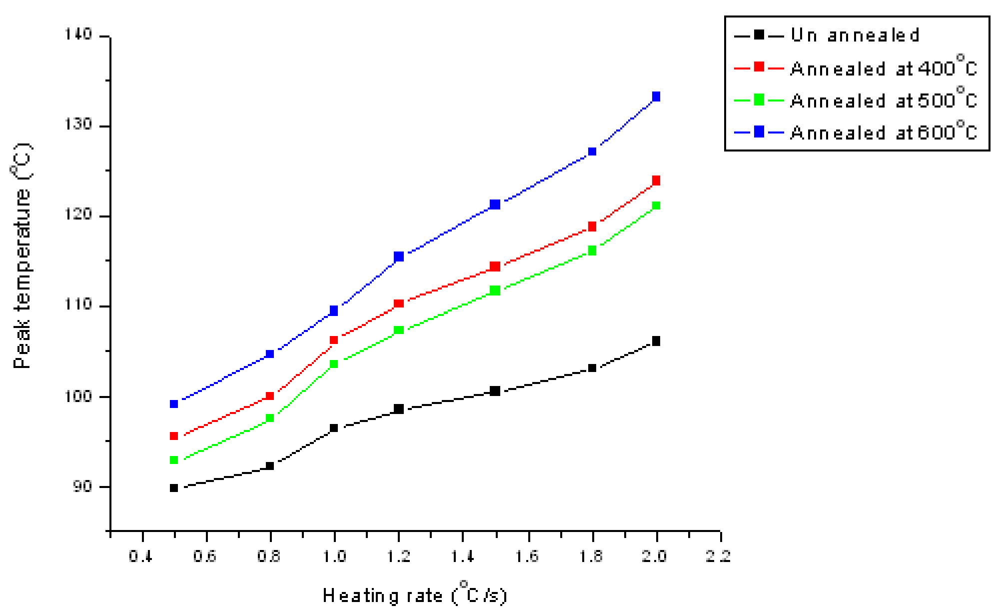

Fig.2: The effect of annealing on the peak temperatures of the glow curves.

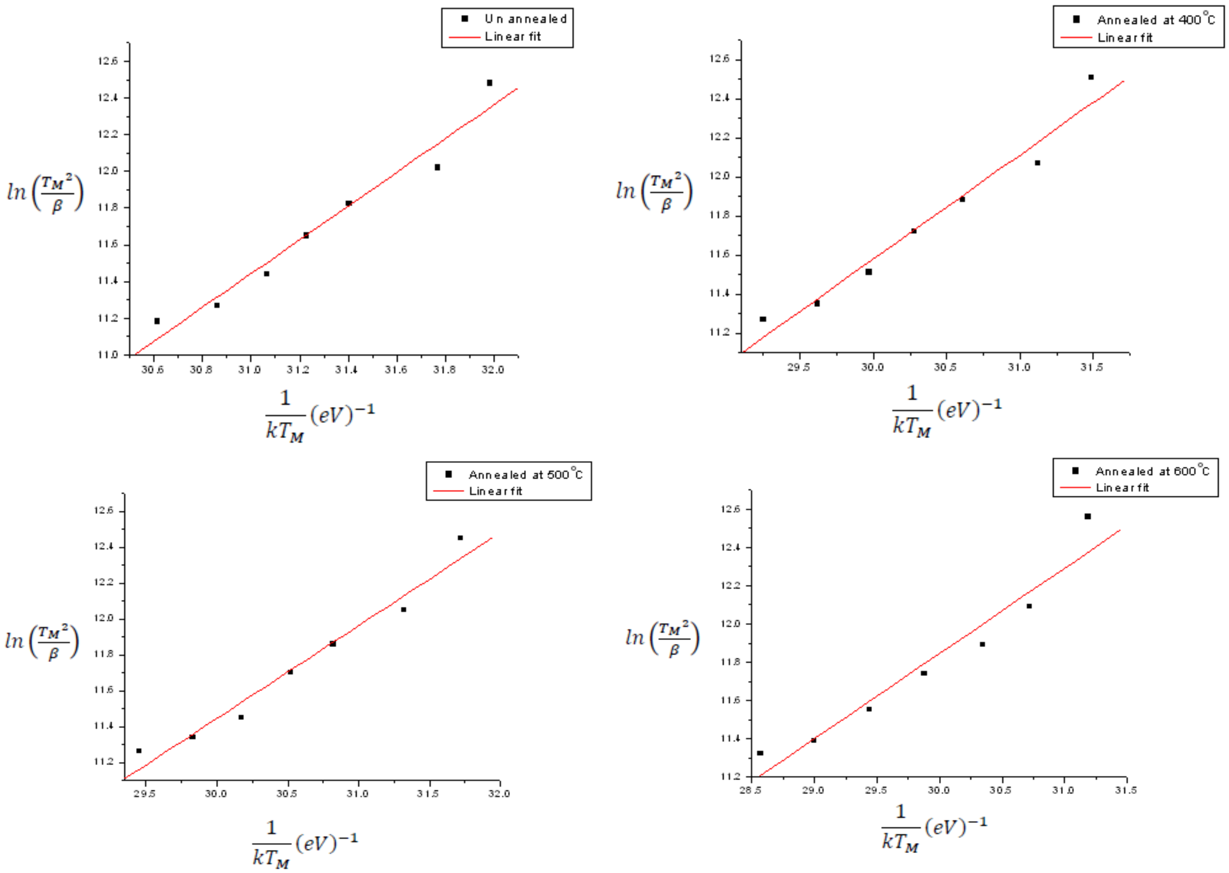

Fig. 3: Calculation of $\frac{1}{kT_M}$ versus $ln\left(\frac{T_M{}^2}{\beta}\right)$ for $KY_3F_{10}:Ho^{3+}$ phosphor powder.

