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Simulations of isothermal processes in the semilocalized transition (SLT) model of thermoluminescence (TL)

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Abstract

Semilocalized transition (SLT) kinetic models for thermoluminescence (TL) contain characteristics of both a localized transition (LT) and of a single trap model. TL glow curves within SLT models typically contain two TL peaks; the first peak corresponds to the intra-pair luminescence due to LTs and the second TL peak corresponds to delocalized transitions involving the conduction band (CB). The latter delocalized TL peak has also been found to exhibit non-typical double-peak structure, in which the main TL peak is accompanied by a smaller peak called the displacement peak. This paper describes the simulation of isothermal luminescence signals using a previously published SLT model. It is found that these simulated isothermal signals exhibit several unusual time characteristics. Isothermal signals associated with the LTs follow first order kinetics and are therefore described by single decaying exponentials. However, isothermal signals associated with delocalized transitions show a non-typical complex structure characterized by several time regions with different decay characteristics. For certain values of the parameters in the SLT model the isothermal signals can also exhibit non-monotonic behaviour as a function of time. Another notable result from the simulations is that isothermal currents (which are proportional to the concentration of electrons in the CB) can persist for very long periods of time, even after the apparent termination of the isothermal luminescence signals. It is concluded that isothermal processes described by the SLT model depend strongly on the presence of SLTs, in contrast to previous studies using Monte Carlo simulations, which showed a weak interdependence of these phenomena. The simulations in this paper suggest that isothermal experiments offer a sensitive method for detecting the presence of SLTs in a dosimetric material.

1. Introduction

Theoretical models of thermoluminescence (TL) are usually based on the assumption of a uniform spatial distribution of traps and recombination centres (RCs), resulting in models known as the single trap model (STM). However, several alternative models of TL are based on the existence of electron-hole (e-h) pairs trapped close to each other, and are known as localized transition (LT) models ([1] and references therein). A third type of model has been proposed in the literature

which is based on the spatial correlation of traps (T) and RCs, and is known as semilocalized transition (SLT) kinetic model. Calculations within these models were carried out initially by using Monte Carlo techniques to simulate thermally stimulated relaxation processes in polycrystalline, one-dimensional and two-dimensional solids [2–9]. The SLT model is also believed to be applicable to physical situations where the excitation is carried out by high-energy radiation, so that traps and RCs are populated along tracks.

Monte Carlo based calculations within the SLT model showed the existence of unusual TL glow curves, in which

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the main TL peak is accompanied by a smaller peak called the displacement peak. In some of these earlier works simulations were carried out with various absolute numbers of carriers in single separate groups and for both isothermal and non-isothermal conditions [2–9]. Phosphorescence decay and thermally stimulated currents (TSCs) were simulated near the TL peak maximum for various parameters in the model [4]. In the case of no retrapping, significant changes were found only for smaller clusters of charge carriers, at high temperatures and for large times. No significant changes were found in simulated phosphorescence decay for large clusters. These simulations were also carried out for isothermal current decay (ICD), which is very sensitive to spatially correlated effects. These earlier published works concluded that TSC/TL phenomena are more sensitive than isothermal experiments to clustering effects.

In a series of later studies Mandowski [6–9] examined spatially correlated systems of traps and RCs, which are thought to be responsible for many anomalous phenomena associated with trap cluster systems. In these studies a completely different approach was used to develop a complex SLT kinetic model for spatially correlated TL systems, which contains elements from both the LT and the STM models. The model can explain unusual phenomena observed in solid state detectors; for example, the SLT model can explain the occurrence of very high frequency factors associated with TL glow curves, by using the mechanism of cascade detrapping (CD).

The complexity of the Mandowski SLT model is partly due to the presence of two distinct activation energies E , the energy gap between the trapped state and the excited state, and E_V , the energy gap between the excited state and the conduction band (CB). The nature of this model is unique since it is based on enumeration of electron and hole states, instead of using global concentrations of electrons and holes, as is commonly done in the LT and STM models. Mandowski [7] recently demonstrated that the SLT model is a natural generalization of the STM and the LT models, and presented kinetic equations for the most general case of SLT.

There are two possible recombination transitions within the model, leading to two TL peaks denoted in the rest of this paper by L_B and L_C . The L_B peak corresponds to the intra-pair luminescence due to LTs and the L_C peak corresponds to delocalized transitions involving the CB. Mandowski [9] found that for certain parameters in the model, the L_C peak can exhibit a non-typical double-peak structure that resembles the TL displacement peaks previously found in spatially correlated systems by Monte Carlo simulations [2–5]. This double-peak structure of the L_C peaks was interpreted as follows: the first L_C peak corresponds to an increasing concentration of charges accumulating in the excited local level and undergoing intra-pair recombination. The second L_C peak was interpreted as relating to carriers wandering in the crystal, and undergoing recombination with holes in the RC via the CB.

Some properties of SLT were analysed also by Pagonis [10] and Kumar *et al* [11]. A similar model for dose dependence studies was also constructed by Horowitz *et al* [12]. Kumar *et al* [13] compared several different models dealing with localized and SLTs, namely the Chen–Halperin,

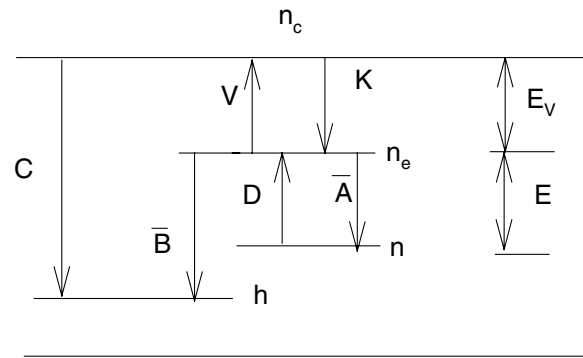


Figure 1. The SLT model developed by Mandowski [9]. The various parameters and transitions in this model are explained in the text.

