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On the mixed order kinetics of the thermoluminescence glow peak

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Abstract. The shape factor of a thermoluminescence (TL) glow peak following mixed order (MO) kinetics has been reinvestigated. A modification of the existing equation of MO kinetics to include the filling factor f ($f = n_0/N$, where n_0 is the initial concentration of trapped electrons and N is the concentration of the electron trap sites) has been proposed. The shifting of peak temperature for a MO kinetics peak with the change of filling factor and hence the irradiation dose absorbed by the sample is reported. A new set of expressions for the evaluation of the activation energy of a TL peak in the light of MO kinetics is presented. The applicability of the MO kinetics model is reported by fitting both the numerically simulated glow peak and the experimentally observed peak of γ-irradiated NaCl:I and x-irradiated BeO.

1. Introduction

Thermoluminescence (TL) appears as a very complex phenomenon. The proposition of a general model accounting for TL can appear as unrealistic because of the wide variety of phenomena occurring in this field. However, simple models [1–3] are generally used to explain some of the salient features of thermoluminescence (TL) glow peaks. The first order kinetics model [1] assumes zero retrapping probability and the shape factor μ_g ($\mu_g = \delta/\omega = (T_2 - T_m)/(T_2 - T_1)$, where T_m is the peak temperature, T_1 and T_2 are the temperatures at half intensity on the rising and falling side of TL respectively) is nearly 0.42 and is independent of the initial concentration of trapped electrons n_0. On the other hand, when there is strong retrapping $A_m m \ll A_n(N - n)$ (A_m and A_n are respectively the probabilities of recombination and retrapping; m, n and N are respectively the concentration of recombination centres, electrons in electron traps and total number of traps) and if $n \ll N$ (low irradiation dose) one obtains the so-called second order kinetics model [2]. The shape factor in this case is nearly 0.52. Both the models [1, 2] are shown to be particular cases of the set of three simultaneous differential equations [4].

In several cases the shape of the glow peak does not follow either the first or the second order glow curves. In order to study a general order (GO) TL peak whose order of kinetics is not necessarily 1 or 2, Chen [3] derived an empirical relation in the form

$$I(t) = -\,\mathrm{d}n/\mathrm{d}t = s' n^b \exp(-E/kT) \tag{1}$$

where b is the order of kinetics and s' is the pre-exponential factor. Gartia *et al* [5] and Rasheedy [6] could independently derive the modified empirical relation of the general order (GO) kinetics in the form

$$I(t) = (n^b/N^{b-1}) s \exp(-E/kT) \tag{2}$$

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which can take into account the filling factor f ($f = n_0/N$). Gartia *et al* [5] could give the theoretical justification of the shifting of peak temperature to the low temperature side with the increase in filling factor and hence to the irradiation dose for those peaks obeying non-first order kinetics ($b \neq 1$).

In spite of the extensive use and application of GO kinetics by a number of research workers, it lacks a physical basis and so a more physical mixed order (MO) kinetics model has been developed by Chen *et al* [7] from the set of three differential equations [4]. Chen *et al* [7] have shown the variation of μ_g with α ($\alpha = n_0/(n_0+c)$, where c is the concentration of trapped electrons or holes not taking part in the TL process in the temperature range considered due to their being in deep traps or in low probability recombination centres). The shape of an MO TL peak depends strongly on α. The value of α of an experimental TL peak can be estimated by knowing the value of μ_g from the variation of μ_g with α presented in this paper. It is important to note here that we can not draw any physically meaningful information from the value of b ($1 < b < 2$) but by knowing the value of α of an experimental TL peak one can have the knowledge of the physically meaningful parameter c. This is one clear advantage of MO kinetics over GO kinetics in the intermediate range $1 < b < 2$ in addition to the undenied fact that the MO kinetics model results directly from the set of three differential equations governing the process by using some mild simplifying assumptions [7]. Recently Yossian and Horowitz [8] have fitted the isolated peak V in LiF:Mg,Ti (TLD-100) following post-irradiation annealing at 165 °C using MO kinetics. Keeping in mind the continuing interest [8] we have reinvestigated in this paper the characteristics of TL peaks following MO kinetics and the method for determination of E of TL peaks. We have introduced in our expression for intensity the filling factor f with which one can study the dosimetric and dating aspects using TL since f is directly related to dose. The prominent shifting of peak temperature (T_m) with the change of α is discussed. We have also shown that the nature of the shifting of T_m with the change of f agrees with that obtained by Gartia *et al* [5]. In this paper we have also presented a new set of expressions involving the MO parameter α to determine the value of activation energy E which agrees well with that obtained by using Chen's formula [3]. An attempt is also made to fit the simulated GO TL peaks and the experimental TL curves of NaCl:I irradiated by γ-rays (2.04 kGy) and thermally cleaned up to 169 and 182 °C and recorded at a constant heating rate of 1.5 °C s^{1} and the TL curve of BeO ($T_m = 160.1$ °C) irradiated with x-rays (5 minutes) [9] with MO kinetics.

