

## PRE-EXPONENTIAL FACTOR IN GENERAL ORDER KINETICS OF THERMOLUMINESCENCE AND ITS INFLUENCE ON GLOW CURVES

C. M. Sunta†\*, W.E. Feria Aytá†, R. N. Kulkarni‡, R. Chen†\* and S. Watanabe†

†Universidade de São Paulo

Instituto de Física, Dept Física Nuclear

CP 66318, 05315-970 São Paulo-SP, Brazil

‡Atomic Energy Regulatory Board

V.S. Bhavan Anushakti Nagar

Bombay-400094, India

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**Abstract** — A model of thermoluminescence kinetics based on a physically meaningful approach shows that the glow curve shapes undergo systematic changes with the change of trap occupancy (dose). In terms of the general order kinetics model it means that the kinetic order changes with sample dose. In parallel to the kinetic order, the pre-exponential factor also changes. In contrast to these results the glow curves calculated from the general order kinetics model show that the peak shape remains nearly constant when the trap occupancy is changed. When appropriately defined, the pre-exponential factor also has a fixed value independent of trap occupancy. In these respects the general order kinetics model, though empirical, seems to describe the glow peak behaviour quite successfully. However, regarding the peak temperature the theoretical results both from the physical as well as the empirical model seem to diverge from the experimental observations when the experimentally determined kinetics is non-first order. In such cases, according to the models, the peak maxima shift with the change in trap occupancy, while in the experimental observations such changes are not evident. The shift in the peak maximum in the non-first order cases of the theoretical models is the result of the pre-exponential factor being effectively changed by the alteration in the trap occupancy. In addition to these findings the paper also includes the possible implications of non-first order kinetics in dosimetric applications.

### INTRODUCTION

May and Partridge<sup>(1)</sup> gave an expression for the intensity  $I$  of thermoluminescence (TL) for the general order kinetics (GOK) as

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

where

- $n$  = concentration of trapped carriers ( $m^{-3}$ );
- $b$  = order of kinetics, which may have any value including a non-integer; the relevant range of  $b$  values, however, is from 1 to about 2;
- $E$  = activation energy of the trap (eV);
- $k$  = Boltzmann constant ( $eV.K^{-1}$ );
- $T$  = temperature of the sample (K);
- $s'$  = a constant quantity called the pre-exponential factor.

Equation 1 is an empirical relation and may not be related to any particular model described by an energy level scheme. The sense in which Equation 1 is general,

however, is that it reduces to the first order kinetics of Randall and Wilkins<sup>(2)</sup> (RW) when  $b = 1$  and to second order of Garlick and Gibson<sup>(3)</sup> (GG) when  $b = 2$ . In the RW model  $s'$  is equal to the basic parameter called frequency factor  $s$  and is in units of  $s^{-1}$ . In the GG model  $s' = s/N$ , where  $N$  = total concentration of the traps. It does not, however, mean that these are the only possible definitions of  $s'$ . One may get other definitions for  $s'$  for first and second order depending on the model used. For example, if one starts with an energy level scheme consisting of one trap and one recombination centre (RC) and imposes the quasi-equilibrium condition in the rate equations, the TL glow peak is expressed by<sup>(4)</sup>

$$I = -\frac{dm}{dt} = \frac{A_m m n s e^{-E/kT}}{(N-n)A_n + mA_m} \quad (2)$$

where  $m$  = concentration of RC ( $m^{-3}$ ) and  $N$  = total concentration of traps ( $m^{-3}$ ).  $A_n$  and  $A_m$  are the capture coefficients of the traps and RC respectively ( $m^3.s^{-1}$ ). Now if further conditions are imposed so that retrapping far exceeds recombination, i.e.  $(N-n)A_n \gg mA_m$  and also  $n \ll N$  and  $m = n$ , Equation 2 reduces to

$$I = s'n^2 e^{-E/kT} \quad (3)$$

This is a second order ( $b = 2$ ) TL relation in which  $s'$  is given by

$$s' = \frac{sA_m}{NA_n} \quad (4)$$

\*Permanent addresses: (C.M.S.) C43/398 Gandhi Nagar (MIG) Bandra (E) Bombay 400051, India.  
 (R.C.) School of Physics and Astronomy Tel-Aviv University, Tel-Aviv, Israel.

