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Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

Monte Carlo simulations of TL and OSL in nanodosimetric materials and feldspars

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HIGHLIGHTS

- A simplified Monte Carlo method for TL and OSL in nanodosimetric materials.
- Method is based on localized model by Jain et al. (2012).
- Small clusters of a few traps can lead to multiple peaks in TL and LM-OSL signals.
- Effects of degree of trap filling and size of clusters are simulated.

ARTICLE INFO

Article history:

Available online xxx

Keywords:

Thermoluminescence

LM-OSL

Monte Carlo simulations

Trap cluster effects

ABSTRACT

The study of luminescent materials consisting of nanoclusters is an increasingly active research area. It has been shown that the physical properties of such nanodosimetric materials can be very different from those of similar conventional microcrystalline phosphors. In addition, it has been suggested that traditional energy band models may not be applicable for some of these nanodosimetric materials, because of the existence of strong spatial correlations between traps and recombination centers. The properties of such spatially correlated materials have been previously simulated by using Monte Carlo techniques and by considering the allowed transitions of charge carriers between the conduction band, electron traps and recombination centers. This previous research demonstrated successfully the effect of trap clustering on the kinetics of charge carriers in a solid, and showed that trap clustering can significantly change the observed luminescence properties. This paper presents a simplified method of carrying out Monte Carlo simulations for thermoluminescence (TL) and optically stimulated luminescence (OSL) phenomena, based on a recently published model for feldspar. This model is based on tunneling recombination processes involving localized near-neighbor transitions. The simulations show that the presence of small clusters consisting of a few traps can lead to multiple peaks in both the TL and linearly modulated OSL signals. The effects of donor charge density, initial trap filling and cluster size are simulated for such multi-peak luminescence signals, and insight is obtained into the mechanism producing these peaks.

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1. Introduction

The physical properties of nanodosimetric materials are known to be different from those of similar conventional microcrystalline phosphors (see for example Salah, 2011; Sun and Sakka, 2014; and references therein). It has been suggested that traditional energy band models may not be applicable for some of these nanodosimetric materials, because of the existence of strong spatial

correlations between traps and recombination centers. The luminescence properties of such spatially correlated materials can be simulated by using Monte Carlo techniques.

Monte Carlo methods for the study of thermoluminescence (TL) were presented in the papers by Mandowski (2001, 2006; 2008), Mandowski and Świątek (1992, 1996; 1998; 2000), Kulkarni (1994) and Pagonis et al. (2014a). These authors suggested that usually the number of carriers in a sample is large and the differential equations used in traditional kinetic models describe the system properly. However, in some solids one must consider clusters of traps as separate systems, since the continuous differential equations are not valid and Monte Carlo methods must be used.

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Additional applications of Monte Carlo simulations in luminescence were presented by Mandowski et al. (2010) for determining the thermal-quenching function, and by Bailey (2004) on an empirical energy band model for quartz. In a different type of Monte Carlo application, Pagonis and Kitis (2012), Pagonis et al. (2011) and Adamiec et al. (2004, 2006) used a method of random variation of kinetic parameters in the study of kinetic models for quartz and other materials.

Recently Pagonis et al. (2014a) presented a simplified method of carrying out Monte Carlo simulations for TL and linearly modulated optically stimulated luminescence (LM-OSL) phenomena, based on the General One Trap (GOT) model, which is a special case of the one trap one recombination center model (OTOR) when quasi-equilibrium conditions (QE) hold. These authors showed that the presence of small clusters consisting of a few traps in a solid can lead to multiple peaks in both the TL and LM-OSL signals. The effects of retrapping and degree of trap filling were simulated for such multi-peak luminescence signals, and the luminescence process within the system of clusters was described in terms of both local and global variables. These authors suggested that the GOT method can be easily generalized for other types of luminescence solids in which the recombination probability varies with time.

The general purpose of this paper is to extend the recent work by Pagonis et al. (2014a) using the model by Jain et al. (2012), which is described in the next section. The specific goals of this paper are:

1. To carry out Monte Carlo simulations of trap clustering phenomena, by applying the simple technique of Pagonis et al. (2014a) to the model by Jain et al. (2012).
2. To show that this simple technique can also be used for simulating LM-OSL phenomena.
3. To obtain physical insight into the nature of the simulated multi-peak TL and LM-OSL signals, by varying the parameters in the model.

2. Simulation of trap clusters within the model of Jain et al. (2012)

2.1. Brief overview of the model by Jain et al. (2012)

The simulations in this paper are based on the model by Jain et al. (2012), who presented a new general kinetic model quantifying localized electronic recombination of donor–acceptor pairs in luminescent materials. This model extended earlier work by several researchers describing ground state tunneling from a random distribution of carriers (Huntley, 2006; Tachiya and Mozumder, 1974; and references therein). In the model by Jain et al. (2012) recombination is assumed to take place via the excited state of the donor, and to occur only between nearest-neighbors within a random distribution of centers. These authors simulated successfully TL and OSL signals as well as the empirical power law behavior for OSL signals. The semi-analytical version of the model by Jain et al. (2012) was examined by Kitis and Pagonis (2013), who obtained analytical solutions for four different experimental modes of stimulation: TL, OSL, linearly modulated OSL (LM-OSL) and isothermal TL processes. Other notable recent work using the model by Jain et al. (2012) was published by Pagonis et al. (2014b) on the TL signals from feldspars, Pagonis et al. (2014c) on IRSL signals from a suite of museum feldspar samples, and Pagonis et al. (2013) and Kitis and Pagonis (2014) on the exact version of the model by Jain et al. (2012).

