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An algorithm for the deconvolution of the optically stimulated luminescence glow curves involving the mutual interactions among the electron traps

K.S. Chung [a,*], H.S. Choe [a], J.I. Lee [b], J.L. Kim [b]

[a] The Research Institute of National Science and Department of Physics, Gyeongsang National University, Jinju 660-701, South Korea
[b] Health Physics Department, Korea Atomic Energy Research Institute, P.O. Box 105, Yuseong, Daejeon 305-600, South Korea

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ABSTRACT

The most of the algorithms reported on the deconvolution of the OSL/TL glow curve is basically based on the one trap one recombination center (OTOR) model. In the OTOR model, each individual trap is considered to be independent with each other (mutually exclusive with each other), and the total glow curve is produced solely by the summation of the glow peaks generated from the luminescence emitted by the electrons in one individual trap when transferring to other trap(s). Therefore, there could be a major difference between the model and real physical process of the OSL/TL mechanism. Because the electrons being excited to be in the conduction band barely have past recollection of the original traps, it is widely believed that electrons in one trap can be easily transferred to other trap via the conduction band. Particularly in case of the OSL, the effects of mutual interactions among the traps could be more dominant than those in case of the TL. An algorithm, which can be used to numerically analyze the OSL/TL curves with reflecting the mutual interactions among the individual traps via the conduction band, is developed. This algorithm is able to promptly generate the glow curves for a system with numerous electron traps and recombination centers. Thus, the algorithm can be used to effectively deconvolute the glow curve of a given measurement data.

1. Introduction

Generally speaking, the luminescence including both the thermoluminescence (TL) and optically stimulated luminescence (OSL) is explained by the transition of electrons between the energy band and impurity energy levels. The TL/OSL is a phenomenon of emission of the light produced from the thermal/optical stimulation of the energy previously absorbed in the trap from the exposure to the radiation (McKeever, 1985; Botter-Jensen et al., 2003). To read the information of the radiation exposure, the dependency of a glow curve on the stimulation information is analyzed by the deconvolution method (Horowitz and Yossian, 1995). A generally adopted deconvolution strategy is that a glow curve is decomposed into the several peaks and then, an individual peak is exclusively generated by one particular trap and one recombination center, respectively (i.e., OTOR) (Chung et al., 2010). But there is a huge difference between the OTOR model and physical reality because the electrons being forced to be jumped into the conduction band almost lose the of their previous status history. So interactions between two individual traps via the conduction band could occur, a glow curve might be composed of several individual peaks and it is unable to be considered that the glow curve is composed of a superposition of the several single peaks (Sakurai, 2001; Basun et al., 2003). Several numerically analyzed results with employing a model of mutually interactive traps show that mutual interactions among the traps have significant impacts on the glow curve even though the traps are located sufficiently far apart with each other or thermally isolated (Marcazzo et al., 2007).

Even though the trap-interacting (TI) model is closer to the physical reality in describing the trap transition phenomena, there are still many unresolved problems left to numerically analyze the glow curve with this model. One of the difficulties is that the intermediate results often tend to show the answers in the unphysical region since the simultaneous equations, which describe the flow of electron phenomena, very sensitively depend on the individual electron concentrations. To avoid this kind of unwanted results, the time interval between the individual recursive calculations can be shortened. However, the shortening the time interval in the recursive calculation requires the extended time period for the calculation. Therefore, it might be almost impossible to deploy the TI model in using the regression method, where the undetermined parameters should be defined.

* Corresponding author. Fax: +82 55 753 1334.
E-mail address: chungkisoo@hanmail.net (K.S. Chung).

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The purpose of the present study is to propose a new algorithm, which can be used in the effective numerical deconvolution of the luminescence glow curve with the TI model. Because the algorithm numerically calculates the relatively adequate solutions fairly fast enough without bringing much difficulty such as a divergence in the recursive calculation, it is estimated that the algorithm can be used in the deconvolution of the glow curves of the TL/OSL materials with multi traps and multi recombination centers.