2. Theory

The intensity of a TL peak obeying MO kinetics can be written as [7]

$$I(t) = -\,\mathrm{d}n/\,\mathrm{d}t = s_1' n(n+c)\exp(-E/kT) \tag{3}$$

with

$$s_1' = sA_m/NA_n. \tag{4}$$

Solving equation (3) under the linear heating profile $T = T_0 + \beta t$, β being the heating rate, one obtains

$$n = n_0(1-\alpha)\Big/\left[\exp\left\{(cs_1'/\beta)\int_{T_0}^{T}\exp(-E/kT')\,\mathrm{d}T'\right\} - \alpha\right] \tag{5}$$

where T_0 is the temperature at time $t = 0$. Differentiating equation (5) w.r.t. T, the expression for TL intensity becomes

$$I(T) = \frac{Nf^2(1-\alpha)s'' \exp(-E/kT) \exp\{(fs''/\beta)\int_{T_0}^{T} \exp(-E/kT')\,\mathrm{d}T'\}}{[\exp\{(fs''/\beta)\int_{T_0}^{T} \exp(-E/kT')\,\mathrm{d}T'\} - \alpha]^2} \tag{6}$$

where

$$s'' = (1-\alpha)sA_m/\alpha A_n. \tag{7}$$

Equation (6) considers both the cases of partially filled ($f < 1$) and completely filled ($f = 1$) traps. The condition for maximum intensity I_m is given by

$$(E/kT_m^2) + (fs''/\beta)\exp(-E/kT_m)\left[\exp\left\{(fs''/\beta)\int_{T_0}^{T_m} \exp(-E/kT')\,\mathrm{d}T'\right\} - \alpha\right]$$
$$= 2(fs''/\beta)\exp(-E/kT_m)\exp\left\{(fs''/\beta)\int_{T_0}^{T_m} \exp(-E/kT')\,\mathrm{d}T'\right\}. \tag{8}$$

Defining dimensionless quantities $u = E/kT$, $u_0 = E/kT_0$ and $u_m = E/kT_m$ and expressing the temperature integral in terms of a second exponential integral [5] one can write equations (6) and (8) as

$$I(T) = \frac{Nf^2(1-\alpha)s'' \exp(-u) \exp[(fs''E/k\beta)\{E_2(u)/u - E_2(u_0)/u_0\}]}{[\exp[(fs''E/k\beta)\{E_2(u)/u - E_2(u_0)/u_0\}] - \alpha]^2} \tag{9}$$

and

$$\{(ku_m^2/E)\} + (fs''/\beta)\exp(-u_m)[\exp[(fs''E/k\beta)\{E_2(u_m)/u_m - E_2(u_0)/u_0\}] - \alpha]$$
$$= 2(fs''/\beta)\exp(-u_m)\exp[(fs''E/k\beta)\{E_2(u_m)/u_m - E_2(u_0)/u_0\}]. \tag{10}$$

Now the fractional intensity x ($x = I/I_m$) can be expressed as

$$I(T) = \frac{B\exp(u_m - u)\exp[(fs''E/k\beta)\{E_2(u)/u - E_2(u_0)/u_0\}]}{[\exp[(fs''E/k\beta)\{E_2(u)/u - E_2(u_0)/u_0\}] - \alpha]^2} \tag{11}$$

with

$$B = 4(fs''/\beta)^2\alpha[(k/E)^2u_m^4\exp(2u_m) - (fs''/\beta)^2]^{-1}. \tag{12}$$

Using the Newton–Raphson method [10] we have found T_m from equation (8) and half intensity points T_1 and T_2 from equations (11) and (12).