The single differential equation arrived at by Kitis and Pagonis (2013) is:

$$\frac{dn_g}{dt} = -\frac{3\rho^{1/3}Az}{s\tau_c} \left(\ln \frac{n_o}{n_g}\right)^{2/3} n_g, \quad (1)$$

where

$$\tau_c = s^{-1} \exp \left[\left(\frac{1}{\rho'} \ln \frac{n_o}{n_g} \right)^{1/3} \right]. \quad (2)$$

In this equation n_g is the instantaneous concentration of electrons in the ground state and n_o is the corresponding initial concentration. The parameter A represents the excitation rate from the ground to the excited state, and is equal to $A = s \exp(-E/kT)$ and $A = \sigma(\lambda)I$ for the cases of thermal and optical excitation correspondingly. Here E = thermal activation energy, s is the frequency factor, λ is the optical stimulation wavelength, $\sigma(\lambda)$ is the optical absorption cross-section and I is the light intensity. Additional parameters are the dimensionless number density of acceptors ρ' , the critical tunneling lifetime τ_c , and $z = 1.8$ is a dimensionless constant introduced in the model, and β is the linear heating rate during a TL experiment. The most important physical parameters in the model are the constant dimensionless number density of acceptors ρ' , and the critical tunneling lifetime τ_c depends on time according to equation (2).

Kitis and Pagonis (2013) obtained the following analytical solutions for the time-dependent concentration $n_g(t)$ and for the luminescence intensity $L(t)$ at time t :

$$n_g(t) = n_o \exp \left[-\rho' [F(t)]^3 \right], \quad (3)$$

$$L(t) = 3n_o\rho'F(t)^2zAe^{-F(t)}e^{-\rho'[F(t)]^3}, \quad (4)$$

with the quantity $F(t)$ defined by:

$$F(t) = \ln \left(1 + z \int_0^t Adt' \right). \quad (5)$$

The simulations in this paper are based on the differential equation (1). For stochastic Monte Carlo simulations, the continuous variable n_g becomes a discrete integer variable.

Table 1

Listing of local and global variables used in the simulations of this paper, and their typical values (see also Fig. 1 for a pictorial presentation of the local variables).

	Description	Typical value
Local variables		
n_{local}	The instantaneous (time dependent) number of remaining filled traps in the cluster.	$n_{\text{local}} = \text{variable} = 3-0$
n_{filled}	The number of initially filled traps per cluster n_{filled} In general $n_{\text{filled}} \leq N_{\text{traps}}$.	$n_{\text{filled}} = 3$
N_{traps}	The total number of traps per cluster	$N_{\text{traps}} = 4$
Global variables		
n	The total instantaneous (time dependent) number of remaining filled traps in the system. This is calculated by summing over all clusters $n = \sum_{\text{all clusters}} n_{\text{local}}$.	$n = \text{variable} = 3 \times 10^5 - 0$
n_o	The total number of initially filled traps in the system. n_o is found from $n_o = N_{\text{clusters}}n_{\text{filled}}$ (with $n_o \leq N$).	$n_o = 3 \times 10^5$
N_{clusters}	The number of trap clusters in the system.	$N_{\text{clusters}} = 10^5$

2.2. The recent modeling work by Pagonis et al. (2014a) – local and global variables for a system of trap clusters

In recent modeling work Pagonis et al. (2014a) carried out Monte Carlo simulations of TL and OSL signals from a system of trap clusters. Specifically these authors studied the general one trap equation (GOT) for TL and LM-OSL intensity, by using both local and

global variables to characterize the system of traps. This subsection summarizes the system of trap clusters and the simulation method used in Pagonis et al. (2014a). The simulation method is a basic “brute force” Monte Carlo technique using differential equation (1). A description of this method is found in many standard textbooks of simulations in Statistical Physics (see for example Gould and Tobochnik, 1995; Landau and Binder, 2013; and references therein).

Pagonis et al. (2014a) introduced local variables describing the internal structure of each cluster, and global variables which describe the whole group of clusters. These local and global variables are listed in Table 1 for easy reference, together with typical numerical values used in this paper. The system simulated in this paper consists of a large number of small clusters of traps and recombination centers. The local physical parameters characterizing each cluster are: the total number of traps per cluster N_{traps} , the number of initially filled traps per cluster n_{filled} (in general $n_{\text{filled}} < N_{\text{traps}}$), and the instantaneous number of remaining filled traps in the cluster denoted by n_{local} . As the system develops in time during the optical or thermal stimulation process, the value of the local variable n_{local} will decrease from its initial value of n_{filled} to zero, as more recombinations take place within the cluster.

In terms of global variables, the system is described by the number of trap clusters in the system N_{clusters} . The total number of available traps in the system is given by the product $N = N_{\text{clusters}}N_{\text{traps}}$, while the total number of initially filled traps n_0 is given by $n_0 = N_{\text{clusters}}n_{\text{filled}}$ (with $n_0 \leq N$). The total instantaneous number of filled traps is denoted by the global variable n , and is calculated as the sum of the locally remaining filled traps n_{local} over all clusters in the system, i.e. $n = \sum_{\text{all clusters}} n_{\text{local}}$. The variables n_{local} , n_{filled} and N_{traps} represent local variables characterizing each cluster in the system, while the global variables N_{clusters} , n_0 , n and N characterize the whole system of trap clusters.

Fig. 1a shows schematically an example of such a system of small trap clusters, in which there are 4 traps in each cluster ($N_{\text{traps}} = 4$ shown as both open and solid circles), with only 3 of them being initially filled ($n_{\text{filled}} = 3$ shown as solid circles). One ensures the charge balance in the system, by assuming the existence of an equal number of 4 luminescence centers (shown as both open and solid stars), 3 of which have been activated (shown as solid stars). In this example one could simulate, for instance, a large number of clusters $N_{\text{clusters}} = 10^5$ in the system, resulting in a total number of $n_0 = N_{\text{clusters}}n_{\text{filled}} = 3 \times 10^5$ initially filled traps. From a physical point of view, the activated luminescence centers in the model by Jain et al. (2012) are assumed to exist in physical proximity to the filled traps, since they both could have been created simultaneously during the irradiation process. As the system of trap clusters in Fig. 1 develops in time, the local variable n_{local} will vary from an initial value of $n_{\text{local}} = n_{\text{filled}} = 3$ to its final value of $n_{\text{local}} = 0$ at the end of the thermal/optical excitation process. Similarly the global variable n will vary from an initial value of $n_0 = 3 \times 10^5$ to its final value of $n = 0$ at the end of the thermal/optical excitation process.

