

A two-stage thermally-assisted optically stimulated luminescence (TA-OSL)

Dianabasi Nkereuwem Akpan ^{1,*}, Mfoniso Udofia Aka ¹, Christopher Iniabasi Effiong ² and Sunday Samuel Ekpo ¹

¹ Department of Physics, University of Uyo, Nigeria.

² Department of Geoscience, University of Uyo, Nigeria.

Abstract

A two-stage thermally-assisted optically stimulated luminescence model is studied. Electrons are thermally and optically raised from the ground state to an excited state and there after they are thermally raised from the excited state to the conduction band. A set of simultaneous differential equations generated from the model was solved numerically using ode 15s MATLAB solver. Three assumed conditions are applied to the model and results from the analytical expressions obtained based on the assumptions were compared with the numerical simulation results. The assumed conditions are : (1) the probability of electron moving from the excited state to ground state (p) is far greater than thermal excitation of electron from the excited state to conduction band ($s_2 e^{-E_2/kT}$) also stimulation light intensity (f) is far greater than thermal excitation of electron from the ground state to the excited state ($s_1 e^{-E_1/kT}$), (2) $p \ll s_2 e^{-E_2/kT}$ also $f \ll s_1 e^{-E_1/kT}$ (3) $p \gg s_2 e^{-E_2/kT}$ also $f \ll s_1 e^{-E_1/kT}$. For the first condition, the effective activation energy (E_{eff}) tends towards the energy of the excited state (E_2), and the effective frequency factor is the ratio of the product of frequency factor for the excited state (s_2) and f to p (i.e. $s_{eff} = \frac{fs_2}{p}$). For the second condition, E_{eff} was the activation energy of the ground state (E_1) and $s_{eff} \approx s_1 e^{-(E_2-E_1)/kT}$, s_1 is the frequency factor for thermal excitation from the ground state to the excited state. For the third condition, E_{eff} was the sum of E_1 and E_2 and $s_{eff} \approx \frac{s_1 s_2}{p} e^{-E_1/kT}$ but when the stimulating light intensities become very small compared to s_1 , then s_{eff} becomes approximately equal to s_2 .

Keywords: Model; Thermally; Optically; Stimulated; Luminescence

1. Introduction

Thermoluminescence (TL) and optically stimulated luminescence (OSL) have several applications in areas such as dating of archaeological and geological samples and in dosimetry [1-7]. Thermally-assisted optically stimulated luminescence (TA-OSL) process can be achieved when the electrons are thermally as well as optically excited from the ground state to the excited state. On reaching the excited state, they are thermally excited into the conduction band. On

*Corresponding author: Dianabasi Nkereuwem Akpan; Department of Physics, University of Uyo, Nigeria.

the other hand, TA-OSL process can also be achieved when the electrons are thermally excited from the ground state to the excited state and then thermally and optically raised into the conduction band. In both cases described above, when the electrons reach the conduction band, they can either be retrapped into the excited state or recombine with holes at the centre to produce Thermally-assisted optically stimulated luminescence (TA-OSL) photons.

Some authors have worked on either stimulation of TL or TA-OSL. For example, Chen *et al.* [8] worked on a two-stage thermal stimulation of TL while Akpan *et al.* [9] worked on a three-stage thermal stimulation of thermoluminescence (TL); Pagonis *et al.* [10] worked on a simplified semi-localized transition (STL) model which was similar to a two-stage thermal stimulation of TL but they introduced a non-radiative transition; Chen and Pagonis [11] worked on modeling TL-like TA-OSL; Chen and Pagonis [12] worked on the role of simulations in the study of TL; Chen and Pagonis [13] studied the stability of the TL and OSL signals and Polymeris [14] overviewed thermally assisted OSL (TA-OSL) from various luminescence phosphors; Chen *et al.* [15] studied a model that described the possibility of simultaneous recombination of two electrons into two-hole centres that produced thermoluminescence.

There have been reports of phosphors which when simulated with light of certain wavelengths did not produce any measurable luminescence, but thermal stimulation produced luminescence. For example, stimulation of quartz with infrared is known not to produce luminescence [16-17] but its thermal stimulation does. It is possible these kinds of phosphors have some of its trap levels at higher energy (excited state) and the light is only able to raise electrons from the ground state to the excited state. It is against this backdrop that we intend to consider a model whereby the first stage of electron elevation is both thermal and optical stimulation and the second stage is only thermal stimulation. The aims of this study are to apply three assumed conditions to the model and compare analytical expressions with numerical simulation results and compare our results with TA-OSL model of Chen and Pagonis [11] and a two-stage stimulation of thermoluminescence (TL) model of Chen *et al.* [8].

