



## On the initial occupancy dependence of thermoluminescence under the OTOR model with different heating profiles

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*Abstract :* The dependence of peak intensity of a thermoluminescence (TL) peak under the one-trap-one-recombination centre (OTOR) model on the initial occupancy of electrons/ holes is considered for different heating schemes. It is observed that for linear heating scheme,  $I_m T_m^2$  ( $I_m$ =peak intensity,  $T_m$  =peak temperature) is proportional to initial concentration of trapped electrons ( $n_0$ ) i.e., to radiation dose. For hyperbolic heating scheme,  $I_m$  is proportional to  $n_0$ . These results will be of significance in TL dosimetry and dating.

### 1. Introduction

Thermoluminescence (TL) is the emission of light from an irradiated (natural or artificial) insulator or semiconductor and can be observed when the solid is heated. A thermoluminescent material is a material which during exposure to ionizing radiation absorbs some energy which is stored in the material. The stored energy is released in the form of visible light when the material is heated. The energy storage capability of a TL material makes it suitable for dosimetric application[1,2]. TL has also important application in dating[3]. In the present paper, we study the dependence of a suitable combination of peak intensity and peak temperature on the initial occupancy of traps under OTOR (one trap one recombination centre) model for linear and hyperbolic heating schemes.

### 2.Theory

Within the framework of OTOR model, the basic theories of first, second and general order thermoluminescence (TL) equations were proposed respectively by Randall - Wilkins [4], Garlick- Gibson[5] and May-Patridge[6].

The rate equations in the case of the first-order ( $b=1$ ) and non-first order kinetics ( $b \neq 1$ ) are[1]

$$-\frac{dn}{dt} = ns \exp\left(-\frac{E}{kT}\right)$$

(1)

$$-\frac{dn}{dt} = \frac{n^b}{N^{b-1}} s \exp\left(-\frac{E}{kT}\right) \quad (2)$$

Where  $n$ =concentration of trapped electron at time  $t$  ( $\text{cm}^{-3}$ ),  $N$ =conc. of traps ( $\text{cm}^{-3}$ ),  $s$  =frequency factor ( $\text{s}^{-1}$ ),  $E$ =activation energy(eV),  $k$ =Boltzmann constant,  $T$  =temperature at time  $t$ ,  $b$ =order of kinetics.

In the present study we consider two types of heating schemes namely linear heating scheme and hyperbolic heating scheme. The equation for linear heating scheme is given by

$$T = T_0 + \beta t$$

Where  $T_0$  and  $T$  are respectively initial temperature and temperature at time  $t$ ;  $\beta$  is the constant heating rate.

For linear heating scheme the solution of eqn.(1) and (2) are given by



$$n = n_0 \exp\left[-\left(\frac{s}{\beta}\right) \int_{T_0}^T \exp\left(-\frac{E}{kT'}\right) dT'\right]$$

$$= 1) \quad (3)$$

and  $n = n_0 / \left[1 + \frac{(b-1)s}{\beta} f^{b-1} \int_{T_0}^T \exp\left(-\frac{E}{kT'}\right) dT'\right]$

$$1) \quad (4)$$

The equation for TL intensity for  $b=1$  and  $b \neq 1$  are given by

$$I(T) = n_0 s \exp\left(-\frac{E}{kT}\right) \exp\left(-\frac{s}{\beta} \int_{T_0}^T \exp\left(-\frac{E}{kT'}\right) dT'\right)$$

$$= 1) \quad (5)$$

and

$$I(T) = N f^b s \exp\left(-\frac{E}{kT}\right) \left[1 + (b-1) s f^{b-1} \int_{T_0}^T \exp\left(-\frac{E}{kT'}\right) dT'\right]^{\frac{-b}{b-1}}$$

$$\neq 1) (6)$$

where  $f = n_0/N$  is the filling factor,  $N$  being the total concentration of traps. The conditions for maximum glow intensity  $I_m$  occurring at a peak temperature  $T_m$  are

$$\frac{E}{kT_m^2} = \frac{s}{\beta} \exp\left(-\frac{E}{kT_m}\right)$$

$$= 1) \quad (7)$$

and  $1 + (b-1) \left(\frac{s}{\beta}\right) f^{b-1} \int_{T_0}^{T_m} \exp\left(-\frac{E}{kT}\right) dT = b \frac{ks}{E\beta} f^{b-1} T_m^2 \exp\left(-\frac{E}{kT_m}\right)$  ( $b \neq 1$ ) (8)