Mandowski and Braunlich–Scharmann models. These authors studied the dose dependence of the glow peak temperature T_m under conditions of strong retrapping. They found that under conditions of low retrapping the TL glow curves from these localized models were similar. However, differences were found when high retrapping was present in the models. Sunta *et al* [14] developed a simple model of TL termed the localized recombination model (LRM). These authors derived analytical expressions for this model and compared them with the numerical solutions of the equations in the model. A different approach to SLT models was used by Piters and Bos [15] who incorporated defect reactions in the rate equations of the kinetic model.

The above studies have focused mostly on the properties of TL glow curves, while much less attention has been given in the literature to the properties of isothermal signals calculated within the SLT model. The purpose of this paper is to perform a detailed study of isothermal signals using the most recently published version of the SLT model [6, 9]. To the best of our knowledge, there have been no other published simulations of isothermal processes using this SLT model. It is found that isothermal signals associated with LTs in the SLT model follow first order kinetics, and therefore can be described by single decaying exponentials. However, isothermal signals associated with delocalized transitions show a non-typical complex structure characterized by several regions with different decay characteristics. The simulation results also show that isothermal currents (which are proportional to the concentration of electrons in the CB) can persist for very long periods of time, even after the apparent termination of the isothermal luminescence signals.

2. The SLT model

In this section we summarize the main assumptions and kinetic equations of the SLT model [6, 9]. The energy diagram and possible electronic transitions for the SLT model are shown in figure 1. Since some of the transitions shown in figure 1 depend on the instantaneous number of carriers occupying traps (T) and RCs, it is not possible to write kinetic equations

using global concentrations of charge carriers in each level. In order to overcome this difficulty, Mandowski [6] introduced the following notation to denote the occupation of all states in a single trap–recombination centre (T–RC) unit,

$$\begin{Bmatrix} \bar{n}_c \\ \bar{n} \\ \bar{h} \end{Bmatrix}, \quad (1)$$

where \bar{n}_c is the number of electrons in the local excited level, \bar{n} is the number of electrons in the trap level T and \bar{h} is the number of holes in the RC.

By using this notation, the following concentrations of states can be defined

$$\begin{aligned} H_0^0 &= \begin{Bmatrix} 0 \\ 0 \\ 1 \end{Bmatrix} & H_1^0 &= \begin{Bmatrix} 0 \\ 1 \\ 1 \end{Bmatrix} & H_0^1 &= \begin{Bmatrix} 1 \\ 0 \\ 1 \end{Bmatrix} & H_1^1 &= \begin{Bmatrix} 1 \\ 1 \\ 1 \end{Bmatrix}, \\ E_0^0 &= \begin{Bmatrix} 0 \\ 0 \\ 0 \end{Bmatrix} & E_1^0 &= \begin{Bmatrix} 0 \\ 1 \\ 0 \end{Bmatrix} & E_0^1 &= \begin{Bmatrix} 1 \\ 0 \\ 0 \end{Bmatrix} & E_1^1 &= \begin{Bmatrix} 1 \\ 1 \\ 0 \end{Bmatrix}, \end{aligned} \quad (2)$$

where $H_m^n(t)$, $E_m^n(t)$ are the concentrations of states with full and empty RCs, respectively. It is assumed that within this model creation of states with two active electrons (H_1^1 and E_1^1) is unlikely. As a result of this assumption, the model does not contain differential equations for H_1^1 and E_1^1 .

The bar variables \bar{A} and \bar{B} (s^{-1}) in figure 1 represent the localized trapping and recombination probability densities for LTs. The corresponding delocalized trapping and recombination probability densities for delocalized transitions are represented by K and C (in $\text{cm}^3 \text{s}^{-1}$). The retrapping coefficient r is defined as $r = \bar{A}/\bar{B}$ and the global concentration of traps in the crystal is denoted by N (cm^{-3}).

In figure 1 the activation energy within the T–RC pair is denoted by E , the energy barrier is E_v and the corresponding frequency factors are ν and ν_v . The parameters $D = \nu \exp(-E/kT(t))$ and $V(t) = \nu_v \exp(-E_v/kT(t))$ describe the thermal excitation probabilities in the SLT model; $D(t)$ represents the thermal excitation probability from the ground state to the excited state of the e–h pair, while $V(t)$ represents the thermal excitation probability from the excited state into the CB. Note that in this paper, we study isothermal processes, hence V and D are functions of time.

In later work Mandowski [8] pointed out that the states H_m^n and E_m^n are very likely physically different from each other. Specifically, H_m^n states represent an electron with a neighbouring hole, while E_m^n are states in which the neighbouring hole has already recombined with an electron from the CB. It is then reasonable to assume that the thermal activation coefficients $D = \nu \exp(-E/kT)$ and $V = \nu_v \exp(-E_v/kT)$ for these two types of electronic states might have different values. Consequently, the states H_m^n in this later version of the SLT model are assigned the following thermal activation coefficients:

$$D_1 = \nu_1 \exp(-E_1/kT), \quad (3)$$

$$V_1 = \nu_{V_1} \exp(-E_{V_1}/kT) \quad (4)$$

and the states E_m^n are assigned the coefficients

$$D_2 = \nu_2 \exp(-E_2/kT), \quad (5)$$

$$V_2 = \nu_{V_2} \exp(-E_{V_2}/kT). \quad (6)$$

From a physical point of view, one may expect that an electron in the state E_m^n will be less bound to the trap than an electron in a state H_m^n , since this latter state is closer to a hole. Therefore one can expect that the thermal activation energies in the model would satisfy the inequalities $E_2 \leq E_1$ and $E_{V_2} \leq E_{V_1}$. Mandowski [8] pointed out that these inequalities lead to the phenomenon of CD, which is one of the important results of the SLT model. This phenomenon can be described as a chain of detrapping events caused by the lowering of the thermal activation energy for an electron weakly bound to the trap. This chain of detrapping events during heating of the sample can lead to very narrow TL peaks shifted towards lower temperatures.