If the condition  $m = n$  is not imposed and instead  $m = n + C$  and further  $C \gg n$  is assumed, Equation 2 reduces to

$$I = s'ne^{-E/kT} \quad (5)$$

This is a first order ( $b = 1$ ) TL equation with  $s'$  defined as

$$s' = \frac{sA_m C}{NA_n} \quad (6)$$

where  $C$  is a fixed high concentration of RC which may arise because of the presence of thermally disconnected deep traps<sup>(4,5)</sup>. Similarly there may be other models and conditions under which different expressions may be obtained for  $s'$  which may fit into Equation 1 along with  $b = 1$  or  $b = 2$ ; but when neither  $b = 1$  nor  $b = 2$  applies in this Equation, an expression for  $s'$  which has a constant value cannot be found from these physically meaningful models. For example if no special conditions as stated above are imposed in Equation 2 it cannot be reduced to the form of Equation 1. The glow peak obtained from Equation 2, however, may be fitted into Equation 1 with fixed values of  $s'$  and  $b$ . The  $b$  values may be different from 1 and 2, including non-integral numbers. The values of these fitted parameters may, however, change with the trap occupancy (dose). An example of such a behaviour is shown in Figure 1 in

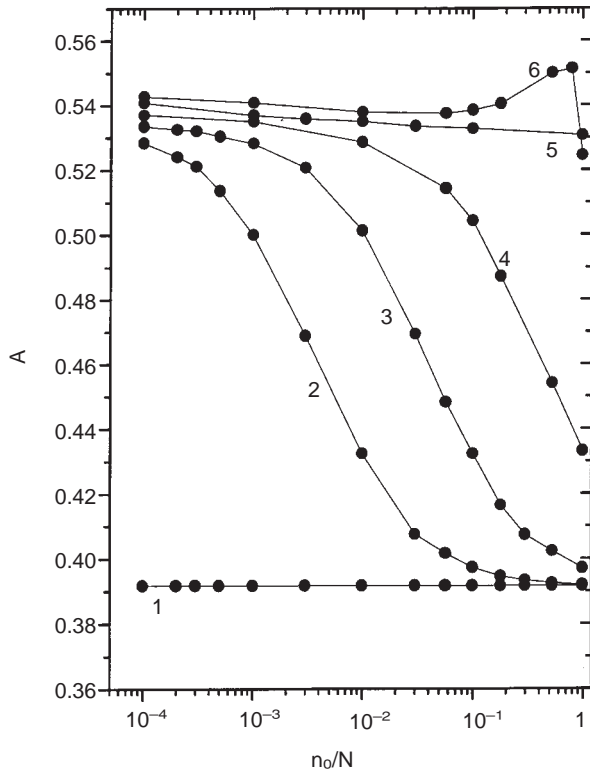


Figure 1. Shape factor  $A$  of the glow peaks calculated from Equation 2 as a function of  $n_0/N$  for various values of  $A_n/A_m$ . Curves 1 to 6 are for  $A_n/A_m = 0, 10^{-3}, 10^{-2}, 10^{-1}, 1$  and  $10$  respectively. The parameter values used in the glow curve calculations are  $E = 1 \text{ eV}$ ,  $s = 10^{12} \text{ s}^{-1}$ ,  $N = 10^{12} \text{ m}^{-3}$  and the heating rate  $\beta = 1 \text{ K.s}^{-1}$ .

