One-dimensional thermoluminescence kinetics

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Abstract

A one-dimensional model of thermoluminescence (TL) and related phenomena is examined. It corresponds to some physical situations, e.g. when the excitation is done by a high-energy radiation so that traps and recombination centres are populated along tracks. This case requires specific computational methods based on the Monte Carlo technique. For this purpose, several models are proposed. Example calculations show some peculiarities of one-dimensional TL kinetics. For example, an additional peak may be formed in the high-temperature region. It relates to charge carriers that avoided nearest-neighbour recombination with adjacent recombination centres. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Thermoluminescence (TL); Trapping; Recombination; Monte Carlo methods

1. Introduction

Thermoluminescence (TL) is a relaxation process of emitting light during thermal stimulation. Initially, at appropriately low temperature the solid under study has to be excited to fill charge carriers' traps with carriers. In dosimetric applications the non-equilibrium initial occupation of traps is generated by high-energy radiation. A series of peaks appearing on TL curve may be attributed to trap levels characterised by different activation energies. The theoretical description of TL usually assumes uniform spatial distribution of traps and recombination centres (the simple model—for review see Chen and McKeever, 1997). Another extreme case is the model of localised transitions by Halperin and Braner (1960) regarding hole–electron pairs trapped close to each other. The model was later modified by Land (1969) and Chen (1976). Only for the two cases it was possible to formulate differential equations describing charge carriers kinetics. Intermediate cases with different types of spatial correlation between traps and recombination centres were studied by using Monte Carlo methods (Mandowski and Świątek, 1992, 1997). It was shown that in these cases one

gets unusual TL spectra that cannot be described in terms of the above standard models. Consequently, the spatial arrangement of traps could change the kinetics of trapping and recombination of carriers. A non-homogeneous distribution of traps in thermoluminescent detectors was suggested by many authors (e.g. Faïn and Monnin, 1977; Fields and Moran, 1974; Horowitz et al., 1996; Townsend and Rowlands, 1999).

When a solid is subjected to a high-energy radiation traps and recombination centres are populated along tracks. This way, each track represents a one-dimensional (or quasi-one-dimensional) structure. To study these cases several mathematical models for TL are suggested. These models can be used for the calculation of TL in one-dimensional (1-D) systems using Monte Carlo technique. Example calculations were performed for various kinetic parameters. It was found that for a certain range of parameters an additional peak may be formed in the high-temperature region of TL.

2. Basic models for 1-D TL

Five models of 1-D TL are presented in Fig. 1. It is assumed that hole–electron pairs are created and localised