Plots of the variables $u_1/(u_1 - u_m)$, $u_2/(u_m - u_1)$ and $u_1u_2/u_m(u_1 - u_2)$, where $u_1 = E/(kT_1)$ and $u_2 = E/(kT_2)$ against u_m ($10 \leqslant u \leqslant 40$) for a particular value of α are found to be linear so that one can write

$$u_m = C_1u_1/(u_1 - u_m) + D_1 \tag{13}$$
$$u_m = C_2u_2/(u_2 - u_m) + D_2 \tag{14}$$
$$u_m = C_3u_1u_2/u_m(u_1 - u_2) + D_3. \tag{15}$$

Equations (13)–(15) can be recast in terms of activation energy and temperature as

$$E_\tau = C_1kT_m^2/\tau + D_1kT_m \tag{16}$$
$$E_\delta = C_2kT_m^2/\delta + D_2kT_m \tag{17}$$
$$E_\omega = C_3kT_m^2/\omega + D_3kT_m \tag{18}$$

where $\tau = T_m - T_1$.

The coefficients C_j and D_j ($j = 1$–3) occurring in equations (13)–(18) depend on α. By using the method of non-linear least-squares regression [11], each of the coefficients C_j and D_j can be expressed as a quadratic function of α ($0 \leqslant \alpha \leqslant 1$) as

$$C_j = C_{j0} + C_{j1}\alpha + C_{j2}\alpha^2 \tag{19}$$

$$D_j = D_{j0} + D_{j1}\alpha + D_{j2}\alpha^2. \tag{20}$$

The coefficients C_{jk} and D_{jk} ($j = 1$–3, $k = 0$–2) occurring in equations (19) and (20) are presented in table 1.

Table 1. Coefficients C_{jk} and D_{jk} ($j = 1$–3, $k = 0$–2) occurring in equations (19) and (20).

j	C_{j0}	C_{j1}	C_{j2}	D_{j0}	D_{j1}	D_{j2}
1	1.4411	0.5256	−0.1947	−2.0763	1.1869	0.2053
2	0.9861	0.3234	0.5607	−0.3134	1.7857	−2.3585
3	2.4246	0.8274	0.3824	−1.3170	0.4891	−0.9844

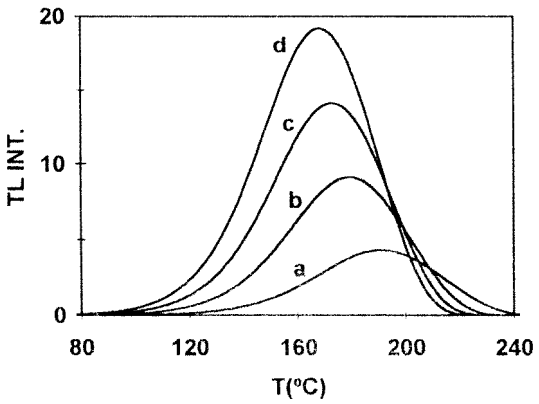


Figure 1. Computed MO TL glow curves ($E = 1.0$ eV, $s = 10^{10}$ s^{-1}, $\beta = 10\,^{\circ}$C s^{-1}, $\alpha = 0.6$, $N = 10^{10}$ cm^{-3}). Curves (a) to (d) are for different values of $f = 0.25$, 0.50, 0.75 and 1.0 respectively.

Equations (16)–(18) can be used to determine the activation energy of a TL glow curve.

3. Results and discussion

Figure 1 shows the numerically simulated MO TL peaks for the same values of trapping parameters, namely $E = 1.0$ eV, $s = 10^{10}$ s^{-1}, $\alpha = 0.6$, $\beta = 1.0\,^{\circ}$C s^{-1} and for different values of filling factor $f = 0.25$, 0.50, 0.75 and 1.0 respectively. It is clear from this figure that with the increase in f the peak intensity increases and the peak temperature decreases. This behaviour is similar to that of the non-first-order ($b \neq 1$) peaks under GO kinetics model. The shifting of peak temperature towards the low temperature side with increase of f for different values of α ($\alpha = 0.2$, 0.4, 0.6, 0.99) is shown in figure 2.

