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Thermoluminescence Studies of Ba_2SO_4 by CGCD Method

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Abstract

Computerized Glow Curve Deconvolution (CGCD) method is applied to decode the trapping parameters of the glow curves of $BaSO_4$ in the framework of kinetic formalism. It was observed that all the glow curves irradiated at different doses of -irradiation can be deconvoluted into five constituent peaks and the activation energy is in the range 0.68 – 1.2 eV and frequency factor ~ $10^8 – 10^{11}$ s^{-1}.

Key words: Trapping parameters, activation energy and frequency factor.

1 Introduction

Thermoluminescence (TL) is a temperature stimulated light emission from a crystal after removal of excitation, and is probably one of the direct evidences of existence of trapping level[1]. A complete TL glow curve of a thermoluminescent material may be approximated by a linear combination of glow peak intensities[2]. A glow curve essentially represents the spectroscopy of traps of a solid in a coded form. Analysis of glow curves is nothing but a sort of decoding the uncoded glow pattern. Excellent texts have dealt this challenging area and a large number of papers have appeared in literature [3-7]. Several methods have been developed in decoding the trapping parameters of the glow peaks by many workers [8-10]. With the easy availability of computerized data acquisition and management system, Computerized Glow Curve Deconvolution (CGCD) has been a quite popular to decode a glow curve in the framework of kinetic formalism[11]. This technique is the curve fitting of TL curves consisting of one or more TL peaks and widely used in various area of TL studied and is well documented [12-14]. In the fitting process, the number of minima of the second derivative plot of glow curve can guide the location of the glow peak temperatures. This method can give a mathematical description of thermoluminescence phenomenon and many experimental glow curves can be described with reasonable degree of confidence.

In the present work, we carried out spectroscopic investigation of the TL glow curves of Ba_2SO_4 by the CGCD method. $BaSO_4$ based materials being high sensitive TL materials were used in TL dosimetry [15]. The TL intensity of $BaSO_4$ phosphor increases by the presence of impurities. Intensity increased about 9 times by the presence of Cu as impurity but about 3 times by the presence of Mn when compared with that of undoped $BaSO_4$[16]. Due to its high sensitivity and stability, doped $BaSO_4$ with Cu & Mn was used as personal and environment radiation monitors.

2 Experiments

All the glow curves reported in this work are recorded at Thermoluminescence Dating Laboratory, Manipur University, using the commercial TL/OSL reader (Model TL/OSL – DA -15) in flowing nitrogen atmosphere. In each measurement 20mg of the sample was taken and spread uniformly over the disk to minimize thermal lag at higher heating rate. The photomultiplier tube used is EM19635 in the operating voltage of 950 volts with optical filter Schott UG -11 in conjunction with Schott BG - 39 for filtering out the unwanted radiation. The combination allows transmission in the wavelength range 300 ~ 400 nm. The rate of heating in all reading is 2°C/s, the maximum temperature is set to 575°C. A second readout was performed to record the background radiations which included the black body radiation. The data presented in the present work are all with background correction.

3 Method of Analysis

All the glow curves are analyzed by using the CGCD method in the kinetics formalism. Computing and fitting of the glow peaks followed the general order kinetics [17] including the famous first order [18] and second order equation [19] was done from CGCD program given in the classical text of Chen and Krish [3], the modification incorporated here is the selection of the peak temperature within ± 2°C. The fitting quality in the program is judged from the minimum value of root mean square deviations.

In the CGCD program, the general order kinetics for the TL intensity I(T) depending in the absolute temperature T as developed by Chen [17] is given as

$$I(T) = -\frac{dn}{dt} = s'n^{b} \exp\left(-\frac{E}{kT}\right) \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

where $s(= s'n_o^{b-1})$ is the frequency factor, n_o the initial concentration of trapped electrons, b the kinetic order, E the activation energy and k the Boltzman's constant.

For a linear heating profile,

$$T=T_o + \beta t \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (2)$$

where T_o is the initial temperature, T the temperature at time t and β the linear heating rate.

