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An algorithm for the integrated deconvolution of radioluminescence and thermally/optically stimulated luminescence glow curves

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HIGHLIGHTS

- RL, TL and OSL glow curve deconvolution employing interacting model.
- Simulation both irradiation and TL/OSL readout stages for various dose level.
- Application in the identification OSL kinetics of Al_2O_3:C.

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ABSTRACT

Radioluminecscence (RL), the light emitted from a material immediately upon ionizing radiation, has been used to detect the dose rate. On the other hand, thermoluminescence (TL) and optically stimulated luminescence (OSL) by the stimulation with the heat and light after irradiation have been used to find out the cumulated radiation dose. Because it was considered as effective to handle these three phenomena in integration to estimate the energy band structure of the material precisely, an algorithm for calculating the glow rapidly and consistently was developed in this study when three types of stimulations including irradiation are applied concurrently or sequentially. The deconvolution using this algorithm can decide the properties contained in the material more precisely because the glows from different stimulations are related frequently to the different aspects of the properties. The computer program to realize the deconvolution by means of these schemes was also developed and it was applied to the glow curves of RL and OSL from Al_2O_3:C to evaluate the efficiency of the developed algorithm.

1. Introduction

Radioluminescence (RL), thermoluminescence (TL) and optically stimulated luminescence (OSL) are the luminescence commonly related to the energy level of materials (Chen and Pagonis, 2011). These are the emission of light from a crystalline material such as insulator or semiconductor by recombination of the electrons and holes created by the irradiation immediately (RL), with heat stimulation (TL) or with light stimulation (OSL). The energy structure of the crystal can be found by analyzing the glow curves corresponding to specific stimulations. Provided that the electrons and holes are stably captured within the trap positioned at the forbidden band of the crystal, the absorbed dose on the material can be estimated by means of TL and OSL. Accordingly, these phenomena are being broadly used in dosimetry as well as in dating of archaeological samples (McKeever, 1985; Botter-Jensen, 2003). However, RL is used to measure ionizing radiation in real time by detecting the light emerged from a material when it is irradiated. Even though the mechanisms of RL, TL and OSL are identically related to the energy band structure, they have been studied independently in most cases.

Because the traps are created by the presence of impurities or crystal defects in crystal, it is necessary to find the trend of differing the energy level structure depending on the complication of impurities injected into the material or the manufacturing process in order to develop an efficient dosimetric material. For these purposes, the glow curve deconvolution analyzing the trap information from the glow curve has been used. Many algorithms and computer programs for the glow curve deconvolution have been developed for dosimetric purposes (Chen and Pagonis, 2011). But most of these algorithms are based on the assumption that each trap

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independently composes one glow curve. This assumption is based on a simplified model of one trap and one recombination center (OTOR) which make a glow peak. The superposition of these peaks makes the whole glow curve consisting of many overlapping peaks. It is general to interpret a certain glow curve by making further assumptions (Halperin and Braner, 1960). But this model does not reflect the physical reality because the interaction possibility of the electrons or holes moving between traps via the conduction band or the valence band is excluded (Sakurai, 2001). Therefore, we developed the appropriate numerical analysis method to solve the flow equations reflecting all possible flows of electrons and holes through the conduction band and through the valence band. By applying this method, the glow curves of TL and OSL was analyzed efficiently (Chung et al., 2011, 2012, 2013, 2014). However, the glow curves are not analyzed easily which require several types of trap because there are many parameters in each trap, and the flow equations of the electrons and holes are closely related each other. It is due to the fact that particular parameters are situated to affect the glow less or two or more parameters affect to the glow in combination under the measuring conditions. A set of the glow curves measured by wide range of radiation dose and various profiles of stimulation shall be analyzed on the same specimen integrally in order to avoid such ambiguities. Nevertheless, this method is limited in use because the repeated measuring on the same specimen may change the physical properties such as the accumulation of the electrons in deep trap and etc.

The algorithm analyzing glow curves emitted from the concurrent or sequential application of ionizing radiation, thermal stimulation and light stimulation was developed in this study. The computer program realizing and investigating this algorithm was developed as well. The cases of continuous measuring of RL and OSL, i.e. after applying the irradiation for certain duration and the light stimulation after a short relaxation time on the specimen, could be taken as an example. It was expected that each parameter can be calculated more precisely when two glow curves were analyzed integrally because many parameters on the trap contributed to RL and OSL differently in this case.