In GO kinetics the shape factor of a TL glow peak strongly depends on the order of kinetics b [3]. In order to show a strong dependence of the shape factor μ_g on α we have numerically computed normalized MO TL glow peaks with the same values of $E = 1.0$ eV,

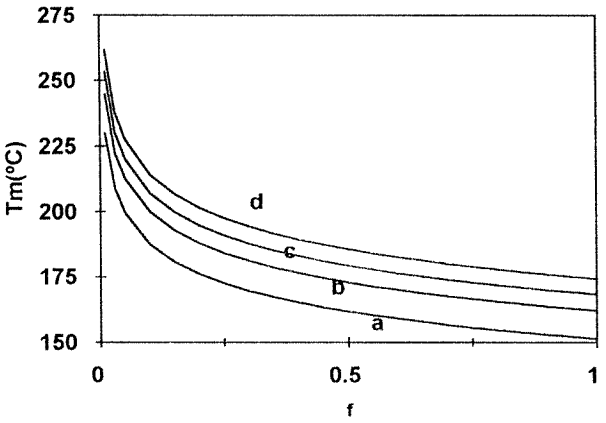


Figure 2. Variation of peak temperatures T_m (°C) with filling factor f ($E = 1.0$ eV, $s = 10^{10}$ s^{-1}, $\beta = 1.0\,^{\circ}$C s^{1}, $N = 10$ cm^{-3}) for different values of α. Curves (a) to (d) are for $\alpha = 0.2$, $\alpha = 0.4$, $\alpha = 0.6$ and $\alpha = 0.99$ respectively.

$f = 1.0$, $N = 10^{10}$ cm^{-3}, $\beta = 1.0\,^{\circ}$C s^{-1} and different values of $\alpha = 0.1$, 0.5, 0.7, 0.99 so as to merge peak temperatures at 130 °C (figure 3). The frequency factors used in the above computations are 2.34×10^{10} s^{-1}, 1.22×10^{11} s^{-1}, 1.68×10^{11} s^{-1} and 2.14×10^{11} s^{-1} respectively. From figure 3 it is clear that the full width ω increases with the increase in α. The rising portion of the curves for different values of α appears to merge although there is a little increase in the values of τ when α increases. On the other hand there is a well defined separation along the falling side of the curves when α increases. The increase in δ is faster than that of τ with the increase in α resulting in an increase in the value of μ_g which is in agreement with the results obtained by Chen *et al* [7]. The values of shape factors at half intensity points in figure 3 are respectively 0.424, 0.458, 0.487, 0.519. It is worthwhile to mention here that although the shape factor μ_g has been calculated by choosing $f = 1$ we have verified that the variation of μ_g with the filling factor f ($0.1 \leqslant f \leqslant 1.0$) for a particular value of α is only in the third decimal place and hence is quite insignificant in the actual measurement.

Table 2. Calculated values of T_m, τ, ω, μ_g and E_τ (eV) of MO TL peaks generated with $E = 1.0$ eV, $s = 10^{10}$ s^{-1}, $N = 10^{10}$ cm^{-3}, $f = 1$ and $\beta = 1.0\,^{\circ}$C s^{-1} for different values of α.

					E_τ (eV)	
α	T_m (°C)	τ (°C)	ω (°C)	μ_g	Present	Chen
0.1	141.4272	20.5253	35.6503	0.4243	0.9883	0.9839
0.2	151.4806	21.9988	38.6441	0.4307	0.9886	0.9696
0.3	157.6403	23.1333	41.2074	0.4386	0.9898	0.9586
0.4	162.1039	24.1066	43.6937	0.4483	0.9916	0.9519
0.5	165.5575	24.9572	46.2173	0.4600	0.9938	0.9503
0.6	168.2938	25.6837	48.8105	0.4738	0.9959	0.9547
0.7	170.4603	26.2717	51.4297	0.4892	0.9978	0.9649
0.8	172.1469	26.7089	53.9005	0.5045	0.9991	0.9790
0.9	173.4261	26.9952	55.8208	0.5164	1.0000	0.9919

