Radiation Measurements 134 (2020) 106307

Contents lists available at ScienceDirect

Radiation Measurements

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

Simulation of thermoluminescence dose response in cluster systems with deep traps

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ARTICLE INFO

Keywords:
Thermoluminescence kinetics
Cluster system
Monte Carlo method
Deep traps
Competition effects
Superlinearity of TL dose Response

ABSTRACT

The TL dose response was simulated within the framework of a kinetic model with cluster defects which contain electron TL-active traps and deep traps. It was shown that superlinearity of the TL dose dependence arises due to the temperature-dependent competition between the intracluster capture of electrons by deep traps and their thermal release into the conduction band. The regularities of the influence of the main parameters of the model (heating rate, activation energies of the ground and excited levels of TL-active traps) on the TL yield and nonlinearity of the dose dependence were found.

1. Introduction

Non-linear dose dependences of thermoluminescence (TL) response are observed in many dosimetric materials (McKeever et al., 1995; Mahajna and Horowitz, 1997; Kortov et al., 2007; Kafadar et al., 2009; Kafadar and Majeed, 2014; Druzhyna et al., 2017). One of the mechanisms of nonlinearity formation is the processes of competition involving various types of defects. Generally, such processes are theoretically considered within the framework of models with uniformly distributed defects. Chen et al. (1996) studied competition between electron TL-active traps and deep traps at the stages of irradiation and heating. Filling of deep traps with increasing dose reduces the probability of their capture of electrons. This results in increasing number of acts of radiative recombination at the luminescence centers and the occurrence of superlinearity of TL dose response. Competition between recombination centers at the irradiation stage can also cause superlinearity (Chen et al., 1996).

Another well-known approach to describe TL kinetics is the simulation of the so called spatially correlated systems. In such systems, defects form clusters within which the transfer of charge carriers is possible bypassing the delocalized bands. Within the framework of the reported models, the following effects are observed: the appearance of a multipeak structure of the TL curve in the presence of traps of the same type (Mandowski and Swiatek, 1998), anomalous heating rate effect (Mandowski and Bos, 2011), isothermal TL build-up (Pagonis and Kulp, 2010) and dose-rate effect (Orzechowski and Mandowski, 2010). So far, the competing effect of deep traps on the nonlinearity of TL dose response in such models has not been studied. One can expect that such investigation will reveal new features of competing processes between traps and TL dose dependences.

The aim of this work was to simulate TL kinetics within the framework of a cluster model with deep electron traps, as well as to analyze the influence of the model parameters on the TL yield and nonlinearity of its dose response.

2. The model

The band diagram of the cluster model with electron deep traps is shown in Fig. 1. The model includes a luminescence center (H) and a large number of independent cluster defects. Each cluster consists of TL-active electron traps (N) which have ground (n) and excited (n_e) states, and competing deep electron traps (M).

Fig. 1a shows transitions at the irradiation stage. X is the transition which is related to the formation of electron-hole pairs under irradiation; D is capture of holes from the valence band by the luminescence center; α is trapping of electrons from the conduction band by the cluster (on the n_e level); B is a radiative recombination of an electron of the conduction band and a hole in the luminescence center, A is an electron transfer from the excited state of trap N in the main state; C is the capture of an electron by a deep trap within the cluster; γ is trapping of a delocalized electron from the conduction band by a deep trap in the cluster.

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https://doi.org/10.1016/j.radmeas.2020.106307
Received 30 October 2019; Received in revised form 23 January 2020; Accepted 9 March 2020
Available online 14 March 2020
1350-4487/

The excited state n_e is part of the trap structure but it is not a new kind of trap. Specifically during the irradiation stage, the energy level for n_e acts as an intermediary stage for electrons transitioning from the CB to the either to the ground state of the trap, or to the deep trap (transitions A, C and α in Fig. 1a). Even though the irradiation stage takes place at room temperature, it is possible that transitions A and C take place, because they are not associated with thermally stimulation, but rather they represent electronic relaxation processes from the CB to n_e (transition α), from n_e to the ground state (transition A) and from n_e to M (transition C).