By replacing the global continuous variables n and n_0 with the local discrete variables n_{local} and n_{filled} correspondingly, the differential equation (1) becomes the following difference equation:

$$\Delta n_{\text{local}} = - \frac{3\rho^{1/3}Az}{\exp\left[\left(\frac{1}{\rho} \ln \frac{n_{\text{filled}}}{n_{\text{local}}}\right)^{1/3}\right]} \left(\ln \frac{n_{\text{filled}}}{n_{\text{local}}}\right)^{2/3} n_{\text{local}} \Delta t, \quad (6)$$

The luminescence intensity from the overall system of trap clusters will consist of the sum of luminescence intensities over all clusters in the system, i.e.:

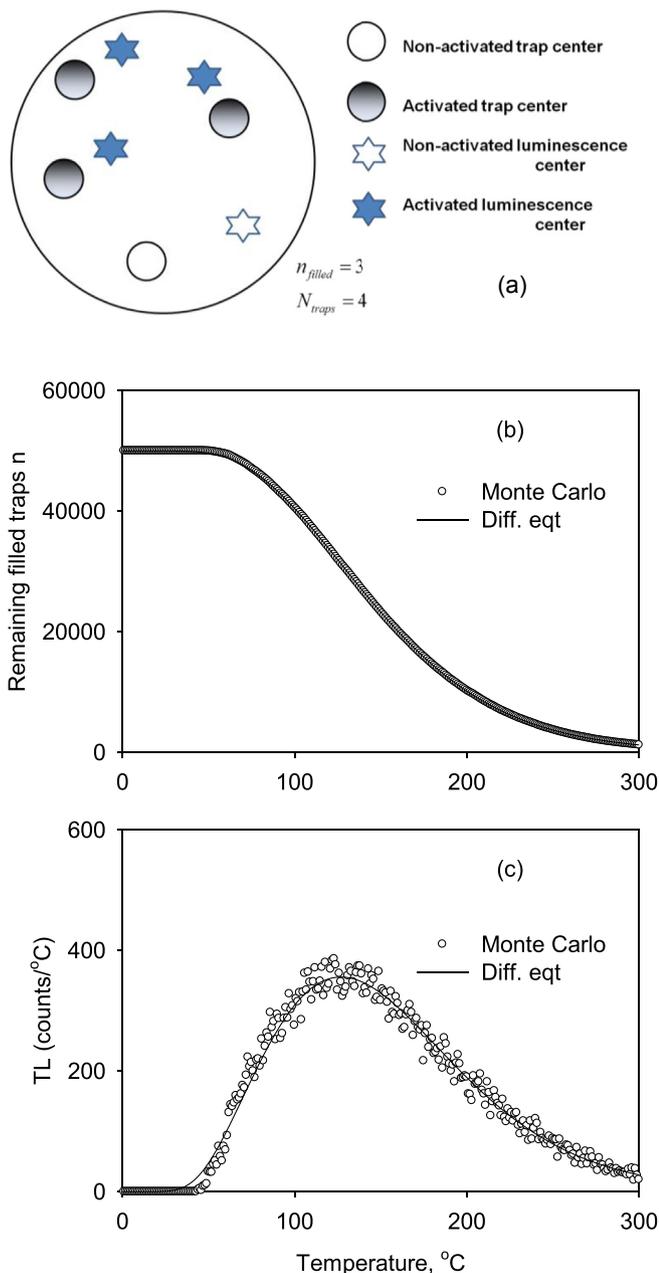


Fig. 1. (a) Schematic representation of a small trap cluster, consisting of a total of four traps in each cluster ($N_{\text{traps}} = 4$ shown as both open and solid circles). Only 3 of these traps are initially filled ($n_{\text{filled}} = 3$ shown as solid circles). An equal number of 4 luminescence centers is assumed (shown as both open and solid stars), 3 of which have been activated (shown as solid stars). The solid is assumed to consist of a large number of such clusters (e.g. $N_{\text{clusters}} = 10^5$). (b) The number of remaining filled traps $n(T)$ as a function of the temperature T . The kinetic parameters are $s = 10^{12} \text{ s}^{-1}$, $E = 0.8 \text{ eV}$, $\beta = 1 \text{ K/s}$ and $\rho' = 10^{-3}$. The simulation is for a system of 5 very large clusters, each cluster consisting of 10^4 initially filled traps, i.e. $n_{\text{filled}} = 10^4$. (c) The TL signal simulated using equation (6). The solid line in (b) and (c) represents the analytical solutions of the differential equation (1).

$$I_{TL}(T) = \sum_{\text{all clusters}} \left(-\frac{1}{\beta} \frac{\Delta n_{\text{local}}}{\Delta t} \right), \quad (7)$$

and

$$I_{LM-OSL}(t) = \sum_{\text{all clusters}} \left(-\frac{\Delta n_{\text{local}}}{\Delta t} \right). \quad (8)$$

A fixed time interval $\Delta t = 1$ s was used in all simulations presented in this paper. The simulations were also repeated for a smaller time interval $\Delta t = 0.1$ s, and the results of the simulations were unchanged.

The detailed software implementation of equations (6)–(8) was described in Pagonis et al. (2014a) and will not be repeated here. An implicit physical assumption in this description of the system is that each cluster acts as an independent entity as far as the luminescence process is concerned, since each electron participates only in local processes within the cluster.

