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On the Suitability of Peak Shape Method for the Analysis of Thermoluminescence in Different Models

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Abstract

In the present paper we have adjudged the suitability of widely used Peak Shape method for determination of activation energy by considering computer generated thermoluminescence (TL) peaks obtained by using One Trap One Recombination (OTOR) model and Interactive Multi Trap System (IMTS) model. We have found that in both OTOR and IMTS models these methods fail when the trap occupancy approaches saturation. Finally we have considered the application of these methods to experimental TL peaks of γ *-irradiated Albite.*

KeywordS: Thermoluminescence (TL), One Trap One Recombination (OTOR),InteractiveMulti Trap System (IMTS), activation energy

I Introduction

Thermoluminescence (TL) is a phenomenon exhibited by an insulating or semiconducting solid following the previous absorption of energy when the solid is irradiated with ionizing radiation such asX-rays, γ-rays or β-rays. It is distinct from blackbody radiation emitted by a solid when it is heated to incandescence. For TL heat is the only stimulator and not the cause of emission of TL because TL material cannot be made to emit light by simply cooling the sample and reheating it. TL has important applications in dosimetryand dating [IX], [X]. TL emission is graphically represented as a glow curve which shows the variation of TL intensity usually as a function of temperature. Different trapping levels lying in the forbidden energy gap of the solid are responsible for the glow curve and these levels give rise to different peaks in the glow curve. So the individual TLpeaks in the glow curve provide information regarding different trapping levels. The conventional trapping parameters in General Order Kinetics (GOK) model [IX], [X]of TL are respectively the activation energy (E), frequency factor(s) and order of kinetics (b). But the GOK model cannot explain all the characteristics of TL glow curve because it does not include the effect of Thermally Disconnected Deep Traps (TDDTs) [IX], [X]. In view of these, different models are developed which are intimately connected to the band picture of the solid. These models are known as One Trap One Recombination (OTOR) model and Interactive Multi Trap System (IMTS) model [IX], [X]. In IMTS model the effect of TDDTs and their interaction are taken into account. Peak Shape method is one of the most popular methods for thedetermination of activation energy in TL developed by Chen [VII], [VIII]. Later on using a novel technique developed by Christodoulides [II] and Singh et al [XVII] modified the Peak Shape method of Chen [VII], [VIII]. In the present paper we consider the suitability of Peak Shape method in the light of OTOR and IMTS models. Apart from the half intensity points, here we have used points in the glow curve corresponding to fractional intensities 2/3 and 4/5,because the upper portion of a peak is usually less affected by overlapping due to satellite peaks [XII]. Finally we consider the application of thismethodtosome experimental TL peaks of Albite irradiated by different doses of γ radiation [XIV].

II Methodology

In peak shape method (PS) the activation energy is determined by using a set of expressions connecting the peak temperature T_m and temperaturesT_{y1} and T_{y2} corresponding to the points in the rising and falling sides of TL peak for which fractional intensity $I/I_m=y$ (say), in which $I_m$is the intensity at peak temperature T_m .The expression for activation energy can be expressed as [XVII], [XV]

$$E = \frac{C_x k T_m^2}{x} + D_x k T_m \quad (1)$$

where x is one of the quantities τ, δ or ω defined by

$\tau = T_m - T_{y1}$, $\delta = T_{y2} - T_m$, and $\omega = T_{y2} - T_{y1}$

The symmetry factor $\mu_g(y)$ corresponding to the fractional intensity y is given by

$$\mu_g(y) = \frac{T_m - T_{y1}}{T_{y2} - T_{y1}} \quad (2)$$

The coefficients C_x and D_x appearing in Eq. (1) can be represented as [XVII]

$$C_x = C_0 + C_{1x} b + C_{2x} b^2 + \cdots \quad (3)$$

$$D_x = D_0 + D_{1x} \mathrm{b} + D_{2x} b^2 + \cdots \quad (4)$$

It is well known that the occurrence of an isolated TL peak is very rare. In complex TL curve there can be interference from other peaks both in the rising and falling sides. So it is better to calculate activation energy not only using only half intensity points($y=\frac{1}{2}$) but also the points of other fractional intensity say 2/3 and 4/5 as mentioned by Sunta [III]. In the present work we consider not only the fractional intensity points of $y=\frac{1}{2}$ but also the points with $y = \frac{2}{3}$and $\frac{4}{5}$. The values of

C_x and D_x for $y = \frac{1}{2}$ have been calculated by Singh et al [XVII]. In the present paper we have calculated the values of C_x and D_x for $y = \frac{2}{3}$ and $\frac{4}{5}$ (Table -1) following the technique adopted by Singh et al [XVII], [XV].

