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Development of a new curve deconvolution algorithm for optically stimulated luminescence

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ABSTRACT

A new computerized glow curve deconvolution (CGCD) algorithm for thermoluminescence(TL) and optically stimulated luminescence (OSL) is presented. The proposed approach can be adopted in a numerical curve fitting for obtaining the relevant trapping parameters of a set of glow data taken with both thermal and optical stimulation. This method is based on the one trap one recombination center (OTOR) model with minimal simplifying assumptions. To demonstrate the ability of the method, a new computer program has been developed and tested with some synthetic OSL data.

1. Introduction

Various kinds of computerized glow curve deconvolution (CGCD) algorithms for thermoluminescence (TL) have been developed (Horowitz and Yossian, 1995; Puchalska and Bilski, 2006). Recently, a new CGCD algorithm has been developed by adopting a method called general approximation (GA) which has fewer simplifying assumptions to solve the approximate values of parameters relating TL phenomenon, and used for analysis of glow curves from some TL materials (Chung et al., 2008). By using this algorithm, the understanding of TL traps and TL mechanisms for some TL materials has been improved significantly. However, in the case of optically stimulated luminescence (OSL), the curve deconvolution analysis is carried out mostly within the framework of first order kinetics (1OK) or most generally within the general order kinetics (GOK) (Chen et al., 2009 and Bos and Wallinga, 2009). Note that there has been less investigation on the analysis of OSL curves.

In this study, we have developed a new curve deconvolution algorithm for analysis of TL and OSL curves. This algorithm includes most of all known kinetic models. We have also prepared an efficient computer program with the algorithm which could be used for analysis not only of all the curves measured by the existing OSL measuring modulation such as CW-OSL, LM-OSL, but also the curves for a mode of stimulation light formed with arbitrary functions.

2. Kinetic model and analysis algorithm

Our model treated here is based on the one trap and one recombination center model (OTOR). In OTOR, the time evolutions of concentration of trapped electrons (n), trapped holes (m) and free electrons in the conduction band (n_c) are governed by three coupled rate equations. With physically realistic assumptions, i.e. that the value and the rate of change of n_c are negligible compared with those of n, TL/OSL intensity is expressed as (Halperin and Braner, 1960)

$$I(t) = -\frac{dn}{dt} = \frac{nmp}{m + R(N - n)} \tag{1}$$

where R is ratio of retrapping coefficient (A_n) and recombination coefficient (A_m), N is the concentration of trapping states and p is defined as the probability per unit time of the release of an electron from a trap. There are two possibilities to reduce the number of parameters by applying more simplifying assumptions. One of them is assuming $n << N$ and $RN << m$ or $R = 1$ named mixed order kinetics (MOK) since it contains 1OK and second order kinetics (2OK) in the two extreme circumstances. Another one is assuming $n_0 = m_0$ which means that the initial electron and the initial hole are just created in pairs. We previously carried out

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a research with the second scheme and developed a software package (Chung et al., 2007).

The function $p(t)$ induced by the thermal and optical stimulation intensity is expressed as,

$$p(t) = se^{-E/kT(t)} + \sigma\varphi(t) \quad (2)$$

with s = frequency factor, E = activation energy, σ = photo-ionization cross section, $T(t)$ = temperature of the material at time t and $\varphi(t)$ = stimulation light intensity at time t. We now describe how to find out the solutions of TL/OSL light intensity $I(t)$ from Eq. (1) with $p(t)$ and

$$f(t) = \int_0^t p(\tau)\mathrm{d}\tau = s\int_0^t e^{-E/kT(\tau)}\mathrm{d}\tau + \sigma\int_0^t \varphi(t)\mathrm{d}\tau \quad (3)$$

where $f(t)$ is related to the cumulated stimulation intensity until time t. The charge neutrality condition could be expressed by

$$m(t) = n(t) + q \quad (4)$$

where q is the excess hole concentration owing to the presence of thermally disconnected traps and maintain constant value in the course of TL/OSL emission. Equation (1) can be analyzed by using the inverse function, analogously to that described by Chung et al. (2007):

$$g(n) \equiv -\left(1+\frac{RN}{q}\right)\ln\frac{n}{n_0} + \left(R+\frac{RN}{q}\right)\ln\left(\frac{n+q}{n_0+q}\right) \quad (5)$$

The solution of Eq. (1) is given by

$$g(n) = f(t) \ \ or \ \ n(t) = g^{-1}[f(t)] \quad (6)$$

Even if n is expressed as a function of time t and eventually TL/OSL intensity $I(t)$ also could be derived, two problems are encountered. One is in expressing the function $f(t)$ explicitly and the other is in calculating the inverse function of $g(n)$. These two problems are analogous to the case of $n_0 = m_0$ $(q = 0)$ (GA) treated by Chung et al. (2007).