2. The model

The model shown in Fig. 1 is a system possessing several traps and recombination centers assuming that the transitions solely occur via the conduction band. The equations governing the process during stimulations are as follow (Shenker and Chen, 1972):

$$\dot{m}_j = -A_{mj} m_j n_c, \quad j = 1, \ldots, P, \tag{1}$$

$$\dot{n}_i = -p_i n_i + A_{ni}(N_i - n_i) n_c, \quad i = 1, \ldots, Q, \tag{2}$$

$$\dot{n}_c = \sum \dot{m}_j - \sum \dot{n}_i. \tag{3}$$

Where m_j is the concentration of holes in the j-th type recombination center (RC), n_i is the concentration of electrons in the i-th type trap, N is the concentration of the i-th type trap, n_c is the concentration of free electrons in the conduction band and A_{mj} and A_{ni} are probabilities of recombination with the j-th RC and retrapping with the i-th traps respectively. And p_i is the rate of stimulation of electrons from the i-th trap and is related to the temperature $T(t)$ and the photon flux $\Phi(t)$ and is described by as follows:

$$p_i(t) = s_i \exp[-E_i/kT(t)] + \sigma_i \Phi(t), \tag{4}$$

Where E_i, s_i and σ_i as the activation energy, the pre-exponential factor and photoionization cross section of the i-th trap, respectively. The luminescence intensity is $I = \alpha \sum \dot{m}_j$ where the summation is done over only the radiative RCs.

This set of $(i+j+1)$ nonlinear simultaneous equations could be solved by the traditional numerical method. But there is a lot of difficulties to solve these equations. One of difficulties is the "blow up" in calculating the solution at a certain time (Shenker and Chen, 1972). This is caused by the dependency of n_c in Eqs. (1)–(2).

3. The numerical method

The set of equations such as Eqs. (1)–(3) is inadequate for the derivation of numerical solution by the traditional way such as the Runge-Kutta method (RKM). To avoid a blow up of solutions caused by the small relative errors, some parameters set to fixed value at an initial time t_0 as follow:

$$\dot{m}_j(t) \approx -\gamma_{j0} m_j(t), \tag{5}$$

$$\dot{n}_i(t) \approx \rho_{i0} - \eta_{i0} n_i(t), \tag{6}$$

$$\dot{n}_c(t) \approx \alpha_0 - \beta_0 n_c(t). \tag{7}$$

Where γ_{j0}, ρ_{i0}, η_{i0}, α_0, and β_0 have all positive values and are treated as the constants defined at an initial time t_0 as follow:

$$\gamma_{j0} = A_{mj} n_c(t_0), \tag{8}$$

$$\rho_{i0} = A_{ni} N_i n_c(t_0), \tag{9}$$

$$\eta_{i0} = p_i(t_0) + A_{ni} n_c(t_0), \tag{10}$$

$$\alpha_0 = \sum p_i(t_0) n_i(t_0), \tag{11}$$

$$\beta_0 = \sum A_{ni}[N_i - n_i(t_0)] + \sum A_{mj} m_j(t_0). \tag{12}$$

Eqs. (5)–(7) can be solved by,

$$m_j(t_0 + \Delta t) \approx m_j(t_0) e^{-\gamma_{j0}\Delta t}, \tag{13}$$

$$n_i(t_0 + \Delta t) \approx n_i(t_0) e^{-\eta_{i0}\Delta t} + \frac{\rho_{i0}}{\eta_{i0}}\left(1 - e^{-\eta_{i0}\Delta t}\right), \tag{14}$$

$$n_c(t_0 + \Delta t) \approx n_c(t_0) e^{-\beta_0 \Delta t} + \frac{\alpha_0}{\beta_0}\left(1 - e^{-\beta_0 \Delta t}\right). \tag{15}$$

Solving the equations by this kind of method is to derive the approximate solutions while several parameter values are prefixed with the constant values. We often call this kind of method of solving the simultaneous equations to derive the solutions as the Quasi-Static Approximation (QSA). The QSA method never "blows up" compared to other traditional methods in deriving the solutions. As seen in Fig. 2, the concentration functions show the behaviors of the exponential decay modes. Thus, the solutions do not have the negative values or diverge into an infinite value in the calculation. Therefore, individual solutions for the concentration of electrons or holes never approach to the values in the unphysical region even though we extend the values of the time interval Δt in the recursive calculation.

