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Thermoluminescence glow curve deconvolution of LiF:Mg,Cu,Si with more realistic kinetic models

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HIGHLIGHTS

- TL glow curve deconvolution employing generalized model.
- Validation of the model through an artificially generated glow curve.
- Application in the identification TL kinetics of LiF:Mg,Cu,Si TLD.

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ABSTRACT

Thermoluminescence (TL) glow curves of LiF:Mg,Cu,Si were deconvoluted with the introduction of enhanced physical model which envisages that both electrons and holes, produced by ionization radiation and trapped at the respective traps, can be thermally released into the conduction and the valence band, respectively and the holes may also radiatively recombine with electrons at the electron recombination centers. The model is more generalized than the ordinary trap interaction model which only permits the traffic of electrons through the conduction band. An effective numerical analysis method was developed to calculate the glow curve to be compatible with the measured curves. The validity of the numerical method was verified through artificially generated TL glow curves for a wide range of trap parameters. In order to identify TL kinetics of LiF:Mg,Cu,Si with higher accuracy, its glow curves were deconvoluted for two more generalized models, namely, the Schön–Klasens model and the Chen–Pagonis–Lawless model as well as the ordinary trap interactive model. The parameters in the more generalized multi-trap multi-recombination center (MTMR) model were found to be consistent with the quasi-static approximation(QSA) method.

1. Introduction

Thermoluminescence (TL) glow curves acquired with a linear time–temperature profile usually consist of several overlapping peaks. Deconvolution of complex glow curves into their individual components is used to obtain information on the luminescence mechanism (Chen and Pagonis, 2011). A widely accepted model for the deconvolution of TL glow curves is based on the one trap one recombination center (OTOR) model. In the OTOR model, each peak in the glow curve is a result of the flow of charge from the trap to the recombination center (RC) through the conduction band. In reality, once the trapped electrons are released into the conduction band due to thermal stimulation, the electrons lose the memory of the traps and may get re-trapped into other traps. Accordingly, the multi-trap multi-recombination center (MTMR) model which allows the interaction among the traps could be considered to be more realistic. However, it is difficult to use the MTMR model for routine deconvolution as considerable amount of time would be spent to process the required equations for the flow of the electrons and the holes.

Chung et al. (2011) arrived at an efficient method of numerical analysis for the simultaneous equations without adding to the additional constraints such as quasi-steady assumptions. The results of new analysis were demonstrated on the TL glow curves of LiF:Mg,Ti (TLD-100) by adopting 5 traps and 1 RC (Chung et al., 2012). The aim of this study is to explore the analysis method by introducing more generalized models as a continuation of the aforementioned study and to report the analysis results for the TL glow curve of LiF:Mg,Cu,Si TLD. The models introduced here are similar to the ordinary MTMR model except that the holes from

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the hole traps could also be excited to valence band by thermal stimulation and recombined with the trapped electrons leading to the emission of photons. Two new models of such potentiality are introduced into the present study along with the one used previously. One is the Schön–Klasens model in which both electrons and holes could be released from their respective traps during thermal stimulation and both electron–hole (e–h) and hole–electron (h–e) recombination transition are radiative (McKeever et al., 1985). The other is the Chen–Pagonis–Lawless model, which introduced a distinct luminescence center (Chen et al., 2008). In the case of Chen–Pagonis–Lawless model, the direct transition of the holes from the valence band to the electron traps did not exist and the specific hole traps were introduced as the luminescence center.

2. Model and method

Fig. 1 shows a schematic energy level diagram of a general MTMR model with several electron and hole traps. The TL process owes to the presence of the traps between valence and conduction bands. In most of the cases including ordinary MTMR model, the TL is explained by the release of the trapped electrons on thermal stimulation and their recombination with the holes at the recombination centers (RC) via conduction band. In the general MTMR model, an inverted situation of release of holes and their recombination with the electrons via valence band can also take place. In fact, the co-existence of both of the processes of release of electrons leading to e-h recombination and release of holes leading to h-e recombination should also be possible. Evidently, the difference between the ordinary MTMR model and the general MTMR model is that the ordinary MTMR model does not envisage the mobility of hole unlike general MTMR model. RC could be either a hole RC or an electron RC. Here, n_i, m_j, n_c and m_v are the electron concentration for the i-th electron trap, the hole concentration for the j-th hole trap, electron concentration in the conduction band and the hole concentration in the valence band, respectively. The parameters (N_i, A_{ni}, A_{pi}, E_i, s_i, σ_i) belong to the i-th electron trap. They represent the concentration of the traps (N_i), the re-trapping probability of electrons (A_{ni}), the recombination probability of electrons with holes (A_{pi}), the activation energy (E_i), the pre-exponential factor (s_i) and photoionization cross section of the trap (σ_i). The parameters (M_j, A_{qj}, A_{mj}, E_j, s_j, σ_j) belong to the j-th hole trap are similar to the parameters of electron trap as indicated in Fig. 1. The equations governing the process during the stimulation are,