From equations (5) and (7) the expression for TL peak intensity  $I_m$  for  $b=1$  becomes

$$I_m = n_0 s \exp(-E/kT_m) \exp\left(-\frac{s}{\beta} \int_{T_0}^{T_m} \exp(-E/kT) dT\right)$$

$$(b \neq 1) \quad (9)$$

$$I_m \neq n_0 \frac{\beta E}{kT_m^2} \exp[-u_m e^{u_m} E_2(u_m)]$$

$$(10)$$

In which  $E_2(u_m)$  is the standard second exponential integral defined as [7]

$$T_m E_2(u_m) = \int_0^{T_m} \exp(-E/kT) dT$$

$$(11)$$

For non-first order kinetics the expression for peak intensity  $I_m$  becomes

$$I_m = N s \exp(-E/kT_m) \left[ b \frac{ks}{E\beta} T_m^2 \exp(-E/kT) \right]^{\frac{b}{b-1}}$$

$$(12)$$

Equation (8) can be expressed as

$$\frac{s}{\beta} f^{b-1} \frac{T_m}{u_m} e^{-u_m} \gamma(u_m) = 1$$

$$(13)$$

Where

$$\gamma(u_m) = b - (b-1) u_m e^{u_m} E_2(u_m)$$

$$(14)$$

Using equation (13) equation (12) can be written as



$$I_m T_m^2 = n_0 \left( \frac{\beta E}{k} \right) \left[ b^{-b} \gamma(u_m) \right]^{\frac{1}{b-1}}$$

(15)

Since  $u_m$  lies between 20 to 40 for most of the observed TL peaks,  $\gamma(u_m) \approx 1$ . Assuming that the absorbed radiation dose  $D$  by a thermoluminescent material is directly proportional to the initial number or electron/holes not trapped in the respective centres, a plot of  $I_m T_m^2$  against dose  $D$  will be a straight line. It may be noted that the linear dependence between the experimentally observable quantity  $I_m T_m^2$  and the absorbed dose holds for all orders of kinetics  $b$ . Hence it provides an order independent way of estimating the applied dose.

It is evident from eqn.(8) that non-first order TL peak is characterized by the shifting of peak temperature with the variation of  $f$  and hence with the variation of absorbed dose. From equation (12) one gets

$$\frac{dT_m}{df} = \frac{\beta (1-b) f^{-b}}{s (2b+u_m)} u_m \exp(u_m) = \frac{(1-b)\gamma(u_m) T_m}{(2b+u_m) f}$$

(16)

Again it can be shown that

$$\frac{dI_m}{dT_m} = \frac{(2b+u_m) I_m}{(1-b) T_m}$$

(17)

From equation (16) and (17)

$$\frac{dI_m}{df} = \frac{dI_m}{dT_m} \frac{dT_m}{df} = \gamma(u_m) \frac{I_m}{f}$$

(18)

For  $b > 1$ , the right hand side of equation (15) is negative which implies that the peak temperature  $T_m$  shift towards low temperature with the increase of fractional filling  $f$ .

$$I_m \sim f^{\gamma(u_m)}$$

(19)

Where  $\gamma(u_m)$  is given by eqn.(14).

Using properties of  $E_2(u_m)$  [7], for large values of  $u_m$ ,  $\gamma$  will be asymptotically approach unity. The growth of TL intensity will be sub linear or super linear depending on whether  $b$  is smaller or greater than unity.

For first order kinetics  $I_m$  is directly proportional to filling factor and hence to the initial occupancy of traps.