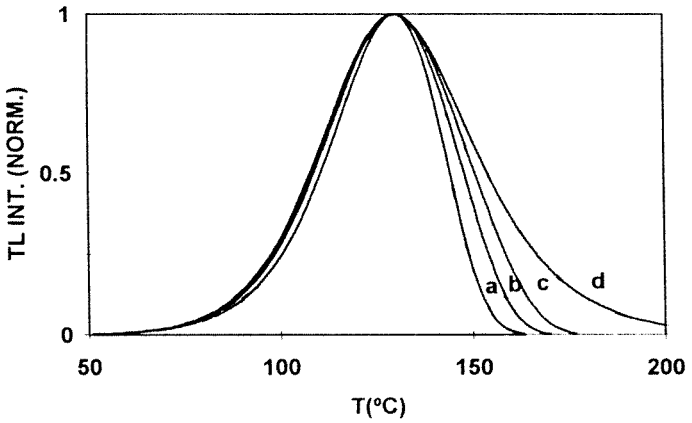


Figure 3. Computed normalised MO TL curves with the same peak temperature $T_m = 130\,^{\circ}\mathrm{C}$ ($E = 1.0$ eV, $f = 1.0$, $N = 10^{10}$ cm^{-3} and $\beta = 1.0\,^{\circ}\mathrm{C\ s}^{-1}$) showing the variation of peak shape for different values of α. Curves (a) to (d) correspond to $\alpha = 0.1$, 0.5, 0.7 and 0.99 with different frequency factors.

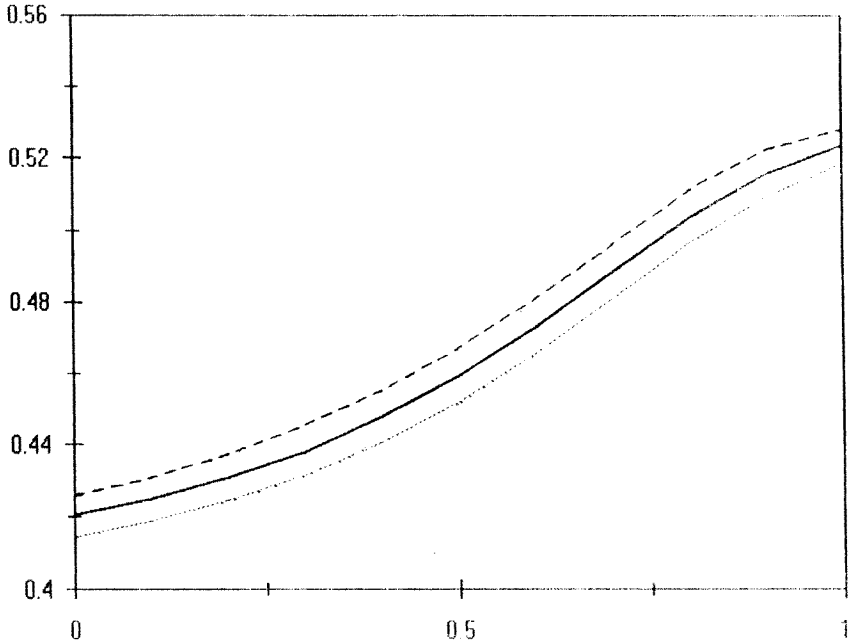


Figure 4. Variation of μ_g as a function of α. – – – for $u_m = 20$, ····· for $u_m = 40$ and —— for an average of the values of μ_g at $u_m = 20$ and $u_m = 40$.