which the shape factor  $A$  of the glow curve (defined as the ratio of the fall side half area to the full area of the glow peak) derived from Equation 2 is plotted as a function of the initial trap occupancy  $n_0/N$ . The individual curves in the figure are for different values of  $A_n/A_m$ . The value of  $A$  is calculated by integrating the area of the full glow peak and that of the fall side half. The glow curves are calculated numerically using the Runge-Kutta method. The parameter values used in the calculation are given in the figure caption. Each of the plotted points in the curves of Figure 1 represents the  $n_0/N$  value at which the glow curve is calculated. As before it is assumed that  $m = n$ . The figure essentially shows that the shape factor  $A$  is dependent on  $n_0/N$ . Since in the GOK the principal influencing factor for  $A$  is the order value  $b$ <sup>(6)</sup>, the figure demonstrates the variation of  $b$  with  $n_0/N$ . This variation is conspicuous when  $0 < A_n/A_m < 1$ . This range of  $A_n/A_m$  ratio leads to  $1 \leq b \leq 2$ .

In parallel to the variation of  $b$  with  $n_0/N$ , there are changes also in the effective value of the pre-exponential factor. An example of the change in this factor with change in  $n_0/N$ , may be seen by examining curve 3 in Figure 1 in which  $A_n/A_m = 10^{-2}$ . In this curve, when  $n_0/N$  is about 0.5 to (near saturation dose) the  $b$  value is seen to be about 1 ( $A \approx 0.40$ ) and  $s'$  is nearly equal to  $s$ . Under this type of trap occupancy, the retrapping rate remains much smaller than the recombination rate until the traps are emptied almost up to 99%. This may be visualised directly from Equation 2 by substituting  $m = n$ ,  $A_n/A_m = 10^{-2}$  and  $n > 10^{-2}N$ . On the other hand, when  $n_0/N \ll 10^{-2}$ , the  $b$  value becomes equal to about 2 ( $A \approx 0.53$ , see Figure 1, curve 3) and  $s' = sA_m/NA_n$  (see Equation 4). Speaking in terms of pure numbers it means a change in  $s'$  from  $10^{12}$  to  $10^2$  for the set of parameters used ( $s = 10^{12} \text{ s}^{-1}$ ,  $N = 10^{12} \text{ m}^{-3}$  and  $A_m/A_n = 10^{-2}$ ) as  $n_0/N$  changes from 1 to  $< 10^{-3}$ . The numbers, however, cannot be compared with each other, since they do not have the same dimensions. None the less it is noteworthy that the change in the character of  $s'$  with change in  $n_0/N$  is commensurate with the change in the value of  $b$  obeyed by the glow peak. This may be visualised directly from Equation 2. When  $A_m m \gg (N - n)A_n$ ,  $s' = s(s^{-1})$  and  $b = 1$ . When  $(N - n)A_n \gg mA_m$ ,  $s'$  has the units of  $\text{m}^3.\text{s}^{-1}$  and  $b = 2$ . This change in the character of  $s'$  in parallel with the change in the value of  $b$  may be seen also in GOK (see Equation 7).

Notwithstanding the above, the non-integral values of  $b$  reported for various materials in the published literature are given without specifying the sample dose. The  $b$  and  $s'$  values in fact seem to be reported as a fixed parameter of a given glow peak in a given material<sup>(7-14)</sup>. However, there are examples of measurement of  $b$  at different doses. Kathuria and Sunta<sup>(15)</sup> and Satinger *et al*<sup>(16)</sup> find that in LiF: Mg,Ti (TLD-100) the  $b$  value is non-integral (between 1 and 2) and is independent of dose under identical conditions of sample

treatment. The determination of  $b$  was carried out using peak shape as well as using isothermal decay of TL. This means that in practice it is possible to encounter glow peaks which have the same  $b$  ( $b \neq 1$ ) when  $n_0/N$  varies. In this paper it is shown that this behaviour of glow peaks may be described by the GOK Equation 1 in which  $b$  and  $s'$  may be given a fixed value regardless of the trap occupancy  $n_0/N$ . It is further shown that the condition of  $b$  and  $s'$  being independent of  $n_0/N$  holds when  $s'$  is defined as