Now we consider the hyperbolic heating function given by [8]

$$\frac{1}{T} = \frac{1}{T_0} - \beta' t$$

(20)

where  $\beta'$  has the unit  $K^{-1}s^{-1}$  and the heating rate at peak temperature is given by  $\left( \frac{dT}{dt} \right) = \beta' T_m^2$

The expression for TL intensity for  $b=1$  is [9]

$$I(T) = n_0 s \exp(-E/kT) \exp \left[ - \left( \frac{sk}{\beta' E} \right) \right] \{ \exp(-E/kT) - \exp(-) \}$$

(21)

The condition for maximum peak intensity is

$$\frac{sk}{\beta' E} = \exp(E/kT)$$

(22)

So the peak intensity  $I_m$  will be

$$I(T) = n_0 (E\beta'/k) \exp \left[ - \{ 1 - \exp(u_m - u_0) \} \right]$$

(23)



Again the TL intensity for non-first order ( $b \neq 1$ ) kinetics can be written as [9]

$$I_m(T_m) = Nf^b n_0 s \exp(-E/kT) \left[ 1 + (b-1)f^{b-1} \left( \frac{sk}{\beta' E} \right) \left\{ \exp(-u_m) - \exp(-u_0) \right\} \right]^{\frac{b}{b-1}} \quad (24)$$

The condition for peak intensity is

$$\left[ 1 + (b-1)f^{b-1} \left( \frac{sk}{\beta' E} \right) \left\{ \exp(-u_m) - \exp(-u_0) \right\} \right] = \left( \frac{bsk}{\beta' E} \right) f^{b-1} \frac{T_m}{f} \exp(-u_m) \quad (25)$$

The TL peak intensity for non-first order kinetics is

$$I_m(T_m) = Ns \exp(-u_m) \left[ \left( \frac{bsk}{\beta' E} \right) \exp(-u_m) \right]^{\frac{b}{b-1}} \quad (26)$$

Eliminating  $s$  from equation (25) using equation (24) one gets

$$I_m = n_0 (E\beta'/k) \left[ 1 + (b-1) \left\{ \exp(u_m - u_0) \right\} \right]^{\frac{1}{b-1}} \quad (27)$$

From equation (17) and (21) it is clearly seen that irrespective of the order of kinetics dose response curve between the peak intensity and  $n_0$  and hence the absorbed dose is a straight line which has a very good implication for TL dosimetry and dating.

For non-first order kinetics ( $b \neq 1$ ) the variation of peak temperature  $T_m$  with filling factor is

given by

$$\frac{dT_m}{df} = (1-b) \frac{[1 - (b-1) \exp(u_m - u_0)] T_m}{u_m f} \quad (28)$$

The variation of  $I_m$  with  $T_m$  can be expressed as

$$\frac{dI_m}{df} = [1 - (b-1) \exp(u_m - u_0)] \frac{I_m}{f} = \left( \frac{bsk}{\beta' E} \right) f^{b-1} \frac{T_m}{f} \exp(-u_m) \quad (29)$$

Then, the dependence of  $I_m$  on  $f$  can be expressed as

$$I_m = f^{\kappa(u_m)} \quad (30)$$

### **3. Results and Discussion:**

#### **3.1 Applicability to numerically computed TL glow peaks :**

The peak temperature  $T_m$  of the glow peaks corresponding to linear heating scheme has been obtained solving eqns ( ) and ( ) by Newton-Raphson method following Singh et al[10] for  $b=1$  and  $b \neq 1$  respectively. Peak intensities  $I_m$  for  $b=1$  and  $b \neq 1$  have evaluated using eqns. (10) and (12) respectively. Using the values of  $T_m$  and  $I_m$  so obtained,  $I_m T_m^2$  has been computed for first order and non-first glow curves. As a check of our computer code, we have also computed  $I_m T_m^2$  by using eqns.(10) and (15) for  $b=1$  and  $b \neq 1$  respectively. A close agreement of the values of  $I_m T_m^2$  as calculated by two different methods as mentioned earlier have been obtained. In fig. 1 we have plotted  $I_m T_m^2$  against  $f$  for different values of the order of kinetics. The synthetic curves are simulated with  $E=1$  eV,  $s=10^{12}$  sec<sup>-1</sup>, heating rate 1Ksec<sup>-1</sup>. In all the cases we have obtained a linear correlation which has very good implication for TL dosimetry and dating.