There is an important physical and mathematical difference between the OTOR model used in Pagonis et al. (2014a), and the localized model of Jain et al. (2012). In the OTOR model it is possible to have all traps in a cluster initially filled, i.e. it is possible to have $n_{\text{filled}} = n_{\text{local}} = N_{\text{traps}}$ at time $t = 0$. This condition is not possible in the model by Jain et al. (2012), since using $n_{\text{filled}} = n_{\text{local}} = N_{\text{traps}}$ in equation (6) results in the right hand side being equal to zero, and nothing will happen within each independent trap cluster. In physical terms, one must have initially at least one empty trap inside each cluster in the Monte Carlo version of the model by Jain et al. (2012). Therefore the initial condition $n_{\text{filled}} < N_{\text{traps}}$ is used in all simulations presented in this paper.

3. Simulation results

In this section the effect of trap clustering on the TL and LM-OSL signals is simulated, and the underlying physical processes are discussed.

3.1. Simulations of TL for clusters of different sizes: trap clustering effects

Fig. 1bc shows the results of a Monte Carlo simulation for the TL signal using equation (7). The kinetic parameters used in these simulations are $s = 10^{12} \text{ s}^{-1}$, $E = 0.8 \text{ eV}$, a linear heating rate $\beta = 1 \text{ K/s}$ and the dimensionless density $\rho' = 10^{-3}$. The example in Fig. 1bc shows the simulation of a system with large clusters, each cluster consisting of 10^4 initially filled traps and one initially empty trap, i.e. $n_{\text{filled}} = 10^4$ and $N_{\text{traps}} = n_{\text{filled}} + 1$. The system consists of 5 such clusters ($N_{\text{clusters}} = 5$), resulting in a total number of $n_o = N_{\text{clusters}} n_{\text{filled}} = 5 \times 10^4$ initially filled traps.

Fig. 1b shows the number of remaining filled traps $n(T)$ as a function of the temperature T , while Fig. 1c shows the corresponding TL signal, obtained by using the difference equation (7). Inspection of Fig. 1bc shows that the relative statistical fluctuations in the number of remaining trapped carriers $n(T)$ in Fig. 1b, are much smaller than the corresponding fluctuations in the TL signal shown in Fig. 1c. This is due to the fact that $n(T)$ is calculated directly from equation (6), while the TL signal is calculated from the differences $\Delta n/\Delta T$ between successive values of $n(T)$, according to equation (7). The Monte Carlo statistical errors and their sources were discussed in some detail by Kulkarni (1994) and more recently in Pagonis et al. (2014a).

The solid line in Fig. 1b and c represents the analytical solution $n(T)$ and $I_{TL} = -dn/dt$ of the differential equation (1), indicating that in the case of large clusters simulated in Fig. 1bc, the results of the

Monte Carlo simulation approach the results from a uniform distribution of particles in the system (Mandowski and Świątek, 1998; Pagonis et al., 2014a).

Fig. 2a shows the results of repeating the simulation of Fig. 1 for two different systems, in which the size of the clusters is changed, while the total number of initially filled traps is kept the same. Specifically Fig. 2a shows the simulation of two large clusters with the corresponding numbers of initially filled traps $n_{\text{filled}} = 10^3, 10^4$

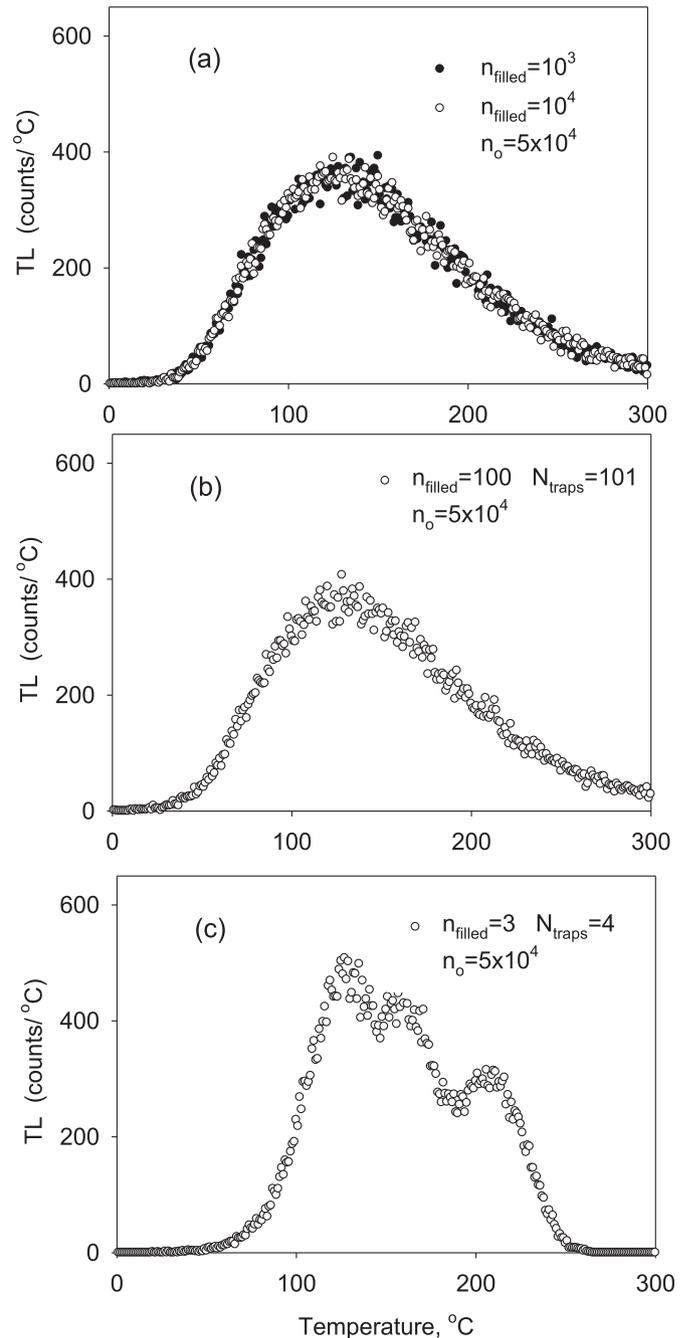


Fig. 2. (a) Monte Carlo simulations of two large clusters with $n_{\text{filled}} = 10^3, 10^4$ and $N_{\text{traps}} = n_{\text{filled}} + 1$. The kinetic parameters are the same as in Fig. 1 and the total number of particles in the system is kept constant at $n_o = 5 \times 10^4$. (b) The same simulations as in (a) repeated with a system of medium size trap clusters with $n_{\text{filled}} = 100$. (c) The same simulations as in (a) for a system of very small size clusters with $n_{\text{filled}} = 3$ and $N_{\text{traps}} = 4$. In all three examples in (a)–(c) the total number of initially filled traps is the same ($n_o = 5 \times 10^4$) and the kinetic parameters are the same. A three-peak structure is seen clearly in Figure (c).