$$s' = \frac{s}{N^{b-1}} \quad (7)$$

This definition of  $s'$  has been given earlier by Rasheedy<sup>(17)</sup>. It facilitates balancing the units of  $s'$  ( $m^{3(b-1)} \cdot s^{-1}$ ) in terms of the physically meaningful parameters  $s$  and  $N$ . The definition applies to the case of a two-level scheme in which the concentration of the trapped carriers involved in the TL emission is equal to that of the available recombination centres (i.e.  $m = n$  in the scheme of Equation 2). The energy level scheme assumed thus is like that of the GG model except that the order is not equal to 2. Such a scheme could be applicable even in glow curves with multiple TL peaks if there is some kind of pairing between the traps and the RC so that within each pair one has  $m = n$ . Such a possibility has been suggested earlier by Chen *et al*<sup>(18)</sup>. As will be seen in the following, the use of the definition of Equation 7 avoids the need to know the absolute values of  $n_0$  to calculate and the glow curves. The calculations may be handled in terms of the initial trap occupancy  $n_0/N$  which may be related to the sample dose. In the following, glow curves are calculated using this definition in Equation 1. The calculated glow curves in particular show that the shape factor remains nearly unchanged with variation in  $n_0/N$ , (see Table 1) thus showing that the effective value of  $b$  is independent of dose. It means that  $s'$  too, as given by Equation 7, is independent of dose. The behaviour of the glow curves derived from the GOK Equation 1 thus is in contrast to that from Equation 2.

GLOW CURVES AND THEIR CHARACTERISTICS

Integration of Equation 1 using  $s' = \frac{s}{N^{b-1}}$  leads to

$$I = n_0 s \left(\frac{n_0}{N}\right)^{b-1} \exp\left(-\frac{E}{kT}\right) \left[ (b-1) \frac{s}{\beta} \left(\frac{n_0}{N}\right)^{b-1} \int_{T_0}^T \exp\left(-\frac{E}{kT'}\right) dT' + 1 \right]^{-b(b-1)} \quad (8)$$

This expression reduces to that of GG when  $b = 2$  and to that of RW when  $b \rightarrow 1$ . Figure 2 shows the com-

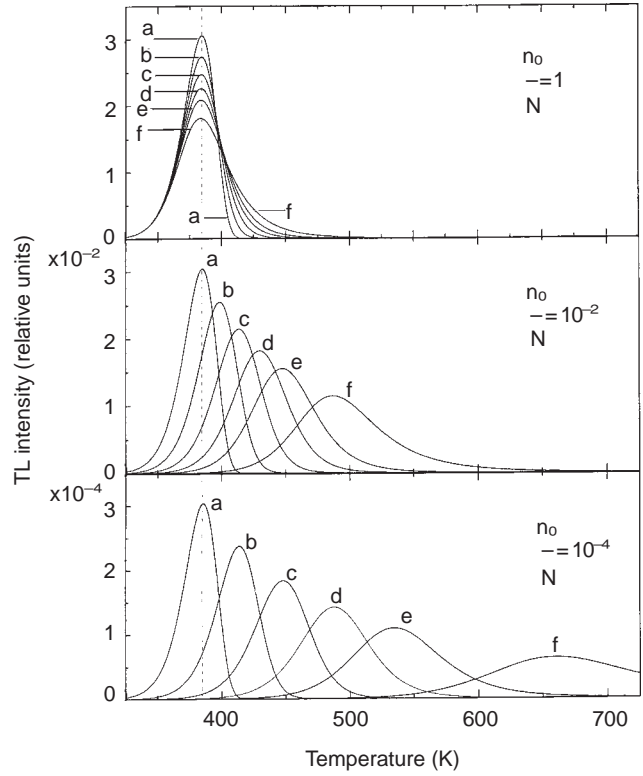


Figure 2. Glow curves obtained by varying  $b$  and  $n_0/N$  as shown. Curves a, b, c, d, e and f correspond to  $b = 1.01, 1.25, 1.5, 1.75, 2$  and  $2.5$  respectively. The values of the parameters  $E, s$  and  $\beta$  used are the same as in Figure 1.

