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Effect of self absorption on thermoluminescence (TL) glow curves: Theoretical studies

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Abstract
The influence of self absorption on thermoluminescence (TL) glow curves have been investigated theoretically for wide range of parameters. The model based on charge carrier transport via conduction band has been assumed in the present study. It has been found that re-absorption causes shift in glow curve position/peak temperature, integrated TL intensity and order of kinetics. Further it is speculated that re-absorption may also be one of the culprit behind the non-reproducibility of TL glow curves in a dosimetry programme based on TL phenomena. Further the effect of re-absorption on activation energy measured using peak shape methods is investigated and it has been found that huge errors in estimation of activation energy may be present.

Keywords: Thermoluminescence, Re-absorption/self absorption, Glow curves and Order of kinetics.

1.0 INTRODUCTION

The emission of light from a thermoluminescencent phosphor is influenced by large number of parameters. Some of the parameters arise from thermoluminescence (TL) instrumentation whereas others may arise from the TL detector [1-6]. One of the parameters originating from TL detector, which generally influences the shape of TL glow curve is re-absorption of the emitted signal. The re-absorption causes the TL glow curve to differ from actual shape which would have obtained without re-absorption. The presence of re-absorption may also cause statistical fluctuations in the resultant signal [4, 6].

As is well known that the electrons can be released from traps optically as well as thermally. Thus the trapped electrons can absorb photons and be promoted to conduction band and this is manifested in optically stimulated luminescence (OSL), phototransferred TL (PTTL) and optical bleaching[4]. Further, if light can release the trapped charge, one must consider the possibility that re-absorption of TL may occur during the readout process. This in turn will give rise to the optical release of the trapped electrons in addition to the thermal component. Generally re-absorption arises when the emitted light can be absorbed either by the dopant or impurities or various defect sites present in the TL crystal and this may be a dominant effect when the wavelength of emitted TL signal lies in the absorption bands of these impurities/defects or dopants. Further in the presence of re-absorption, there can be errors in the estimation of kinetic parameters such as activation energy, frequency factor, order of kinetics as well as in defect characterization carried out using TL technique. In literature, limited information is available about re-absorption and its influence on glow curves and on estimation of various kinetic parameters from TL glow curves. In this paper, an attempt has been made to comprehensively investigate the effect of re-absorption on shape of glow curves and kinetic parameters.

2. The Model

The rate equations describing the transport of electrons via delocalized/ conduction band as described in Halperin and Braner's model assuming single electron trap and single hole centre are [4, 7]

$$\frac{dm}{dt} = -A_m m n_c \quad (1.1)$$

$$\frac{dn}{dt} = -ns\exp(-E/kT) + A_n(N-n)n_c \quad (1.2)$$

$$\frac{dn_c}{dt} = \frac{dm}{dt} - \frac{dn}{dt} \quad (1.3)$$

where N is the total number of traps (m^{-3}), m is the concentration of holes in recombination centres (m^{-3}); n is the concentration of electrons in traps (m^{-3}) and n_c is the concentration of electrons in the conduction band (m^{-3}). A_m, the recombination probability (m^3s^{-1})

and is equal to $\sigma_m v$ where σ_m is recombination cross-section (m^2) and v is the velocity of the charge carrier (ms^{-1}); A_n, the retrapping probability (m^3s^{-1}) and is equal to $\sigma_n v$ where σ_n is retrapping cross-section (m^2) s, the frequency factor (s^{-1}); E, the activation energy (eV); k, the Boltzmann's constant (eVK^{-1}); T, the absolute temperature (K). Recombination of free electrons with trapped holes gives rise to emission of wavelength, λ described by TL intensity $A_m\ m\ n_c$. However by considering the fact that the trapped electrons can absorb at this wavelength by undergoing a transition to conduction band, the above equations are modified as [4, 6]

$$\frac{dm}{dt} = -A_m m n_c \qquad (1.4)$$

$$\frac{dn}{dt} = -ns\exp(-E/kT) + A_n(N-n)n_c - (1-\exp(-\alpha n))A_m m n_c$$
(1.5)