2. Model

The energy level of the TA-OSL model is shown in fig. 1. The first stage is a combination of thermal and optical stimulation while the second stage is only thermal stimulation. This process consists of the trapping state with concentration N (cm^{-3}), instantaneous occupancy n (cm^{-3}), the stimulating light intensity f (s^{-1}) and the excited state n_e for movement of electrons from the ground state to excited state, the activation energy is E_1 (eV) and the frequency factor is s_1 (s^{-1}). For movement of electrons from the excited state into the conduction band, the activation energy is E_2 (eV) and frequency factor is s_2 (s^{-1}). In the excited state, there also exists a probability of electrons being retrapped into the ground state. The probability of retrapping the electrons into the ground state is p (s^{-1}). The instantaneous concentration of electrons in the conduction band is denoted by n_c (cm^{-3}). Once the electrons reach the conduction band, it can be retrapped into the excited state with a probability coefficient of A_n ($\text{cm}^3 \text{s}^{-1}$) or recombine with a hole at the

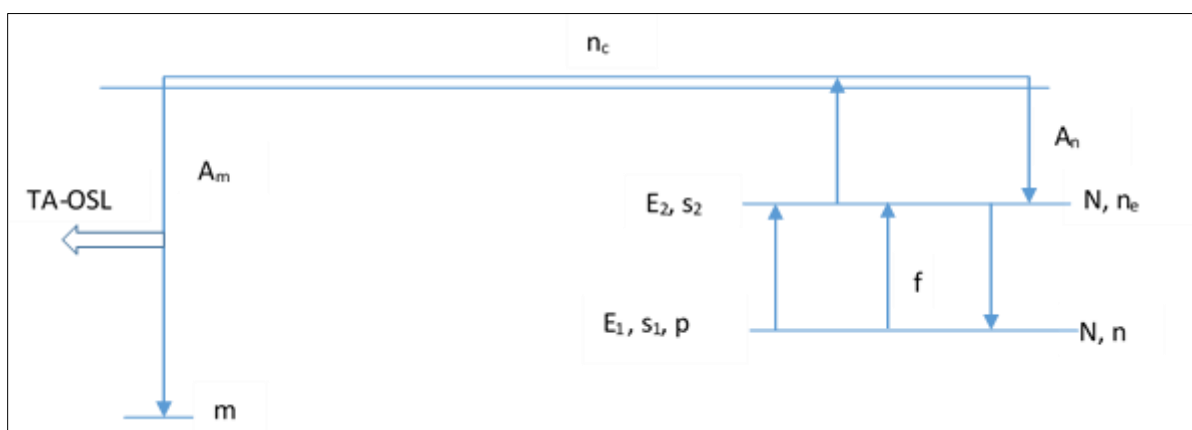


Figure 1 Model - Energy level diagram of TA-OSL. Here the first stage of electron elevation is thermal and optical and the second stage is thermal

Luminescence centre with a probability coefficient of A_m ($\text{cm}^3 \text{s}^{-1}$). It is this recombination process of electrons and hole that produces TA-OSL photons with instantaneous intensity I . At the end of excitation, the number of trapped holes (m_o) must be equal to the total number of trapped electrons ($n_o + n_{eo}$). Since the excitation is always carried out at a relatively low temperature, n_{eo} will therefore be relatively very small compared to n_o . As a result of this, the number of trapped holes approximately equals the number of trapped electrons. i.e. ($m_o \approx n_o$). According to Halperin and Braner [18], applying the detailed-balance principle and neglecting electronic degeneracies to this model, for the first transition state, the sum of the values of frequency factor, s_1 and stimulating light intensity, f must be equal to the retrapping probability, p , that is $s_1 + f = p$.

A set of differential equations governing the process during heating stage:

$$\frac{dn}{dt} = -s_1 n \exp(-E_1 / kT) - fn + pn_e \quad (1)$$

$$\frac{dn_e}{dt} = A_n (N - n - n_e) n_c + s_1 n \exp(-E_1 / kT) - s_2 n_e \exp(-E_2 / kT) + fn - pn_e \quad (2)$$

$$I = -\frac{dm}{dt} = A_m n n_c \quad (3)$$

$$\frac{dn_c}{dt} = \frac{dm}{dt} - \frac{dn}{dt} - \frac{dn_e}{dt} \quad (4)$$

3. Analytical considerations

From eq. (2), we have

$$\frac{dn_e}{dt} + (p + s_2 \exp(-E_2 / kT)) n_e = A_n (N - n - n_e) n_c + s_1 n \exp(-E_1 / kT) + fn \quad (5)$$

Since the excited states relax quite rapidly at the time scales of TL experiments, we therefore apply this approximation:

$\frac{dn_e}{dt} \ll (p + s_2 \exp(-E_2 / kT)) n_e$ and using quasi-equilibrium assumption: $(N - n) \gg n_e$. Applying these conditions to eq. (5), we have

$$n_e = \frac{A_n (N - n) n_c + s_1 n \exp(-E_1 / kT) + fn}{p + s_2 \exp(-E_2 / kT)} \quad (6)$$

Putting eq. (6) into (1)

$$\frac{dn}{dt} = -s_1 n \exp(E_1 / kT) - fn + p \left[\frac{A_n (N - n) n_c + s_1 n \exp(-E_1 / kT) + fn}{p + s_2 \exp(-E_2 / kT)} \right] \quad (7)$$

$$\frac{dn}{dt} = -\frac{[s_1 \exp(-E_1 / kT) + f] s_2 n \exp(-E_2 / kT) + p A_n (N - n) n_c}{p + s_2 \exp(-E_2 / kT)} \quad (8)$$

$$\frac{dn}{dt} = -n s_{\text{eff}} \exp(-E_2 / kT) + A_{n, \text{eff}} (N - n) n_c \quad (9)$$