2. Model and numerical analysis

Electron-hole pairs are created when the electrons in the valence band are excited by the irradiation on the crystal with the energy level model shown in Fig. 1. The level under the conduction band caused by the imperfection of crystal acts for the electron trap because it is initially empty and can accept the electrons. On the other hand, the level just above the valence band acts for the hole trap because it is initially filled with electron and can accept the holes. The electrons and holes captured with these traps could keep the position stably when there is no other stimulation. The set of coupled differential equations governing the process during the excitation and stimulation by heat or light is

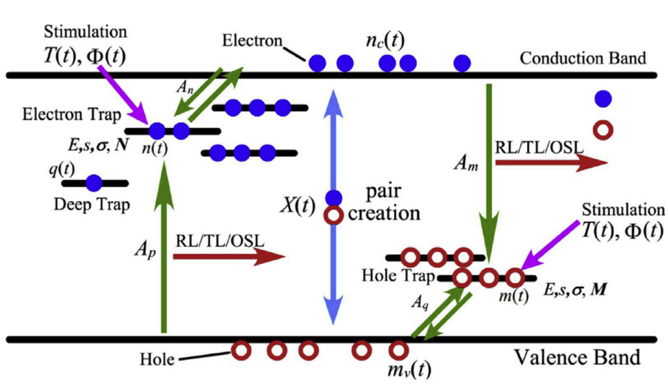

Fig. 1. Schematic energy–level diagram with various kind of electron trap and hole trap. Transitions of electron and hole occur both during excitation (X) and during stimulation by heat (T) or light (Φ).

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

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

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

$$\frac{dm_v}{dt} = X + \sum_j p_j m_j - \sum_j A_{qj}(M_j - m_j)m_v - \sum_i A_{pi}n_i m_v. \tag{4}$$

Here, n_i, n_c, m_j and m_v are the occupancies for the i-th electron trap, for the conduction band, the j-th hole trap and for the valence band respectively and all of them are functions of time. The coefficients (N_i,A_{ni},A_{pi}) related to the i-th electron trap are the concentration, the trapping probability from conduction band and the recombination probability for capturing holes from the valence band, respectively. Actually the finally mentioned recombination process is the transition of electrons from this trap to the holes in the valence band and light could be emitted from there as shown for a horizontal arrow in Fig. 1. (M_j,A_{qj},A_{mj}) related to the j-th hole trap are similar to the parameters of the electron trap. The final recombination process is also the transition of electrons in the conduction band to the holes in the hole trap and light could be emitted also. X denotes the transition probability of electrons from the valence band to the conduction band and it is proportional to the dose rate of excitation. It is understood as the production of electron–hole pairs. p_i (p_j) is the transition probability rate of electrons (holes) stimulated out of the i-th electron traps (j-th hole trap) per unit time. This rate is related to the temperature $T(t)$ and the photon flux $\Phi(t)$ and it is described by the following:

$$p_{i,j} = s_{i,j}\exp(-E_{i,j}/kT) + \sigma_{i,j}\Phi. \tag{5}$$

Here, E_i, s_i and σ_i are the activation energy, preexponential factor and photoionization cross section of the i-th electron trap respectively. (The subscript j notes for the hole trap). X or p is the factor creating or exciting the electron and hole, and eventually induced the flow of charge. Table 1 displays how these factors are related to three processes of irradiation, relaxation and stimulation. The luminescence from the irradiation process is RL and the lights from the stimulation process are TL and OSL.

The emitted RL/TL/OSL light, shown as the horizontal arrows in Fig. 1, is related to the rate of recombination of free electrons with holes, which is formulated by

$$I = \sum_i A_{pi}n_i m_v \eta_i + \sum_j A_{mj}m_j n_c \eta_j. \tag{6}$$

Here, η_i as luminescence efficiency associated with the thermal-

Table 1
The existence of X and p in each irradiation, relaxation and stimulation stage.

Stage	Irradiation	Relaxation	Stimulation
X	$X \neq 0$	$X = 0$	$X = 0$
p_i	$p_i = 0$	$p_i = 0$	$p_i \neq 0$