By using the variables $H_m^n(t)$, $E_m^n(t)$ defined above, Mandowski developed the following set of equations:

$$\dot{H}_1^0 = -(D + Cn_c)H_1^0 + \bar{A}H_1^0, \quad (7)$$

$$\dot{H}_0^1 = DH_1^0 - (\bar{A} + \bar{B} + V_1 + Cn_c)H_0^1 + Kn_cH_0^0, \quad (8)$$

$$\dot{H}_0^0 = V_1H_0^1 - (K + C)n_cH_0^0, \quad (9)$$

$$\dot{E}_1^0 = Cn_cH_1^0 - D_2E_1^0 + \bar{A}E_1^0, \quad (10)$$

$$\dot{E}_0^1 = Cn_cH_0^1 + D_2E_1^0 - (\bar{A} + V_2)E_0^1 + Kn_cE_0^0, \quad (11)$$

$$\dot{E}_0^0 = \bar{B}H_0^1 + Cn_cH_0^0 + V_2E_0^1 - Kn_cE_0^0, \quad (12)$$

$$\begin{aligned} \dot{n}_c &= -Cn_c(H_1^0 + H_0^1 + H_0^0) + V_1H_0^1 + V_2E_0^1 \\ &\quad - Kn_c(H_0^0 + E_0^0). \end{aligned} \quad (13)$$

The variables $H_m^n(t)$, $E_m^n(t)$ are related to the standard global variables used in the LT model and the single trap model (STM) of TL by the following equations:

$$n = H_1^0 + E_1^0, \quad (14)$$

$$n_c = H_0^1 + E_0^1, \quad (15)$$

$$n_c = H_0^0 - E_0^1 - E_1^0, \quad (16)$$

$$h = H_1^0 + H_0^1 + H_0^0. \quad (17)$$

Here n , n_e , n_c and h denote as usual the concentrations of carriers in the trap level, excited level, CB and RC, correspondingly. The system of seven differential equations (3)–(13) was solved by using the differential equation solver in the software package *Mathematica*. The typical CPU time for solving this system of equations on a desktop computer and for producing graphs of $H_m^n(t)$, $E_m^n(t)$, $n(t)$, $n_e(t)$, $n_c(t)$, $h(t)$ was less than 1 min. A linear heating rate β is assumed during simulations of the TL process within this model so that the temperature varies with time t according to $T = T_o + \beta t$, while the heating rate β is equal to zero during simulations of isothermal processes.

The two recombination transitions within the model lead to two possible isothermal luminescence signals L_B and L_C with their corresponding intensities given by

$$L_B = \bar{B}H_0^1, \quad (18)$$

$$L_C = Cn_c h = Cn_c(H_1^0 + H_0^1 + H_0^0). \quad (19)$$

The numerical values of the parameters used in this paper are $\nu_1 = \nu_2 = \nu_{v1} = \nu_{v2} = 10^{10} \text{ s}^{-1}$, $\beta = 1 \text{ K s}^{-1}$, $C = 10^{-10} \text{ cm}^3 \text{ s}^{-1}$, $\bar{B} = 10^3 \text{ s}^{-1}$, $K = 0$ and the retrapping ratio $r = \bar{A}/\bar{B} = 0$. The thermal activation energies E_1 , E_2 , E_{V_1} , E_{V_2} appearing in equations (3)–(13) are varied within the simulations, in order to demonstrate the various possible behaviours of the isothermal signals. The initial concentration of trapped carriers is assumed to be $H_1^0(0) = n_0$ and all the other initial concentrations $H_m^n(0)$, $E_m^n(0)$ are assumed to be zero. Furthermore, it is assumed that all states are initially full so that $n_0 = N$, where $N = 10^{17} \text{ cm}^{-3}$ is the global concentration of traps in the crystal. In the next sections we present several simulations of isothermal experiments within the SLT model by varying the numerical values of the activation energies E_1 , E_2 , E_{V_1} , E_{V_2} . Due to the complexity of this model, one might expect unusual and different results from those obtained using the widely studied STM and LT models. Three such simulated examples producing unusual physical results are given in the next section.

3. Results and discussion

Simulation #1: Isothermal signals when $E_2 = E_1 = 0.9 \text{ eV}$ and $E_{V_2} = E_{V_1} = 0.7 \text{ eV}$. We start by investigating isothermal signals for the choice of parameters $E_2 = E_1 = 0.9 \text{ eV}$, $E_{V_2} = E_{V_1} = 0.7 \text{ eV}$ and $r = 0$. The TL signals resulting from this set of parameters have been previously studied in some detail [9, 10]. The simulated TL glow curves using this set of E -values are shown in figure 2(a), in which the model produces single L_B and L_C TL peaks, with no apparent double-peak structure. However, Pagonis [10] demonstrated the presence of an underlying double-peak structure for the L_C TL peak, by simulating several standard methods of analysis for TL glow curves. When higher values of the retrapping ratio r are used in the model, the L_C peaks demonstrate clear double-peak structures [9, 10, 13].