From equations (1) and (2) we get Randall and Wilkins [18] equation (b=1)

$$I(T) = n_o s \exp\left(-\frac{E}{kT}\right) \exp\left[-\frac{s}{\beta}\int_{T_o}^{T} \exp\left(-\frac{E}{kT'}\right) dT'\right] \ldots\ldots\ldots\ldots\ldots (3)$$

and general order kinetic equations (b≠1)

$$I(T) = n_o s \exp\left(-\frac{E}{kT}\right)\left[1+\frac{s(b-1)}{\beta}\int_{T_o}^{T} \exp\left(-\frac{E}{kT'}\right) dT'\right]^{\frac{-b}{b-1}} \ldots\ldots (4)$$

with successive integration by parts over a limited number of terms, Chen and Winer [20] approximated the integral as

$$\int \exp\left(-\frac{E}{kT'}\right) dT' \approx \frac{kT^2}{E} \exp\left\{\left(-\frac{E}{kT}\right)\left(1-2\frac{E}{kT}\right)\right\} \ldots\ldots\ldots\ldots\ldots (5)$$

Then the TL intensity may be written [21] on the general form as

$$I(T) = I_m b^{\frac{b}{b-1}} \exp\left(\frac{E}{kT}\right)\left(\frac{T-T_m}{T_m}\right) \times \left[(b-1)\left(1-2\frac{kT}{E}\right)\left(\frac{T^2}{T_m^2}\right) \times \exp\left(\frac{E}{kT}\right)\left(\frac{T-T_m}{T_m}\right) + \left\{1+\frac{2kT_m(b-1)}{E}\right\}\right]^{\frac{-b}{b-1}} \ldots\ldots\ldots\ldots\ldots (6)$$

where, I_m and T_m are the maximum intensity and peak temperature respectively.

And one may deduce the frequency factor s also as

$$s = \left(\frac{\beta E}{kT_m^2}\right)\left(\frac{1}{1+\frac{(b-1)2kT_m}{E}}\right) \exp\left(\frac{E}{kT_m}\right) \ldots\ldots\ldots\ldots\ldots\ldots\ldots (7)$$

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Though Chen's general order kinetics formalism [17] does permit order of kinetics 'b' beyond two (i.e. b > 2), conventional analysis reported in the literature [22] shows $1 \le b \le 2$.

The goodness of fit of the measured TL glow curve was again tested using standard statistical χ^2-test of normality [23-24] which measured goodness of fit in term of normality of error distribution. As a cross check, Figure of Merit (FOM) [25-26] was also calculated.

A simple method of heating correction [27] used to determine the exact effective heating rate (β_{eff}) between the heating element and the thermoluminescent sample during the TL readout in the reader by using the equation,

$$\beta_{eff} = \frac{(T_g - T_0 - \Delta T)}{(T_g - T_0)}\beta = \frac{(T_m - T_0)}{(T_g - T_0)}\beta \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (8)$$

where $\Delta T = T_g - T_m$, T_g is the observed peak temperature (K) and T_m is the real peak temperature (i.e., with thermal lag correction), T_o is room temperature (25°C).

4 Results and Discussion

The TL glow curves of $BaSO_4$ subjected to various doses (1, 2, 3, 4, 5, 6, 7, 8, 9 and10 Gy) of β-irradiation at a constant heating rate of 2°C/s is shown in Figure 1a. Another set of glow curve with higher doses (5, 10, 15, 25, 50 and 100Gy) is also shown in Figure 1b. In all cases same pattern of glow curves are observed with most intense peak at about 190°C.

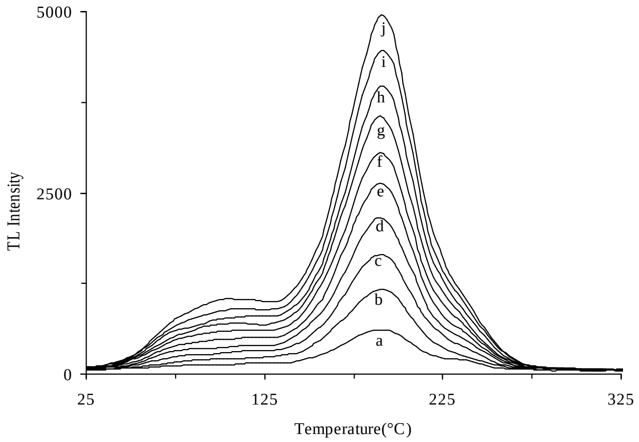

Figure 1a. Glow curves of $BaSO_4$ subjected to different dose o β-irradiation with constant heating rate $2°Cs^{-1}$