$$\frac{dn_c}{dt} = \frac{dm}{dt} - \frac{dn}{dt} \qquad (1.6)$$

where α is the absorption probability of photons by trapped electrons (m^3) and the meaning of other symbols are same as described above. In this case, the measured TL intensity is given by that proportion of photons generated during recombination which are not reabsorbed by the trapped electrons and is

$$I_{TL} = A_m\, m\, n_c - (1-\exp(-\alpha n))\, A_m m n_c$$
$$= A_m\, m\, n_c \exp(-\alpha n) \qquad (1.7)$$

From Equation (1.7), it is seen that the normally used expression for TL intensity is modified by the term *exp* ($-\alpha n$). For α = 0, the set of Equations (1.4-1.6) reduces to the set of Equations (1.1-1.3).

3. Numerical Study

The set of Equations (1.1-1.3) and (1.4-1.6) are solved numerically for wide range of parameters. The parameters assumed theoretically unless otherwise mentioned are E = 0.90 eV, s = 10^{10} Hz, A_n = 10^{-18} m^3s^{-1}, A_m =10^{-16} m^3s^{-1}, heating rate (β) = 1 K s^{-1}, T_0 = 300 K and N = 10^{23} m^{-3}. The studies are carried out for various values of α viz. 10^{-18} m^3, 10^{-19} m^3, 10^{-20} m^3, 10^{-21} m^3 and 10^{-22} m^3 for fixed dose. Further studies are also carried out for α = 0. In addition, dose dependence of TL glow curves is checked for different values of α. In this study, the variation in dose (number of electrons and holes (m^{-3}) produced by the ionizing radiation) is assumed from m_0 = n_0 = 10^{18} m^{-3} – 10^{22} m^{-3} and n_{c0} = 0. The simulation time is chosen in such a way that complete glow curve is acquired. The effect of reabsorption on TL glow curves is manifested with the variation of glow peak temperature T_m with α and the order of kinetics as reflected by the values of the shape factor μ_g (The shape/geometrical factor μ_g is defined by the ratio of [δ/ω = $(T_2\text{-}T_m)/(T_2\text{-}T_1)$] where T_m is the temperature corresponding to maxima of the glow peak (I_m) and T_1 and T_2 are the value of the temperature for the rising and falling portions of the glow curve at I_m /2. In fact (T_2-T_1) represents full width at the half maximum (FWHM). It is worth mentioning that the order of kinetics determined using the values of μ_g is one if the value of the shape/geometrical factor μ_g lies in the range 0.38-0.42, whereas for second order kinetics, the value of shape/geometrical factor μ_g lies in the range 0.48-0.52. For the shape/geometrical factor μ_g values in the range 0.42-0.48, the order of kinetics is between one and two).

4. Results and Discussion

The TL glow curves obtained from the solution of the set of Equations (1.1-1.3) and (1.4-1.6) are shown in Figure 1 for different values of absorption coefficient, α. The order of kinetics reflected from the value of shape/geometrical factor μ_g depends on the value of α and the variation of μ_g with the absorption coefficient, α is given in Table-1. For m_0 = n_0 =10^{22} m^{-3}, the value of shape factor, μ_g changes from 0.44 for α = 0 (no absorption of the emitted signal) to 0.46 for α = 10^{-22} m^3. It has been found that the increase in the self absorption factor causes the glow curve to shift towards higher temperatures and the glow peak height/maximum value of TL intensity also decreases. In addition, the order of kinetics measured from the value of shape factor μ_g also changes.

5. Conclusions

In this study it has been demonstrated theoretically that self absorption of the emitted light during heating stage by the TL detector influences the shape of the glow curve. Further shift in the maximum value of the peak temperature, integrated TL intensity and order of kinetics may also be present. In addition to above, the influence of variation of dose for glow curves with and without self absorption is studied and the values of the shape/geometrical factor, μ_g may differ for glow curves simulated by assuming the presence of self absorption even for same dose. Further for high value of α, the simulated glow curves do not differ much from each other for high dose values and resultant glow curves exhibit stable T_m and μ_g values. Hence the self absorption of the emitted signal influences various kinetic parameters of the glow curve.