Where

$$s_{eff} = \frac{[s_1 \exp(-E_1 / kT) + f]s_2}{p + s_2 \exp(-E_2 / kT)} \quad (10)$$

and

$$A_{n,eff} = \frac{pA_n}{p + s_2 \exp(-E_2 / kT)} \quad (11)$$

In this case the exact behavior of the TA-OSL peak has to do with the relation between p and $s_2 \exp(-E_2 / kT)$ and also between f and $s_1 \exp(-E_1 / kT)$. Therefore, applying the following conditions to eqs. (10) and (11): condition 1: $p \gg s_2 \exp(-E_2 / kT)$ and $f \gg s_1 \exp(-E_1 / kT)$, condition 2: $p \ll s_2 \exp(-E_2 / kT)$ and $f \ll s_1 \exp(-E_1 / kT)$ and condition 3: $p \gg s_2 \exp(-E_2 / kT)$ and $f \ll s_1 \exp(-E_1 / kT)$. We have

3.1. Condition 1

From eq. (10), if $p \gg s_2 \exp(-E_2 / kT) \sim$ i.e. comparing the probability of electron moving from the excited state to the ground state and to the conduction band.

$$s_{eff} = \frac{[s_1 \exp(-E_1 / kT) + f]s_2}{p} \quad (12)$$

If $f \gg s_1 \exp(-E_1 / kT) \sim$ i.e. comparing the probability of optical excitation and thermal excitation to the excited state.

$$s_{eff} = \frac{fs_2}{p} \quad (13)$$

From eq. (11), if $p \gg s_2 \exp(-E_2 / kT)$

$$A_{n,eff} = A_n \quad (14)$$

Putting eqs. (13) and (14) into (9), we have

$$\frac{dn}{dt} = - \frac{fs_2}{p} n \exp(-E_2 / kT) + A_n (N - n)n_c \quad (15)$$

3.2. Condition 2

From eq. (10), if $p \ll s_2 \exp(-E_2 / kT)$

$$s_{eff} = \frac{[s_1 \exp(-E_1 / kT) + f]s_2}{s_2 \exp(-E_2 / kT)} \quad (16)$$

If $f \ll s_1 \exp(-E_1 / kT)$

$$s_{eff} = s_1 \exp[(E_2 - E_1) / kT] \quad (17)$$

From eq. (11), if $p \ll s_2 \exp(-E_2 / kT)$

$$A_{n,eff} = \frac{p}{s_2} \exp(E_2 / kT) A_n \quad (18)$$

Putting eqs. (17) and (18) into (9), we have

$$\frac{dn}{dt} = -s_1 n \exp(-E_1 / kT) + \frac{p}{s_2} \exp(E_2 / kT) A_n (N - n) \quad (19)$$

3.3. Condition 3

From eq. (10), if $p \gg s_2 \exp(-E_2 / kT)$, we obtain eq. (12)

From eq. (12), if $f \ll s_1 \exp(-E_1 / kT)$, we have

$$s_{eff} = \frac{s_2 s_1 \exp(-E_1 / kT)}{p} \quad (20)$$

From eq. (11), if $p \gg s_2 \exp(-E_2 / kT)$

$$A_{n,eff} = A_n \quad (21)$$

Putting eqs. (20) and (21) into (9), we have

$$\frac{dn}{dt} = -\frac{s_2 s_1}{p} n \exp[-(E_1 + E_2) / kT] + A_n (N - n) \quad (22)$$

4. Results and discussion

4.1. Analytical expressions

The analytical expression obtained based on the assumption of condition 1 (eq. (15)) reveals that the effective activation energy is that of thermal excitation from the excited state to the conduction band (E_2). The effective frequency factor, s_{eff} , is the ratio of the product of the stimulating light intensity and frequency factor in the excited state to the retrapping probability in the ground state of the trap as shown in eq. (15). At high stimulating light intensities, s_{eff} tends to the value of the frequency factor, s_2 , for thermal excitation out of the excited state. This means that s_{eff} is directly proportional to the stimulating light intensity. The implication of this is that kinetic analysis of glow curves obtained with different stimulation light intensities will produce similar activation energy but different frequency factors.

As shown in eq. (19) for condition 2, the effective activation energy is that of thermal excitation from the ground state to the excited state (E_1) and the effective frequency factor is equivalent to that of the ground state (s_1). Therefore, unlike what is obtained with condition 1, the two kinetic parameters are independent of value of stimulation light intensity as long as condition 2 is satisfied. $A_{n,eff}$ is dependent on the retrapping probability, frequency factor of the excited state and an exponential temperature term (as shown in eq. (19)). When similar condition ($s_2 \exp(-E_2/kT) \gg p$) was applied to a two-stage thermal stimulation of thermoluminescence (TL) of Chen et al [8], the resulted analytical expression yielded effective activation energy and effective frequency factor expressions obtained in this study.

The final analytical expression for condition 3 is shown in eq. (22). The effective activation energy, based on this condition, is approximately $E_1 + E_2$, while the effective frequency factor is approximately the ratio of the product of frequency factors for thermal excitation from the ground state and the excited state to the electron retrapping

probability of the ground state (i.e. $S_{eff} = \frac{s_1 s_2}{p}$). Since $s_1 + f = p$, when the stimulating light intensities becomes very small compared to s_1 , s_{eff} becomes approximately equal to s_2 . This case where $f \ll s_1$ is similar to the model of two-stage thermal stimulation of TL of Chen *et. al.* [8] for which the effective activation energy and effective frequency factor were also $E_1 + E_2$ and s_2 respectively.

4.2. Numerical results and discussion

The model in fig. 1 was solved numerically using eqs. (1) – (4) by appropriately chosen set of trapping parameters. The numerical simulation was performed using MATLAB ode 15s program. The heating rate used in all the simulation was 1Ks^{-1} .