Fig. 1b shows transitions at the heating stage. Here transitions which describe thermal release of an electron from the TL-active trap to the excited level n_e (transition P) and the one from the excited level n_e to the conduction band (transition P_{ne}) are considered.

In principle, there may be a direct delocalization from the ground state into the conduction band. However, our model is a two-stage transition model, similar to several other two-stage models described in the literature (Chen et al., 2012). In this type of model, the release of the carriers is assumed to take place via an intermediate localized excited state. Electrons are thermally stimulated from the trap into an excited state, and then thermally released into the conduction band. So even though such a ground state-to CB transition is in principle possible, it is not part of the equations in the model.

In our model we assume that tunneling from either the ground state or the excited state has a much smaller probability than the electronic transitions shown in Fig. 1. In general, one would expect that the electronic transitions shown in Fig. 1 will be much slower than tunneling transitions. At the same time, it is assumed that the shallow and deep traps may be very close together because they are part of the same cluster.

We used Monte Carlo method to calculate TL curves and dose dependences of its yield (Mandowski and Swiatek, 1992). In this method, a random time is generated for each charge carrier transition according to the exponential probability distribution. The shortest time is selected from the obtained times, and the transition with the shortest time is considered to be completed. The population of the levels changes in response to the resulting transition. Then, the next iteration occurs, etc. The calculation method is described in more detail in the paper (Merezhnikov et al., 2019). The novelty of our proposed approach is the two-stage generation of random numbers while choosing a transition. First, the type of charge carrier transition that occurred for all the clusters is selected. Then, we choose the cluster which "participated" in the transition. This approach allows us to significantly reduce the amount of generations of random numbers and the calculation time. TL was simulated in three successive stages: irradiation, relaxation, and heating.

The parameters used in the calculations are shown in Table 1. To calculate TL curves, we used irradiation time $\tau = 500$ s.

3. Results and discussion

We chose the initial number of holes in the luminescence centers to

Table 1
Model parameters.

Notation	Description	Value
Z	Total number of clusters	10^5
N	The number of the TL-active (main) traps per cluster	1
M	The number of deep traps per cluster	2
H	The number of emission centers	10^9
s	The frequency factor for an electron in the ground state of the main trap	10^{10} s^{-1}
s_{ne}	The frequency factor for an electron in the excited state of the main trap	10^{10} s^{-1}
E	The energetic distance from the ground state of the main trap to the excited state of the main trap (trap activation energy)	0.7 eV
E_{ne}	The energetic distance from the excited state of the main trap to the conduction band (cluster activation energy)	0.7 eV
γ	Coefficient of an electron capture by the deep trap from conduction band	10^{-3} s^{-1}
D	Probability coefficient of a hole capture by an emission center	10^{-3} s^{-1}
α	Coefficient of an electron capture to the excited state of the main trap from conduction band	$2 \cdot 10^{-3}$ s^{-1}
B	Probability coefficient of radiative recombination	10^{-8} s^{-1}
C	Probability coefficient of local trapping by a deep trap	0.5 s^{-1}
A	Probability coefficient of local trapping by the main trap	1 s^{-1}
β	Heating rate	5 K/s
X	The efficiency of electron-hole pair's creation	100 s^{-1}