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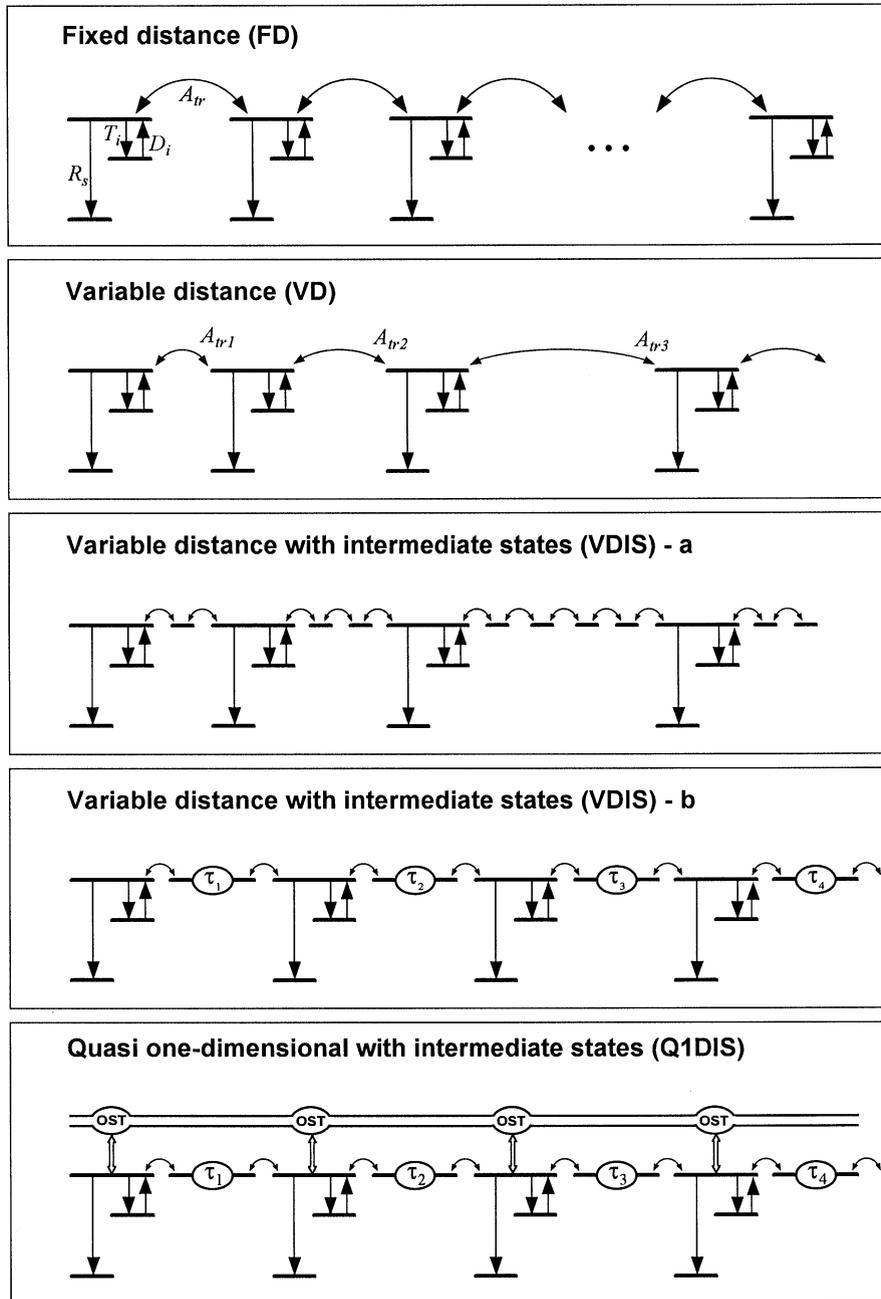*E-mail address:* a.mandowski@wsp.czest.pl (A. Mandowski).

Fig. 1. One-dimensional models for thermoluminescence kinetics. T_i—trapping, D_i—detrapping, R_s—recombination, A_{tr}—transition rate, τ_i—mean lifetime of a charge carrier in the intermediate shallow trap states, OST—outer shallow traps.

along the tracks. Each pair has a common excited state. The probability densities per unit time for a single carrier for detrapping (D_i), trapping (T_i) and recombination (R_s) are given by common formulas:

$$D_i = v_i \exp\left(\frac{-E_i}{kT}\right), \tag{1}$$

$$T_i = A_i(N_i - n_i), \tag{2}$$

$$R_s = B_s m_s, \tag{3}$$

where E_i and v_i are the activation energies and frequency factors, espectively; N_i, n_i, and m_s denote the concentrations of trap states, electrons trapped in 'active' traps

and holes trapped in recombination centres. As there are no more electrons and holes in traps than one, the last two variables are constant (or simply $=0$ when the trap is occupied). For the sake of simplicity it was assumed that all electron traps have the same activation energies $E = 0.9\,\mathrm{eV}$, also the recombination centres have the same parameters. The simplest 1-D model presented in Fig. 1 is the fixed distance (FD) model. Electrons in excited states can move to neighbouring pairs. The transition probability A_{tr} could be assumed to be thermally activated, i.e.:

$$A_{tr} = v_{tr} \exp\left(\frac{-E_{tr}}{kT}\right) \tag{4}$$

or not activated: $A_{tr}=\mathrm{const}$. E_{tr} represents the height of potential barrier between two adjacent states. In general, the coefficient v_{tr} could depend on the distance between trap levels. This situation is presented in the next diagram in Fig. 1—the variable distance (VD) model. Here, the transition probabilities may vary along the track.

The first two models (FD and VD) force an excited electron to move only through 'active' excited states, i.e. states from which the carrier could be captured by a recombination centre or an 'active' trap. Therefore, these models do not take into account the influence of other shallow traps that are also created along the track. If a carrier spends significant amount of time in these places it should be also considered by an appropriate model. The variable distance model with intermediate states (VDIS-a) presented in the next diagram looks complex due to many unknown parameters characterising spatial and energy distribution of the shallow traps. Nevertheless, each chain of traps between 'active' excited states can be well approximated by an additional level with the average charge carriers lifetime τ_i (VDIS-b). Thus, the computation method does not differ significantly from the VD model. An exact VDIS-b model should include a 'memory' function build into the probability distribution of times $\tau_i(t)$ to remember where the incoming electron comes from. However, numerical experiments show that neglecting this effect does not change the global kinetics considerably.

