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Study of Thermoluminescence Kinetics of Dy Doped Limgbo$_3$ Phosphor

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ABSTRACT

In this study Dy doped $LiMgBO_3:xDy^{3+}$ phosphor has been synthesized by conventional solid state reaction method for different concentrations of Dy (x = 0, 0.0005, 0.001, 0.002, 0.005, 0.01 and 0.02). The synthesized phosphor was characterized for its TL properties that included TL glow curves and TL response for different concentrations of Dy after exposure of 700 Gy gamma rays. Further the kinetic parameters: Activation energy (E) and Order of kinetics (b), were determined using Chen's Peak Shape method and theoretical curves were generated using GCD glow fit. Good match of kinetic parameters have been observed between two methods. Simple glow curve structure and good TL response can make this phosphor a better candidate for future work on radiation dosimetry.
Keywords: $LiMgBO_3$; thermoluminescence; peak shape method; kinetic parameters

I. INTRODUCTION

Borates are of great interest in thermoluminescence (TL) dosimetry, because the borates are relatively stable chemical compounds and respond without serious problems for attempt to dope them with TL sensitizers such as rare earth, copper or manganese ions. Materials such as $Li_2B_4O_7$, MgB_4O_7 have a close tissue equivalence and are thus worth considering for their TL properties (Chopra et al., 2013; Singh et al., 2011; Lochab et al.,2007). Moreover low Z tissue equivalent $LiMgBO_3:xDy^{3+}$ shows excellent TL properties. Initially the monoclinic form of $LiMgBO_3$ was synthesized and its crystal structure was studied by single crystal X-Ray diffraction methods (Norrestam, 1989). The polycrystalline $LiMgBO_3:xDy^{3+}$ material was synthesized by novel solution combustion method and was studied for its luminescence characterstics (Bajaj et al., 2014). Further $LiMgBO_3$ nanoparticles were prepared by Pechini process and their structural, electrical conductivity studies were investigated (Parsad et al., 2016). Recently, the dielectric properties have also been studied for nanocrystalline $LiMgBO_3$ particles (Parsad et al., 2017). To the best of our knowledge, the material has not been synthesized using solid state reaction method earlier. So, in the present study we have synthesized $LiMgBO_3:xDy^{3+}$ phosphor for different doping concentrations of Dy. Further their TL glow curves and response have been noticed and finally kinetic parameters have been focoused using different methods of deconvulation.

II. EXPERIMENTAL

$LiMgBO_3:xDy^{3+}$ (x = 0, 0.0005, 0.001, 0.002, 0.005, 0.01 and 0.02) were synthesized by conventional solid state reaction method. AR grade Li_2CO_3, MgO, H_3BO_3 and Dy_2O_3 were used as starting materials. All the precursors were weighed as per the stoichiometric ratio. The chemicals were pulverized in a porcelain mortar using pestle and transferred into porcelain crucibles. The contents along with the crucibles were kept for heating at 350 ^oC for three hours. After cooling, the contents of the crucibles were crushed and annealed at 700 ^oC for 7 hours. After cooling, the phosphors were collected and characterized for their thermoluminescence (TL)

properties. The samples were irradiated using Co^{60} gamma ray source for dose 700 Gy and then their TL glow curves were recorded on Nucleonix TL1009I TL reader at heating rate of 5 K/sec.