and with $N_{\text{traps}} = n_{\text{filled}} + 1$. The kinetic parameters used in these simulations are the same as in Fig. 1 and the total number of particles in the system is kept constant at $n_0 = N_{\text{clusters}}n_{\text{filled}} = 5 \times 10^4$, while the size of the clusters is changed by varying the number of clusters N_{clusters} in the system. Fig. 2a shows that the TL intensity from the two systems of different large cluster sizes is the same, indicating that the luminescence signal from systems of large clusters is independent of the size of the cluster. The situation is seen to be different in Fig. 2b and c, which shows the same simulations as in Fig. 2a, repeated with clusters of smaller size, while the total number of initially filled traps is kept fixed at $n_0 = 5 \times 10^4$. Specifically Fig. 2b shows the TL signal from a system of medium size trap clusters, in which each cluster consists of a total of 100 initially filled traps ($n_{\text{filled}} = 100$ and $N_{\text{traps}} = 101$). Fig. 2c shows TL from a system of very small size clusters, in which each cluster consists of 3 initially filled traps with $n_{\text{filled}} = 3$ and $N_{\text{traps}} = 4$. In all three examples in Fig. 2abc, the total number of initially filled traps is the same, $n_0 = 5 \times 10^4$ and the values of the rest of the parameters also remain the same. As the size of the clusters is decreased from Fig. 2a to c, the shape of the TL glow curves changes and a three-peak structure is seen clearly in Fig. 2c. The appearance of additional peaks in the TL glow curves has been attributed previously to trap clustering effects for the case of TL (Mandowski, 2008; Pagonis et al., 2014a).

The effect of the cluster size on the TL signal and the systematic increase of the number of peaks in the TL glow peak are shown in Fig. 3, for progressively larger clusters (while the total number of particles is kept fixed at $n_0 = 5 \times 10^4$). Specifically Fig. 3(a)–(d) show trap clustering effects for $n_{\text{filled}} = 1, 2, 3, 100$ and with $N_{\text{traps}} = n_{\text{filled}} + 1$. Fig. 3 is consistent with previous simulations by Mandowski and Świątek (1998) and Pagonis et al. (2014a), who showed that the number of constituent TL peaks is always equal to the number of filled traps per cluster n_{filled} , in this case equal to 1, 2, 3, 100.

As will be shown in the next subsection, these trap clustering effects can occur also for LM-OSL signals, and they are more likely

to occur for conditions of high numerical values of the charge density ρ' .

3.2. Simulations for variable charge density ρ'

This subsection presents a systematic study of the effect of the charge density ρ' on the trap clustering effects for TL signals. Fig. 4 shows the variation of the number of remaining filled traps $n(T)$ as a function of temperature T , and for variable ρ' in the range $\rho' = 5 \times 10^{-4} - 2 \times 10^{-3}$. Fig. 5 shows the corresponding TL signals. In this example there are four initially filled traps per cluster ($n_{\text{filled}} = 4$), and the total number of initially filled traps in the

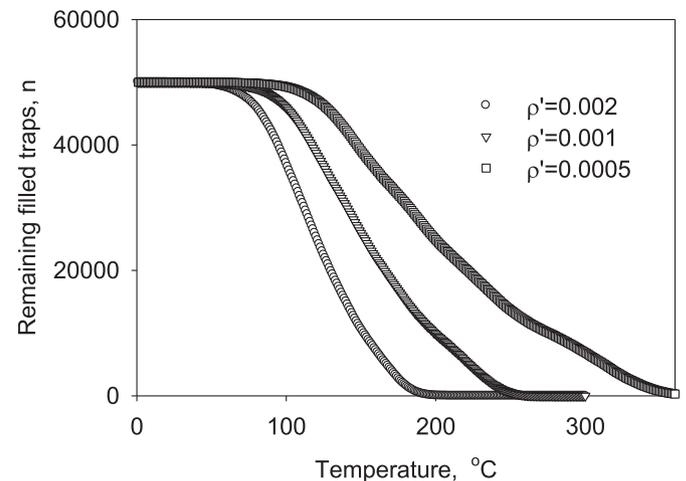


Fig. 4. The effect of the charge density ρ' on the trap clustering effects for TL signals. The variation of the number of remaining filled traps $n(T)$ as a function of temperature T is shown for the range $\rho' = 5 \times 10^{-4} - 2 \times 10^{-3}$. Fig. 5 shows the corresponding TL signals. As the value of ρ' is decreased, $n(T)$ follows a “four-step” variation (squares), with the charge carriers recombining at progressively higher temperatures.

