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# An algorithm for unified analysis on the thermoluminescence glow curve

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## HIGHLIGHTS

- TL glow curve deconvolution employing interacting model.
- Simulation both irradiation and TL readout stages for various dose level.
- Application in the identification TL kinetics of LiF:Mg,Cu,Si TLD.

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## ABSTRACT

An algorithm was developed to integrally handle excitation by radiation, relaxation and luminescence by thermal or optical stimulation in thermoluminescence (TL) and optically stimulated luminescence (OSL) processes. This algorithm reflects the mutual interaction between traps through a conduction band. Electrons and holes are created by radiation in the beginning, and these electrons move to the trap through the conduction band. These holes move to the recombination center through a valence band. The ratio of the electrons allocated to each trap differs with the recombination probability and these values also relevant to the process of luminescence. Accordingly, the glow curve can be interpreted by taking the rate of electron–hole pairs created by ionizing radiation as a unique initial condition. This method differs from the conventional method of interpreting the measured glow curve with the initial electron concentration allocated to each trap at the end of irradiation. A program using the Visual Studio's C# subsystem was made to realize such a developed algorithm. To verify this algorithm it was applied to LiF:Mg,Cu,Si. The TL glow curve was deconvoluted with a model of five traps, one deep trap and one recombination center (RC).



## 1. Introduction

Thermoluminescence (TL) and optically stimulated luminescence (OSL) are phenomena of light emission that take the form of a glow curve, which is produced by thermal and optical stimulation of pre-irradiated crystalline or polycrystalline materials (Chen and Pagonis, 2011). To extract detailed information about radiation exposure, recorded glow curves are usually analyzed by computerized deconvolution methods (Horowitz and Yossian, 1995). A widely accepted model for the deconvolution of TL glow curves is based on the one trap-one recombination center (OTOR) model. Recently, a new algorithm was developed (Chung et al., 2011) that has been used to numerically analyze the TL glow curves of LiF:Mg,Ti (TLD-100) (Chung et al., 2012) and LiF:Mg,Cu,Si TLD materials. These experimental glow curves were analyzed by assigning five interacting traps, one thermally disconnected deep trap and one recombination center (RC) (Chung et al., 2013). The deconvolution algorithms or computer programs are up to date and focus on resolving the initial concentration of electrons in active traps without considering the irradiation process of the material. However, the concentration of the electrons and holes in the traps during the course of irradiation are also governed by the physical parameters of the traps. Accordingly, it is meaningful and necessary to comprehensively consider the radiation excitation process and the stimulation process through heating. This study is an extension of the previous study of Chung et al. (2013), who considered only the TL glow process.

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## 2. Model and numerical method

The algorithm for the unified deconvolution of TL glow curves from the radiation stage to the read-out stage is based on the model shown in Fig. 1. Fig. 1 also shows a schematic energy level diagram of the general interacting model with several traps and RCs of multi-trap-multi-recombination (MTMR). In this algorithm, (1) the creation of electrons and holes is paired with a rate $X$, (2) the procedure of relaxation with sufficient time passing as $n_c$ and $m_v$ decays to negligible values, and (3) the readout procedure of the glow curve by thermal and/or optical stimulation is sequentially considered. The equations governing the process during irradiation, relaxation and stimulation are as follows:

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

$$\frac{dm_j}{dt} = A_{qj}(M_j - m_j)m_v - A_{mj}m_j n_c, j = 1, \cdots, Q, \tag{2}$$

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

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

Here, $n_i$, $N_i$ and $A_{ni}$ denote the occupancy, concentration and retrapping probability of the $i$-th electron trap, respectively. $m_j$, $M_j$, $A_{qj}$ and $A_{mj}$ related to the $j$-th RC are similar to the parameters of the electron trap, as indicated in Fig. 1. $p_i$ is the transition probability rate per unit time of electrons stimulated out of the $i$-th traps. This rate is related to the temperature $T(t)$ and the photon flux $\Phi(t)$ and is described by the following:

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

Here, $E_i$, $s_i$ and $\sigma_i$ are, respectively, the activation energy, pre-exponential factor and photoionization cross section of the $i$-th electron trap. The TL/OSL/RL emission, which is shown in Fig. 1 by a horizontal arrow, is associated with the recombination of thermally or optically released electrons with the holes in the RCs and can be expressed as follows:

$$I = \sum_j A_{mj}m_j n_c. \tag{6}$$

All of the four concentrations of electrons and holes ($n_i$, $n_c$, $m_j$, $m_v$) have negatively proportional time dependencies of their own.