In order to calculate symmetry factors, activation energies and frequency factors, different formulae used by Chen [19] were used in this work. Such formulae include:

4.2.1. For the symmetry factor, μ_g

$$\mu_g = \frac{\delta}{\omega} \quad (23)$$

Where $\delta = T_2 - T_m$ and $\omega = T_2 - T_1$; T_1 is the low temperature of half intensity, T_2 is the high temperature of half intensity and T_m is the temperature at maximum intensity. Note that for a symmetry factor $\mu_g \approx 0.42$; the glow peak is a typical first-order peak while for a symmetry factor $\mu_g \approx 0.52$; the glow peak is a typical second-order peak.

4.2.2. For effective activation energy, E_{eff} , for first-order peak

$$E_{eff} = kT_m \left(2.52 \frac{T_m}{\omega} - 2 \right) \quad (24)$$

4.2.3. For effective activation energy, E_{eff} , for second-order peak:

$$E_{eff} = kT_m \left(3.54 \frac{T_m}{\omega} - 2 \right) \quad (25)$$

4.2.4. For frequency factor, s :

$$s = \frac{\beta E}{kT_m^2} \exp \left(\frac{E}{kT_m^2} \right) \left[1 + (b-1) \frac{2kT_m}{E} \right]^{-1} \quad (26)$$

Where E is the activation energy and b is order of kinetics. Other parameters have their usual meaning as earlier stated above.

4.3. Application to first-order kinetic glow peaks

The set of parameters used in fig. 2 are: $E_1 = 0.6\text{eV}$, $E_2 = 0.4\text{eV}$, $s_1 = 10^{12}\text{s}^{-1}$, $p = s_1 + f$, $s_2 = 10^{13}\text{s}^{-1}$, $A_n = 10^{-12}\text{cm}^3\text{s}^{-1}$, $A_m = 10^{-7}\text{cm}^3\text{s}^{-1}$, $N = 1.1 \times 10^{10}\text{cm}^{-3}$, $n_0 = m_0 = 10^{10}\text{cm}^{-3}$, $n_{e0} = 0$, $n_c = 0$ and $f = 10^5, 10^7, 10^9$ and 10^{11}s^{-1} . In this case the recombination probability coefficient was higher than the retrapping probability coefficient by 5 orders of magnitude and the parameters.

Were chosen so that condition 1 is satisfied. In fig. 2, curves (a), (b), (c) and (d) represent glow curves obtained with $f = 10^5, 10^7, 10^9$ and 10^{11}s^{-1} respectively. The peak symmetry for all the peaks resulting from the different stimulation light intensities was a typical first-order TA-OSL peak with $\mu_g \approx 0.42$. The corresponding effective activation energy, E_{eff} for the peaks was approximately 0.40eV (the energy of the excited state, E_2) and the effective frequency factors, s_{eff} were $1.0 \times 10^6, 1.0 \times 10^8, 1.0 \times 10^{10}$ and $8.3 \times 10^{11}\text{s}^{-1}$ for curves (a), (b), (c) and (d) respectively. This again showed proportionality between stimulating light intensity and effective frequency factor. The effective retrapping probability

coefficient, $A_{n,eff}$ was approximately A_n . The above results for E_{eff} , s_{eff} and $A_{n,eff}$ were in good agreement with what is expected from the analytical expression in eq. (15). The proportionality between stimulating light intensity and effective frequency factor can be attributed to the fact that increase in stimulating light intensity increases the number of electrons transferred from the ground state to the excited state. Consequently, the number of electrons raised per second from the excited state to the conduction band increases with light intensity. As shown in fig. 2, the peak position of the glow peak shifts toward the lower temperature region as f increases. This is expected because the increase in f makes more electrons available in the excited state and therefore decreases the retrapping probability. Applying similar conditions ($p \gg f + s_2 \exp(-E_2 / kT)$ and $f \gg s_2 \exp(-E_2 / kT)$) to the models of Chen and Pagonis [11] – where the first stage is thermal and the second stage is both thermal and optical stimulation by applying similar conditions showed that the effective activation energy was that of the ground state. s_{eff} , in this case, was equivalent to the stimulating light intensity. The effective retrapping probability coefficient is approximately A_n . The implication of this comparison is that the activation energy values obtained from glow curves of materials with characteristics similar to the ones in this study are dependent on whether the light stimulation is only able to move electrons from the ground state to the excited state or from the excited state to the conduction band.