Figures 2(b) and (c) show the results of simulating an isothermal experiment carried out at temperatures $T = 370$ – 390 K . Figure 2(b) shows the isothermal luminescence signal L_B originating from electronic transitions from the excited state of the e–h pair directly into the luminescence centre, while figure 2(c) shows the signal L_C resulting from transitions occurring from the CB into the luminescence centre.

Several previous studies have shown that TL and isothermal signals originating from localized L_B transitions follow first order kinetics, at least in cases of small retrapping within the e–h trap [1, 9, 10]. According to first order kinetics, the isothermal L_B signals shown in figure 2(b) can be expected to decay exponentially with time in accordance with the expression

$$L_B(t) = A \exp(-\lambda t), \quad (20)$$

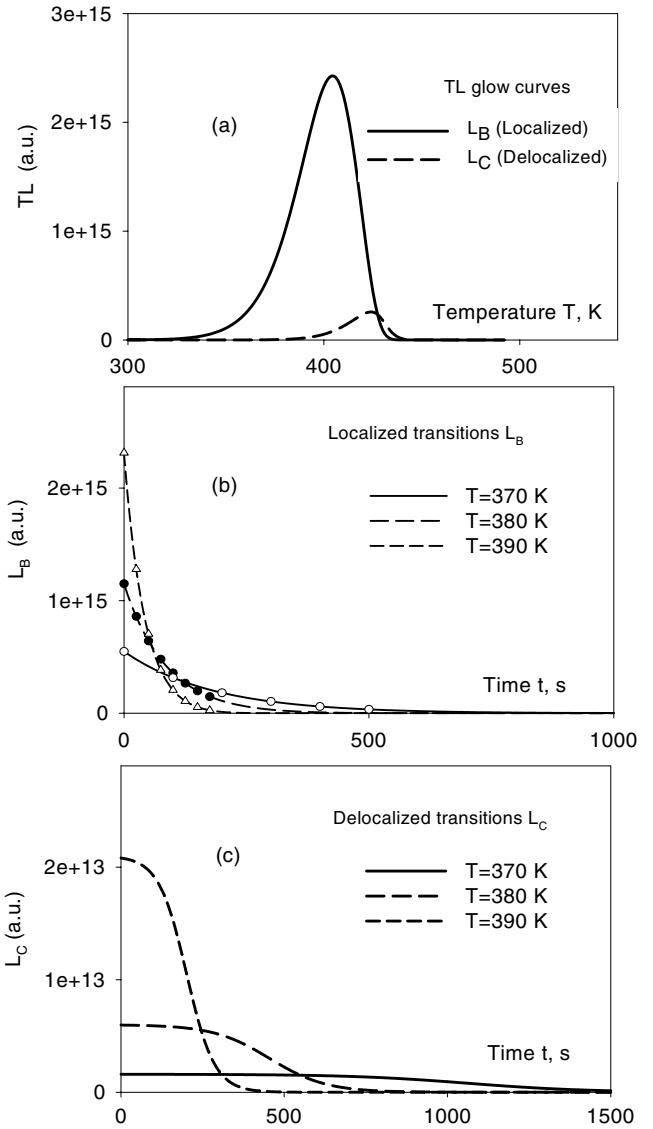


Figure 2. (a) Simulated TL glow curves due to localized (L_B) and delocalized transitions (L_C). (b) Simulated isothermal curves L_B at temperatures $T = 370$ – 390 K (solid lines). The individual points shown indicate single exponential decays calculated from equations (20) and (21). (c) Simulated isothermal curves L_C at temperatures $T = 370$ – 390 K . These luminescence signals correspond to delocalized transitions in the model. The numerical values of the parameters used in the model are given in the text.

with the expected decay constant λ or these LTs given by

$$\lambda = 1/\tau = \nu \exp(-E/kT). \quad (21)$$

Here λ represents the decay constant (s^{-1}) at the isothermal temperature $T(\text{K})$, τ is the corresponding decay time (s), k is the Boltzmann constant and E is the thermal activation energy for raising electrons from the trapped state into the excited state of the e–h pair. In the numerical example presented in this section the values of the kinetic parameters are $E = E_1 = E_2 = 0.9 \text{ eV}$ and $\nu = \nu_1 = \nu_2 = 10^{10} \text{ s}^{-1}$. The individual points in figure 2(b) represent the $L_B(t)$ values calculated by using these values of the parameters in equations (20)–(21). Very good agreement can be seen between the simulated L_B

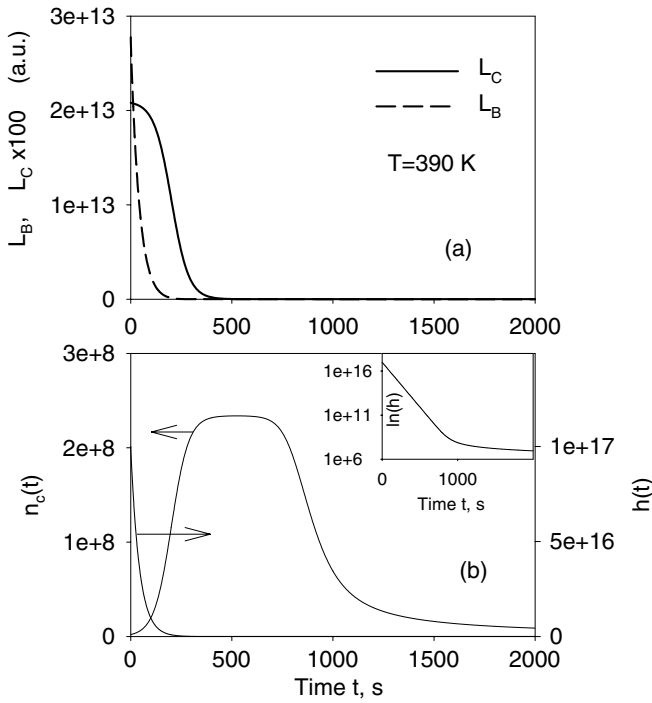


Figure 3. (a) The simulated $L_B(t)$ and $L_C(t)$ isothermal luminescence signals during the isothermal process at a constant temperature $T = 390$ K. (b) The time variations of the number of electrons $n_c(t)$ in the CB and of the number of holes $h(t)$ in the RC for the simulated data in (a). Note the double y-axis in this figure. The inset shows the $h(t)$ simulated data on a semilog scale.

data and the analytical expression in equation (20), verifying that the localized isothermal signals $L_B(t)$ follow first order kinetics.