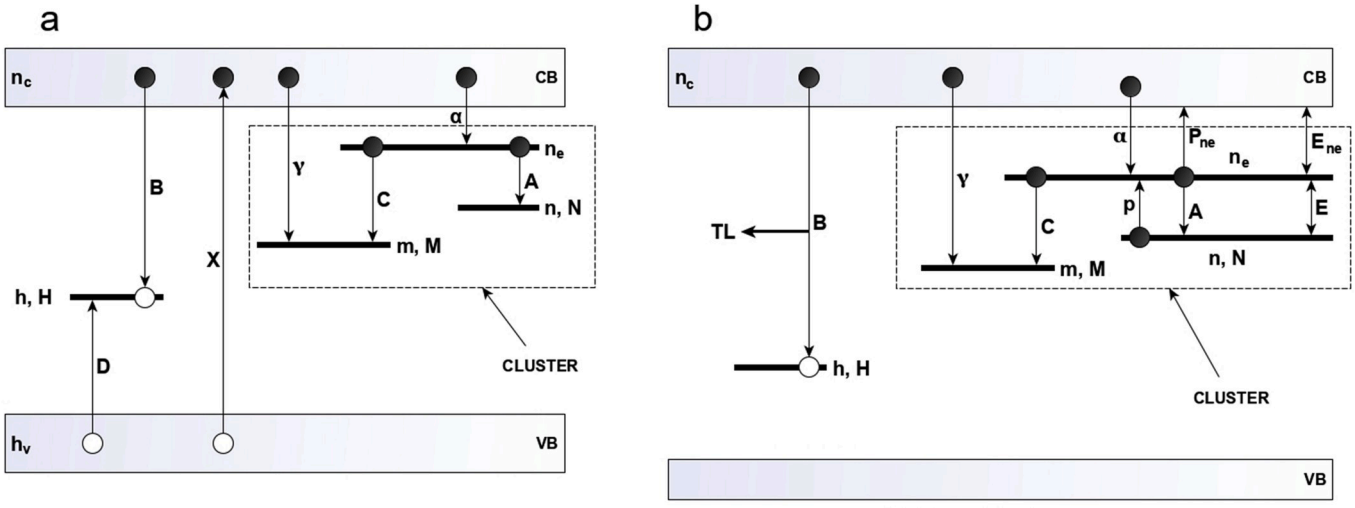

Fig. 1. Energy band diagram for TL cluster model with deep electron traps for the irradiation (a) and heating (b) stages. In this figure N is the number of the TL-active traps per cluster, M is the number of deep traps per cluster, H is the number of emission centers, γ is the coefficient of an electron capture by the deep trap from conduction band, D is the probability coefficient of a hole capture by an emission center, α is the coefficient of an electron capture to the excited state of the main trap from conduction band, B is the probability coefficient of radiative recombination, C is the probability coefficient of local trapping by a deep trap, A is the probability coefficient of local trapping by the main trap, X is the efficiency of electron-hole pair's creation, P is the probability of thermal release of an electron from the TL-active trap to the excited level n_e, P_{ne} is the probability of thermal release of an electron from excited level n_e to the conduction band, E and E_{ne} are the activation energies of transitions P and P_{ne}, respectively.

be large enough to ensure that the reported number changes only insignificantly during all stages of the formation of TL response (irradiation, relaxation, heating), Thus, in the model under discussion, the main factor which determines the regularities in changes in carrier numbers at the localized levels at the irradiation stage was the competition between TL-active (N) traps and deep electron traps (M). One can expect that the mechanism of competition between TL-active traps and deep traps within the framework of our model at the irradiation stage differs slightly from that for the model with uniformly distributed traps, thus it will not be considered in detail. It is known that such competition causes superlinear growth of TL yield with increasing radiation dose when the deep traps are filled faster than the TL-active ones (Chen et al., 1996). In the TL dose dependence, the initial linear region is replaced by a superlinear one, as the deep traps are filled. This region continues until the deep traps are completely populated. Further, the superlinear region turns into a linear one, and only then the dose dependence is saturated.

The simulation of the irradiation and relaxation stages resulted in certain numbers of filled localized levels, which were later used as initial conditions to simulate the heating stage.