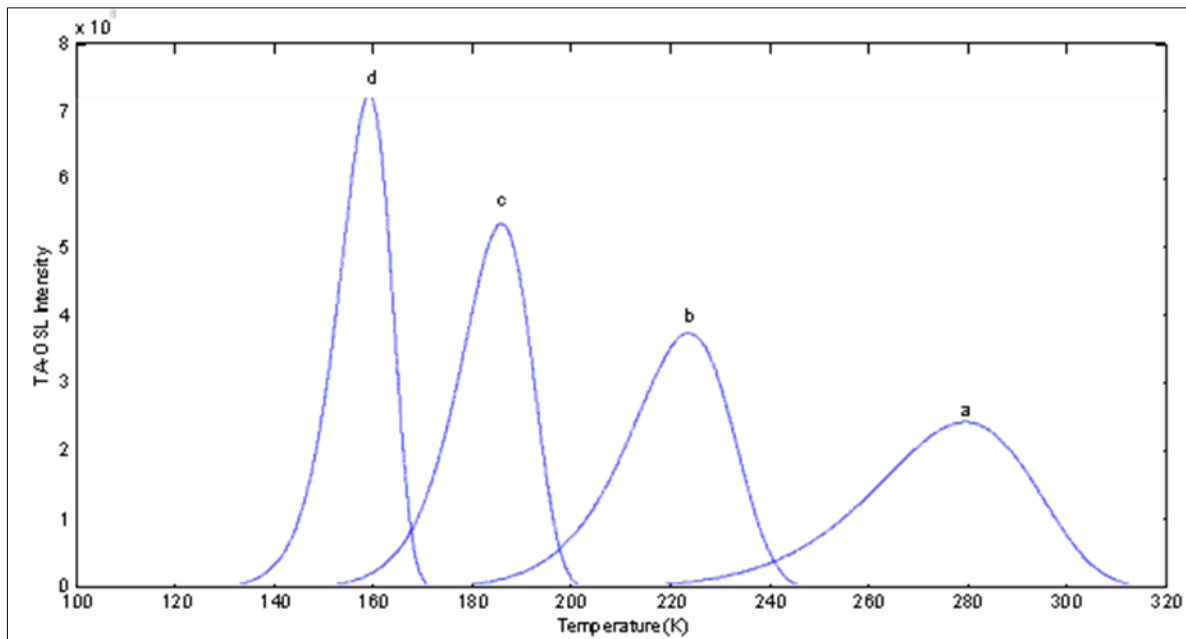


Figure 2 Simulated results of TA-OSL peaks using eqs. (1) – (4) for different values of f . The parameters used were: $E_1 = 0.6\text{eV}$, $E_2 = 0.4\text{eV}$, $s_1 = 10^{12}\text{ s}^{-1}$, $p = s_1 + f$, $s_2 = 10^{13}\text{ s}^{-1}$, $A_n = 10^{-12}\text{ cm}^3\text{ s}^{-1}$, $A_m = 10^{-7}\text{ cm}^3\text{ s}^{-1}$, $N = 1.1 \times 10^{10}\text{ cm}^{-3}$, $n_0 = m_0 = 10^{10}\text{ cm}^{-3}$, $n_{e0} = 0$, $n_c = 0$ and $f = 10^5, 10^7, 10^9$ and 10^{11} s^{-1}

In fig. 3, the same set of trapping parameters as in fig. 2 was used except $s_1 = 10^6\text{ s}^{-1}$, $s_2 = 10^{14}\text{ s}^{-1}$ and $f = 10^{-6}, 10^{-7}$ and 10^{-8} s^{-1} for condition 2 to be achieved. Only glow curve obtained with $f = 10^{-6}\text{ s}^{-1}$ is shown in fig. 3 for clarity purpose. The symmetry factor, μ_g for the glow peaks generated with the different stimulation light intensities was estimated to be approximately 0.43. The corresponding effective activation energy is approximately 0.60eV, the value for thermal excitation from ground state to the excited state. The effective frequency factor is approximately $s_1 = 1.0 \times 10^6\text{ s}^{-1}$. The effective retrapping probability coefficient exhibited an exponential dependence on temperature. These values of effective activation energy and frequency factor were in good agreement with what is expected from the analytical expressions in eq. (19). With this condition 2 for which $f \ll s_1 \exp(-E_1 / kT)$, the present model becomes almost equivalent to the two-stage thermal stimulation model of Chen *et al.* [8]. It is therefore not surprising that eq. (19) is similar to the eq. (15) of the model of Chen *et al.* [8]. Hence the expressions for effective activation energy for condition 2 of this study and that of the model of Chen *et al.* [8] are the same. The same is true for the effective frequency factor. It should also be noted, in eq. (19) of this study, that effective retrapping coefficient is temperature dependent. As temperature increases during readout the retrapping coefficient decreases. The explanation for this is that as temperature increases electrons in the excited state traps increase since thermal excitation from the ground state to the excited state is more pronounced than optical excitation and also because retrapping into the ground state is very small.

Another set of simulation is shown in fig. 4 which uses the same set of parameters as in fig. 2 except $s_1 = 10^{13} \text{ s}^{-1}$, $s_2 = 10^9 \text{ s}^{-1}$ and $f = 10^1, 10^2, 10^3$ and 0 s^{-1} for condition 3 to be.