The procedure to solve Eq. (6) is as follows: (i) $f(t)$ as a function of t, depends on the three trapping parameters E, s and σ. Both E and s are related to thermal stimulation (TL case) and σ is related to optical stimulation (OSL case). Under optical stimulation, it is easy to deal with $f(t)$ because the parameter σ is just a multiplication constant. From the measured $\varphi(t)$ the cumulated stimulation light intensity is evaluated in the first stage and can be used as a basic function in the course of deconvolution. (ii) $g(n)$, as a function of $n(t)$, depends not only on the three trapping parameters R, N and q but also on the initial trapped electron concentration n_0. Because this function shows a monotonously decreasing behavior and its second derivative is positive, the inverse function $g^{-1}(a)$ can be evaluated quickly by Newton's method which is the most widely used technique for finding the roots of functions. (iii) For a given t, $f(t)$ is calculated and subsequently $n(t)$ is evaluated from $g^{-1}(f)$. Once $n(t)$ is calculated as a function of t, the intensity of the TL/OSL emission, i.e., the glow curve $I(t)$ is calculated as a function of t by applying this result to Eq. (1).

To distinguish from GA, our method in this paper is called as 'GA+'. In GA+, similar to MOK, q is one of the crucial parameters. We use the parameter α instead of q as in the case of MOK, defined by

$$\alpha = \frac{n_0}{n_0+q} \quad (7)$$

Thus, GA+ would go to MOK in the limit $n << N$ and $RN << m$ or $R = 1$ and would go to GA in the limit $\alpha = 1$. GOK and MOK also would go to the first order kinetics (1OK) and the second order kinetics (2OK), respectively, in appropriate limiting conditions. Table 1 summarizes the undetermined TL/OSL parameters for various kinetic models including full iteration (FI) where no simplifying assumptions are made with the OTOR.

3. Computer program

An efficient computer program has been developed in order to deconvolute the peaks from the TL and OSL curves. This program is an extended version of TLanal in some respect (Chung et al., 2007). First, MOK and GA+ algorithms are appended to the program, so the parameter a can be retrieved. With a, the excess hole concentration q can be easily evaluated. Secondly, this program is capable of treating both types of stimulation. That is, if some material has traps which can be stimulated thermally and/or optically, it can be analyzed properly. In a separate paper, we have reported the application of MOK and GA+ algorithms to TL (Chung et al., 2008).

Here, we restrict our attention to pure OSL. As previously stated, the $p(t)$ and $f(t)$ have favorable behavior only in the optical stimulation case. On the other hand, it is not easy to handle $f(t)$ when the thermal stimulation is applied because $f(t)$ contains one trapping parameter E, which should be retrieved by optimization procedure, inside the integral. When a certain OSL data set is utilized, the cumulated stimulation light intensity,

$$\Phi(t) = \int_0^t \phi(t)\mathrm{d}\tau \quad (8)$$

is tabulated as a function of t and will have an important role in calculating the curve. When we treat any function in numerical analysis, there is no crucial difference between the function that could be expressed in an explicit form and the function that could be evaluated from a tabulated data array. Moreover, the applied stimulation light intensity can be measured and recorded over a finite time interval as is the OSL signal. Even if the OSL signal is measured using some well-known stimulation modes such as linear (LM-OSL) or hyperbolic (HL-OSL), it is better to use the measured data as the source of stimulation because some differences might exist between the control signal and the response in the real OSL instrument. For example, in pulsed modulation OSL (POSL) the stimulation light has some time lag and there may be some distortion. However, our algorithm uses actual measurement data regardless of the stimulation light modulation type.

4. Computational result and discussion

In order to test our algorithm, reference glow data is generated by using a fully numerical iteration of the three simultaneous equations without any assumption (FI). In this work, we also have generated test data by the FI with variation of σ, R and a. In using

Table 1
Undetermined TL/OSL parameters for various kinetic models

Abbreviation	Kinetic model	Undetermined parameter
1OK	1st Order Kinetics	n_o, E, s, σ
2OK	2nd Order Kinetics	n_o, E, s, σ
GOK	General Order Kinetics	n_o, E, s, σ, b
MOK	Mixed Order Kinetics	n_o, E, s, σ, α
GA	General Approximation	n_o, E, s, σ, R, N
GA+	GA Plus	n_o, E, s, σ, R, N, α
FI	Full Iteration	n_o, E, s, σ, R, N, A_m, α, n_{c0}