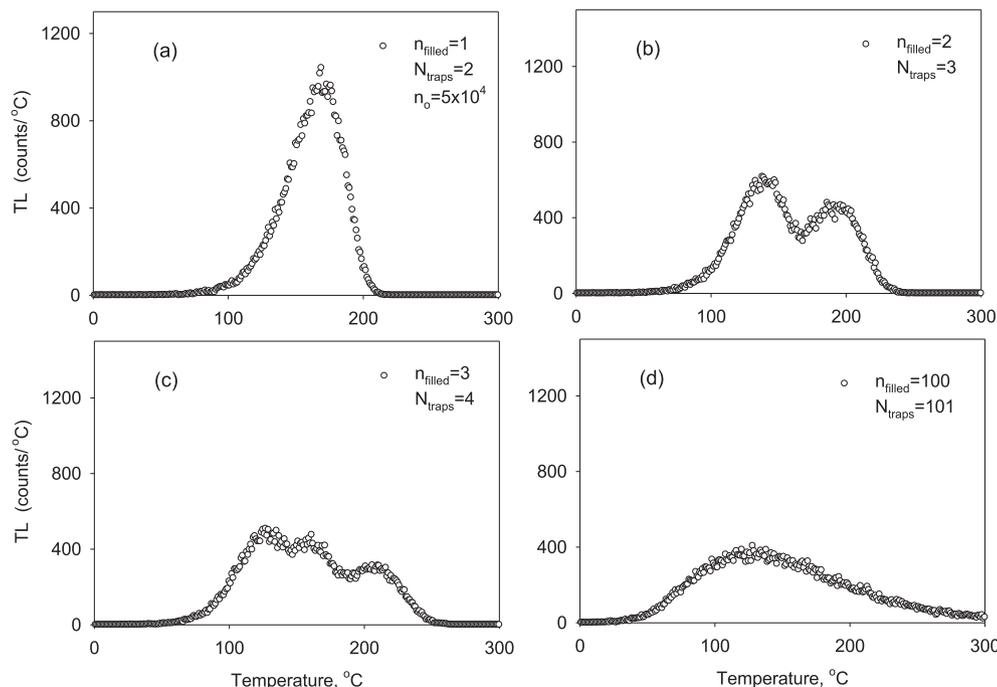


Fig. 3. (a)–(d) Study of the effect of the cluster size on the TL signal for progressively larger clusters ($n_{\text{filled}} = 1, 2, 3, 100$ and $N_{\text{traps}} = n_{\text{filled}} + 1$), while the total number of particles in kept fixed at $n_0 = 5 \times 10^4$. The number of constituent TL peaks is equal to the number of filled traps per cluster $n_{\text{filled}} = 1, 2, 3, 100$.

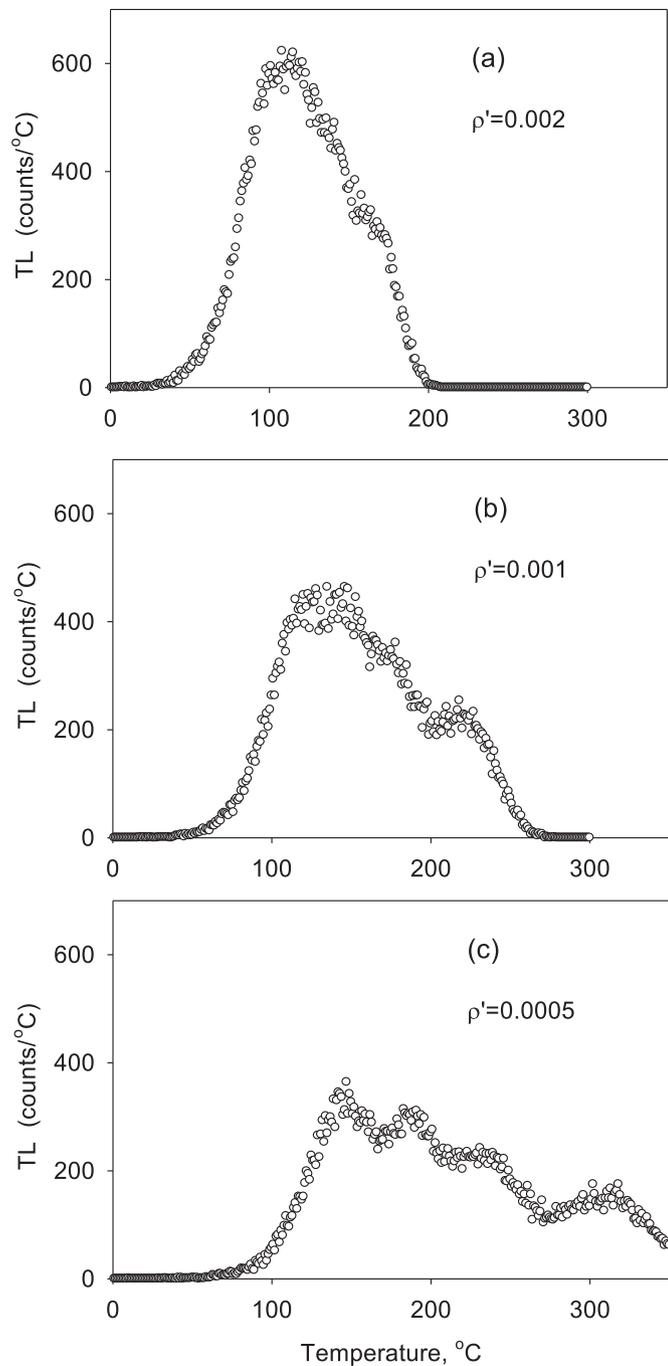


Fig. 5. TL signals for the simulations shown in Fig. 4. The TL glow curve consists of 4 underlying peaks, which become more separated in temperature as ρ' decreases. This is consistent with the number of constituent TL peaks being equal to the number of filled traps per cluster $n_{\text{filled}} = 4$.

simulated system is again $n_o = 5 \times 10^4$.

At high values of $\rho' = 2 \times 10^{-3}$ (circles in Fig. 4), the number of carriers $n(T)$ decreases continuously at all temperatures T , and the corresponding TL signal in Fig. 5a consists of a broad TL peak with several apparent overlapping peaks. However as the value of ρ' is decreased in Fig. 4, it is clear that $n(T)$ follows a “four-step” variation (squares in Fig. 4), with the charge carriers recombining at progressively higher temperatures as ρ' is decreased. The corresponding TL intensities in Fig. 5bc show clearly that the TL glow curve consists of 4 underlying peaks, which become more

separated in temperature as ρ' decreases. This is consistent with the previous statement that the number of constituent TL peaks should be equal to the number of initially filled traps per cluster, in this case $n_{\text{filled}} = 4$.

