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On the computation and curve fitting of TL glow curves using MATLAB

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Abstract

Thermoluminescence glow peaks obeying first order and non-first order kinetics have been constantly used to explain the experimental glow curves relevant to dating and dosimetry. Analysis of glow peaks for numerically simulated peaks using MATLAB software has been investigated. The applicability of computerized glow curve deconvolution using MATLAB software has also been reported

Keywords: thermoluminescence, activation energy, order of kinetics, deconvolution.

1.0 INTRODUCTION

Most insulating or semiconducting materials exhibit thermoluminescence (TL) glow curve ususally with a number of peaks when charge carriers are released. After a substance has absorbed energy from exposure to ionizing radiation such as X-rays or γ-rays etc., its glow curve is extracted by heating the substance under a controlled heating scheme and measuring the glow intensity as a function of temperature. The shape of the glow curve is related to the orders of kinetics of the thermoluminescence peaks. Different traps lying within the valence band and the conduction band of the solid are characterized by various trapping parameters viz. activation energy (E) i.e. the trap depth, frequency factor (s) and the order of kinetics (b) [1,2]. Controlled measurement of the emitted light from the TL material is normally used for the determination of the radiation dose absorbed by it. To understand the behaviour of the material, it is necessary to analyse the glow curve and evaluate the trapping parameters. Glow curve is also used to study the dependence of TL on linear energy transfer (LET) from the radiation [3,4], predicting the fading in TL [5] and to determine the life time of glow peaks.

The computerized glow curve deconvolution (CGCD) into the individual glow peaks have recognized to be of major importance in the analysis of TL. The information obtained from CGCD enables one to understand the mechanism of TL in different materials. Capabilities of several computer codes for CGCD and their assessment of glow curve parameters were adjudged by Glow Curve Analysis Intercomparison (GLOWCANIN) project [6-8], a joint venture of IRI at Delft, The Netherlands and CIMAT at

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Madrid, Spain. In the present work, we attempt to develop a new computer code for the deconvolution of the TL glow curves using MATLAB software. We have considered a number of models : i) 1st order kinetics model of Randall and wilkins [9], ii) General Order Kinetics (GOK) model [10] which reduces to the Garlick and Gibson model for b = 2 [11], iii) One Trap One Recombination centre model (OTOR) [12], iv) Interactive multitrap system [IMTS] model [12]. In the first step, we compute TL curves in different models and finally we apply the present technique to both computer generated and experimental glow peaks.

2.0 METHODOLOGY

We start from OTOR model [12] which can be represented by a set of coupled differential equations

$$\frac{dn}{dt} = -nse^{-\frac{E}{kT}} + n_c(N-n)A_n \tag{1}$$

$$\frac{dn_c}{dt} = nse^{-\frac{E}{kT}} - n_c(N-n)A_n - n_c n_h A_h \tag{2}$$

The charge neutrality condition [12] in OTOR model is given by

$$n_h = n + n_c \tag{3}$$

where, n and n_c are the concentration of electrons in traps and in the conduction band respectively. n_h is the concentration of holes in the recombination centres. A_n and A_h are retrapping and recombination coefficients respectively. N is the total concentration of the electron traps, k is the Boltzmann constant and T is the temperature. The TL intensity I is given by

$$I(t) = -\frac{dn_h}{dt} = n_c n_h A_h \tag{4}$$

The set of differential equations (1) and (2) cannot be solved exactly. Now invoking the quasi-equilibrium (QE) approximation [1,2] according to which

$$\left|\frac{dn_c}{dt}\right| << \left|\frac{dn}{dt}\right| \tag{5}$$

$$\left|\frac{dn_c}{dt}\right| << \left|\frac{dn_h}{dt}\right| \tag{6}$$

$$n_c << n \tag{7}$$

one can write

$$n_c = \frac{nse^{-\frac{E}{KT}}}{A_n(N-n) + n_h A_h} \tag{8}$$

$$n_h \approx n \tag{9}$$

$$\frac{dn}{dt} \approx -nse^{-\frac{E}{kT}}\left[1 - \frac{A_n(N-n)}{A_n(N-n) + n_h A_h}\right] \tag{10}$$

So, in the OTOR model, using the QE approximation the expression for TL intensity is given by

$$I \approx -\frac{dn}{dt} = nse^{-\frac{E}{kT}}\left[1 - \frac{A_n(N-n)}{A_n(N-n) + n_h A_h}\right] \tag{11}$$