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References:

1. A. Ege, Y. Wang and P. D. Townsend, Systematic errors in thermoluminescence, **Nuclear Instruments and Methods in Physics Research A** 576, 411-416 (2007).
2. Munish Kumar, L. C. Prasad and R. K. Kher, What ails Thermoluminescence (TL) Dosimetry? **J. Med. Phys.**, 30 (2005) 303.
3. Munish Kuamr, L. C. Prasad and R. K. Kher, Parameters influencing the performance of Thermoluminescence Dosimetry System, **Proc. Annual Conference of Indian Nuclear Society (INSAC-2005)**, Mumbai, 2005.
4. Chen R. and McKeever S. W. S., ***Theory of Thermoluminescence and Related Phenomena***, (Singapore: World Scientific) (1997).
5. Munish Kumar, G. Chourasiya, B.C. Bhatt & C.M. Sunta, Dependence of peak height of glow curves on heating rate in thermoluminescence, **Journal of Luminescence**, 130 (7), 1216-1220 (2010).
6. Munish Kumar, K. Gomati and G. Chourasiya, Investigation of effect of self absorption on thermoluminescence glow curves: theoretical studies, **Proc National Conference on Luminescence and its Applications** (organized by Luminescence Society of India (LSI)), 97, 2010.
7. Munish Kumar, G. Chourasiya, R. K. Kher, B. C. Bhatt and C. M. Sunta, Theoretical investigations of localized, semi-localized and unified models of thermally stimulated luminescence: A review, **Indian Journal of Physics**, Vol. 85, 527-550 (2011).

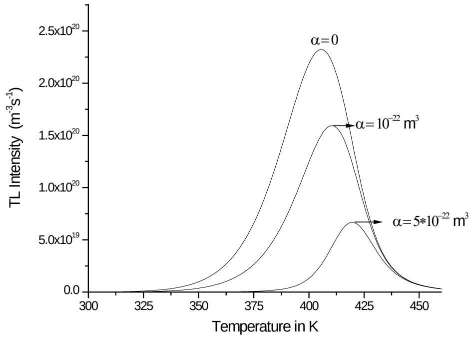

Fig. 1: TL glow curves obtained for a) no self absorption (α = 0), b) with self absorption. The other parameters assumed theoretically are $m_0 = n_0 = 10^{22}$ m^{-3}, n_{c0} = 0, E = 0.9 eV, $N = 10^{23}$ m^{-3}, s = 10^{10} Hz, $A_m = 10^{-16}$m^{3}s^{-1} $A_n = 10^{-18}$m^{3}s^{-1}, T_0 = 300, β = 1 Ks^{-1}.

Table 1: Variation in the values of glow peak temperature(T_m) and shape factor (μ_g) for different doses and self absorption (α) coefficients.

α (m^3)	$m_0 = n_0$ & $n_{c0} = 0$	T_m (K)	μ_g
0	10^{18}	533.73	0.54
	10^{19}	486.08	0.49
	10^{20}	441.40	0.48
	10^{21}	413.88	0.48
	10^{22}	405.45	0.44
10^{-22}	10^{18}	533.73	0.54
	10^{19}	486.08	0.52
	10^{20}	441.40	0.51
	10^{21}	413.88	0.50
	10^{22}	409.77	0.46
10^{-20}	10^{18}	533.73	0.54
	10^{19}	486.08	0.51
	10^{20}	450.58	0.51
	10^{21}	445.03	0.50
	10^{22}	447.09	0.50
10^{-18}	10^{18}	547.09	0.59
	10^{19}	537.11	0.58
	10^{20}	537.27	0.56
	10^{21}	537.32	0.52
	10^{22}	537.68	0.50