The physical picture behind the “four-step” variation of $n(T)$ in Fig. 4 can be interpreted as follows. As the charge density ρ' decreases, the critical tunneling lifetime τ_c increases according to equation (2). This increase in the value of τ_c causes a “delay” in the recombination processes within each cluster, resulting in the multiple peaks appearing in the TL glow peak at progressively higher temperatures, as shown in Figs. 4 and 5. A similar multistep variation of $n(T)$ was demonstrated by Pagonis et al. (2014a) in their study of the GOT model of luminescence.

3.3. Trap clustering effects for LM-OSL signals

Fig. 6 shows a Monte Carlo simulation for an LM-OSL experiment, based on equations (6) and (8), with A representing now the optical excitation probability. It is preferable to simulate LM-OSL experiments instead of continuous wave OSL (CW-OSL), because the latter is a featureless signal that does not lend itself to easy visualization of the various components in the signal. The parameters used in the simulation of Fig. 6 are the same as in the TL simulation of Fig. 1, with the additional parameter in equation (6)

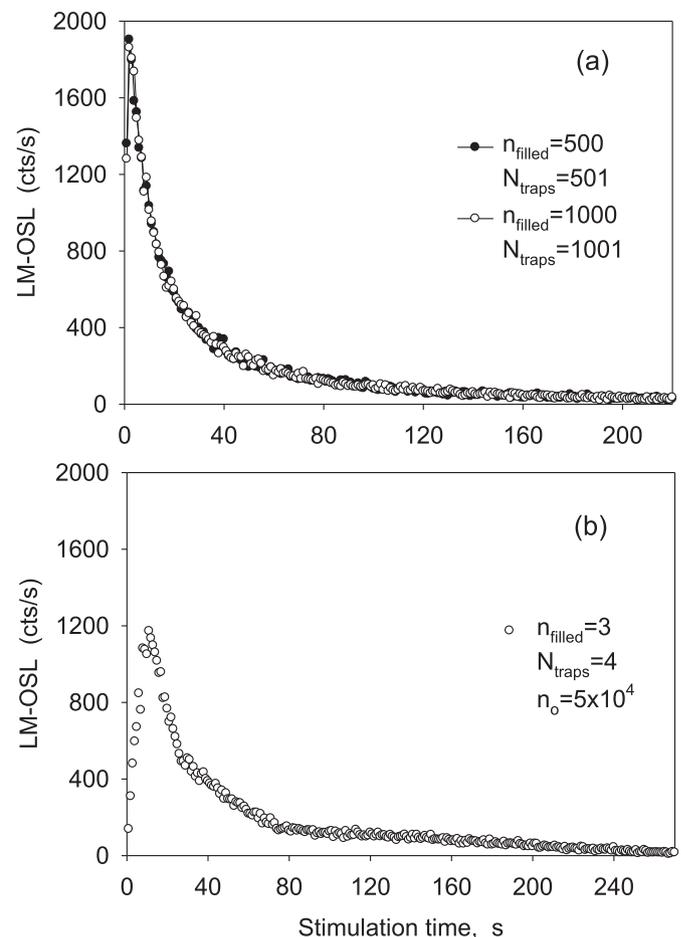


Fig. 6. Monte Carlo simulation for an LM-OSL experiment based on equations (6) and (8) and with the parameters used in Fig. 1. The optical stimulation probability is $A = 10 \text{ t} (\text{s}^{-1})$. (a) The results for two large systems of clusters with $n_{\text{filled}} = 500, 5 \times 10^3$ with the total number of particles in the system kept constant at $n_o = 5 \times 10^4$. (b) The same simulations as in (a), for a system of very small size clusters with $n_{\text{filled}} = 3$ and $N_{\text{traps}} = 4$. A three-peak structure is seen clearly.

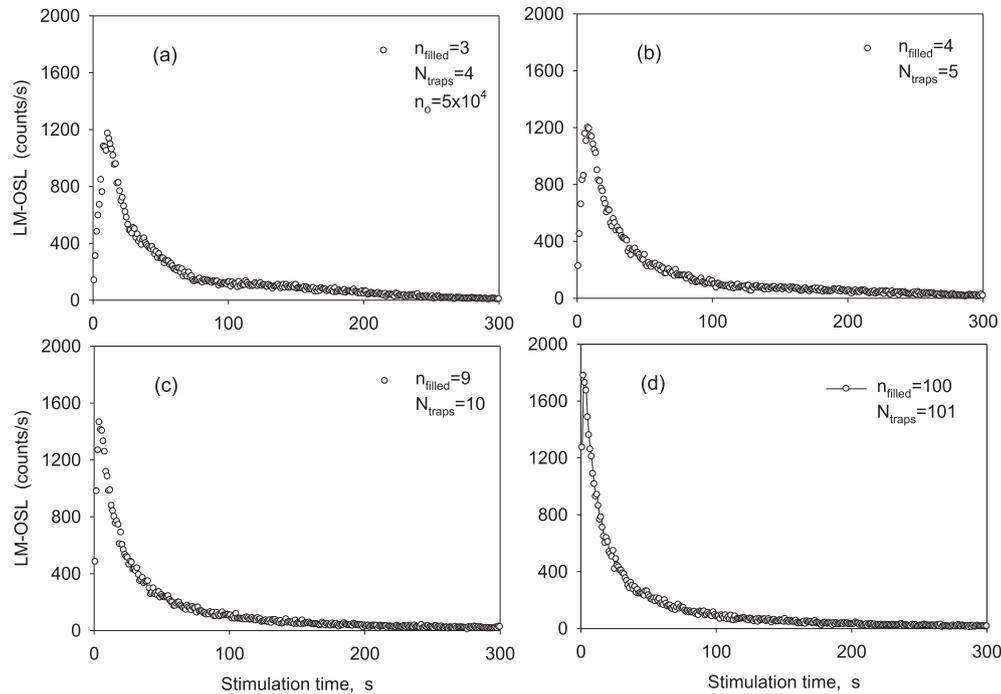


Fig. 7. The results of systematically increasing the size of clusters in the system, while keeping the total number of particles in the system fixed. LM-OSL curves are shown for several sizes of clusters $n_{\text{filled}} = 3, 4, 9, 100$.