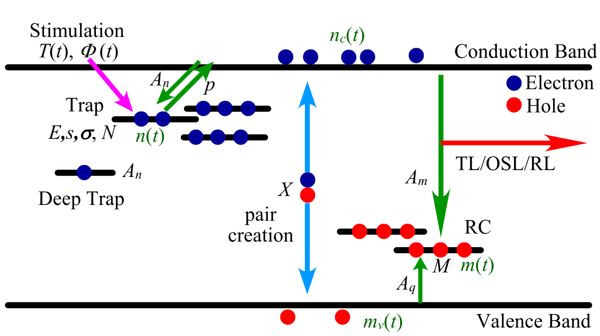


**Fig. 1.** A schematic energy level diagram of the general MTMR model with several electron traps, one deep trap and several recombination centers (RC). The excitation rate by ionizing radiation is given by $X$. In this diagram, $n(t)$, $m(t)$, $n_c(t)$ and $m_v(t)$ may be changed depending on time.

Therefore, the flow equations of these concentrations are basically formulated with the following:

$$\frac{df}{dt} = \alpha - \beta f. \tag{7}$$

In the quasi-static approximation (QSA) scheme, $\alpha$s and $\beta$s are regarded as constants $\alpha_0$ s and $\beta_0$ s, which are the values defined at the initial time, $t_0$. The above equation can be solved by the following:

$$f(t_0 + \Delta t) \approx f_0 e^{-\beta_0 \Delta t} + \frac{\alpha_0}{\beta_0}\left(1 - e^{-\beta_0 \Delta t}\right). \tag{8}$$

A trap has five unknown parameters, including the initial concentration in the TL process. In the case of a RC, it has two unknown parameters. If a material has $P$ traps, one deep trap and $Q$ RCs, $(5P + 2Q + 2)$ unknown parameters have to be decided in the traditional MTMR method considering only the glow process by thermal stimulation. On the contrary, $(4P + 3Q + 3)$ unknown parameters, including $X$ and $A_q$, exist when the analytical method suggested in this paper is used. The quantity of the unknown parameters is reduced because the quantity of traps is usually bigger than the quantity of RCs.

## 3. Deconvolution result of LiF:Mg,Cu,Si

The program that created the algorithm suggested in this paper was made with the C# programming language. The procedures for performing deconvolution for one set of measured data are classified into four parts, as follows. (1) Read the measured data containing information about the time, temperature, flux of stimulation light and TL/OSL glow intensity. At this time, if there is a datum of radioluminescence (RL), which is a glow from the irradiation process, this can be used for the analysis too. The said algorithm can function regardless of the existence of $X$ or $p$, even with the coexistence of both. (2) Decide how many traps and RCs are involved in the TL process by utilizing all information, such as the shape of glow curve, TL/OSL spectrum and photo transferred TL (PTTL). (3) Set the rate of electron–hole pairs created by ionizing radiation, ($X$), irradiation time and relaxation time. At this moment, $X$ will usually be the unknown parameter. The irradiation time and relaxation time will be the known parameter. (4) Perform the regression analysis on the given unknown parameters.

Fictitious TL glow curves with physically reasonable parameters were created and analyzed with QSA to verify the rationale of this algorithm. A value within 0.1% of the initially set value could usually be found. And the glow curves of LiF:Mg,Cu,Si TLD developed at the Korea Atomic Energy Research Institute (Lee et al., 2006) were deconvoluted using the QSA method introduced in this paper. To obtain experimental glow curves of LiF:Mg,Cu,Si, six TLD samples irradiated to various dose levels were used. Dose levels in the range of 50–500 mGy were delivered using $^{137}$Cs gamma rays. The samples did not have any prior thermal or irradiative treatment and were assumed to be in their manufacturing state. The glow curves were recorded on a Harshaw 4500 TLD reader using a heating rate of 1.0 °C/s from room temperature to 300 °C.

The obtained LiF:Mg,Cu,Si glow curve was deconvoluted with five traps, one deep trap and one RC, as shown in the study by Chung et al. (2013). This (deep trap) can be imagined naturally with PTTL measurements utilizing 250 nm UV LED for the light of re-stimulation, glow spectrum measurements and shape of the glow curves. The analyzed results also showed that such an assumption was quite reasonable. The initial concentration of