Table -1: Coefficients C_0, C_1, C_2 and D_0, D1, D2 occurring in Eqs. (2) and (4) for x $= 2/3$ and $4/5$

y	x	C0	C1	C2	D0	D1	D2
	τ	0.682	0.428	-0.051	-0.718	-1.19	0.095
2/3	δ	0.147	0.687	-0.045	0.181	-0.435	-0.092
	ω	0.828	1.111	-0.102	-0.524	-0.611	-0.028
	τ	0.451	0.339	-0.045	-0.478	-1.187	0.087
4/5	δ	0.154	0.491	-0.039	0.182	-0.610	-0.063
	ω	0.603	0.832	-0.083	-0.295	-0.779	-0.005

The literature [IX], [X]points towards the necessity of model beyond GOK. We have already mentioned two such models OTOR and IMTS [IX], [X]. These models have been based on the band picture of solids and the concept of trapping of charge carriers. The set of coupled differential equations in OTOR model is given by [XVI]

$$\frac{dn_t}{dt} = -n_t s exp[-E/kT] + n_c(N - n_t)A_n \tag{5}$$

$$\frac{dn_c}{dt} = n_t s exp[-E/kT] - n_c(N - n_t)A_h - n_c n_h A_h \tag{6}$$

where N is the total concentration of electron traps in the crystal, n_t is the concentration of filled electrons held by the traps, n_c is the concentration of electron in the conduction band and n_h is the concentration of holes in the recombination centre. E is the activation energy corresponding to the trap under consideration. A_n and A_h are respectively capture coefficients of trap and recombination centres. The charge neutrality condition is given by [I]

$$n_h = n_t + n_c \tag{7}$$

In the OTOR model the effect of Thermally Disconnected Deep Traps (TDDT) is not considered. The importance of TDDTs in TL process has been pointed out by Fain et al [V], [VI].

In IMTS model the effect of TDDTs have been considered. The IMTS model is described by the following set of coupled differential equations [XVI]

$$\frac{dn_t}{dt} = -n_t s exp\left[-\frac{E}{kT}\right] + n_c(N - n_t)A_n \quad (8)$$

$$\frac{dm}{dt} = n_c(M - m)A_m \quad (9)$$

$$\frac{dn_c}{dt} = n_t s exp\left(-\frac{E}{kT}\right) - n_c(N - n_t)A_n - n_c(m - M)A_m - \frac{dn_h}{dt} \quad (10)$$

The charge neutrality condition is given by [XVI]

$$n_h = n_t + m + n_c \quad (11)$$

The TL intensity is given by

$$I = -\frac{dn_h}{dt} = A_h n_c n_h \quad (12)$$

In the above equations (8) – (12) M and m are respectively the concentration of total and filled TDDTs. A_m is the capture coefficients of TDDTs. Other symbols have the same meaning as in OTOR model. To convert the OTOR and IMTS equations from time domain to temperature domain we use linear heating technique. The equation govern linear heating technique is given by

$$T = T_0 + \beta t \quad (13)$$

III Results and discussion

TL intensity I in OTOR and IMTS models have been obtained by solving relevant set of coupled differential equations [Eqs. (5)-(7) for OTOR model and Eqs. (8)- (12) for IMTS model] numerically by using modified version of the fourth order Runge-Kutta Method modified by Gill [XIII]. The solutions are very much sensitive to step size (temperature difference) ΔT used in the computation. The step size is varied from 0.005K to 0.001K. As a check of our computer code we reproduced the OTOR and IMTS results as reported by Pagonis et al [XVI]. The values of n_0 and m_0 have been obtained by assuming that traps are filled up with dose according to saturating exponential function and their filling rate constants are proportional to A_n and A_h respectively[IV]. We have computed numerical TL curves in OTOR and IMTS models and applied peak shape method (PSM) to these numerical curves in order to investigate the applicability of peak shape method to an arbitrary TL curve. In Figs. 1 and 2 we have presented two TL curves corresponding to OTOR and IMTS models, respectively. Using both the models OTOR and IMTS we have computed the proportional errors in the determination of activation energy denoted by

$\Delta E_x (= \frac{|E_{in} - E_x|}{E} \times 100\%), E_{in}$ is the input value of activation energy, E_x is the activation energy as calculated by peak shape method where $x = \tau, \delta$ and ω .In Fig. 3 we depict the variation of ΔE_ω with filling ratio $f\left(= n_0 / N\right)$ for OTOR model, where n_0 is the initial concentration of trapped charge carriers. It is evident from Fig. 3 that ΔE_ω increases with increasing f , as f increases from 10^{-6} to 1, ΔE_ω increases from about 2% to 11%.