Real tracks in TL detectors are not strictly one-dimensional. They have a finite thickness. Thus, escaped charge carriers may omit nearest neighbours to recombine with a farther recombination centre. To include this important feature into the model an additional set of outer shallow traps (OST) is suggested. Carriers go to these traps through the local excited state. This situation is presented in the last diagram of Fig. 1. Let us note that it is not necessary to consider additional transitions between intermediate states and OST. The whole charge carrier traffic coming to/from OST through intermediate states can be included into the account of transition rates between OST and excited states.

3. Numerical calculations

The principles of application of the Monte Carlo method to the calculation of TL kinetics have been described in some previous papers (Mandowski and Świątek, 1992, 1997; Mandowski, 1999). To apply the algorithm for 1-D models presented in Fig. 1, one has to the consider additional transitions between localised pairs—e.g. given by Eq. (4). In such a case, the transition times are represented by a non-homogeneous exponential distribution function (Mandowski and Świątek, 1992). It is essential that the whole track has to be considered simultaneously during a single computational run. In the present calculations long chains consisting of 250 localised hole–electron pairs were considered. Periodic boundary conditions were applied to avoid difficulties with tracks which are too short. For the sake of simplicity it was assumed in all calculations that 'active' traps have the same activation energy $E = 0.9\,\mathrm{eV}$ and the frequency factor $v = 10^{10}\,\mathrm{s}^{-1}$. Therefore, according to standard TL models such a system should produce glow curve consisting of a single peak. Below some characteristic features of the first two models shown in Fig. 1 are discussed.

The fixed distance model is the simplest one. Nevertheless, it clearly demonstrates some properties typical for all models presented in Fig. 1. In Fig. 2 three TL curves are shown calculated for the FD model with a thermally activated transition rate (4). For the first two cases (Figs. 2A, and B) one can easily distinguish two peaks. As the A_{tr} increases (Fig. 2C) those peaks merge into a single second-order peak. This property is typical also for a constant transition rates $A_{tr}=\mathrm{const}$. Two example curves of this kind are shown in Fig. 3.

The explanation of the high-temperature peak is very simple. It comes from charge carriers that avoided nearest-neighbour recombination by escaping from its native electron–hole system. The carriers recombine in another place occupying a different recombination centre. Therefore, residual charge carriers have to look for a free recombination centre along the track.

For very low retrapping coefficients the additional high-temperature peak decreases significantly and is almost invisible. Explanation of this phenonenon is obvious. With no retrapping the travel of a charge carrier along track is much faster so, it is easier to find a free recombination centre. The appearance of the transport (or 'displacement') peak is common for all models (Fig. 1).

Calculations performed for VD model show another interesting property. Due to variable transition probabilities some of the localised pairs tend to join together forming clusters. TL properties in such systems were extensively studied (Mandowski and Świątek, 1997; Mandowski, 1999), therefore there is no need to repeat the conclusions. As the separation between hole–electron pairs is a random variable, the clusters created along the track have different size.
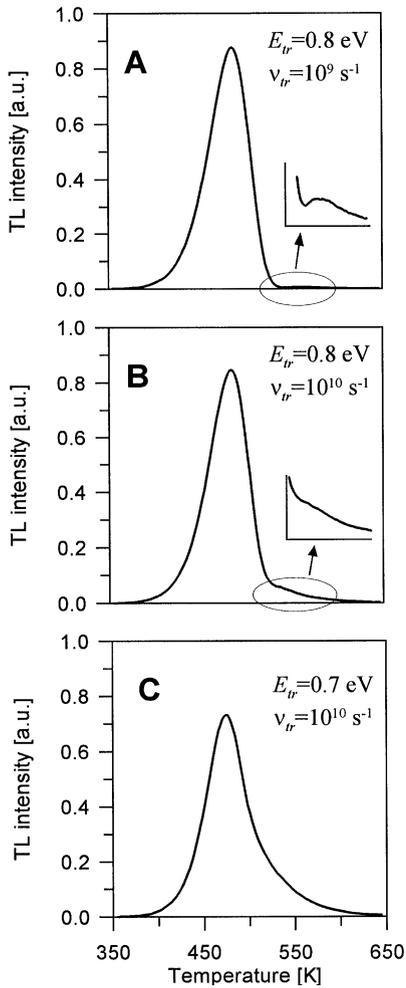
Fig. 2. TL curves calculated in the framework of the fixed distance model (Fig. 1) with thermally activated transition rate. Trap parameters: $E = 0.9$ eV, $v = 10^{10}$ s^{-1} and the retrapping coefficient $r = 100$. Parameters of transition rates are shown in the diagrams.