Table 1. List of  $s'', T_m$  and  $A$  for various  $n_0/N$  and  $b$  obtained from the calculations used in the glow curves of Figure 2.

$n_0/N$ $b$	1			$10^{-2}$			$10^{-4}$		
	$s''(s^{-1})$	$T_m(K)$	$A$	$s''(s^{-1})$	$T_m(K)$	$A$	$s''(s^{-1})$	$T_m(K)$	$A$
1.01	$10^{12}$	384.50	0.393	$10^{11.98}$	385.1	0.392	$10^{11.96}$	385.6	0.394
1.25	$10^{12}$	384.34	0.436	$10^{11.5}$	398.6	0.435	$10^{11}$	413.9	0.436
1.5	$10^{12}$	384.18	0.472	$10^{11}$	413.6	0.475	$10^{10}$	447.8	0.477
1.75	$10^{12}$	383.96	0.503	$10^{10.5}$	429.8	0.505	$10^9$	487.6	0.510
2.0	$10^{12}$	383.86	0.531	$10^{10}$	447.3	0.535	$10^8$	534.6	0.541
2.5	$10^{12}$	383.48	0.575	$10^9$	486.6	0.583	$10^6$	660.1	0.596

puted glow curves for various values of  $b$  for three cases of trap occupancies, namely  $n_0/N = 1, 10^{-2}$  and  $10^{-4}$ . The parameter values used are described in the figure caption. The results may be summarised as follows:

- (i) As a general rule the glow curve shifts to higher temperature as  $b$  increases and  $n_0/N$  decreases. As an exception, however the peak remains at nearly the same position irrespective of changes in  $b$  when  $n_0/N = 1$  and irrespective of change in  $n_0/N$  when  $b \rightarrow 1$ . The lowest temperature that a given glow peak of  $b \neq 1$  may attain is when  $n_0/N = 1$ . The temperature of maximum intensity,  $T_m$ , under this condition nearly coincides with that of the  $b = 1$  case. It is easy to comprehend that the peak shift to higher  $T$  is the consequence of increased retrapping which takes place due to increased  $b$  and/or decreased  $n_0/N$ .
- (ii) The shape factor  $A$  of the glow curve is influenced almost solely by changes in  $b$  and is independent of trap occupancy  $n_0/N$  (dose). Glow curves of general order kinetics have been calculated earlier by Chen<sup>(6)</sup> for different values of  $E$ ,  $b$  and  $s''$ , where  $s'' = s'n_0^{b-1}$ . The graphical shape with varying  $b$  has been shown by Kirsh<sup>(19)</sup>. The glow curves given in Figure 2 are more informative than those using a fixed  $s''$  (compare for example with Figure 2 in Kirsh<sup>(19)</sup>, which depict the glow peaks at the same temperature of maximum  $T_m$  when  $b$  is varied). Table 1 lists  $T_m$  and  $A$  for different values of  $b$  and  $n_0/N$ . The values of  $s''$  are also listed. In terms of Equation 8,  $s''$  is given by

$$s'' = s \left( \frac{n_0}{N} \right)^{b-1} \quad (9)$$

It may be seen that when  $s''$  is fixed,  $T_m$  also becomes nearly fixed (see Table 1). The effect of change in  $b$  on  $T_m$  is negligible once  $s''$  is fixed. The condition under which  $s''$  and  $T_m$  are independent of  $b$  in the present formalism of general order kinetics is when  $n_0/N = 1$ . Under this condition  $s''$  becomes equal to the frequency factor,  $s$ , in this formalism. We may now retrospectively<sup>(6,19)</sup> state that the results depicting a fixed  $T_m$  for varying values of  $b$  would always apply to the specific case of  $n_0/N = 1$ , i.e. when all the samples having different  $b$  (but the same  $E$  and  $s$ ) are given a dose to saturate their TL. One may, however, produce glow peaks with nearly the same  $T_m$  at non-saturation doses ( $n_0/N < 1$ ) also by selecting the individual values of  $s$  and  $b$  such that the total quantity  $s (n_0/N)^{(b-1)}$  is equal to a constant; but this would sacrifice the information on the effects of changes in  $s$ ,  $b$  and  $n_0/N$  individually.