Table 2 shows the calculated values of T_m, τ, ω and μ_g with various values of α from 0.1 to 0.9 for the values of trapping parameters used in the computation of glow peaks in figure 1 but using the value of $f = 1$. The shifting of T_m with the change in α in the MO kinetics model is enormously large ($\approx$32 °C in table 2) compared to the change of only around 3 °C in the case of GO kinetics model when the order of kinetics b is varied from

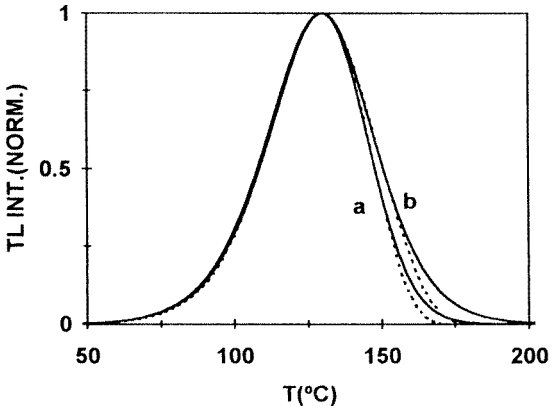
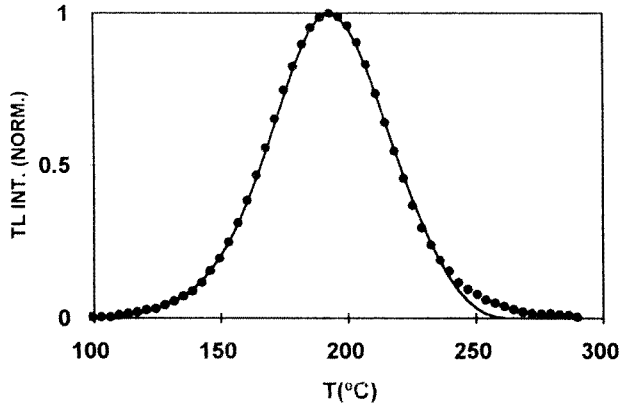


Figure 5. Curve fitting of MO peaks with GO peaks. The dotted curves (a) and (b) are MO TL peaks with $\alpha = 0.5$ and 0.7 respectively corresponding to curves (b) and (c) of figure 3. The continuous curves are the best fitted GO TL peaks. The fitted trapping parameters are $E = 0.936$ eV, $b = 1.27$, $s = 3.30 \times 10^{10}$ s^{-1} for curve (a) and $E = 0.951$ eV, $b = 1.56$, $s = 5.16 \times 10^{11}$ s^{-1} for curve (b) respectively.

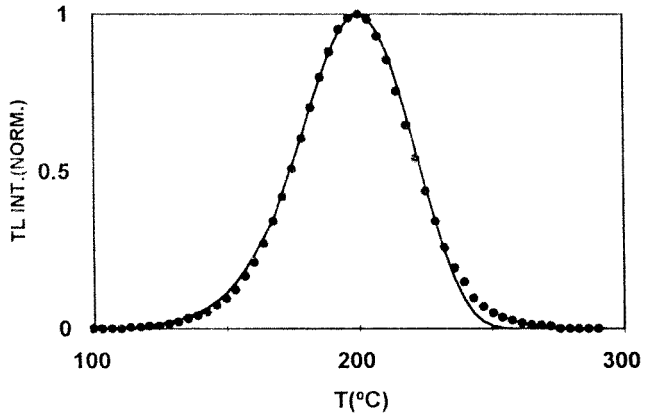
0.7 to 2.5 for a wide range of activation energies (0.1 to 1.6 eV) and frequency factors (10^5 s^{-1} to 10^{13} s^{-1}).

In order to determine E_j ($j = \tau, \delta, \omega$) using the set of expressions (16)–(18) we require the value of the MO parameter α of a TL peak which can be estimated from the μ_g–α curve [7]. Chen *et al* [7] observed that the curve is modified slightly with change in E and s'', i.e. with u_m since T_m can be found if s'' is known for a particular value of E. Since we do not know the value of u_m of an experimental TL peak, the average values of μ_g for $u_m = 20$ and $u_m = 40$ are obtained for different values of α and are plotted as a function of α (solid line in figure 4). Variations of μ_g with α for $u_m = 20$ and $u_m = 40$ are also shown in figure 4. The average μ_g–α curve can now be used as a preliminary estimation of α to find E with a maximum error of 3%. It is to be noted that for many experimental TL peaks u_m lies between 20 and 40 except for a limited number of peaks. In order to check the suitability of using equations (16)–(18) we have calculated E_j ($j = \tau, \delta, \omega$) of synthetic peaks for different values of α and these are found to be in good agreement with the input value and that obtained by using Chen's GO formula [3]. The calculated values of E_τ using the present set of expressions and Chen's formula [3] for different values of α are reported in table 2.