Therefore, the resulting glow curve is a sum of individual curves coming from single electron–hole pairs and groups containing two, three and more pairs. The resulting peak is usually broad and does not reveal its internal structure. Typically, the peak is followed by a transport peak. For the sake of clarity, in Fig. 4 is a very special curve of this kind is presented. It comes from a system consisting of only two-pair clusters. Transitions are thermally activated according to Eq. (4).

4. Conclusions

Thermoluminescence phenomena in radiation detectors are complex and still far from being well understood.
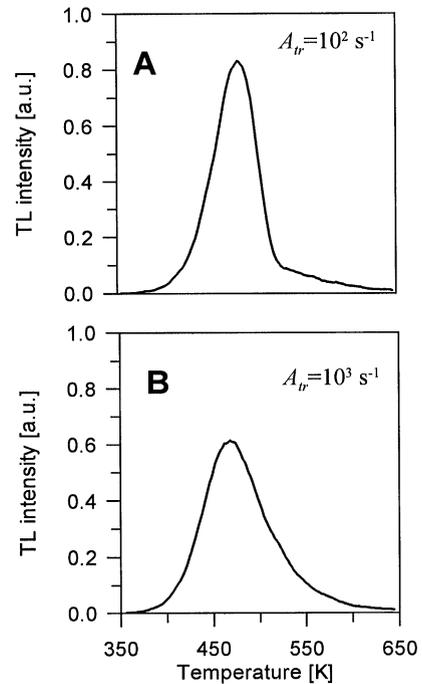

Fig. 3. TL curves calculated in the framework of the fixed distance model (Fig. 1) with a constant transition rate. Trap parameters the same as in Fig. 2. Parameters of transition rates are shown in the diagrams.

The one-dimensional kinetic models presented here could help in a more realistic description of charge carrier relaxation in TL detectors. Numerical analysis performed for the FD and VD models indicates two important features of 1-D TL kinetics: the possibility of formation of a high-temperature transport peak, and the possibility of clustering of hole–electron pairs. Each of these features affects the measured TL glow curves. Applying standard analysis (e.g. glow curve deconvolution) to the obtained data one gets results that do not relate to any real parameters of the studied system. Kinetic parameters of the transport peaks have usually enormously high values of the activation energies. For example, the fitted activation energy of the transport peak shown in Fig. 2A is $E_{tr} = 1.51$ eV. The best fit was obtained using second-order kinetics. Assuming that conductivity of the considered system is proportional to the density of electrons in excited states, it can be shown that similar properties one can notice in TSC measurements. Moreover, the high-temperature TSC peaks are much higher than TL ones (Mandowski and Świątek, 2000). To apply the models to the experimental data it is also necessary to include many additional factors such as the occurrence of various trap levels and non-radiative centres. It is essential also to construct an analytical model of TL dependent
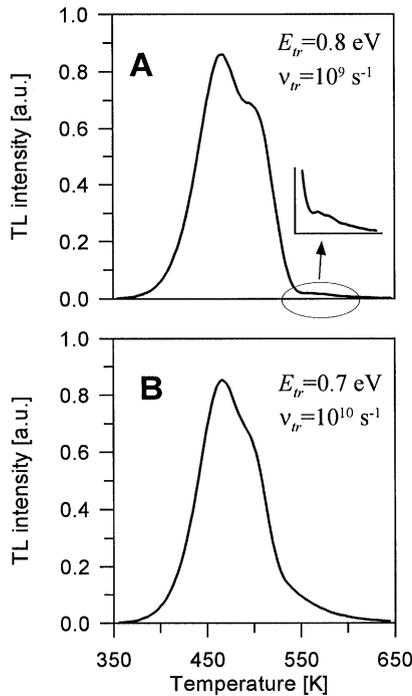
Fig. 4. TL curves calculated in the framework of the variable distance model (Fig. 1) with thermally activated transition rate. The system is characterised by two thermally activated transition rates: the faster one $E_{tr1} = 0.6$ eV, $v_{tr1} = 10^9 \, s^{-1}$ and the slower one with parameters given in the diagrams. Trap parameters the same as in Fig. 2.

on the spatial distribution of traps and recombination centres.

Acknowledgements

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