In this paper we analyzed TL dose dependences, with a special focus on the processes of competition between different localized states at the heating stage. During the heating stage the thermally released electrons from traps N can be captured by deep traps M or radiatively recombine with the holes in luminescence centers H. The probability of electrons to be released into the conduction band depends significantly on the ratio between the probabilities of thermal release (transition P_{ne}) and intracluster capture of electrons by deep traps (transition C). When the intracluster transition C dominates, the electron is relatively rarely released in the conduction band, which reduces the total number of radiative recombination acts. In the contrary case, the electron is more likely to be released in the conduction band, from where it can radiatively recombine with the hole in the luminescence center (transition B). In accordance with this reasoning, it should be expected that the total TL yield rises, as the temperature increases, since the probability of P_{ne} transition increases. In this way, within the framework of the model, the processes of competition between luminescence centers and deep electron traps depend on the recombination temperature. One can expect that the nonlinearity of TL dose dependences will also depend on this parameter. It is known that with an increase in the heating rate, the TL peak shifts to the higher temperature region (the recombination temperature increases) (McKeever et al., 1995). Therefore, within the framework of the model, the heating rate will affect the TL yield and the nonlinearity of its dose dependence.

The influence of the heating rate on the TL dose response is known from some experimental works. Bos et al. (2010) observed anomalous heating rate effect for YPO_4:Ce^{3+}, Sm^{3+} samples. Kafadar and Majeed (2014) studied the effect of the heating rate on the dose dependences of $CaSO_4$:Dy (TLD-900) samples. In this paper, the nonlinearity coefficient at $D > 1$ Gy decreased with increasing heating rate. For TLD-200, as the heating rate increased, the linear dose range expanded into the high dose region for the low-temperature and high-temperature peaks (Kafadar et al., 2009). At a heating rate of 1 °C/s, the upper limit of the linear range of the TL yield of both peaks was 1 Gy, whereas at a heating rate of 10 °C/s, the range expanded to 10 Gy and 5 Gy for the low-temperature and high-temperature peaks, respectively. A similar regularity was observed for TLD-400 (Kafadar et al., 2009). The linear range of the dose dependence was observed up to 10 Gy at 1 °C/s and up to 100 Gy at 10 °C/s. The influence of the heating rate on the dose at which the TL yield was saturated, was also discussed in the above-mentioned paper. Thus, for TLD-200 with increasing heating rate, the saturation dose decreased, and for TLD-400 remained unchanged.

Fig. 2a shows the TL curves calculated within the framework of the model shown in Fig. 1, at different heating rates. It is seen that with increasing heating rate, the TL yield increases due to the shift of the peak into higher temperature region, where the probability of thermal release P_{ne} of electrons from the localized states n_e in the conduction band is higher. Moreover, Fig. 2b shows that at high doses, due to the filling of deep traps, the severity of the effect decreases. It is also seen that as the heating rate increases, the super-linearity in the high-dose region decreases.