This expression for I is known as general one trap (GOT) expression for TL emission [13]. For no retrapping, $A_n = 0$. So the expression for I reduces to

$$I \approx -\frac{dn}{dt} = nse^{-\frac{E}{KT}} \tag{12}$$

Equation (12) is the well-known first order kinetics model of Randall and Wilkins [9]. Now for equal probability of recombination and retrapping *i.e.* $A_n = A_h$, equation (11) can be expressed as

$$I \approx -\frac{dn}{dt} = \frac{n^2}{N}se^{-\frac{E}{kT}} \tag{13}$$

Equation (13) is the well-known second order kinetics model of Garlick and Gibson [11]. Based on the equations (12-13), May and Partridge [10] suggested a general order kinetics model of TL given by

$$I \approx -\frac{dn}{dt} = \frac{n^b}{N^{b-1}}se^{-\frac{E}{kT}} \tag{14}$$

Although equation (14) reduces to equation (12) and (13) for b = 1 and 2 respectively, it cannot be obtained from the general one trap equation (11). So the GOK equation may be considered as an empirical equation. Solving equations (12-14), one obtains the expression for TL intensity as a function of temperature (T) for the linear heating profile given by

$$T = T_0 + \beta t \tag{15}$$

where T_0 is the initial temperature and β is the heating rate. The expressions for TL intensity for different orders of kinetics are as follow.

For b = 1

$$I(T) = n_0 sfe^{-\frac{E}{kT}}\exp\left[-\frac{s}{\beta}J\right] \tag{16}$$

For b = 2

$$I(T) = n_0 sf^2 e^{-\frac{E}{kT}}\left[1 + \frac{sf}{\beta}J\right]^{-2} \tag{17}$$

For $b \neq 1$

$$I(T) = n_0 sf^{b-1}e^{-\frac{E}{kT}}\left[1 + \frac{(b-1)sf^{b-1}}{\beta}J\right] \tag{18}$$

Here $J = \int_{T_0}^{T} e^{-\frac{E}{kT'}}dT'$ is the temperature integral and $f = \frac{n_0}{N}$ is the filling ratio. The corresponding maxima

conditions are provided here. The corresponding maxima conditions are given here.

For $b = 1$

$$\frac{\beta E}{kT_m^2} = se^{-\frac{E}{kT_m}} \tag{19}$$

For $b = 2$

$$\frac{\beta E}{kT_m^2} = 2sf^2 e^{-\frac{E}{kT}}\left[1+\frac{sf}{\beta}J\right]^{-1} \tag{18}$$

For $b \neq 1$

$$\frac{\beta E}{kT_m^2} = bsf^{b-1} e^{-\frac{E}{kT}}\left[1+\frac{(b-1)sf^{b-1}}{\beta}J\right]^{-1} \tag{21}$$

Equation (18) reduces to equation (17) for $b = 2$ and in the limit $b \to 1$, we get equation (16).

Now we come over to the IMTS model [12] in which the effect of thermally disconnected deep traps (TDDT) are taken into account. The importance of TDDTs in TL has been discussed by Fain *et al.* [14,15]. The set of coupled differential equations for IMTS model can be written as

$$\frac{dn}{dt} = -nse^{-\frac{E}{kT}} + N_c(N-n)A_n \tag{22}$$

$$\frac{dn_c}{dt} = nse^{-\frac{E}{kT}} - n_c(N-n)A_n - n_c(M-m)A_m - n_c n_h A_h \tag{23}$$

$$\frac{dm}{dt} = n_c(M-m)A_m \tag{24}$$

Here, the charge neutrality condition is given by

$$n_h = n + n_c + m \tag{25}$$

Subsequently, *I(t)* is derived from equation (4). Here *M* and *m* are the concentrations of TDDTs and electrons trapped in TDDTs respectively. A_m is the capture co-efficient for TDDTs.

3.0 RESULTS AND DISCUSSIONS

The temperature integral $J = \int_{T_0}^{T} e^{-\frac{E}{kT'}} dT'$ occurring in equations (16-18) cannot be evaluated analytically. Now, setting the value $T_0 = 0$ and integrating by parts, we can write [16]

$$\int_0^T e^{-\frac{E}{KT'}} \approx Te^{-\frac{E}{KT}} + \frac{E}{k}E_i\left(-\frac{E}{kT}\right) \tag{26}$$

where *Ei(x)* is the exponential integral function with argument *x* [17].

We have computed TL curves of different orders of kinetics *viz.* b = 1, 1.5 and 2 using MATLAB software and the curves are presented in figures (1-3) respectively.