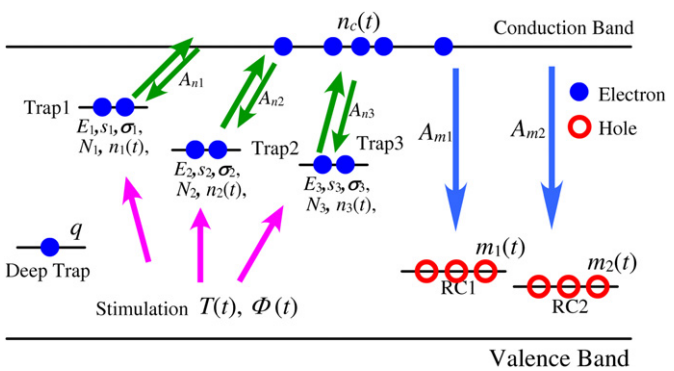

Fig. 1. The energy-level diagram of the multi traps and multi recombination centers model at the presence of thermally disconnected deep trap.

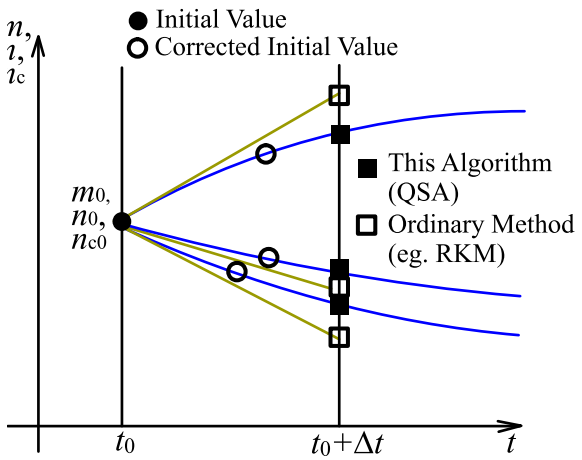

Fig. 2. The behaviors of individual concentrations calculated by the QSA in the course of one time step Δt.

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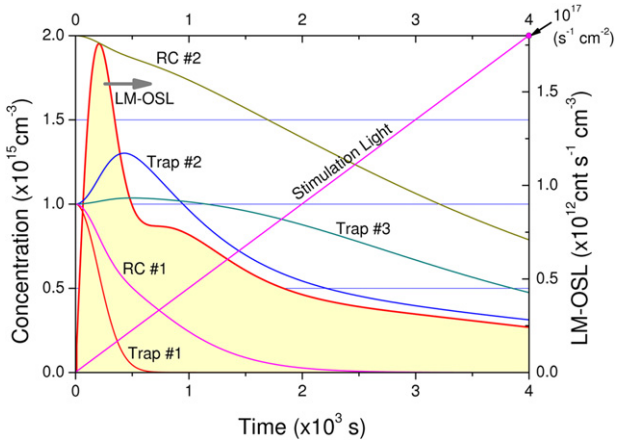

Fig. 3. The time evolution of n_i, n_c, m_j and LM-OSL glow curve by the QSA method with 3 traps and 2 RCs. The input parameters are $n_{10} = 10^{15}$ cm^{-3}, $\sigma_1 = 10^{-18}$ cm^2, $A_{n1} = 10^{-12}$ cm^3 s^{-1}, $N_1 = 10^{18}$ cm^{-3}, $n_{20} = 10^{15}$ cm^{-3}, $\sigma_2 = 10^{-19}$ cm^2, $A_{n2} = 10^{-9}$ cm^3 s^{-1}, $N_2 = 10^{17}$ cm^{-3}, $n_{30} = 10^{15}$ cm^{-3}, $\sigma_3 = 10^{-20}$ cm^2, $A_{n3} = 10^{-11}$ cm^3 s^{-1}, $N_3 = 10^{18}$ cm^{-3}, $m_{10} = 10^{15}$ cm^{-3}, $A_{m1} = 10^{-7}$ cm^3 s^{-1}, $m_{20} = 2 \times 10^{15}$ cm^{-3}, and $A_{m2} = 10^{-8}$ cm^3 s^{-1}.