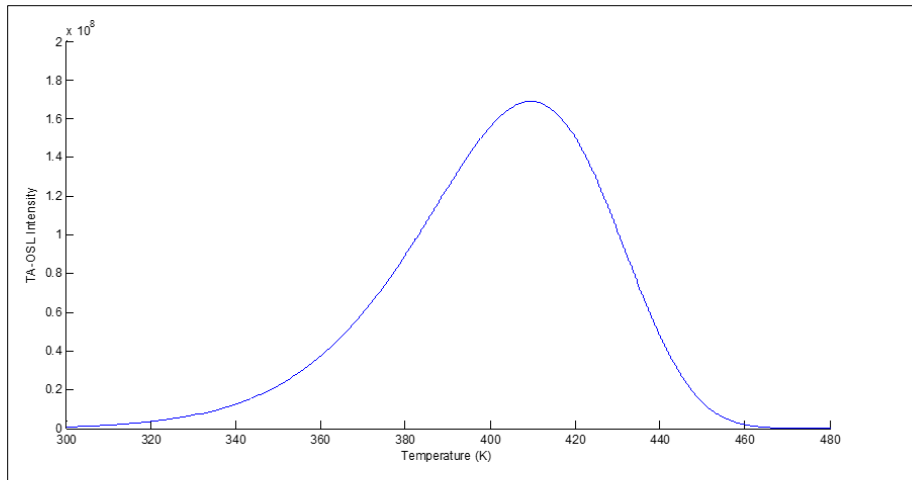


Figure 3 Simulated results of a TA-OSL peak using eqs. (1) – (4) for different values of f . The same set of parameters was used as in fig.2 except $s_1 = 10^6 \text{ s}^{-1}$, $s_2 = 10^{14} \text{ s}^{-1}$ and $f = 10^{-6}, 10^{-7}$ and 10^{-8} s^{-1} . Note that only $f = 10^{-6} \text{ s}^{-1}$ is shown in the figure for clarity purpose

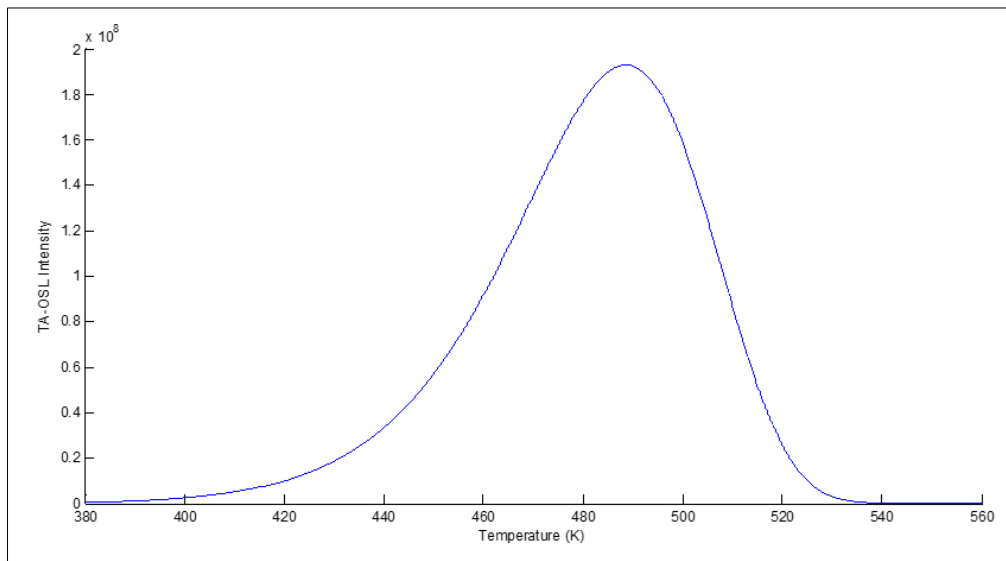


Figure 4 Simulated results of a TA-OSL peak using eqs. (1) – (4) for different values of f . The same set of parameters was used as in fig.2 except $s_1 = 10^{13} \text{ s}^{-1}$, $s_2 = 10^9 \text{ s}^{-1}$ and $f = 10^1, 10^2, 10^3$ and 0 s^{-1} . Note that only $f = 10^1 \text{ s}^{-1}$ is shown on the graph for clarity purpose

Achieved. The glow curves obtained for all the stimulating light intensities are similar therefore for clarity purpose only the one for $f = 10^1 \text{ s}^{-1}$ is shown in fig. 4. The peak obtained for all the stimulating light intensities was a typical first-order TA-OSL peak with $\mu_g \approx 0.42$. For all values of f , the effective activation energy is approximately 1.0eV, sum of the activation energy values for thermal excitation out of the ground state and the excited state. On the other hand, the effective frequency factor depends on f in a form which is in agreement with the effective frequency factor expression

$\frac{s_1 s_2}{p}$ in eq. (22). The values of the effective activation energy were also in good agreement with corresponding values expected from the analytical expressions in eq. (22). The effective retrapping probability coefficient was approximately

A_n . At low stimulating light intensities (e.g. $f = 10^1 \text{ s}^{-1}$ and 0 s^{-1}), the estimated effective frequency factor approached s_2 ($1.0 \times 10^9 \text{ s}^{-1}$). This indicates a perfect agreement with the analytical expression because when f is small, then from $p = s_1 + f$, it implies that $p = s_1$. Hence the expression $\frac{s_1 s_2}{p} = s_2$. Even when the stimulating light intensity was switched off ($f = 0 \text{ s}^{-1}$), s_{eff} was still tending to s_2 . Therefore in this limit of low stimulating light intensity, E_{eff} and s_{eff} expressions for the present model are the same with those reported by Chen *et. al.* [8] for their two-stage thermal stimulation model.