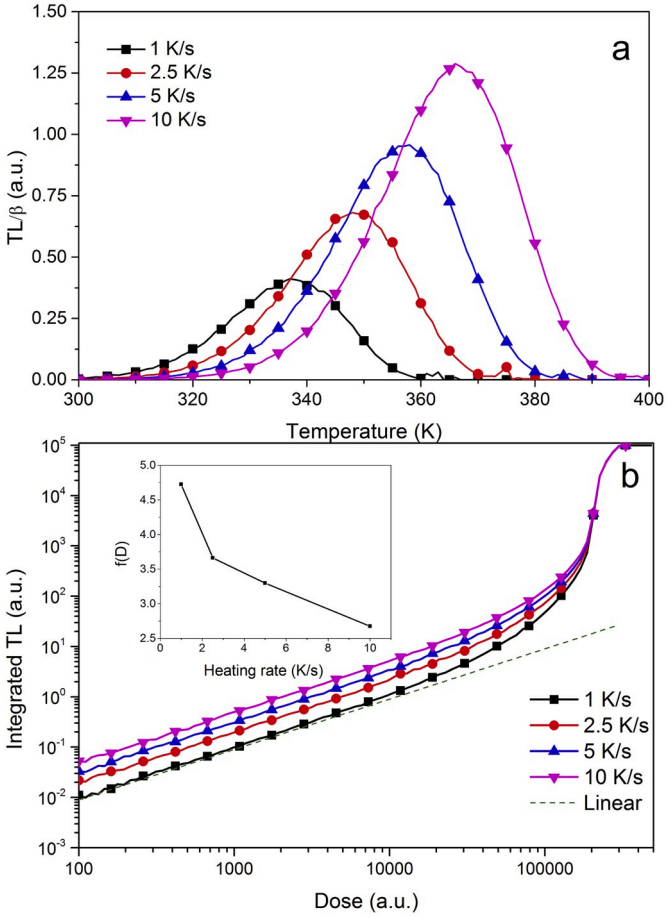

Fig. 2. TL curves (a) and TL dose dependencies (b) calculated with different heating rates. The inset shows the dependence of f(D) at $D = 10^5$ a.u. on the heating rate.

The superlinearity can be quantitatively estimated by using the normalized dose response function f(D) (Mahajna and Horowitz, 1997):

$$f(D) = \frac{F(D)}{D} \Big/ \frac{F(D_L)}{D_L} \quad (1)$$

where F(D) is the TL yield at dose D, $F(D_L)$ and D_L are the TL yield and dose respectively, selected in the linear region. We used $D = 10^5$ a.u. for calculation f(D). The inset in Fig. 2b shows the dependence of the normalized dose response on the heating rate. It can be seen that this value decreases from 4.75 down to 2.70 with an increase in the heating rate from 1 up to 10 K/s.

We also studied the influence of some other parameters of the model on the superlinearity of the TL dose dependence and yield. Fig. 3 shows the TL curves and dose dependences obtained at different depths of the trap N, characterized by the activation energy E (Fig. 1b). As the depth of the TL trap increases, the peak shifts to higher temperatures at which the probability of thermal release from the excited level n_e into the conduction band is greater (transition P_{ne}). Therefore, the TL output increases and the superlinearity decreases as the competing influence of deep traps in clusters weakens. In addition, the inset of Fig. 3b shows that f(D) undergoes the biggest drop (from 10 to 2.25) when the activation energy changes from 0.6 to 0.7 eV.

Fig. 4 shows the TL curves and dose dependences obtained at

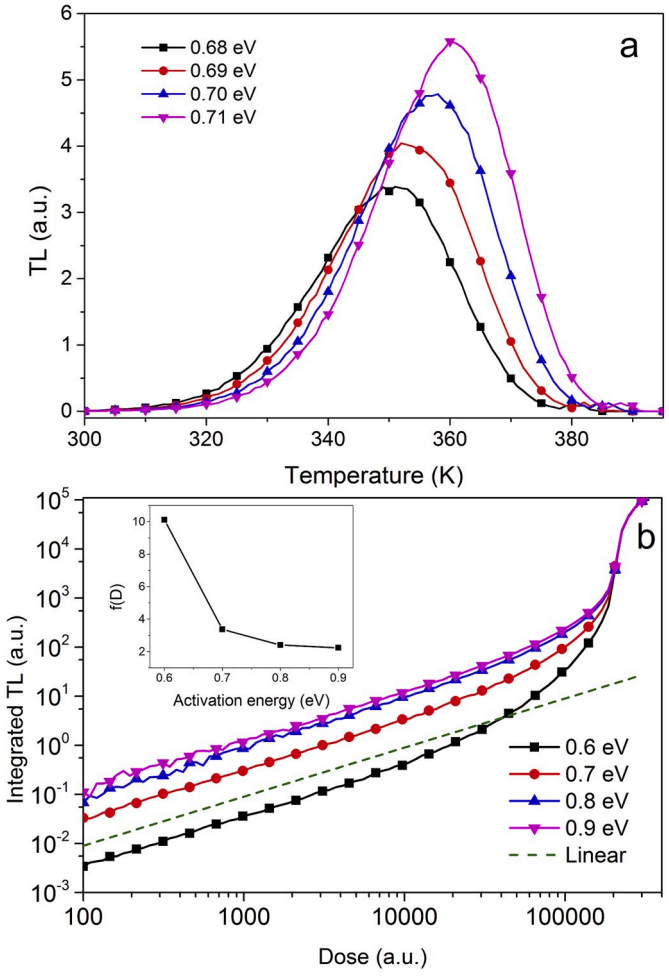

Fig. 3. TL curves (a) and TL dose dependencies (b) calculated with different trap activation energies E, E_{ne} = 0.7 eV. The inset shows the dependence of f(D) at D = 10^5 a.u. on the activation energy E.