Figure 2(c) shows the luminescence signals L_C originating through delocalized electronic transitions from the CB into the luminescence centre. These signals exhibit a different behaviour from the exponentially decaying L_B signals in figure 2(b). During an initial time interval the L_C signal varies slowly, followed by a decaying luminescence signal; obviously the overall shape of the L_C signals cannot be described by exponential decays. We can obtain some physical insight into the non-typical shape of the L_C signals by considering in some detail the simulation results for a specific temperature of $T = 390$ K, as shown in figure 3. Figure 3(a) shows the isothermal L_B and L_C signals; the isothermal L_B signal decreases to a negligible value after ~ 250 s, while the corresponding L_C signal lasts almost twice as long for a time interval of ~ 500 s. Figure 3(b) shows the corresponding time variations of the number of electrons $n_c(t)$ in the CB and of the number of holes $h(t)$ in the RC. The inset of figure 3(b) shows the simulated $h(t)$ data on a semilog scale.

The quantity $n_c(t)$ shown in figure 3(b) is proportional to the *isothermal currents*, which in principle can be measured experimentally in a dosimetric material. The behaviour of $n_c(t)$ shown in figure 3(b) is rather surprising, indicating that isothermal currents can persist for very long periods of time (~ 2000 s), even after the apparent termination of the isothermal luminescence signals L_B and L_C . This unusual broad peak behaviour for $n_c(t)$ was found in all simulated cases

presented in this paper, and seems to be typical of the SLT model. The width and magnitude of these isothermal currents depend on the temperature at which the isothermal experiment is carried out. The slow decay of the isothermal currents at large times is consistent with the previous interpretation of L_C signals, in which carriers are raised into the CB and are subsequently wandering in the crystal, in search of available holes to recombine [6, 9].

Figure 3(a) shows that for $t > 250$ s the L_B signal reaches negligible values, and therefore the LT pathway L_B becomes negligible. We can therefore expect that the delocalized transition pathway L_C becomes more important for $t > 250$ s. Furthermore, figure 3(b) shows that during the time interval 250 s $< t < 750$ s the concentration $n_c(t)$ remains constant. The physical interpretation of this constant $n_c(t)$ is that a dynamic equilibrium is established between electrons being raised into the CB from the excited state of the e-h pair and electrons being removed from the CB via the L_C pathway. As a result of this dynamic equilibrium the rate of change of the number of electrons in the CB becomes negligible, i.e. $dn_c/dt \sim 0$, and therefore $n_c \sim \text{constant}$ for the time interval 250 s $< t < 750$ s. At large times $t > 750$ s the isothermal current $n_c(t)$ decreases slowly, due to the very small remaining number of holes which can act as RCs. This is consistent with the time variation of $\ln(h)$ shown in the inset of figure 3(b).

Simulation #2: Isothermal signals when $E_1 = E_2 = E_{V_1} = E_{V_2} = 0.9$ eV. As a second example of an isothermal simulation we examine a case in which all activation energies in the SLT model are equal, i.e. $E_1 = E_2 = E_{V_1} = E_{V_2} = 0.9$ eV. As discussed in a previous section, the CD mechanism is expected to become important when the activation energies satisfy the inequalities $E_2 \leq E_1$ and $E_{V_2} \leq E_{V_1}$. The case studied in this section therefore represents a limiting case in which the activation energies satisfy the *equal* signs in these relations. The rest of the parameters used in this simulation are unchanged.

Figure 4(a) shows the corresponding simulated TL glow curves for this numerical example and figure 4(b) shows the results of simulating isothermal experiments carried out at temperatures $T = 410$ – 430 K. The simulated delocalized L_C signals in figure 4(b) show a very unusual shape with multiple stages. Specifically these signals show an initial fast decay, followed by a time interval in which the L_C signal is almost constant and finally by a slowly decaying luminescence signal which persists for large times.

In figure 5 we show the isothermal simulation results for a specific temperature of $T = 420$ K, using the same parameters as in figure 4. Figure 5(a) shows the isothermal L_B and L_C signals, while figure 5(b) shows the corresponding time variations of $n_c(t)$ and $h(t)$. The inset of figure 5(b) shows the derivative dn_c/dt as a function of time. Inspection of figure 5(b) shows that n_c is negligible for $t < 30$ s, then increases between 30 and 120 s and finally decays slowly for large times $t > 120$ s. The unusual shape of the $L_C(t)$ and $n_c(t)$ graphs shown in figures 5(a) and (b) can be explained by considering once more the various competing physical processes as follows.