III. RESULTS AND DISCUSSION

3.1 TL properties

The TL properties of synthesized $LiMgBO_3:xDy^{3+}$ phosphor examined in this study includes TL glow curves, TL response at different concentrations of dopant Dy. **Fig. 1** shows the TL glow curves of synthesized $LiMgBO_3:xDy^{3+}$ for different values of x (x = 0, 0.0005, 0.001, 0.002, 0.005, 0.01 and 0.02) for 700 Gy exposure of gamma rays. It is observed that the phosphor shows an isolated peak at around 443 K and the intensity of TL peak keeps on increasing with increase in the concentration of dopant. The TL intensity of pure host material ($LiMgBO_3$) was found to be 100 times lesser than the $LiMgBO_3:0.02Dy^{3+}$. So, the dopant plays an important role in defining the TL properties of phosphor. Doping with Dy introduces predominantly deeper trapping levels in $LiMgBO_3$ and also the enhancement in TL intensity. The change in trap distributions may be due to the lattice perturbation caused by incorporation of Dy in $LiMgBO_3$ (Kumar et al., 2009). Further the effect of dopant concentration on the TL response (peak intensity) has been investigated and is shown in Figure 2. It is observed that the TL intensity increases with the increase in concentration of dopant. An activator which acts as a luminescent centre is surrounded by the non luminescent host centres. Therefore the released charge carriers cannot recombine directly with the luminescent centres. Most probably the energy is transferred non-radiatively through the host lattice to the activator, which on recombination gives characteristic emission (Kumar et al., 2009).

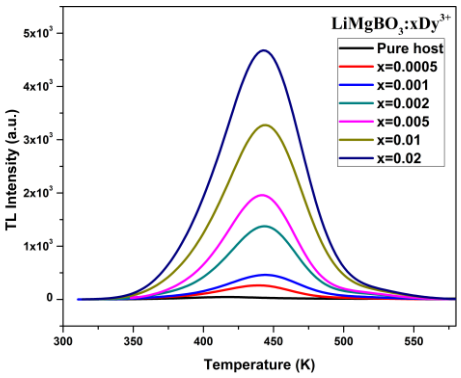

Figure 1. Effect of dopant concentration on TL glow curves of $LiMgBO_3$: Dy samples exposed to 700 Gy gamma rays

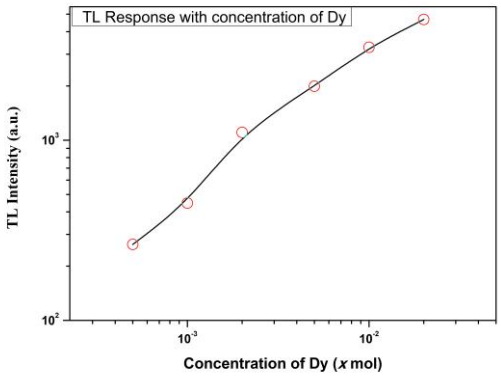

Figure 2. Effect of dopant concentration on TL response of $LiMgBO_3$: Dy samples exposed to 700 Gy gamma rays

3.2 Calculations of trapping parameters

The glow curve is related to the trap levels that lie at different depths in the band gap between the conduction and the valence bands of a solid. These trap levels are characterized by different trapping parameters. Hence, several peaks are found to appear in the TL glow curve at different temperatures. In the present work, the trapping parameters associated with the glow peaks have been calculated using glow curve deconvolution (GCD) glow fit and peak shape method.

(a) Glow Fit Method

The TL glow curve of $LiMgBO_3$: x Dy (x=0.02), exposed to gamma dose of 700 Gy has been deconvolved first by using a computer program, GlowFit (Puchalska and Bilski, 2006) and then the trapping parameters have been obtained. **Fig. 3** shows the composite glow curve and the peaks isolated by using glow fit method. It has been found that the composite glow curve comprises of two peaks. Both deconvoluted peaks (at 442 K and 475 K) show first-order kinetics, i.e. the probability of electron re-trapping during thermoluminescence process

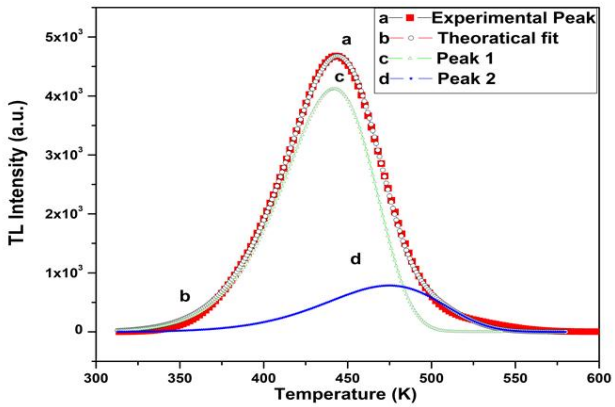

Figure 3. Deconvulation of TL glow curve of $LiMgBO_3$: Dy exposed to 700 Gy of γ-dose.