4.4. Application to second-order kinetic glow peaks

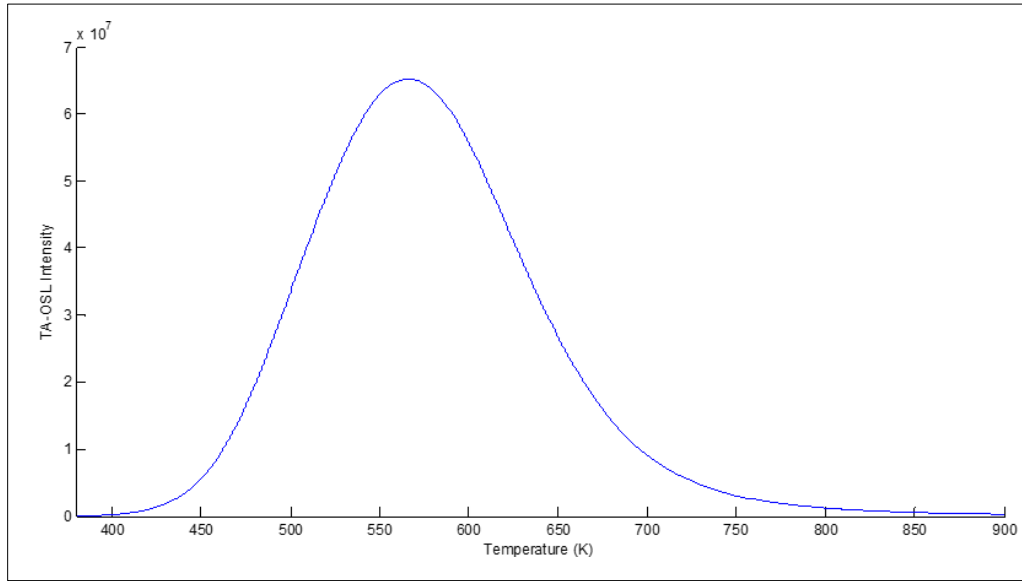


Figure 5 Simulated results of a TA-OSL peak using eqs. (1) – (4) for different values of f . The same set of parameters was used as in fig.3 except that $A_n = 10^{-3} \text{ cm}^3\text{s}^{-1}$ was significantly higher than $A_m = 10^{-10} \text{ cm}^3\text{s}^{-1}$. Note that only $f = 10^{-6} \text{ s}^{-1}$ is shown in the figure for clarity purpose

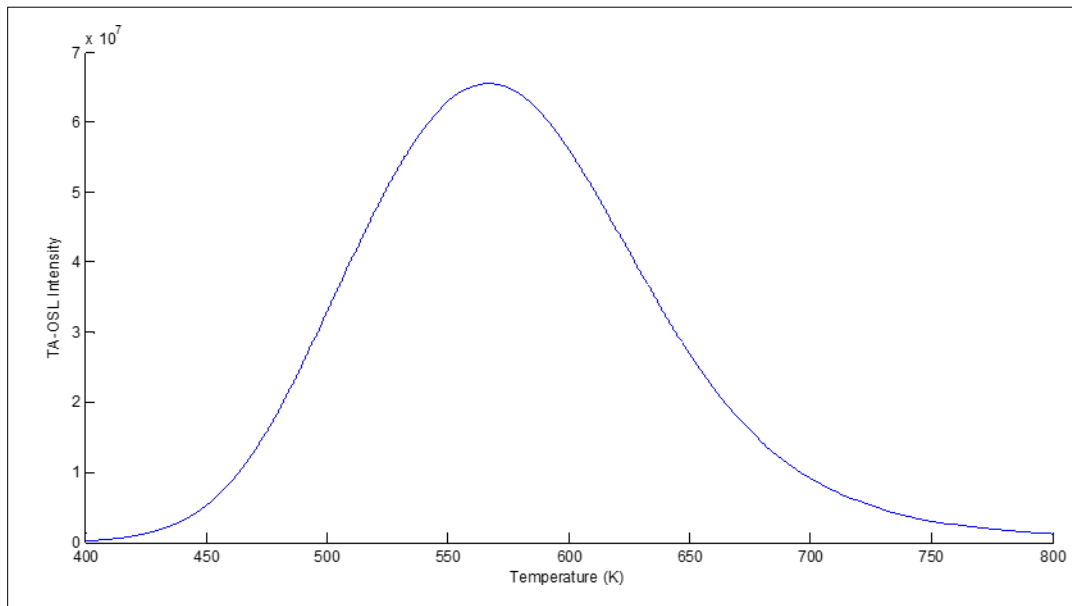


Figure 6 Simulated results of a TA-OSL peak using eqs. (1) – (4) for different values of f . The same set of parameters was used as in fig.4 except that $A_n = 10^{-8} \text{ cm}^3\text{s}^{-1}$ was higher than $A_m = 10^{-10} \text{ cm}^3\text{s}^{-1}$. Note that only $f = 10^1 \text{ s}^{-1}$ is shown in the figure for clarity purpose

Table 1 A summary of the simulated and analytical results for activation energies and frequency factors for the first and second order kinetics