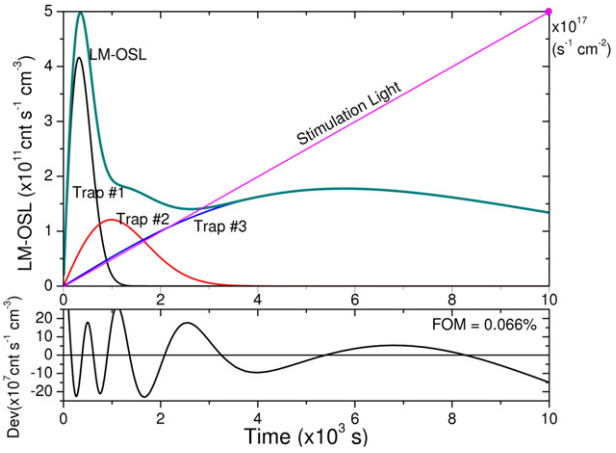

Fig. 4. Glow curve fitting with OTOR scheme and its residue for a glow curve generated by the conditions given in Table 1.

However, these kinds of solutions derived with the QSA method have some weak points so that the charge conservation is not automatically satisfied. To overcome this kind of difficulty, we introduced two proposed methods in the algorithm. One of them is to apply the predictor-corrector method in the calculation. That is we firstly take the initial values of γ, ρ, η, α and β with those values at "t_0" and then evaluate these values in $t_0 + \Delta t$ by Eqs. (13)–(15). The time average values of γ, ρ, η, α and β at the time interval of $t_0 \sim t_0 + \Delta t$ are applied as initial values once again.

The other method is that we countercheck whether the charge conservation is kept or not at an individual calculation step. If the degrees of deviations in the charge conservation increase more during the calculation, then we decrease the value of the Δt and increase the value vise versa when the charge conservation continues. As results of applying the processes of increasing or decreasing the time intervals in the calculations, we could obtain a speed of few tens of milliseconds in generating the glow curve with a common personal computer in case of complex situations where a total sum of the numbers of traps and RCs becomes about a value of ten. And we maintained a ratio of 1 ppm in a break of the charge conservation rule.

4. Result of numerical analysis and deconvolution

To prove the validity of the QSA method proposed in the study, we could generate the glow curves by imposing several physical conditions. When we compare the resultant solutions calculated from Eqs. (1)–(3) with using the derivation processes by the 6th order Runge-Kutta method and the QSA method, the solutions from the both calculations during the entire processes are consistent with each other within 1 ppm. It was found that we could calculate the solutions more than 100 times faster by using the QSA method than those by using the Runge-Kutta method. From this fact, it is convinced that the application of the QSA algorithm in the glow curve is more helpful in the calculation.

Fig. 3 shows the LM-OSL glow curve of sample materials with three traps and two RCs. It also shows the time evolution of n_i and m_j. The corresponding generation conditions of the glow curve are depicted in the caption of the figure.

The data generated in other conditions was calculated by deconvoluting the glow curve (Chung et al., 2007) with the OTOR and QSA method. The given values and resultant calculating values are listed in Table 1 and they are depicted in Figs. 4 and 5. Although three traps could be identified by the calculation method with using the OTOR model while showing a result of the existence of three RCs, it is proved that the calculation results are far apart from the physical reality. Based on other individual parameters and the values of the FOM estimated by the two methods, it is proved that the QSA method derives the more sophisticated values in the estimation and more physically adequate resultant values than those calculated by the OTOR method.

5. Discussion

A set of simultaneous equations governing the processes of the TL/OSL has been solved with the quasi-static approximation (QSA) method. The resultant values are used to examining the validity of

Table 1
Values of the parameters found by the OTOR and QSA deconvolution method for the glow curves generated from full iteration. The first column shows the input parameters.