Table 1. Values of trap depth (E) and order of kinetics for isolated peak calculated by GCD glow fit method and Peak shape method

S. No	Deconvulation Method	PEAK No	Peak T_m (K)	Activation Energy E (eV)	Geometrical form factor (μ_g)
1.	GCD Glow fit method	Peak 1	442	0.60	0.42 (I^{st} order kinetics)
2.	Peak Shape method	Peak 1	442	0.63	0.43 (I^{st} order kinetics)

is negligible. The parameter describing the quality of fitting, known as Figure of Merit (FOM) was found to be 2.71 %. Glow curves with FOM values in excess of 5% are subjected to further investigation to determine the reasons for the poor fit. Since the intensity of peak at 475 K is five times less than the intensity of peak at 442 K, so only prominent peak at 442 K is considered for parameter calculations. The values of trapping parameters; trap depth (E), frequency factor (s) of peak at 442 K have been calculated by GCD glow fit method and are shown in Table 1. Since the dosimetry information stored in the materials after irradiation strongly depends on the position of trapping levels within the forbidden gap, i.e. trap depth. Hence, the trap depths of isolated peaks was also calculated by peak shape method in order to verify the values obtained by GCD glow fit method (Chopra et al., 2013).

(b) Peak Shape Method

The Peak shape method is commonly known as Chen's method (Chen and Winer, 1970) that does not make use of any iterative procedures and does not require knowledge of the kinetic order for calculating the activation energy. The shape of the peak is considered in this method in order to ascertain the trap depth *E* and order of kinetics *b*. TL glow peaks defines the following parameters

T_M = The peak temperature at the maximum

T_1 and T_2 are, respectively, the temperatures on either side of T_M, corresponding to half intensity

$\tau = T_M - T_1$ is the half-width at the low temperature side of the peak

$\delta = T_2 - T_M$ is the half-width toward the fall-off side of the glow peak

$\omega = T_2 - T_1$ is the total half-width

$\mu = \delta/\omega$ is the so-called geometrical shape or symmetry factor.

The kinetic order (b) is found by using the symmetry factor μ from the peak shape. The equations for finding activation energy (E) can be summed up as

$$E_\alpha = C_\alpha \left(\frac{KT_m^2}{\alpha}\right) - b_\alpha (2KT_m) \quad (1)$$

where α is τ , δ, or ω. The values of c_α and b_α are summarized as below:

$c_\tau = 1.510 + 3.0(\mu - 0.42)$, $\quad b_\tau = 1.58 + 4.2(\mu - 0.42)$

$c_\delta = 0.976 + 7.3(\mu - 0.42)$, $\quad b_\delta = 0$

$c_\omega = 2.52 + 10.2\,(\mu - 0.42)$, $\quad b_\omega = 1$

where $\mu = \delta / \omega$ is the symmetry factor that determines the order of kinetics (if μ is close to 0.42 , then this is

the case of first-order TL glow peaks, and if μ is close to 0.52 , then this is the case of second-order peaks). Both the peaks show first order kinetics in the present study. The values of trapping parameters of prominent peak at 442 K have been calculated by peak shape method and are shown in Table 1. It has been observed that the values of trap depth calculated by peak shape method shows a good agreement with the values of trap depth using GCD glow fit method.

IV. CONCLUSIONS

$LiMgBO_3$ doped with Dy was successfully prepared by using conventional solid state reaction method for different concentrations of Dy (x = 0, 0.0005, 0.001, 0.002, 0.005, 0.01 and 0.02) and was found to show single peak TL glow curve at 443 K. The TL response was found to increase with increase in dopant concentration. Further the glow curve was deconvolved to two peaks using GCD glow fit software and both the peaks were found to show first order kinetics. The trapping parameter results were also matched with Chen's Peak shape method. So, the Simple glow curve structure and good TL response can make this phosphor a better candidate for future work on radiation dosimetry.

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