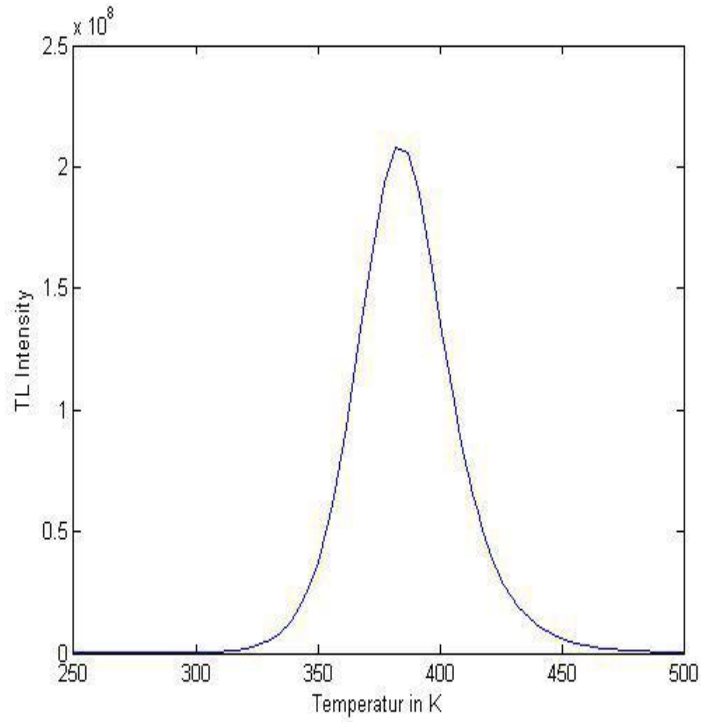

Fig. 1: TL curve for OTOR model; E=1.0 eV, $s=10^{12}\ s^{-1}$, $N=10^{10}\ cm^{-3}$, $A_n=10^{-7}\ cm^3\ s^{-1}$

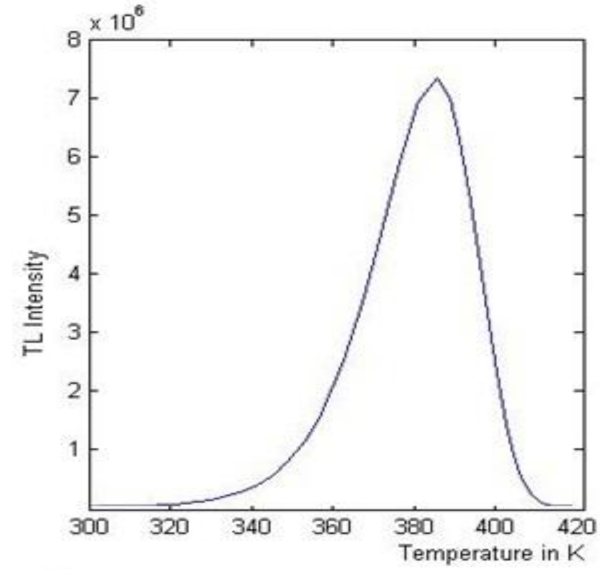

***Fig. 2**: TL curve for IMTS model; E=1.0 eV, $s=10^{12}\ s^{-1}$, $N=10^{10}\ cm^{-3}$, $M=10^{10}\ cm^{-3}$, $A_n=10^{-7} cm^3s^{-1}$, $Am=10^{-5}\ cm^3s^{-1}$,*