Model	n_0 (10^{15} cm^{-3})	σ (10^{-20} cm^2)	N (10^{16} cm^{-3})	A_n (10^{-16} cm^3/s)	m_0 (10^{15} cm^{-3})	A_m (10^{-13} cm^3/s)	FOM (%)
Generation	1.000	100.0	10.00	10.00			
	1.000	10.00	1.000	1.000	3.000	1.000	–
	1.000	1.000	10.00	100.0			
OTOR	0.224	103.5	23.23	0.5091	0.6037	2.411	
	5.681	6.004	2.403	171.5	0.1964	1.001	0.066
	2.615	1.168	17.444	3672	2.573	52.77	
QSA	1.001	100.0	10.03	10.02			
	1.002	9.998	1.012	0.999	2.997	0.999	0.009
	1.001	1.000	9.995	100.0			

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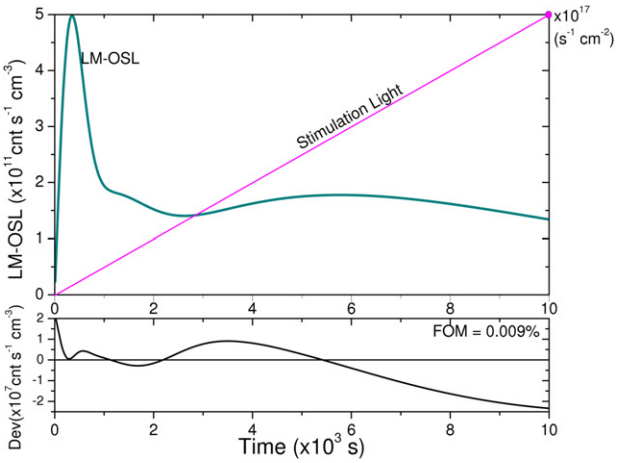

Fig. 5. Glow curve fitting with QSA scheme and its residue for a glow curve generated by the conditions given in Table 1.

the glow data generated by full iterations with the 6th order Runge-Kutta method without imposing additional priori assumptions on the original equations. It is proved that the QSA method could derive the adequate resultant solutions for the practical physical situations with very fast calculation. At the same time, we conducted the deconvolution of the glow curve with employing the QSA method and it is proved that the method almost completely well estimates the glow curve generation conditions. Thus, it is strongly believed that we could apply this kind of algorithm in generating the glow curve of materials with multi traps, multi RCs, and, at the same time, thermally disconnected deep trap.

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References

Basun, S., Imbusch, G.F., Jia, D.D., Yen, W.M., 2003. The analysis of thermoluminescence glow curves. J. Lumin. 104, 283–294.
Botter-Jensen, L., McKeever, S.W.S., Wintle, A.G., 2003. Optically Stimulated Luminescence Dosimetry. Elsevier, Amsterdam.
Chung, K.S., Choe, H.S., Lee, J.I., Kim, J.L., 2007. A new method for the numerical analysis of thermoluminescence glow curve. Radiat. Meas. 42, 731–734.
Chung, K.S., Park, C.Y., Lee, J.I., Kim, J.L., 2010. Development of a new curve deconvolution algorithm for optically stimulated luminescence. Radiat. Meas. 45, 320–322.
Horowitz, Y.S., Yossian, D., 1995. Computerised glow curve deconvolution: application to thermoluminescence dosimetry. Radiat. Prot. Dosimetry 60, 1–110.
Marcazzo, J., Santiago, M., Spano, F., Lester, M., Ortega, F., Molina, P., Caselli, E., 2007. Effect of the interaction among traps on the shape of thermoluminescence glow curves. J. Lumin. 126, 245–250.
McKeever, S.W.S., 1985. Thermoluminescence of Solids. Cambridge University Press, Cambridge.
Sakurai, T., 2001. Fatal defect in computerized glow curve deconvolution of thermoluminescence. J. Phys. D: Appl. Phys. 34, L105–L107.
Shenker, D., Chen, R., 1972. J. Comput. Phys. 10, 272–283.

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