Since both GO kinetics and MO kinetics models can have shape factors lying between 0.42 and 0.52 one may consider a GO kinetics model with a particular value of b to arrive at the TL glow curve following the MO kinetics model. In order to check this we have used the curve fitting technique [12]. For this purpose we have considered the numerically generated TL peak under MO kinetics as a synthetic peak and it has been fitted by GO kinetics. The whole of both the curves (a) and (d) in figure 3 can be nicely fitted with GO kinetics. The trapping parameters which can fit curve (a) are $E = 0.983$ eV, $b = 1.031$ and $s = 1.38 \times 10^{12}$ s^{-1}. Similarly the best fitted values of the curve (d) are $E = 1.001$ eV, $b = 2$ and $s = 2.23 \times 10^{11}$ s^{-1}. The fitted values of b are in fair agreement with that of Chen *et al* [7] who have reported that for $\alpha = 0$ ($n_0 \ll c$) MO kinetics tends to first-order kinetics and for $\alpha = 1$ ($n_0 \gg c$) to second-order kinetics. In order to check the viability of using MO kinetics as an alternative to GO kinetics in the intermediate range of b ($1 < b < 2$)



(a)



(b)

Figure 6. (a) Curve fitting of TL peaks of NaCl:I ($\beta = 1.5\,^{\circ}$C s^{-1}) irradiated with γ-rays (2.04 kGy) and recorded after thermal cleaning up to $T_c = 169\,^{\circ}$C. The symbol ● denotes the experimental peak and the continuous curve denotes the fitted curve using MO model. The best fitted trapping parameters are $E = 1.091$ eV, $f = 1.0$, $s = 4.54 \times 10^{10}$ s^{-1} and $\alpha = 0.82$. (b) Same as figure 6(a) but with $T_c = 182\,^{\circ}$C, $E = 1.147$ eV, $f = 1.0$, $s = 9.44 \times 10^{10}$ s^{-1} and $\alpha = 0.63$.

we have made an attempt to fit the whole of the curves (b) and (c) in figure 3 with GO kinetics (figure 5). It is clear from figure 5 that almost all portions of the peaks except the wings on the falling side can be fitted nicely. The best fitted values of E are 0.936 and 0.951 eV with order of kinetics 1.27 and 1.56. It is to be noted that I_m which is assumed to be an important point of a TL glow curve has been nicely fitted except for a little misfit at the wings on the falling side compared to the fitting reported by Yossian and Horowitz [8] who have fitted nicely both the wings of a synthetic MO glow peak ($c/n_0 = 2$) with GO kinetics ($b = 1.15$) at the cost of fitting around I_m.

The applicability of the MO kinetics can be studied by fitting the experimental TL glow peak with mixed order kinetics. For this purpose we have recorded TL of NaCl:I irradiated with γ-rays (2.04 kGy) and x-irradiated (5 minutes) BeO. In order to observe an isolated peak we have thermally cleaned the irradiated sample up to 169 and 182 °C and recorded