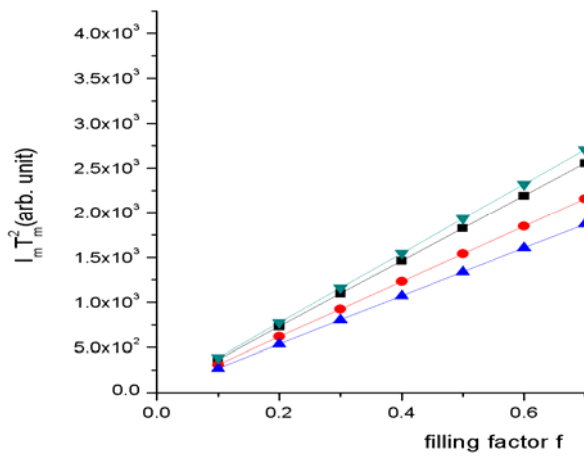


Fig1: plot of  $I_m T_m^2$  against filling factor  $f$  for different values of the order of kinetics 'b'. In all cases  $E=1\text{eV}$ ,  $s=10^{12}\text{ sec}^{-1}$ , heating rate  $1\text{Ksec}^{-1}$ .

Coming over to hyperbolic heating scheme, it is observed from eqn.(23) and (26) that for  $b=1$  and  $b\neq 1$ ,  $I_m$  is directly proportional to  $f$ . So in this case it is sufficient to consider the variation of  $I_m$  with  $f$  for the measurement of dose.

### 3.2 Applicability to Experimental glow peaks :

$\text{BaSO}_4$ , alternately known as 'Barite' or 'Schwerspat' is a natural crystalline solid having orthorhombic, bipyramidal structure. Here  $\text{BaSO}_4$  is doped with Dy and is annealed in a quartz crucible at  $600\text{K}$  for 1 hour. It is then irradiated with different doses of  $\gamma$ -rays ranging from  $100\text{-}500\text{ Gy}$  by using  $\text{Co}^{60}$  gamma irradiator. TL glow curves are recorded in the TL recording system (model TL 1404, supplied by Indotherm Instruments, Mumbai, India). The glow curves for different doses of  $\gamma$ -rays are presented in Fig. 2.

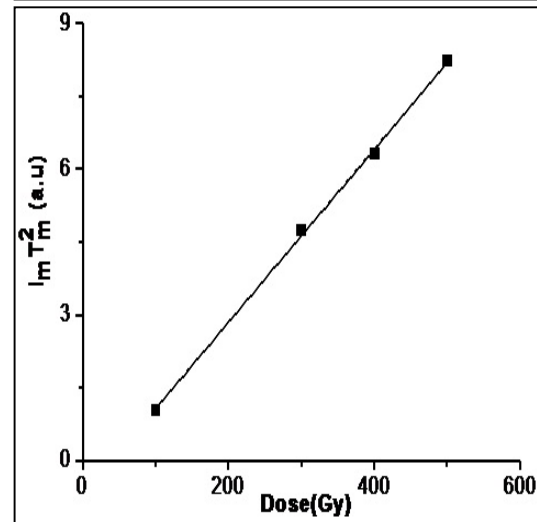
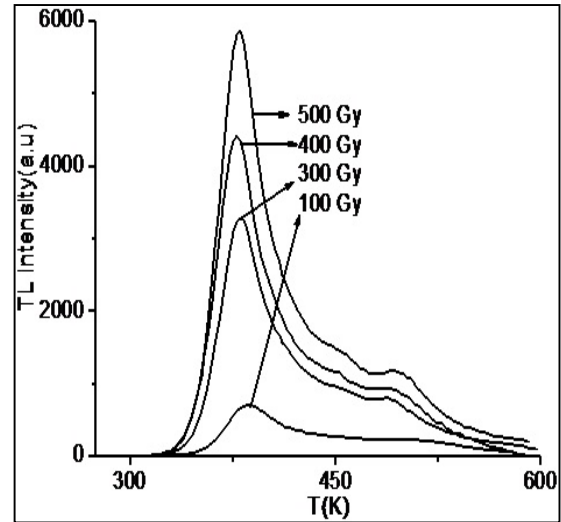