#### IMPLICATION OF $b \neq 1$ IN DOSIMETRIC APPLICATION OF TL MATERIALS

As seen from Figure 2, the non-first order ( $b \neq 1$ ) glow peaks shift in position with the change in trap

occupancy. This is an undesirable property for dosimetry, particularly if multiple overlapping peaks, each having a different  $b$  exist in a glow curve. In such cases the shape of the full glow curve may vary with change in dose, thus complicating the dose-intensity relation. Another property of non-first order glow peaks, according to the GOK model, is that the TL peak intensity grows supralinearly with trap occupancy. This is because the glow peaks become wider due to increased retrapping as the occupancy decreases. The area of the glow peak, however, has a linear relationship with the occupancy under the model used. The relative TL peak intensity ( $I$ ) to occupancy ( $n_0/N$ ) relation is found to be

$$I = (n_0/N)^x \quad (10)$$

before the saturation effect sets in. The value of the growth exponent  $x$  depends on  $b$ . Table 2 shows the  $x$  values corresponding to different  $b$  in the glow curves of Figure 2. At  $b = 2$ , the present value of  $x = 1.07$  matches with  $x = 1.08$  found by Chen *et al*<sup>(18)</sup>.

#### APPLICABILITY OF GOK MODEL

The results presented above show that under the GOK model, a glow peak may be described by fixed values of  $b$  and  $s'$  independent of  $n_0/N$ . However, the glow peak shifts its position with change in  $n_0/N$  when  $b \neq 1$ . This shift is the consequence of change in the value of  $s''$  (see Equations 8 and 9). The quantity  $s''$  may be called the effective pre-exponential factor. Its units being  $s^{-1}$ , its influence on the glow curves is the same as that of the frequency factor  $s$ , in RW model. The value of  $s''$  for an experimentally measured glow peak may be determined by using the following relation (see Equation 13 in Chen<sup>(6)</sup>):

$$s'' = \frac{\beta E \exp(E/kT_m)}{kT_m^2 [1 + (b-1) (2kT_m/E)]} \quad (11)$$

where  $s''$  in the present treatment is given by Equation 9. Being variable with  $n_0/N$ , the value of  $s''$  determined at an unspecified sample dose is of little use. However at  $n_0/N = 1$  (saturation dose),  $s''$  becomes equal to the frequency factor  $s$ . Thus according to the present treatment of the GOK, the measurement of the so called pre-exponential factor should be carried out by giving saturation dose to the sample, at which the obtained result provides the frequency factor  $s$  directly. Care should be taken to select the saturation dose, since too high a dose after reaching the saturation level may damage the sample and change its TL characteristics.

**Table 2. Values of growth exponent  $x$  at different values of  $b$  for the GOK glow curves of Figure 2.**

$b$	1.01	1.25	1.5	1.75	2.0	2.5
$x$	1.001	1.016	1.032	1.05	1.07	1.14



In a variety of samples that have been studied and are found to have  $b \neq 1$ , there is no shift in  $T_m$  with change in dose. The representative examples of such samples may be cited as the well known materials LiF:Mg,Ti<sup>(15,16)</sup>, calcite<sup>(14)</sup> and CaSO<sub>4</sub>:Dy<sup>(7,20)</sup>. In fact such examples may be many. This behaviour of  $T_m$  is in complete divergence from that expected from the GOK model, which shows a systematic shift in  $T_m$  as the trap occupancy is changed (see Figure 2 and Table 1). This means that neither the physically meaningful model given by Equation 2, nor the GOK model given by Equation 1 is able to describe the actual glow curve behaviour both in respect of glow peak shape (i.e.  $b$ )

as well as the glow peak temperature. Obviously the interpretation of the kinetic behaviour of the TL glow peaks may involve phenomena additional to the simple untrapping, recombination and retrapping of charge carriers.

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