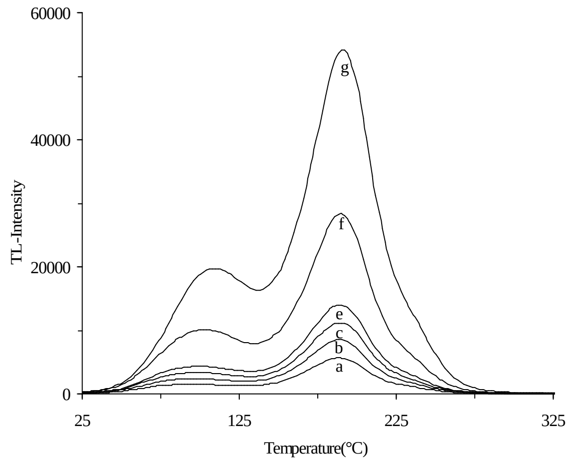

Figure 1b. Glow curves of $BaSO_4$ subjected to different dose of β-irradiation with constant heating rate $2°Cs^{-1}$. (Curves a, b, c, d, e, f and g corresponds

The experimental glow curves show a complex structure with the indication of a number of TL peaks all over the region. The dose responses for the two set of irradiation are shown in Figure 2a & 2b. The results of the measurements show a linear dose up to 100Gy. The glow curves presented in Figure 1b are subjected to CGCD within the framework of kinetic formalism[17-19]. The program takes into consideration the best-fit with variable intensity and temperature.

Table 1. Trapping papramaters for different peaks at different peaks

Trapping parameters	Dose (Gy)	I - Peak	II-Peak	III-Peak	IV-Peak	V-Peak	FOM %	c^2
T_m (°C)	5	68	90	120	186	220.5	0.06	4.12 (d.f =2)
E(eV)		0.7	0.79	0.8	0.91	1.2		
s (s^{-1})		3.09×10^{9}	1.26×10^{10}	2.15×10^{9}	9.84×´10^{8}	2.01×10^{11}		
b		1.04	1.34	1.22	1	1.4		
T_m (°C)	10	74	98	124	186	218	0.09	2.61 (d.f=2)
E(eV)		0.68	0.72	0.8	0.91	1.19		
s (s^{-1})		9.87×10^{8}	7.31×10^{8}	1.68×10^{9}	9.84×10^{8}	1.82×10^{11}		
b		1	1.04	1.06	1	1.48		
T_m (°C)	15	76	106	134	190	224	0.31	1.83 (d.f=3)
E(eV)		0.68	0.69	0.81	1.07	1.15		
s (s^{-1})		8.36×10^{8}	1.67×10^{8}	1.13×10^{9}	9.52×10^{10}	5.1×10^{10}		
b		1.3	1	1.96	1	1.47		
T_m (°C)	25	76	100	126	187	220	0.19	6.40 (d.f=3)
E(eV)		0.68	0.72	0.8	0.9	1.1		
s (s^{-1})		8.55×10^{8}	6.43×10^{8}	1.45×10^{9}	7.16×10^{8}	1.80×10^{10}		
b		1.01	1.01	1.3	1.01	1.47		
T_m (°C)	50	83	106	134	188	219	0.31	5.77 (d.f=3)
E(eV)		0.68	0.75	0.81	0.99	1.19		
s (s^{-1})		5.27×10^{8}	1.14×10^{9}	1.19×10^{9}	7.19×10^{9}	1.71×10^{11}		
b		1.02	1.01	1.3	1.01	1.49		
T_m (°C)	100	88	108	136	190	221	0.34	3.2 (d.f=2)
E(eV)		0.69	0.69	0.82	0.99	1.15		
s (s^{-1})		5.27×10^{8}	1.48×10^{8}	1.28×10^{9}	6.5×10^{9}	5.76×10^{10}		
b		1.02	1	1.85	1.03	1.4		

For correcting the heating rate we followed the works of Kitis and his co-workers[27]. The results of the analysis are presented in Table 1 and some of the fitting are shown in Figure 3a and 3b.

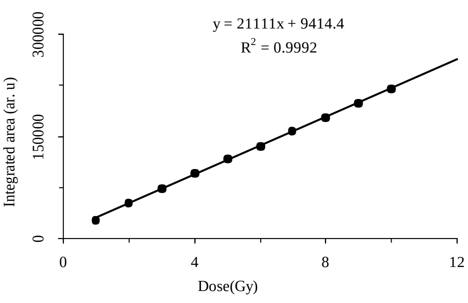

Figure 2a. Dose Response curve for lower dose of irradiation.