Fig2 : TL glow curves of nanocrystalline  $\text{BaSO}_4:\text{Dy}$  Fig 3: Plot of  $I_m T_m^2$  vs dose of  $\gamma$ -rays for

**annealed at  $600\text{ }^\circ\text{C}$**  and irradiated with nanocrystalline  $\text{BaSO}_4:\text{Dy}$  **annealed at  $600\text{ }^\circ\text{C}$**  different doses of  $\gamma$ -rays and irradiated with different doses of  $\gamma$ -rays

In Fig. 3, the plot of  $I_m T_m^2$  with dose is depicted As there is a linear dependence of dose and ' $f$ ', this plot actually shows the linear correlation between  $I_m T_m^2$  with ' $f$ '.

Fig 4 shows TL glow curves  $\text{BaSO}_4:\text{Dy}$  annealed at the same temperature but with larger grain size. It has a multippeak nature having two intense TL peaks at  $390$  and  $454\text{ K}$ . As

expected, TL intensity increases with the increase of absorbed gamma doses. A plot of  $I_m T_m^2$  versus  $\gamma$ -dose in

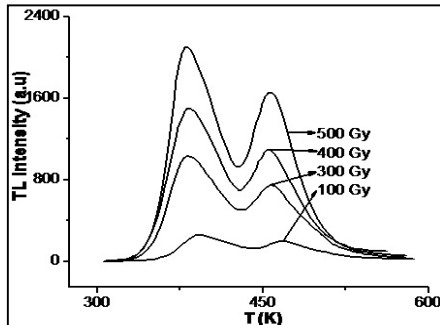


Fig 4: TL glow curves of nanocrystalline BaSO<sub>4</sub>:Dy annealed at 600 °C and irradiated with different doses of  $\gamma$ -rays.

Gy corresponding to the two TL peaks with  $T_m = 390\text{K}$  and  $454\text{K}$  (shown in Fig 4) are displayed at Fig. 5(a) and 5(b). In this case the linear behavior of  $I_m T_m^2$  against dose is not observed. This is due to the overlapping glow peaks.

#### 4. Conclusion

In this paper we have observed that for the case of TL peaks in the framework of OTOR model,  $I_m T_m^2$  is proportional to the filling factor  $f (=n_0/N)$  in a linear heating scheme. Since dose is proportional to  $f$  (or  $n_0$ ), these results can be used for an order independent way of estimating the dose. For hyperbolic heating scheme, the dose can be estimated from linear variation of peak intensity with dose.

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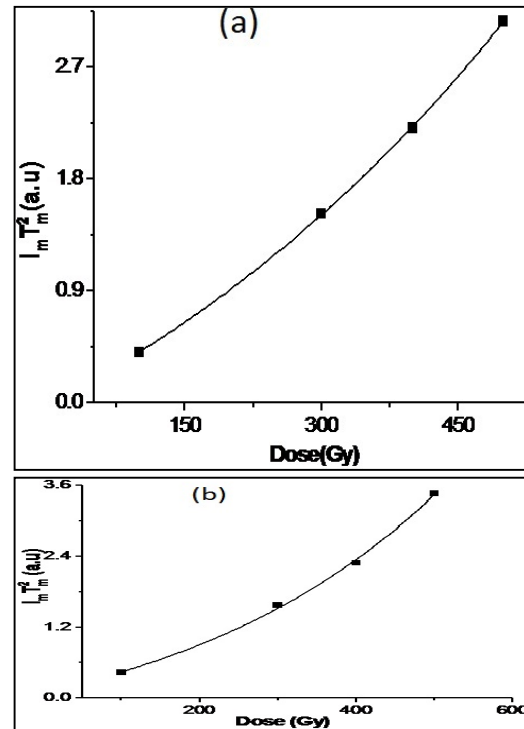


Fig5: Plot of  $I_m T_m^2$  vs. dose of  $\gamma$ -rays for nanocrystalline BaSO<sub>4</sub>:Dy annealed at 600 °C and irradiated with different doses of  $\gamma$ -rays- (a)  $T_m = 390\text{K}$ , (b)  $T_m = 454\text{K}$

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