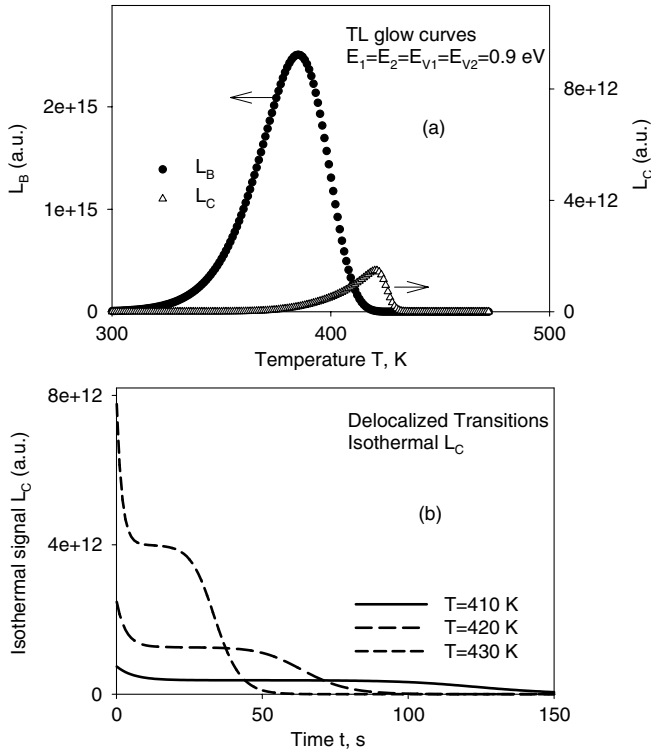


Figure 4. (a) Simulated TL glow curves due to localized (L_B) and delocalized transitions (L_C) for a second set of parameters given in the text. (b) Simulated isothermal curves L_C at temperatures $T = 410$ – 430 K. These delocalized L_C signals have an unusual shape with multiple stages.

These behaviours can be explained by considering equation (19) in the model. The luminescence signal $L_C(t)$ defined in equation (19) is proportional to the product of the two terms $n_c(t)$ and $h(t)$. During the initial time interval $t < 30$ s the number of electrons in the CB $n_c(t)$ remains constant and very small, as shown in figure 5(b), with few electrons being raised into the CB. Meanwhile the function $h(t)$ in figure 5(b) decays very fast with time; as a result, the initial fast decay of the L_C signal is determined mostly by the fast initial decay of $h(t)$. This is consistent with the behaviour of the derivative dn_c/dt shown in the inset of figure 5(b).

From an analytical point of view we can also consider equation (13) which expresses dn_c/dt , the rate of change of the number of electrons in the CB. By substituting the numerical value of $K = 0$, this equation becomes

$$\dot{n}_c = -Cn_c(H_1^0 + H_0^1 + H_0^0) + V_1H_0^1 + V_2E_0^1. \quad (22)$$

For $t > 50$ s the L_B signal reaches negligible values as shown in figure 5(a), and the delocalized pathway creating the L_C signal again becomes dominant. This means that for times $t > 50$ s $L_B = \bar{B}H_0^1 \rightarrow 0$, which also implies that $H_0^1 \rightarrow 0$, i.e. the number of states H_0^1 with one electron in the excited state and one active hole is reduced to zero. As a consequence, the positive term $V_1H_0^1 + V_2E_0^1$ in equation (22) starts decreasing, and this decrease leads to a smaller rate of change dn_c/dt . This is again consistent with the inset of figure 5(b), where for $t > 60$ s the rate of change dn_c/dt decreases continuously with time.

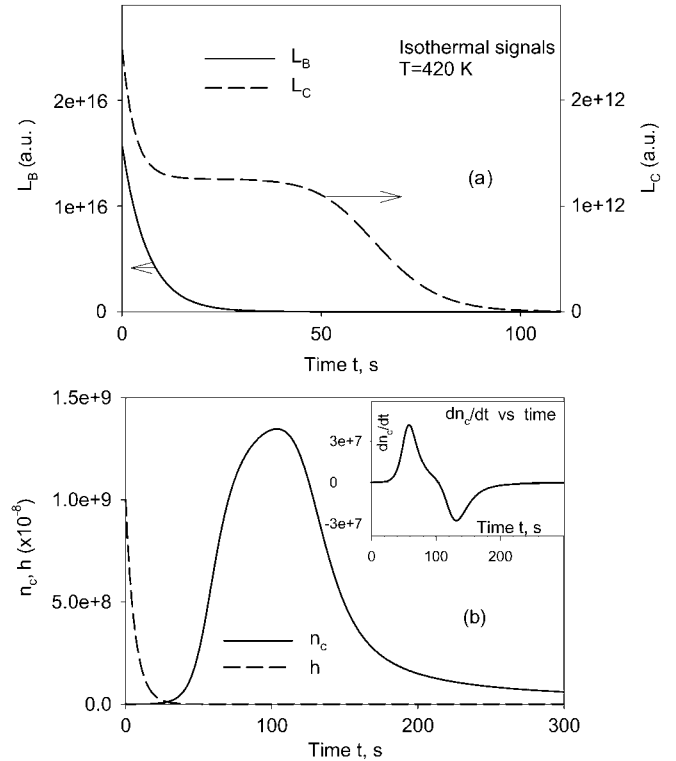


Figure 5. (a) The simulated L_B and L_C signals from figure 4 are shown at a specific temperature of $T = 420$ K. (b) The time variations of the carrier concentrations $n_c(t)$ and $h(t)$ for the data in (a). The inset shows the derivative dn_c/dt as a function of time.

It must be noted that in a real material the presence of additional electron and hole traps will most likely lead to smaller lifetimes of electrons in the CB than those shown in figures 3 and 5, since many more relaxation pathways will exist from the CB into additional electron and hole traps in the material.

