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INTENSITY OF A THE RMOLUMINE SCENCE GLOW CURVE INVOLVING GENERAL ORDER KINETICS

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Abstract: ***Adirovitch set of equations has been modified with an aim to develop a generalized equation for the intensity of a thermoluminescence glow curve. It has been found that the first order kinetics is a recombination dominant process with no retrapping. The probabilities of recombination and retrapping are equal in second order kinetics. With the increasing order of kinetics, the extent of recombination has been found to decrease with a simultaneous increase in the retrapping.***

Key Words: Adirovitch set, generalized equation, thermoluminescence, recombination dominant.

Intensity of a first order thermoluminescence (TL) glow curve is expressed by

$$i_1 = n_0 s_1 \exp[\,(-E_{a1}/kT) - (s_1/b)\int_{T_0}^{T}\exp(-E_{a1}/kT')dT'\,] \qquad (1)$$

where n_0 is the initial concentration of trapped electrons per unit volume, s the pre-exponential factor, k the Boltzmann's constant, T_0 the absolute temperature wherefrom the TL glow curve starts to appear, T' an arbitrary temperature in the range T_0 to T and suffix 1 represents the data related to first order kinetics . It has been pointed out by Randall and Wilkins that the first order kinetics is a recombination dominant process with negligible or no retrapping . When the probabilities of recombination and retrapping are equal, one observes a second order TL glow curve. The intensity of a second order TL glow curve is expressed by

$$i_2 = n_o^2 s_2 \exp[\,(-E_{a2}/kT)]\,.\,[1 + (n_o s_2/b)\int_{T_0}^{T}\exp(-E_{a2}/kT')dT'\,]^{-2} \qquad (2)$$

s_2 is related to s_1 through the equation

$$s_1 = s_2 N \qquad (3)$$

N represents the concentration of total electron traps per unit volume and suffix 2 represents the parameters associated with second order kinetics. It is obvious that s_2 has the dimension m^3s^{-1}. When all the available electron traps are filled initially i.e. when $N = n_o$, the intensities of second and higher order TL glow curves are expressed by

$$i_\iota = n_o s_1 \exp[\,(-E_{a\iota}/kT)]\,.\,[1 + \{ s_1(\iota - 1)/b)\int_{T_0}^{T}\exp(-E_{a\iota}/kT')dT'\,]^{\iota-1}/(\iota-1)\} \qquad (4)$$

where suffix ι represents the order of kinetics involved.

It is obvious that equation (4) fails to represent the intensity of first order TL glow curve whereas it successfully explains the intensities of second and higher order TL glow curves. This anomaly has been resolved by Prakash by suggesting a modified form of equation (4). Later on, it has been found that modifications proposed by Prakash have some inherent demerits. It has, therefore, been suggested that Adirovitch set of equations should be represented as

$$i_\iota = -dm/dt = (1/\iota)\, m\, n_c A_m \qquad (5)$$

$$-dn/dt = n s_\iota \exp[\,(-E_{a\iota}/kT)] - (\iota - 1)\, n_c(N - n)A_n \qquad (6)$$

$$m = n + n_c \qquad (7)$$

where m is the density of recombination centre, n_c the density of electrons in the conduction band, A_m the recombination probability, n the density of electrons in the trap centre and A_n the retrapping probability . The first term on the right hand side of equation (6) represents the rate of release of electrons from their respective traps and the second term represents the rate of their retrapping. It is obvious that for $\iota = 1$, the rate retrapping is zero which is a characteristic feature of the first order TL glow curve.

Similar to the case of retrapping of electrons, it is proposed here that the release of electrons from their respective traps should also be a function of the order of the kinetics involved. Consequently, the rate of release of electrons for ιth order of kinetics should be expressed by

$$N\, s_\iota N^{(\iota - 1)} \exp[(-E_{a\iota}/kT)] \qquad (8)$$

where s_ι is the pre-exponential factor and has the dimensions $m^{3(\iota - 1)}\, s^{-1}$. Pre-exponential factors s_1 and s_ι are related through the expression...

$$s_1 = s_\iota N^{(\iota - 1)} \qquad (9)$$

Obviously, equation (9) changes to equation (3) for $\iota = 2$

Thus, the modified Adirovitch set of equations be represented by equations (5) and (7) and

$$-dn/dt = N\, s_\iota N^{(\iota - 1)} \exp[(-E_{a\iota}/kT)] - (\iota - 1)\, n_c (N - n) A_n \qquad (10)$$

Equations (5) , (7) and (10) characterize the mechanism responsible for the appearance of TL glow curve.

In the situation when

$$n_c << n \quad \text{and} \quad dn_c/dt << dn/dt \qquad (11)$$

One gets from equations (5) , (7) and (10)

$$i_\iota = -dn/dt = -dm/dt$$

$$i_\iota = n\, s_\iota N^{(\iota-1)} \exp[(-E_{a\iota}/kT)][(m\, n_c A_m/\iota) / \{(m\, n_c A_m/\iota) + (\iota - 1)\, n_c (N - n) A_n\}] \qquad (12)$$

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$$i_l = (1/l)n_0^l s_l \exp[(-E_{al}/kT)-(n_o^{(l-1)} s_l/lb)\int_{T_0} \exp(-E_{al}/kT')dT'] \qquad (15)$$

It can be seen that equation (15) changes to equation (1) for l =1. Thus, TL intensities of second and higher order kinetics including the first order can be represented by generalized equation (15).

It is apparent from equation (13) that the occurrence of TL glow curves involving different order of kinetics depends on the extents of recombination and simultaneous retrapping. First order kinetics is a process which involves 100% recombination and no retrapping. For the second order kinetics, the probabilities of recombination and retrapping are 50% each. These conclusions are found to be in accordance with the statements made by Randall and Wilkins for first order of kinetics and by Garlick and Gibson for second order kinetics . Conditions for the appearance of TL glow curves involving different order of kinetics are presented in table 1. It is obvious from the table that the rate of recombination decreases with a simultaneous increase in the rate of retrapping when one pocesses to higher order kinetics from first order.

Condition for the location of peak of TL glow curve can be obtained from equation (15). The location of the peak at T_{ml} of lth order of kinetics is given by

$$(1/l)T_{ml}^2 = [b E_{al}/k s_l n_o^{(l-1)}] \exp[E_{al}/k T_{ml}] \qquad (16)$$

This equation for the first order kinetics i.e. for l = 1changes to

$$T_{ml}^2 = (b E_{al}/k s_l) \exp[E_{al}/k T_{ml}] \qquad (17)$$

suggesting that the location of TL peak at T_{ml} will be independent of the initial concentration of the trapped centers. Whereas for second order kinetics it shows a dependence on initial concentration vide

$$T_{m2}^2 = (2b E_{a2}/k s_2 n_o) \exp[E_{a2}/k T_{m2}] \qquad (18)$$

This conclusion happens to be in qualitative agreement with the results reported by Chen et al. It can be concluded therefore that if T_m does not depend on n_o, it is a process which involves first order kinetics. If T_m is found to depend on n_o, a kinetics higher than first order will be involved.

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Table - 1

Conditions for the appearance of TL glow curves involving different order kinetics

Order of the kinetics involved	Relationship in-between m and n	Extent of recombination	Extent of simultaneous retrapping
First	m = n	100%	0%
Second	m = n	50%	50%
Third	m = n	33%	67%
Fourth	m = n	25%	75%
Fifth	m = n	20%	80%
:	:	:	:
:	:	:	:
:	:	:	:
:	:	:	:
?h	m = n	(100 / ?)%	(100(?-1)/?)%

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