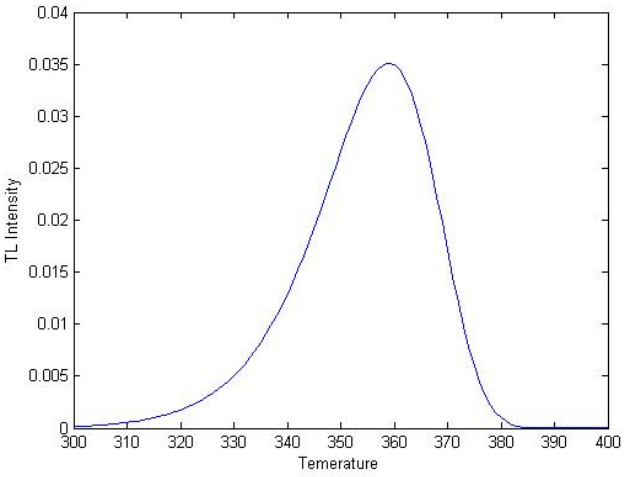

Fig. 1: Glow curve in 1st order kinetics model generated by MATLAB. The input parameters are : b = 1, E = 1 *eV*, $s = 10^{12}$ s^{-1} and $n_0 = N = 10^{12}$/cc.

The shapes of computed curves are in good agreement with those reported by Pagonis et al. [12] which were derived by using a different software `Mathematica'.

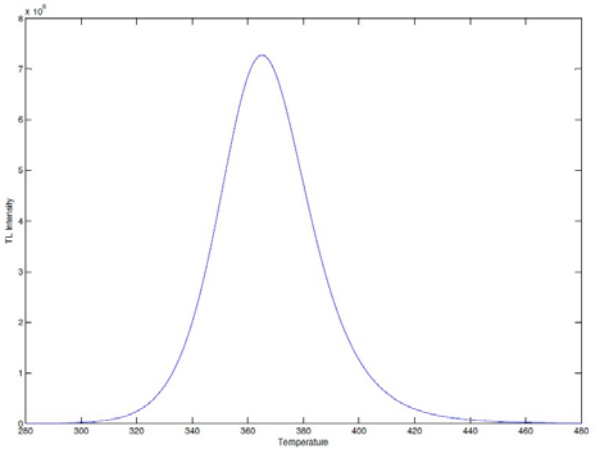

Fig. 2: Glow curve in kinetics order model produced by MATLAB. The input parameters are : b = 1.5, E = 1 *eV*, $s = 10^{12}$ s^{-1} and $n_0 = N = 10^{12}$/cc.

After computing the TL curves in kinetic order formalism, we have used MATLAB software to compute TL curves corresponding to OTOR and IMTS models. At first, we attempt to solve the relevant sets of differential equation by using the 'ode45' solver of MATLAB which is a one-step Runge-Kutta medium order (fourth to fifth) solver. This is to note that the differential equations for different models

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considered here are stiff equations, the 'ode45' solver for such stiff problems. Finally we have used the 'ode15s' solver of MATLAB which is an implicit, multi-step numerical differentiation solver of varying order (first to fifth). This method is suitable for problems that require

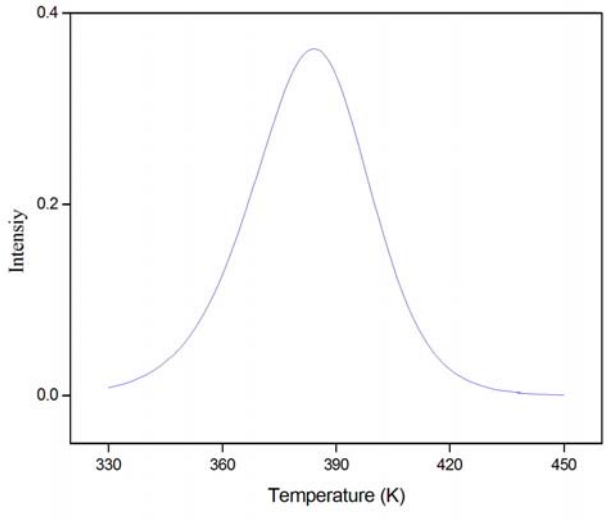

Fig. 3: Glow curve in GOK model produced by MATLAB. The input parameters are : $b = 2$, $E = 1$ eV, $s = 10^{12}$ s^{-1} and $n_0 = N = 10^{12}$/cc.

moderate accuracy. The computed TL curves calculated by using OTOR and IMTS models are presented in figures 4 and 5 respectively.

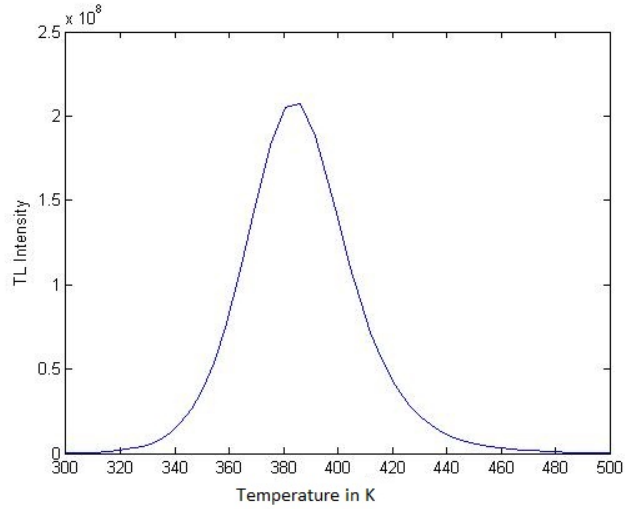

Fig. 4: Glow curve in OTOR model. The input parameters are : $E = 1$ eV $s = 10^{12}$ s^{-1}, $n_0 = N = 10^{12}$/cc, $A_n = 10^{-7}$ cm^3 s^{-1} and $A_h = 10^{-5}$ cm^3 s^{-1}.

