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The influence of competition effects on the initial rise method during thermal stimulation of luminescence: A simulation study

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HIGHLIGHTS

- The IR method is very accurate in single peaks.
- In complex TL glow curves the accuracy of IR depends on the competition between active traps.
- In presence of strong deep trap competitors the competition between active traps is eliminated.
- The IR method is very accurate in absence of competition among active traps.
- The IR method underestimates E of an active trap which acts as competitor.

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ABSTRACT

The initial rise (IR) method is a widely used method to evaluate the activation energy of complex thermoluminescence (TL) glow-curves. It is generally accepted that competition effects take place among the various electrons traps responsible for the TL peaks of a glow curve. In the present study the potential influence of such competition effects on the IR method is studied. The simulation is divided in two parts. In the first part it is assumed that retrapping probabilities are lower than recombination probability. This simulation was carried out assuming either strong or weak competition from a thermally disconnected deep trap (TDDT). In the second part of the simulation the retrapping probabilities are assumed to be greater than the recombination probability. The simulation results showed that competition has serious effects on the resulting glow-curve shapes. Furthermore, the simulation showed that in most cases the IR method is able to reproduce accurately the input values of the activation energies of the three TL peaks. However, it was found that the IR method underestimated the activation energy of the last peak corresponding to the highest temperature by 8–20%, when the retrapping probabilities were higher than the recombination probability.

1. Introduction

The thermoluminescence (TL) glow curves of natural and synthetic materials consist of many TL peaks showing various degrees of overlap. The theoretical description of complex TL glow-curves is attempted by using phenomenological models based on the energy band theory in solids. The main characteristic of all these models is the existence of competition effects among the various trapping levels during both irradiation and during TL readout to record the TL glow curve. However, strictly speaking, when dealing with experimental TL glow-curves, there are no criteria to decide whether competition effects were present during recording of the TL glow curve. On the other hand, the assumption of competition effects is necessary in order to explain various TL effects, especially the superlinear TL dose response in some of the most widely used TL materials, like quartz (Chen et al., 1988) and LiF (Mische and McKeever, 1989).

The experimental TL glow-curves contain all the information about the kinetic parameters and populations of electron traps responsible for each individual glow peak. There are several methods used to extract the kinetic information from glow curves. The initial rise method (IR) is a widely applied method to evaluate

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activation energies of thermoluminescence peaks and is presented in detail in textbooks (Chen and Kirsh, 1981; Chen and McKeever, 1997). Historically the method was introduced by Garlick and Gibson (1948). Later the method was generalized as the fractional glow technique (FGT) by Gobrecht and Hofmann (1966) and Tale (1981). Although the method was widely used, to the best of our knowledge the only simulations available in the literature are those by Kierstead and Levy (1991).

As the IR method involves many partial heatings on the same material, it is logical to assume that competition effects are present at each IR cycle. Taking into account the importance of the IR method itself, as well as the existence of competition effects, it is very useful to simulate the response of the IR method under various degrees of competition.

The goals of this work are:

1. To test the validity of IR on simulated glow curves derived by either elementary phenomenological models (like general order kinetics), or by more complex phenomenological models.
2. To investigate in detail the influence of competition on the initial rise method by varying the parameters in the models.
3. To simulate the potential use of the IR method when one uses an exponential heating function, instead of the commonly used linear heating function.

2. Phenomenological model

A common and very general phenomenological model, which can describe a TL glow-curve consists of a number of electron traps and one recombination center. The system of differential equations in such a model are:

$$\frac{dT}{dt} = \beta, \tag{1}$$

$$\frac{dn_i}{dt} = -n_i\, s_i\, exp\left(\frac{E_i}{k\,T}\right) + A_i(N_i - n_i)n_c \quad i = 1\ldots n \tag{2}$$

$$\frac{dn_d}{dt} = A_d(N_d - n_d)n_c \tag{3}$$

$$\frac{dm}{dt} = A_h(M - m)n_v - A_m\, m\, n_c, \tag{4}$$

$$\frac{dn_v}{dt} = R - A_h(M - m)n_v, \tag{5}$$

$$\frac{dn_c}{dt} = R - \sum_i \frac{dn_i}{dt} + \frac{dn_d}{dt} - A_m\, m\, n_c \tag{6}$$