$A_h=10^{-5}\ cm^3s^{-1}$

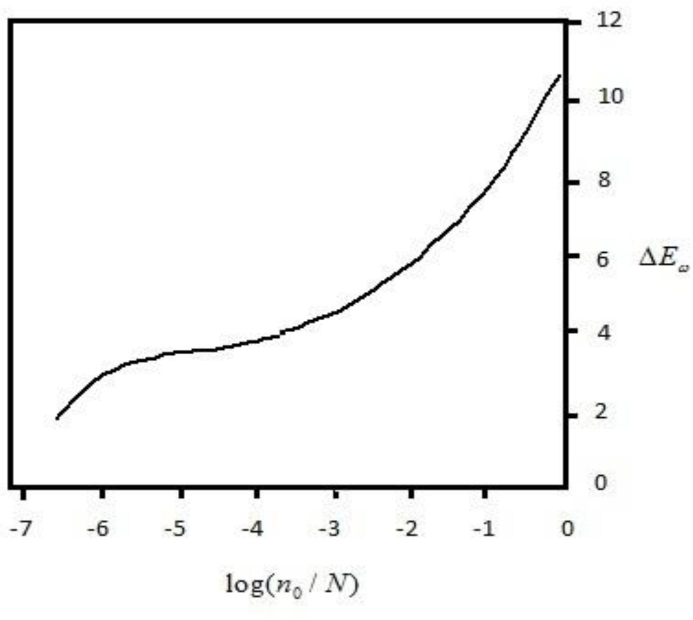

Fig. 3: Variation of ΔE_ω with log (n_0/N) for OTOR model, y=1/2

In Fig. 4 we display the variation of ΔE_{ω} with f in case of IMTS model. However, the results are similar to those of OTOR model. Our findings are in fair agreement with that reported by Sunta [III]. We have obtained similar type of results for ΔE_{ω} in both the models for $y = 2/3$ and $y = 4/5$. These are shown in Figs.5 and 6.Inall these cases we find that ΔE_{ω} increases with increase in the filling ratio f $\left(y = 1/2, 2/3, 4/5\right)$. We have also found that ΔE_{τ} and ΔE_{δ} show similar dependence on the filling ratio.

We now consider experimental TL curve of Albite irradiated by various dose of γ -rays from CO^{60} [XIV]. The peaks occur in the region 383K-387K with dose in the range 177Gy to 468Gy. A close inspection of the symmetry factor μ_g of the peak shows that the peak follows second

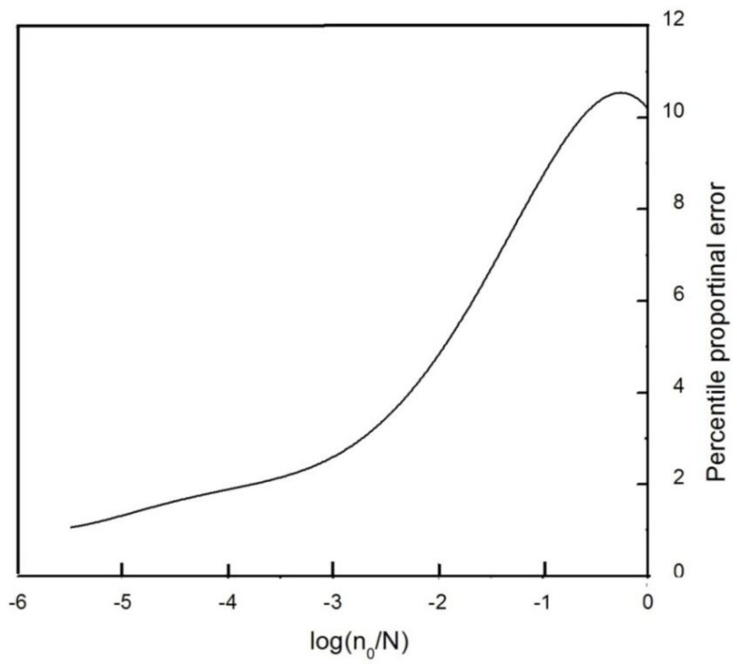

Fig. 4: Variation of ΔE_{ω} with log (n_0/N) for IMTS model, y=1/2

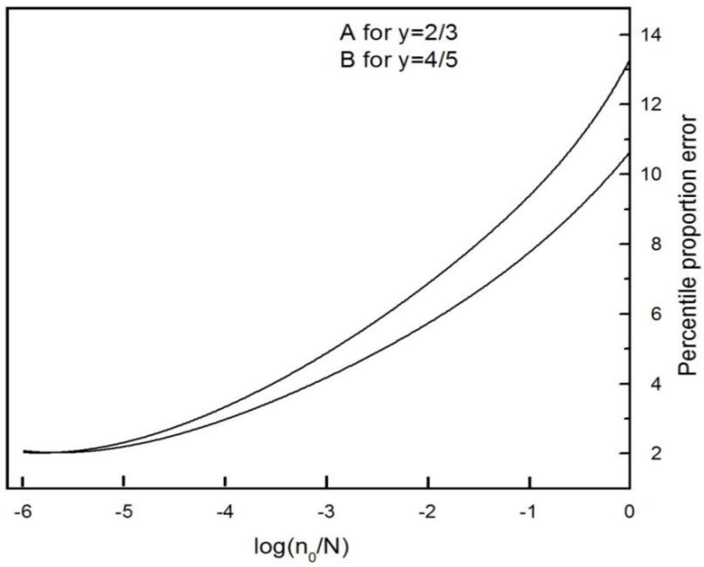

Fig. 5: Variation of ΔE_ω with $\log\left(\frac{n_0}{N}\right)$ for y=2/3 and 4/5 for OTOR model