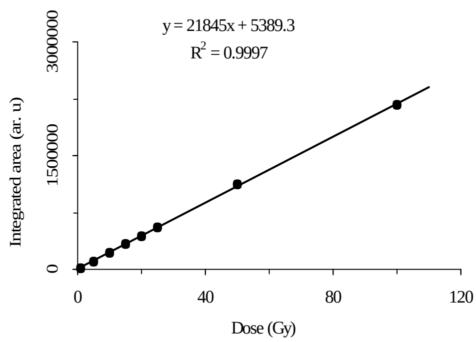

Figure 2b. Dose Response curve for higher dose of irradiation.

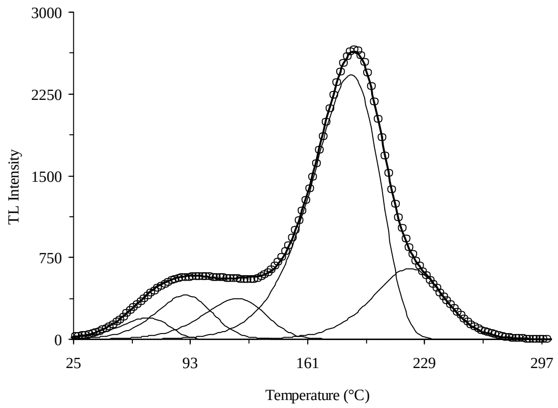

Figure 3a.CGCD of BaSO4 irradiated at 5Gy. (Heating rate = 2°Cs^{-1})

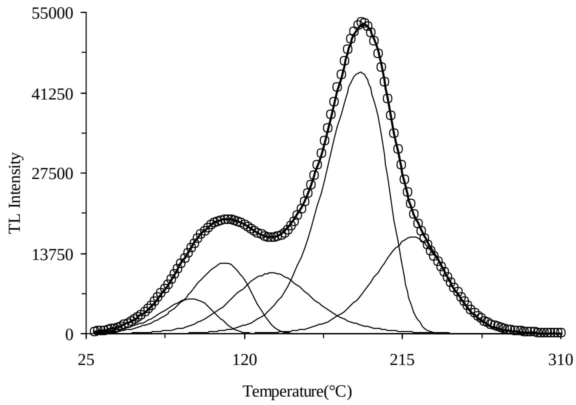

Figure 3b. CGCD of BaSO4 irradiated at 100Gy. (Heating rate = 2°Cs^{-1})

oooooo Experimental curve

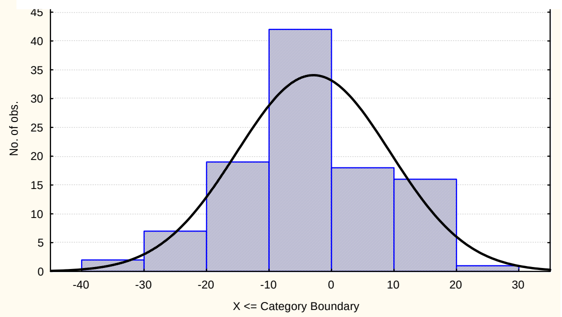

Figure 4a. Error distribution of TL glow curve of BaSO₄ corresponds to the dose of 5Gy.

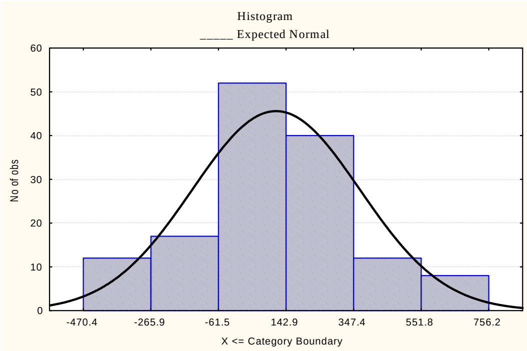

Figure 4b.Error distribution of TL glow curve of BaSO4

corresponds to thedose of 100Gy.

The outcomes of the analysis show that not only the values of the key trapping parameters namely E, s and b are in physically realistic range, but also the fitting are extremely good which are also supported by FOM and χ^2 values. In all cases FOM is less than 1% and χ^2 test passes at 5% level of probability. The histogram of error also presented in Figure 4a and 4b, which shows the normality of deviation (Experimental – Theoretical).

5 Conclusion

Trapping parameters obtained from the rigorous analysis of $BaSO_4$ glow curves irradiated at different dose of β-irradiation show activation energy in the range 0.68 – 1.2 eV and frequency factor in the range $10^8 – 10^{11}$ s^{-1}. Each glow curves can be fitted by five constituent's peaks.

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