Order of Peaks	Assumption	Stimulating light intensities, $f(s^{-1})$	Activation Energy, E(eV)			Frequency Factor, s(s^{-1})		
			Analytical results	Simulation results	%difference	Analytical results	Simulation results	% difference
First	Condition1: $p \gg s_2 \exp(-E_2/kT)$ $f \gg s_1 \exp(-E_1/kT)$	10^5	0.40	0.40	0	10^6	1.0×10^6	0
		10^7	0.40	0.40	0	10^8	1.0×10^8	0
		10^9	0.40	0.40	0	10^9	1.0×10^{10}	0
		10^{11}	0.40	0.40	0	10^{11}	8.3×10^{11}	730
	Condition2: $p \ll s_2 \exp(-E_2/kT)$ $f \ll s_1 \exp(-E_1/kT)$	10^{-6}	0.60	0.60	0	10^6	1.0×10^6	0
		10^{-7}	0.60	0.60	0	10^6	1.0×10^6	0
		10^{-8}	0.60	0.60	0	10^6	1.0×10^6	0
	Condition3: $p \gg s_2 \exp(-E_2/kT)$ $f \ll s_1 \exp(-E_1/kT)$	10^1	1.0	1.0	0	10^9	1.0×10^9	0
		10^2	1.0	1.0	0	10^9	1.0×10^9	0
		10^3	1.0	1.0	0	10^9	1.0×10^9	0
		0	1.0	1.0	0	10^9	1.0×10^9	0
Second	Condition2: $p \ll s_2 \exp(-E_2/kT)$ $f \ll s_1 \exp(-E_1/kT)$	10^{-6}	0.60	0.60	0	10^6	0.4×10^4	99.6
		10^{-7}	0.60	0.60	0	10^6	0.4×10^4	99.6
		10^{-8}	0.60	0.60	0	10^6	0.4×10^4	99.6
	Condition3: $p \gg s_2 \exp(-E_2/kT)$ $f \ll s_1 \exp(-E_1/kT)$	10^1	1.0	0.61	39	10^9	0.5×10^4	100.0
		10^2	1.0	0.60	40	10^9	0.4×10^4	100.0
		10^3	1.0	0.61	39	10^9	0.5×10^4	100.0
		0	1.0	0.61	39	10^9	0.5×10^4	100.0

Fig. 5 depicts the glow curve obtained using the same set of parameters (condition 2) as in fig. 3 except that the retrapping probability coefficient was significantly higher than the recombination probability coefficient (i.e. $A_n = 10^{-3} \text{ cm}^3\text{s}^{-1}$ and $A_m = 10^{-10} \text{ cm}^3\text{s}^{-1}$) to achieve second-order kinetic behaviour. Similar glow curves were produced for all the stimulating light intensities but for clarity purpose only the one for $f = 10^{-6} \text{ s}^{-1}$ is shown in fig. 5. The peak obtained was a typical second-order peak with $\mu_g \approx 0.52$. The effective activation energy was the same as the activation energy for thermal excitation from the ground state to the excited state (0.60 eV) as expected from eq. (19) for condition 2. The effective frequency factor value ($0.4 \times 10^4 \text{ s}^{-1}$) was however smaller than s_1 ($1.0 \times 10^6 \text{ s}^{-1}$) that is expected based on eq. (19).

The reason for the short fall in effective frequency factor is not far-fetched. At low stimulating light intensities, the glow peak shifted to a high temperature region. According to Sunta et al [20], on high temperature side of the glow peak, the quasi-equilibrium condition becomes less satisfied and this has adverse effect on the accuracy of the method used to analyze the glow peak as well the kinetic parameters measured. Moreover, the retrapping effect is strongly dependent on the density of empty traps. The retrapping effect increases with density of empty traps [21]. Therefore, when there is high retrapping, there is likelihood that most methods used to analyze the kinetic parameters will produce inaccurate results [22]. This is why the effective frequency factor was inaccurate.

Fig. 6 shows glow curve which uses the same set of parameters as in fig. 4 except that the retrapping probability coefficient is higher than the recombination probability coefficient by 2 orders of magnitude (i.e. $A_n = 10^{-8} \text{ cm}^3\text{s}^{-1}$ and $A_m = 10^{-10} \text{ cm}^3\text{s}^{-1}$). For clarity purpose only $f = 10^1 \text{ s}^{-1}$ was shown in fig. 6. The symmetry factor for $f = 10^1, 10^3$ and 0 s^{-1} was approximately 0.52 while the symmetry factor for $f = 10^2 \text{ s}^{-1}$ was approximately 0.53. The kinetic parameters for $f = 10^1, 10^3$ and 0 s^{-1} were $E_{\text{eff}} \approx 0.61 \text{ eV}$ and $s_{\text{eff}} \approx 0.5 \times 10^4 \text{ s}^{-1}$. Other kinetic parameters for $f = 10^2 \text{ s}^{-1}$ were $E_{\text{eff}} \approx 0.60 \text{ eV}$ and $s_{\text{eff}} \approx 0.4 \times 10^4 \text{ s}^{-1}$. One had expected to obtain the same kinetic parameters as in fig. 4 but the obtained kinetic parameters fell below the expected results. The reason for the short fall in kinetic parameters could be as reported by Braunlich [23] and Sunta et al [22]: in a one-trap one-recombination centre model (OTOR), if retrapping is strong and if the traps are filled to saturation, the methods used such as peak shape methods as well as the initial-rise method and glow-peak fitting will produce very low effective activation energy (although our retrapping was not strong). Even though these authors did not evaluate the effective frequency factor but there is likelihood that the frequency factor will yield low value. Therefore, this could be responsible for the short fall in kinetic parameters.

The simulated and analytical values for both activation energies and frequency factors for the first and second order kinetics under different assumed conditions are summarized in Table 1.

5. Conclusion

A TA-OSL model in which the first stage is both thermal and optical stimulation and the second stage is thermal stimulation has been studied. Three assumed conditions were applied to the model. The analytical expressions and numerical simulation results were found to be in good agreement in almost all cases. In few cases where the analytical expressions failed to agree with the numerical results especially in second-order peaks due to high retrapping effect, explanation has been offered.

Compliance with ethical standards

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