4 Structural and Morphological Studies

Figure 4 shows the XRD pattern of the sample.It shows monoclinic structure with unit cell dimensions (in Angstrom) a=10.41, b=6.73, c=12.46 match with JCPDS Card No 21-1458. The XRD pattern shows almost single Phase with some impurity. The width of the peak increases as the crystallite size decreases. The Transmission electron microscopy (TEM) images are also given in Fig. 5. TEM image indicates that the particles of $KY_3F_{10}:Ho^{3+}$ extend up to several hundreds of nanometers in size.

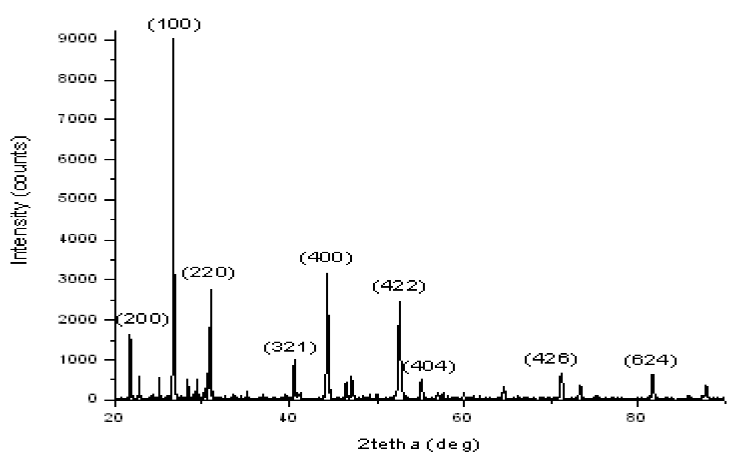

Fig. 4: The XRD pattern of $KY_3F_{10}:Ho^{3+}$ phosphor.

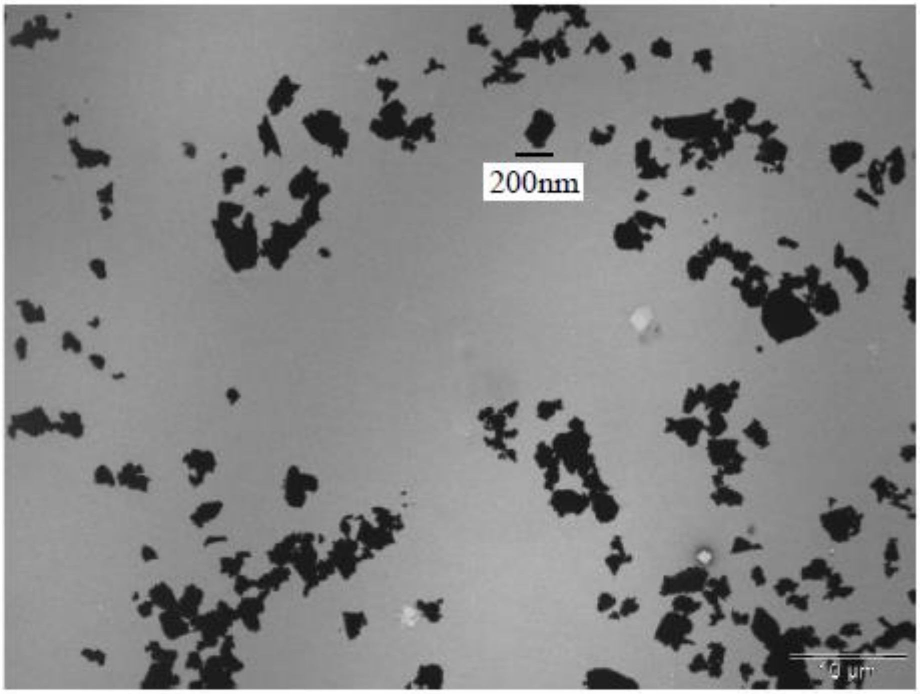

Fig. 5: TEM image of $KY_3F_{10}:Ho^{3+}$phosphor

5 Conclusions

In conclusion, the effect of annealing on the level of the trap within the band gap of $KY_3F_{10}:Ho^{3+}$is studied in detail. It is observed that the trap depth decreases after annealing. Larger decrease in trap depth was observed after annealing at higher temperatures. This study has explicitly shown that the tuning of trap level with in the band gap is possible by the method of thermal annealing. An increment in the intensity of the TL and shifting of the TL peaks towards higher temperature region were also observed following the thermal treatment.

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