being the optical stimulation probability $A = 10t \text{ (s}^{-1}\text{)}$. Fig. 6a shows the results for two large systems of clusters with $n_{\text{filled}} = 500, 5 \times 10^3$ with the total number of particles in the system kept again constant at $n_o = 5 \times 10^4$. Fig. 6a shows that the LM-OSL intensity from the two systems of different cluster sizes is the same, indicating that the luminescence signal from systems of large clusters is independent of the size of the cluster. The situation

is seen to be different in Fig. 6b which shows the same simulations as in Fig. 6a, repeated for a system of very small size clusters, with $n_{\text{filled}} = 3$ and $N_{\text{traps}} = 4$. As the size of the clusters is decreased from Fig. 6a to b, the shape of the LM-OSL signals changes and a three-peak structure is seen clearly in Fig. 6b. The appearance of additional peaks in the LM-OSL signals was also reported by Pagonis et al. (2014a) in their study of the GOT model.

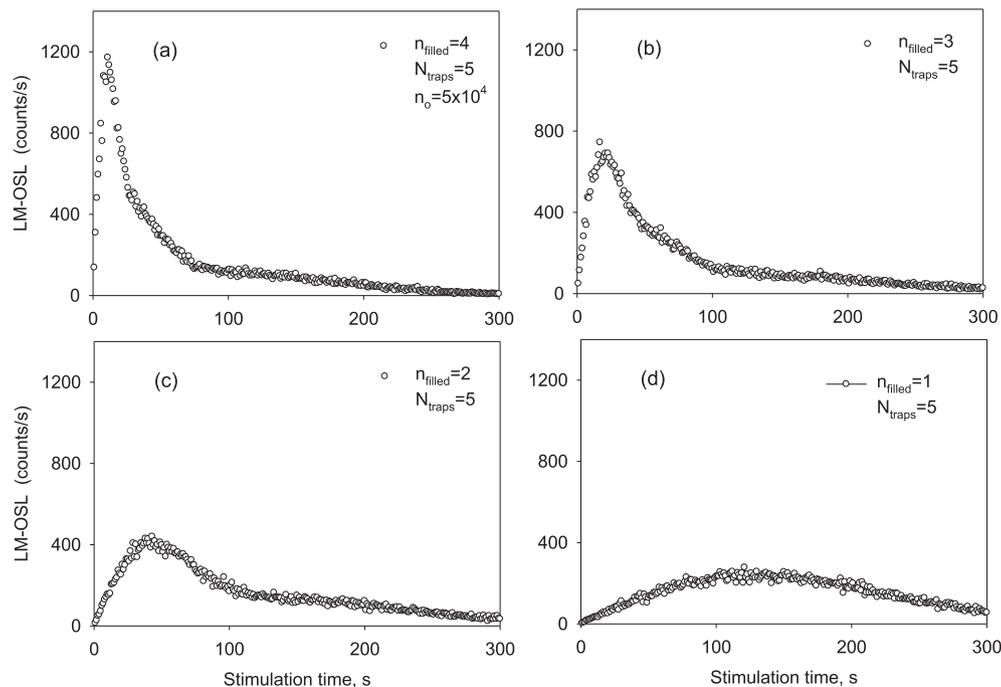


Fig. 8. Trap clustering effects for different degrees of initial trap filling with $n_{\text{filled}} = 4, 3, 2, 1$ while $N_{\text{traps}} = 5$. As the degree of trap filling is increased, the number of constituents peaks is increased, the LM-OSL signal shifts towards lower stimulation times, and the area under the LM-OSL curve remains constant and equal to $n_o = 5 \times 10^4$.

As is well known from the theory of luminescence signals containing multiple components, there exists an important physical difference between the TL and LM-OSL signals shown in this paper. As discussed for example in Kitis and Pagonis (2008), in LM-OSL signals all components start simultaneously at time $t = 0$, while in TL signals the various components appear in general sequentially at different temperatures, depending on the values of the kinetic parameters E, s for each component.

Fig. 7 shows the results of systematically increasing the size of clusters in the system, while keeping the total number of particles in the system fixed. Specifically LM-OSL curves are shown for several cases of clusters $n_{\text{filled}} = 3, 4, 9, 100$.

Fig. 8 shows trap clustering effects for different degrees of initial trap filling with $n_{\text{filled}} = 1, 2, 3, 4$ while $N_{\text{traps}} = \text{fixed} = 5$. As the degree of trap filling is increased, the number of constituents peaks is increased, the LM-OSL signal shifts towards lower stimulation times, and the area under the LM-OSL curve remains constant and equal to $n_0 = 5 \times 10^4$.

4. Discussion

This paper demonstrates trap clustering effects as a stochastic process within the model of Jain et al. (2012). As mentioned previously, the system simulated in this paper consists of many independent clusters of electron–hole pairs. Electrons are released from the traps by either thermal or optical stimulation, and are subsequently recombined radiatively within the same cluster. This is a different luminescence process than the one described by Pagonis et al. (2014a) within the GOT model and under quasi-equilibrium (QE) conditions.

In a material such as feldspar, one might expect that the trap clustering effects could occur by either a localized or a delocalized mechanism, depending on the excitation energy provided to the system. Several recent experimental and modeling papers have contributed to a better understanding of the luminescence and tunneling processes in feldspars and supported the existence of tunneling processes involving localized recombinations with tunneling taking place from the excited state of the trap, as well as charge migration through the conduction band-tail states.

Even though the simplified method presented here will not be applicable for all luminescence systems, it is interesting to note that several of the general conclusions from this paper are very similar to the conclusions reached in the detailed studies by Mandowski (2008) and by Pagonis et al. (2014a).

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