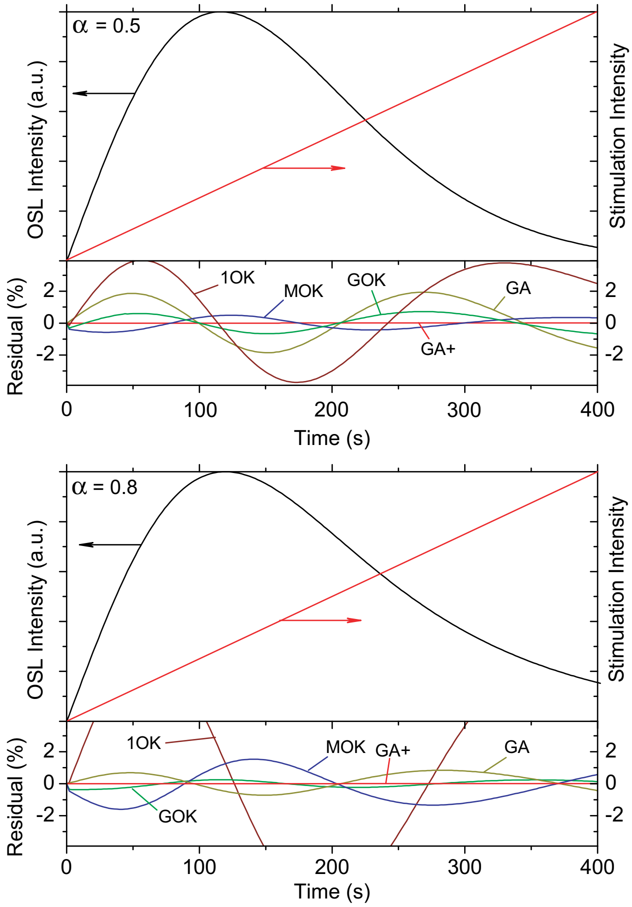

Fig. 1. The reference LM-OSL data generated by FI and the fitting error analyzed by GA+, GA, MOK, GOK and 1OK. The parameters used are given in the text.

the FI, the fifth order Runge-Kutta technique has been employed. Although this program has no restrictions for simulation light profiles and number of traps, LM-OSL mode has been used to synthesize glow peaks for a single trap with the function $p(t) = \sigma\,\gamma\,t$ where $\sigma\,\gamma = 10^{-4}\text{s}^{-2}$. Other parameters used are $n_0 = 1.0 \times 10^9$ cm^{-3}, $R = 0.1$, $N = 1.0 \times 10^{10}$cm^{-3}, $A_m = 1.0 \times 10^{-7}$cm^{3}s^{-1} and $\alpha = 0.1$–1.0. In Fig. 1, several results of fitting are shown for $\alpha = 0.5$ (a) and $\alpha = 0.8$ (b). The GA+ shows the best performance for all the range of generating parameter (FOM = 0.005%). In the GA+ case, the fitted parameters are also nearest to the input parameters. Generally speaking, introducing more undetermined parameters, the quality of the fit will be better. This is natural because GA+ has two extra parameters R and N compared to MOK and has one extra parameter α compared to GA. As seen from the graph which correlates the input a with α_{fit} and b_{fit} (Fig. 2), GA+ has the best performance compared to other methods. Even if in GA+ one neglects the parameter A_m in FI, in most cases, the fitted values almost coincide with the input values.

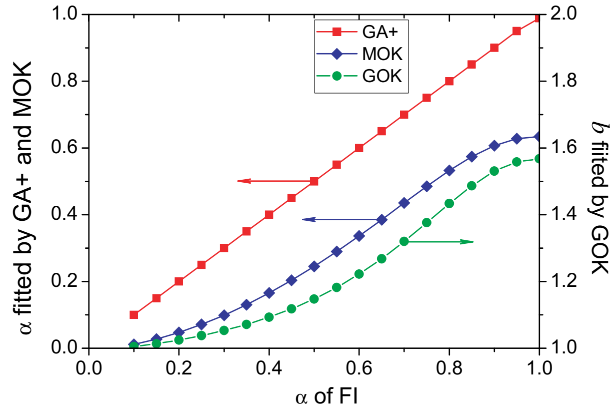

Fig. 2. Correlation between input α and fitted α and fitted b.

5. Conclusion

An efficient computer program, TolAnal, has been developed for the GA+ algorithm. The performance of the program has been enhanced by fast and efficient algorithms. This program is designed to be easily used on any MS Windows-based computer with a graphical user interface and can be used for the deconvolution of the TL/OSL glow curves with not only the GA+ but also the GA and the MOK. There is no restriction in the stimulation profile, i.e., this program can be used in the case where the temperature and/or stimulation light change linearly as well as arbitrarily. A full functional version of this program and example files can be freely downloaded from the web site http://physica.gnu.ac.kr/TLanal.

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