Now we come to the deconvolution process of the TL curves in kinetic order model. We plan to deconvolute the experimental TL peaks of colourless calcite [18] irradiated with 4.08 *KGy* of Co^{60} γ - rays under a linear heating rate of 3.03 Ks^{-1} without any filter. The CGCD results produced by using MATLAB are presented in table-1.

Table 1: Fitting parameters of glow curves of colour-less calcite (without filter) corresponding to heating rate 3.03 Ks^{-1}.

Peak No.	T_m (K)	E_{cf} (eV)	b_{cf}	s_{cf} (sec^{-1})	FOM (%)
1	516	1.50	2	8.3×10^{13}	0.61
2	613	1.65	2	5.3×10^{12}	

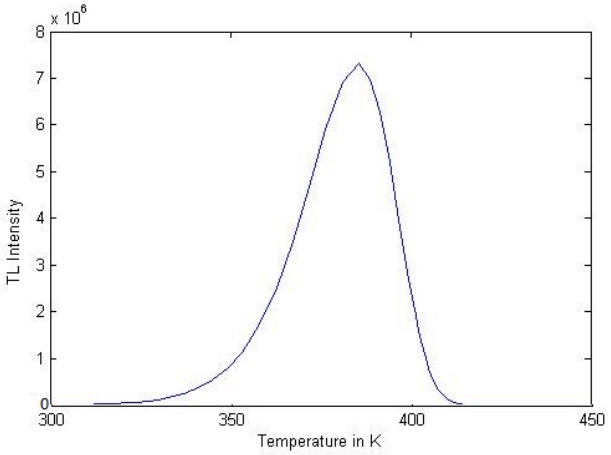

Fig. 5: Glow curve in IMTS model. The input parameters are : $E = 1$ eV $s = 10^{12}$ s^{-1}, $n_0 = N = 10^{12}$/cc, $m_0 = M = 10^{10}$/cc, $A_n = 10^{-7}$ cm^3 s^{-1} and $A_h = A_m = 10^{-5}$ cm^3 s^{-1}.

The deconvoluted glow curves along with the experimental data points are shown in figure-6. It is evident the results that the resultant glow curve is a superposition of two

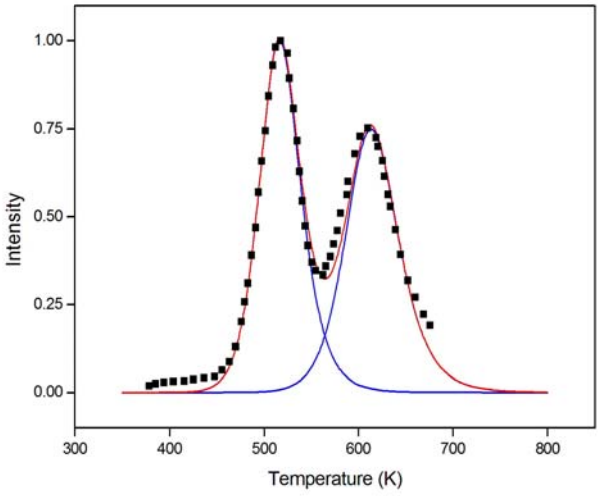

Fig. 6: Deconvoluted glow curve of colourless calcite generated by using MATLAB. The scattered points (black) are the experimental results, the red line is the fitted curve and the blue lines are the deconvoluted peaks.

second-order TL peaks. The figure of merit corresponding to the entire glow curve is about 0.61% indicating reasonably good fit [19,20]. However, we observe that there is a scope for improvement in respect of the second peak.The same glow curve is deconvoluted using a FORTRAN77 algorithm presented by Chen and Kirsh [16] and the fitted parameters E_{cf} , b_{cf} and s_{cf} are in close agreement with the present MATLAB results.

4.0 CONCLUSION

The MATLAB software is quite efficient for analyzing computer generated as well as experimental TL glow curves. The deconvolution parameters obtained by using MATLAB software agree fairly well with the results derived from other conventional codes used for this purpose. Further deconvolution of complex TL glow curves using more sophisticated models such as OTOR and IMTS are in the pipeline.

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