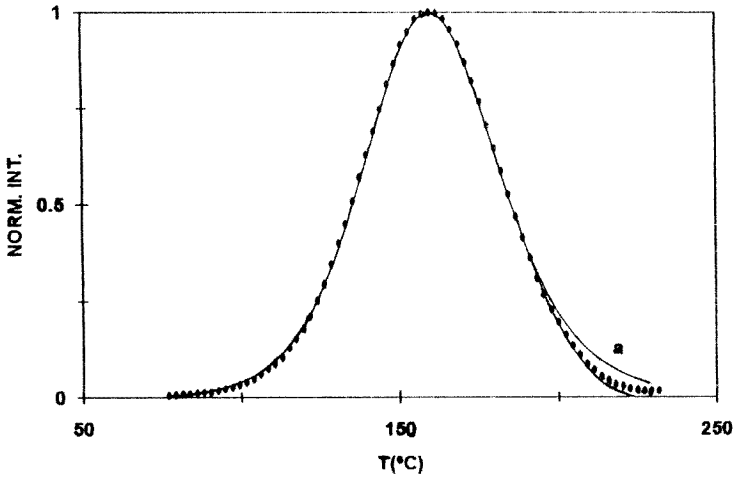


Figure 7. Curve fitting of experimental TL peak (full circles) of BeO ($T_m = 160.1\,^{\circ}$C) with MO kinetics (continuous line) ($E = 1.041$ eV, $s = 6.67 \times 10^{10}$ s^{-1}, $A_m = A_n = 10^{-7}$ s^{-1}, $N = n_0 = 10^{10}$ cm^{-3} and $\alpha = 0.85$) and GO kinetics (continuous line marked with a) ($E = 1.02$ eV, $b = 1.8$ and $s = 4.07 \times 10^{10}$ s^{-1}).

TL glow curve with a linear heating rate $\beta = 1.5\,^{\circ}$C s^{-1}. The peak temperatures of the glow curves are 192.7 and 200 °C with shape factors 0.48 and 0.46. Similar values of μ_g are reported by Mahajan *et al* [13] for the 203 °C peak of NaCl:Ba(T) recorded after γ-ray (8 Gy) irradiation followed by thermal annealing at 150 °C for 30 minutes. Using the curve fitting technique [12] the experimental curve with $T_m = 192.7\,^{\circ}$C can be best fitted to a mixed order kinetics peak with activation energy $E = 1.09$ eV, $\alpha = 0.82$ and $s = 4.54 \times 10^{10}$ s^{-1}, $A_m = A_n = 10^{-7}$ s^{-1}, $f = 1$ and $N = 10^{10}$ cm^{-3} (figure 6(a)). Similarly the other peak ($T_m = 200\,^{\circ}$C) can be best fitted with $E = 1.145$ eV, $\alpha = 0.63$ and $s = 9.44 \times 10^{10}$ s^{-1}, $A_m = A_n = 10^{-7}$ s^{-1}, $f = 1$ and $N = 10^{10}$ cm^{-3} (figure 6(b)). Figure 6(b) has already been fitted with GO kinetics [14]. In spite of the better GO fitting at the wings of the falling side of figure 6 the resulting parameter b has no physical meaning compared to α in the case of MO fitting. The present finding of the activation energies is in agreement with the activation energy E_{IR} obtained by using the initial rise method [13]. Finally we have shown in figure 7 the fitting of the well studied experimental TL peak of BeO ($T_m = 160.1\,^{\circ}$C) [14] irradiated with x-rays with both GO and MO kinetics. The fitted trapping parameters in the case of GO kinetics are $E = 1.02$ eV, $b = 1.8$ and $s = 4.07 \times 10^{10}$ s^{-1} and in the case of MO kinetics $E = 1.041$ eV, $\alpha = 0.85$, $s = 6.7 \times 10^{10}$ s^{-1}, $A_m = A_n = 10^{-7}$ s^{-1}, $f = 1$, $N = 10^{10}$ cm^{-3}. A close inspection of figure 7 reveals that the MO fitting of the experimental TL peak of BeO (figure 7) is better than the GO kinetics fitting.

4. Conclusion

In the present paper we have studied in detail the characteristics of TL peaks following MO kinetics. The MO kinetics is a physically realistic model in contrast to the empirical nature of the GO kinetics equation for TL process. The shifting of peak temperature towards the low temperature side with the increase of filling factor and hence the irradiation dose is one of the important characteristics of a TL peak following MO kinetics, similar to GO kinetics. Unlike the general order kinetics model where there is a maximum shifting of T_m

by about 3 °C when the order is allowed to vary from 0.7 to 2.5 for the same activation energy and frequency factor, one obtains in MO kinetics a large shift of T_m of about 32 °C (table 2) when α is varied from 0.1 to 0.99. We have also presented a new set of expressions for determining the activation energy E. Lastly the TL peaks of γ-irradiated NaCl:I and x-irradiated BeO can be explained in terms of the MO kinetics model.

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