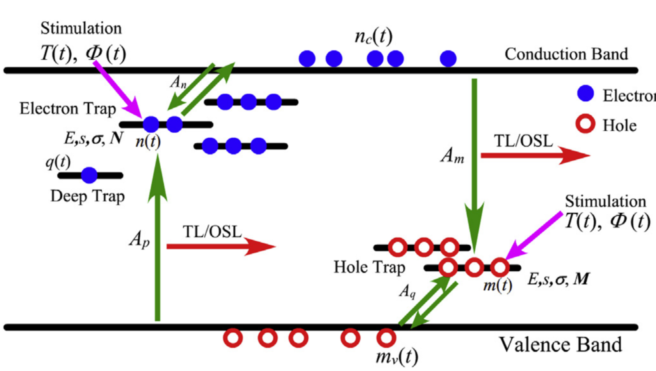

Fig. 1. A schematic energy level diagram of the general MTMR model with several electron hole traps. Both of the traps can also be regarded as thermally and/or optically stimulated simultaneously. Both kinds of traps can also be treated as a luminescence center by capturing the hole in the valence band and the electron in the conduction band respectively.

$$\frac{dn_i}{dt} = -p_i n_i + A_{ni}(N_i - n_i)n_c - A_{pi}n_i m_v, \quad (1)$$

$$\frac{dm_j}{dt} = -p_j m_j + A_{qj}(M_j - m_j)m_v - A_{mj}m_j n_c, \quad (2)$$

$$\frac{dn_c}{dt} = \sum_i p_i n_i - \sum_i A_{ni}(N_i - n_i)n_c - \sum_j A_{mj}m_j n_c, \quad (3)$$

$$\frac{dm_v}{dt} = \sum_j p_j m_j - \sum_j A_{qi}(M_j - m_j)m_v - \sum_i A_{pi}n_i m_v. \quad (4)$$

where p_i (p_j) is the rate of stimulation of electrons (holes) from the i-th electron (j-th hole) trap is related to the temperature $T(t)$ and the photon flux $\Phi(t)$ and is described by

$$p_i = s_i \exp(-E_i/kT) + \sigma_i \Phi. \quad (5)$$

The TL/OSL emission, shown in Fig. 1 by two horizontal arrows, is associated with the recombination of thermally released electrons with the holes and thermally released holes with the electrons and can be expressed as

$$I = \sum_i A_{pi}n_i m_v + \sum_j A_{mj}m_j n_c. \quad (6)$$

The quasi-static approximation (QSA) method developed by Chung et al. (2011) was extended to the generalized models as follows. The flow equation for n_i, m_j, n_c and m_v is basically formulated as

$$\frac{df}{dt} = \alpha - \beta f. \quad (7)$$

Here, f can be any of n_i, m_j, n_c and m_v with positive values of α and β in each of the four cases. For example, when f is n_c, α and β are expressed as,

$$\alpha = \sum_i p_i n_i, \quad (8)$$

$$\beta = \sum_i A_{ni}(N_i - n_i) + \sum_j A_{mj}m_j. \quad (9)$$

In the QSA method, α's and β's are regarded as constants where in α_0's and β_0's are the values defined at the initial time, t_0. Eq. (7) can be solved by,

$$f(t_0 + \Delta t) \approx f_0 e^{-\beta_0 \Delta t} + \frac{\alpha_0}{\beta_0}\left(1 - e^{-\beta_0 \Delta t}\right). \quad (10)$$

The approximate solutions for n_i, m_j, n_c and m_v can be obtained from their initial values and the values of the corresponding α and β. This f in the Eq. (10) does not diverge nor oscillate in the subsequent calculating process as α and β have positive values. The computer program developed by Chung et al. (2012) was extended to realize this algorithm.

For some specific TL models, not every transition for the electrons and holes shown in Fig. 1 is allowed. As indicated earlier, this study attempted to deconvolute the glow curves by using three models, which include (1) the ordinary MTMR model, (2) the Schön–Klasens model and (3) the Chen–Pagonis–Lawless model. The illustration shown in Fig. 1 is for the Schön–Klasens model. The trap parameters related to different kinetic models are summarized in Table 1.