where the index $i = 1..n$ stands for active the electron traps, $E_i(eV)$ is the activation energy, $s_i(s^{-1})$ the frequency factor, $N_i(cm^{-3})$ is the concentration of available electron traps, $n_i(cm^{-3})$ the concentration of trapped electrons, $M(cm^{-3})$ is the concentration of available luminescence recombination centers, $m(cm^{-3})$ concentration of tapped holes. N_d, $n_d(cm^{-3})$ are the concentrations of available and occupied thermally disconnected deep traps (TDDT), $n_c(cm^{-3})$ and $n_v(cm^{-3})$ are the concentration of electrons in the conduction and holes in the valence band, $A_i(cm^3s^{-1})$ are the trapping probabilities in electron traps N_i, $A_m(cm^3s^{-1})$ is the recombination probability, $A_h(cm^3s^{-1})$ the trapping probability of holes in luminescence centers, $A_d(cm^3s^{-1})$ trapping probability in TDDT, $\beta\ (K/s)$ the heating rate and R is the rate of production of electron-hole pairs (e-h) per second $(e-h/s)$ which it is assumed to be proportional to the dose rate.

In the above model all traps having the index i can trap and release electrons by thermal stimulation, and they are referred to as active traps. On the other hand, the trap with index d can only trap electrons but thermal stimulation is not allowed, so these traps are called thermally disconnected deep traps (TDDT).

3. The competition and its consequences

A simulation study using the above general model involves basically three stages: the irradiation stage, the relaxation stage and the heating stage. The irradiation is simulated by creating electron-hole pairs at a rate R. The holes from the valence band are trapped at luminescence centers, whereas all electron traps compete with each other in trapping electrons from the conduction band. After the irradiation the materials is allowed to relax for a relaxation period, to allow electrons and holes in the conduction and valence band to settle into the various energy levels. During the heating stage the trapped electrons are thermally stimulated into the conduction band. The conduction band electrons have three alternatives routes: (a) to be re-trapped in the active trap from which they were thermally released, (b) to be re-trapped in any other existing deeper active trap, as well as to the TDDT, and (c) to recombine with a hole in a luminescence center producing the TL signal.

It is obvious from the above discussion that competition effects take place during the irradiation, relaxation and heating stages. The competition effects are the basis of explaining many experimental results, with the most important one being the TL dose response in natural and synthetic dosimetric materials. In the present work the influence of competition effects on the IR method will be investigated in detail.

4. The initial rise (IR) method

The IR method is the most valuable technique for the evaluation of the activation energy of an electron trap responsible for a TL peak. The method was introduced Garlick and Gibson (1948), and is described in some detail below.

The analytical expression describing a single TL peak following general order kinetics and measured using a linear heating rate β, is (May and Partridge, 1964; Chen and Kirsh, 1981; Chen and McKeever, 1997):

$$I(T) = n_0 \cdot F \cdot p(T) \cdot FST(T) \tag{7}$$

$$p(T) = \frac{s}{\beta} exp\left(-\frac{E}{kT}\right) \tag{8}$$

$$FST(T) = \left[1 + \frac{b-1}{\beta} F\, s \int_{T_0}^{T} exp\left(\frac{E}{kT}\, dT'\right)\right]^{\frac{b}{b-1}} \tag{9}$$

$$F = \left(\frac{n_0}{N}\right)^{b-1} \tag{10}$$

For the sake of simplicity it is assumed in the simulation that $n_0 = N$. Eqs (7)–(9) are plotted in Fig. 1(a) showing how a TL peak is produced (Chen and McKeever, 1997). According to the detailed description in Chen and McKeever (1997), the initial rise method is applied at the very low temperature side of the TL peak, where the population of the trapped electrons (represented by the n/n_0) is negligibly disturbed. From a practical point of view, the theoretical