Simulation #3: Isothermal signals when $E_1 = 0.9$ eV, $E_2 = 0.6$ eV and $E_{V_2} = E_{V_1} = 0.7$ eV. In this section we present a third numerical example which demonstrates yet a different possible behaviour of the L_C signals within the SLT model. Specifically we consider the numerical values of the activation energies used by Mandowski [6] to demonstrate the CD mechanism. These values are $E_1 = 0.9$ eV, $E_2 = 0.6$ eV and $E_{V_2} = E_{V_1} = 0.7$ eV; the assumption here is that the H_m^n states have an activation energy $E_1 = 0.9$ eV, while the E_m^n states have a much smaller activation energy $E_2 = 0.6$ eV. The activation energies for raising both H_m^n , E_m^n states into the CB are assumed to be the same and are also assumed to be lower than the value of the activation energy E_1 , namely $E_{V_2} = E_{V_1} = 0.7$ eV.

The behaviour of the L_B signals in this case can be described by single exponentials, in agreement with first order kinetics. Figure 6 shows that in this numerical example the simulated L_C signals are peak shaped, demonstrating a non-monotonic behaviour. Furthermore, the peak of the L_C signal shifts to lower temperatures as the isothermal temperature is increased.

Once more, this unusual behaviour of the L_C signals can be understood as the result of the combined effect of

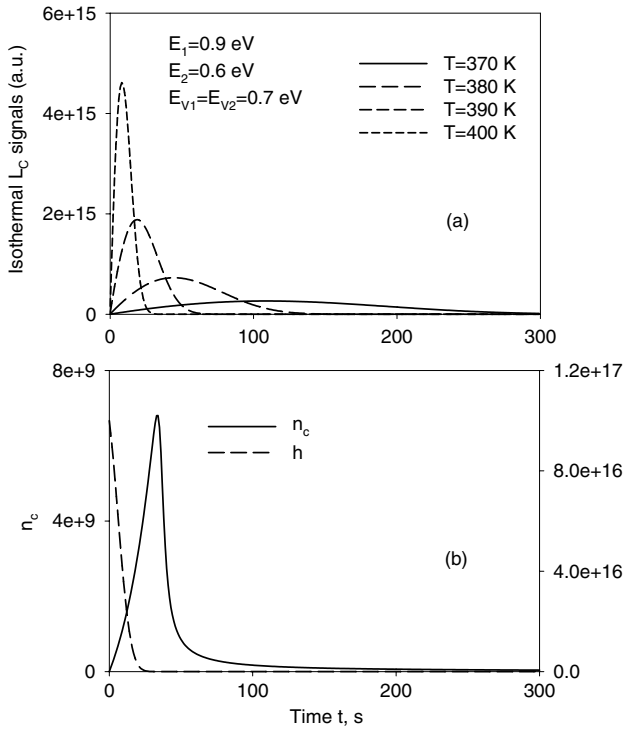


Figure 6. (a) The simulated isothermal L_C signals for the third set of parameters given in the text, for temperatures $T = 370\text{--}400$ K. The L_C signals in this case are peak shaped, demonstrating a very unusual non-monotonic behaviour. As the isothermal temperature is increased, the peak of the L_C signal shifts to lower temperatures. (b) The time variations of the carrier concentrations $n_c(t)$ and $h(t)$ for the simulated data in (a) at a temperature $T = 370$ K.

the monotonically increasing $n_c(t)$ and the corresponding monotonically decreasing function $h(t)$. These simulated functions are shown in figure 6(b) for an isothermal temperature $T = 370$ K. The simulated data in figure 6(b) show that for this set of E -values the function $n_c(t)$ increases initially very fast with time. This initial fast increase competes strongly with the decaying function $h(t)$, resulting in a peak-shaped delocalized isothermal signal L_C .

From a physical point of view this fast initial increase of $n_c(t)$ makes sense, since both activation energies E_{V_1}, E_{V_2} for raising the electrons into the CB are lower than the value of the activation energy E_1 for raising electrons from the ground state to the excited state of the e–h system.

3.1. Phase portraits

We can achieve additional insights into the SLT model by varying the parameters in the model in a systematic fashion. In particular we focus on varying E_V , the energy gap between the excited state and the CB, while keeping the energy gap E between the ground state and the excited state of the e–h pair fixed.

In order to visualize qualitative changes in the dynamics of the model, we use a different type of plot, not usually seen in the TL literature. The most common way to represent the results of a TL model is to plot the luminescence curves as a function of temperature or time. However, in the study of dynamical

systems, it is common to plot one variable or observable as a function of another. For example, in mechanical systems, one typically plots the momentum p of the particle as a function of the position x . This type of plot is known as a phase space plot or a phase portrait. Phase portraits are useful when looking at the qualitative changes in the dynamics of a system while a parameter in the system is varied. For example, in the case of an undamped harmonic oscillator the phase portrait is an ellipse centred at the origin [16]. However, when damping is introduced in this mechanical system, the phase portrait of the damped harmonic oscillator becomes a stable spiral centred at the origin. The qualitative change in the phase portrait of this well-known example from mechanics represents the changing of the system’s behaviour from periodic motion to damped oscillations, eventually decaying into the equilibrium position.

In this paper similar plots are created for the SLT model by generating plots of isothermal L_B versus isothermal L_C signals. We will refer to these L_B versus L_C plots as *isothermal phase portraits*. As will be seen, these phase portraits are useful for illustrating the different qualitative behaviours that arise in the SLT model as the parameter E_V varies.