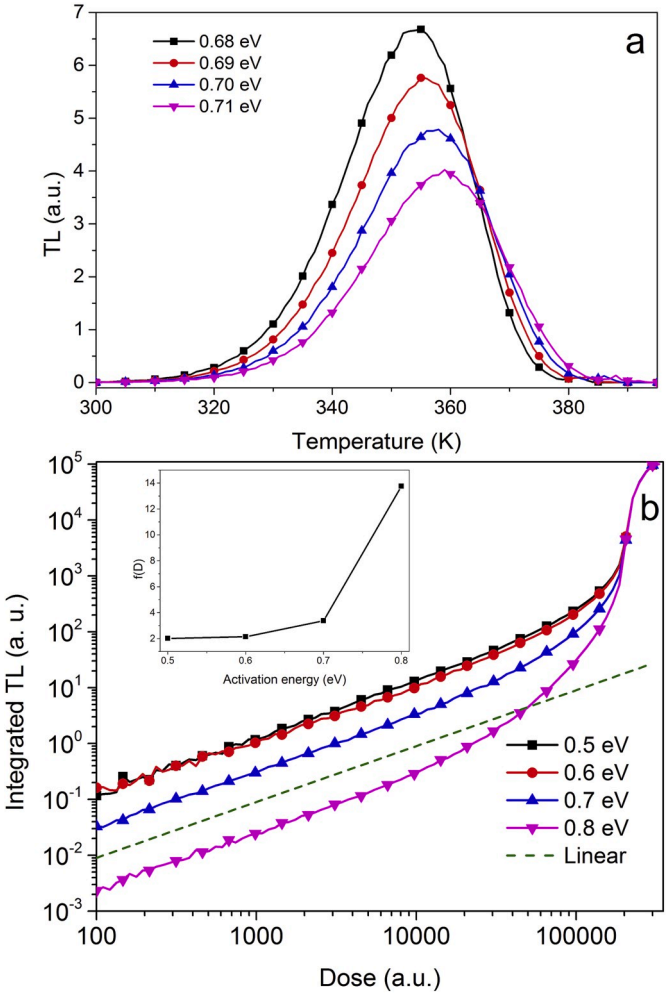

Fig. 4. TL curves (a) and TL dose dependencies (b) calculated with different cluster activation energies E_{ne}, E = 0.7 eV. The inset shows the dependence of f (D) at D = 10^5 a.u. on the activation energy E_{ne}.

different activation energy E_{ne}, characterizing the energy difference between the localized state n_e and the bottom of the conduction band. With a change in the activation energy between 0.68 and 0.71 eV, the probability of thermal stimulation P_{ne} changes by about 2 times compared with the probability of localized capture by a deep trap (C). The maximums of TL peaks in Fig. 4a differ by the same amount. Significant changes in the superlinearity coefficient are observed with more essential changes in the activation energy (0.5–0.8 eV), as shown in Fig. 4b.

4. Conclusions

In this paper the mechanisms of formation of nonlinear TL dose dependence within the framework of the model with clusters which contain deep electron traps are analyzed for the first time. It was found that the formation of the superlinearity of the TL dose response is associated with temperature-dependent competition between two processes: intracluster capture of electrons from the excited level of the TL-active trap by deep centers and their thermal release into the conduction band. It was found that the superlinearity of the TL dose dependence increases, and the TL yield drops when the heating rate or the depth of TL-active traps decreases. The same feature is observed when the energy difference between the excited level of TL-active trap and the bottom of the conduction band increases.

Acknowledgments

This work was financially supported by the Russian Science Foundation (RSF), project No. 18-72-10082.

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