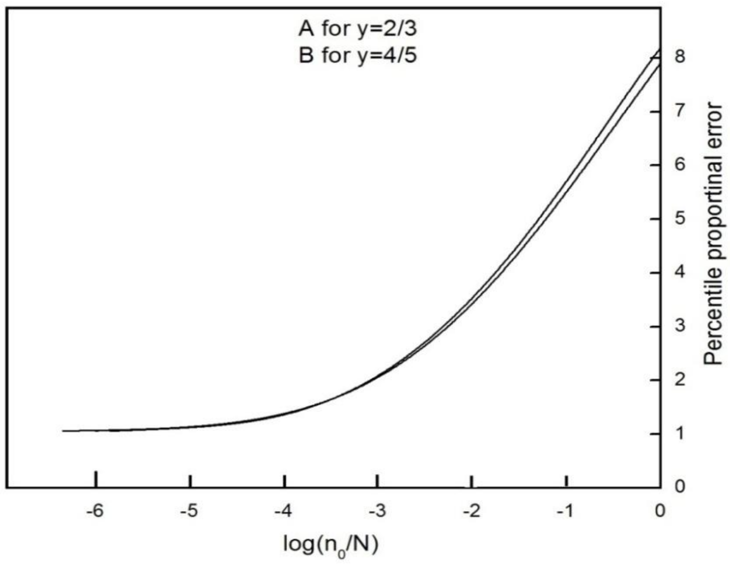

Fig. 6: Variation of ΔE_ω with $\log\left(\frac{n_0}{N}\right)$ for y=2/3 and 4/5 for IMTS model

order kinetics since μ_g lies between 0.50 and 0.52 [IX], [X]. For consistency [III] we have considered not only the half intensity points but also the points having fractional intensities 2/3 and 4/5. In this case $\mu_g(y)$ decreases with increase in y indicating asecond order kinetics (b=2) [III]. Use of temperature at different intensity ratios normally is expected to provide more reliable results from statistical point of view since more number of points are used for the same glow curve. The results of our calculation are presented in Table-2. The results indicate that the values of $E_x(x = \tau, \delta, \omega)$ are not only consistent for a particular dose but also for a wide range of γ- doses. In order to further check the consistency of our results we have determined the values of activation energy and order of kinetics of the peaks by using the rigorous curve fitting method of Chen and Krish [XI]. In order to test the goodness of fit we have also calculated the figure of merit (FOM) [9]. The results are presented in Table-2. The values of FOM indicate a good fit [XV]. The values of *E* and *b* obtained as a result of curve fitting are indicated by E_{cf} and b_{cf}. The values of E_{cf} and b_{cf} are in agreement with the values of activation energy and order of kinetics as obtained by peak shape method.

Table – 2: Determination of activation energies and order of kinetics of peaks of Albitewithdifferent dose

Dose Gy	T_m (K)	y	$\mu_g(y)$	b	E_τ eV	E_δ eV	E_ω eV	E_{cf}	b_{cf}	FOM
		1/2	0.523	2.0	0.976	0.960	0.968			
177	386	2/3	0.500	2.0	0.965	1.024	0.995	0.98	2.0	1.05
		4/5	0.500	2.0	0.989	1.033	1.012			
		1/2	0.523	2.0	0.966	0.950	0.957			
237	384	2/3	0.490	2.0	0.922	1.013	0.968	0.97	2.0	0.98
		4/5	0.500	2.0	1.027	1.070	1.049			
		1/2	0.549	2.0	0.997	0.876	0.929			
296	385	2/3	0.516	2.0	0.993	0.987	0.990	1.06	2.1	1.18
		4/5	0.511	2.0	1.033	1.027	1.030			
		1/2	0.523	2.0	0.997	0.976	0.986			
414	385	2/3	0.508	2.0	1.029	1.053	1.040	1.01	2.0	1.05
		4/5	0.500	2.0	1.033	1.076	1.055			
		1/2	0.511	2.0	0.927	0.960	0.945			
468	386	2/3	0.500	2.0	0.93	0.992	0.963	0.93	1.9	1.02
		4/5	0.522	2.0	1.308	0.988	1.012			

IV Conclusion

In the present paper we have studied the suitability of peak shape method in the light of some existing models of TL namely OTOR and IMTS. It is shown that the percentile proportional error in the determination of activation energy by this method increases with increasing the filling ratio. The methodis found to be suitable for TL peaks for which filling ratio is low. But as the filling ratio increases the method tends to lose its validity. For completeness the peak shape method has been applied to experimental TL peaks of γ-irradiated Albite and encouraging results have been obtained.

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