3.2. The L_B – L_C phase portrait

As mentioned above, we are interested in understanding how the behaviour of the SLT model changes as the energy gap between the CB and the excited state (E_{V_1}, E_{V_2}) varies, while the energy gap E_1, E_2 between the ground and the excited state of the e–h pair remains fixed. Specifically in the example shown in figure 7 the values of E_{V_1}, E_{V_2} vary between 0.8 and 0.93 eV, while the energy gap $E_1 = E_2 = 0.9$ eV remains fixed during the simulations. Figure 7(a) shows the simulated isothermal L_C curves, while figure 7(b) shows the corresponding phase portrait.

The simulated curves in figure 7(a) show a clear transition point in the behaviour of the L_C curves at a value of $E_{V_1} = E_{V_2} \sim 0.89$ eV. For values of E_{V_1}, E_{V_2} smaller than ~ 0.89 eV the L_C curves show a non-monotonic peak-shaped behaviour, while for values of E_{V_1}, E_{V_2} larger than ~ 0.89 eV the L_C curves show an initial fast decay followed by a region of almost constant L_C signal, finally followed by a slow decay at large times. These behaviours were explained in the three cases #1–#3 discussed in the previous sections.

Figure 7(b) shows the phase portraits for the simulated data of figure 7(a) on a log–log scale. The trajectories in figure 7(b) start at the right-hand side of the phase portrait and each trajectory moves towards the left-hand side of the phase portrait. A clear qualitative change occurs in the phase portrait trajectory when $E_{V_1} = E_{V_2} \sim 0.89$ eV. This change in behaviour is very likely related to the onset of the CD mechanism in the SLT model, since this mechanism is assumed to be present for $E_2 \leq E_1$ and $E_{V_2} \leq E_{V_1}$.

4. Summary and conclusions

There are two types of isothermal signals in SLT models, labelled L_B and L_C . The isothermal L_B signals arise from LTs occurring from the excited state of the e–h pair into the RC,

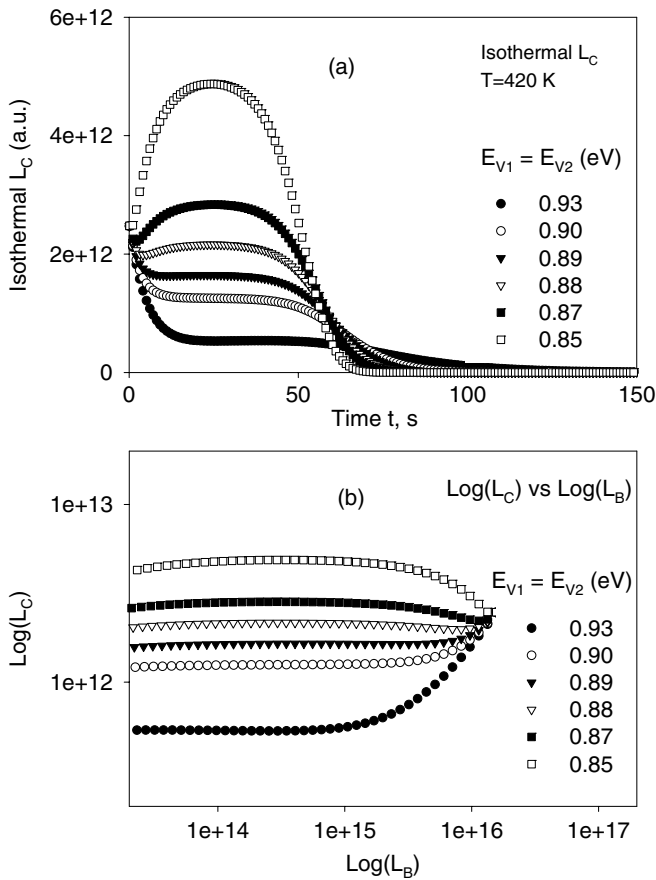


Figure 7. (a) Simulated isothermal L_C curves when the values of E_{V_1} , E_{V_2} vary between 0.8 and 0.93 eV, while the energy gap $E_1 = E_2 = 0.9$ eV remains fixed. The simulations were carried out at a constant temperature $T = 420$ K. For values of E_{V_1} , E_{V_2} smaller than ~ 0.89 eV the L_C curves show a non-monotonic peak-shaped behaviour, while for values larger than ~ 0.89 eV the L_C curves show an initial fast decay followed by an almost constant L_C signal and finally by a slow decay at large times. (b) The corresponding phase portrait of the data in (a) is obtained by plotting L_C versus L_B curves. A clear qualitative change occurs in the phase portrait trajectory when $E_{V_1} = E_{V_2} \sim 0.89$ eV.

and their behaviour can be described using first order kinetics. The isothermal L_C signals arise from delocalized transitions occurring from the CB into the RC, and they show non-typical behaviour. The non-typical behaviour of the L_C signals depends strongly on the numerical values of the activation energies E_1 , E_2 , E_{V_1} , E_{V_2} in the SLT model. A variety of

possible behaviours of the isothermal $L_C(t)$ signals is possible within the model, including very unusual peak-shaped $L_C(t)$ curves. The variety of different behaviours can be understood on the basis of the combined effect of the functions $h(t)$ and $n_c(t)$ in the SLT model.

The simulated isothermal currents (which are proportional to the concentration of electrons in the CB n_c) show rather unusual broad peak behaviours within the SLT model. The width and magnitude of these isothermal currents depend on the temperature at which the isothermal experiment is carried out. These isothermal currents can persist for very long periods of time, even after the apparent termination of the isothermal luminescence signals. The behaviour of these *isothermal currents* within the SLT model can also be understood on the basis of the various competing kinetic processes.

The simulated results in this paper suggest that isothermal experiments may offer a sensitive method for detecting the presence of SLTs in a dosimetric material. This is in contrast to previous studies using Monte Carlo simulations, which showed a weak interdependence of these phenomena.

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