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Thermal Fading Rates in LiF:Mg, Ti Main Thermoluminescence Peaks

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ABSTRACT

Thermal fading (TF) is an important characteristic in choosing appropriate thermoluminescence (TL) materials for particular applications. TF is the process of reducing the capability of producing the response due to radiation exposure. The fading rate of LiF based thermoluminescent (TL) material depends on many experimental parameters such as storage temperature, readout mechanism, annealing and radiation type or time based on the storage time before or after radiation.

The general aim of the present work is to investigate if the activation energy *E*, frequency factor s, maximum temperature, T_m and concentration, n values evaluated from the experimental thermal fading rates glow–curves through a computerized glow–curve deconvolution analysis (CGCD), can simulate the thermal fading glow-curves using a phenomenological model for LiF:Mg, Ti.

Keywords: Thermoluminescence, Thermal fading, Activation energy, Frequency factor, Glow curve, Peak deconvolution

1. Introduction

The fading of the thermoluminescent (TL) output signal is a seriously annoying property of thermoluminescent dosimeters (TLDs) that is important to characterize, in order to accurately relate TL output with amount of radiation exposure[1,14-15]. Previous authors used post-irradiation treatments to empty the low temperature TL traps and remove the low temperature glow peaks before readout to reduce or eliminate this fading [5,10,16-17]. Izak-Biran et al. [5] concluded that the application of post-irradiation procedure significantly reduces fading due to the removal of the minimum temperature peaks.

Wide variation in fading characteristics measured for LiF:Mg,Ti demonstrates the complexity involved in characterizing a fading rate [2-4, 15]. The trapping parameters; the activation energy (E) and frequency factor (s) of the glow-peak 3, 4 and 5 of LiF:Mg, Ti have been extensively studied [3, 6-7, 10].

This work was designed to characterize the post-irradiation fading of the individual peaks of glow curves to better understand the fading of the integrated TL response and also the evaluation of a stable and reproducible integrated signal for LiF:Mg,Ti. The details are described separately in the next section.

The general aim of the present work is to investigate if the activation energy E, frequency factor s, maximum temperature T_m and concentration n values evaluated from the experimental thermal fading rates glow–curves through a computerized glow–curve deconvolution analysis (CGCD), can simulate the thermal fading glow-curves using a phenomenological model for LiF:Mg,Ti.

The study was organized accord to the following steps:

Step 1: The CGCD analysis of glow- curve.
Step 2: Valuate E, T_m, s and n for exact glow peak.
Step 3: Simulate the effect using "Mathematica" software.

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2. Experimental

2.1 Instruments and Measurement Conditions

The sample used in these experiments was LiF with dimensions of 8×4×3 mm (available from Bicron).

The experimental Instrument (TL reader Harshaw model 3500) consists of the Photomultiplier Tube (PM) (EMI 9635QA bialkali), high voltage (HT), amplifier, beta source (The Studsvik Irradiator)., furnace etc., which are the common units of any nuclear spectroscopy system (TL among them).The rest of the equipment consists of a chamber which can be evacuated. Inside the chamber there is a metallic plate which can be heated to a constant temperature. The sample is positioned on the metallic plate in front of the PM window.

All measurements were performed in a nitrogen atmosphere with a low constant heating rate of 2°C/s , in order to avoid significant temperature lag. Samples were heated up to the maximum temperature of 350°C.

2.2 Experimental protocol and method of analysis

The experimental procedure for the study of thermal fading was performed according to the following protocol:

Step 1: Test dose was 400 mGy.

Step 2: TL up to the pre-heat temperature T_i.
i= (140, 150, 160, 170, 180, 190, 200 °C).

Step 3: Cool to room temperature.

Step 4: TL up to 350 °C to obtain the residual.

Step 5: Repeat step 1 to 4 for a new T_i.

The computerized glow–curve deconvolution (GCCD) analysis procedure was performed over experimental data obtained [8]. The general order kinetics analytical equation which have been proposed by Kitis et al. [9] was used, as follows:

$$I(T) = \mathrm{I_m}.b^{b/b-1}.exp\left(\frac{E}{kT}.\frac{T-T_m}{T_m}\right)\times\left[(b-1).(1-\Delta).\frac{T^2}{T_m^2}\times exp\left(\frac{E}{kT}.\frac{T-T_m}{T_m}\right)+Z_m\right]^{-b/b-1} \quad (2.1)$$

where $\Delta=\frac{2kT}{E}$, $\Delta_m=\frac{2kT_m}{E}$, $Z_m = 1+(b-1)\Delta_m$ (2.2)

The goodness of fit was tested by the figure of merit (FOM) [13].

2.3 Analysis of experimental results

To eliminate the need to measure and retain absolute calibration information for LiF, the above protocol experiment should be obeyed. Fig. 1 shows the fading TL glow curves of LiF:Mg,Ti after different pre-heat temperatures (140-200°C). The peaks 1 and 2 were removed as it was mentioned in the introduction, the intensity of the glow curves of the glow peaks 5, 4 and 3 decreases when the temperature increases at 200°C.

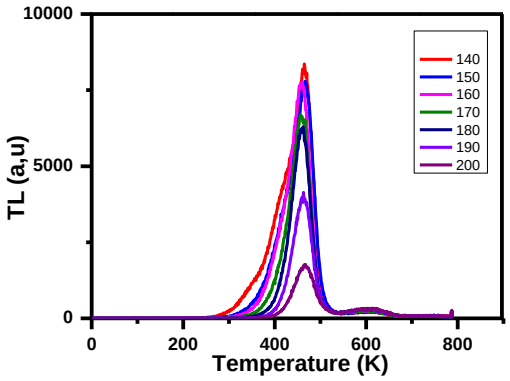


Fig.1 *Thermal treatment for different heating temperatures.*

By fitting the experimental data with Eq. (2.1), the kinetic parameters for each pre-heat temperature was obtained. The typical results of the fitting are shown in Fig. 2 for 140°C (it has to be noted that only first temperature is shown). The calculated kinetic parameters for each of the three peaks are shown in Table 1 and the values are comparable to the Refs [10,11]. The frequency factor was evaluated using the following equation:

$$s_i = \left(\frac{E_i}{Bk\,T_{mi}^2}\right)exp^{\left[\frac{E_i}{(Bk\,T_{mi})}\right]} \quad (2.3)$$

The kinetic order b could be taken as 1.00001 for both peaks as it is lower and small change in T_m for each peak (Fig. 1) cannot be observed easily or accurately during experimental work for all the peaks. In real physical systems consisting of many trapping levels, the competition to the luminescence process will be very strong.

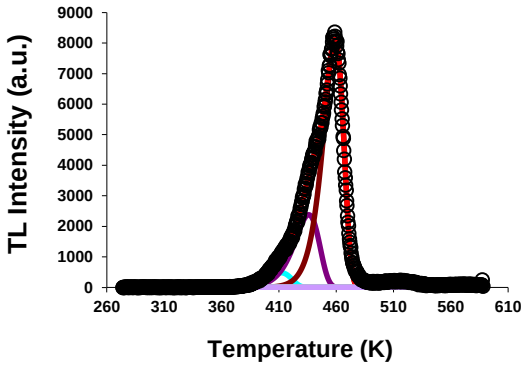


Fig.2 *CGCD of the TL glow curves for LiF (at 5%) at pre-heat temperature of 140°C.*

Figs. 3 and 4 represent real effects of the pre-heat temperature at the intensity and peak integral respectively.

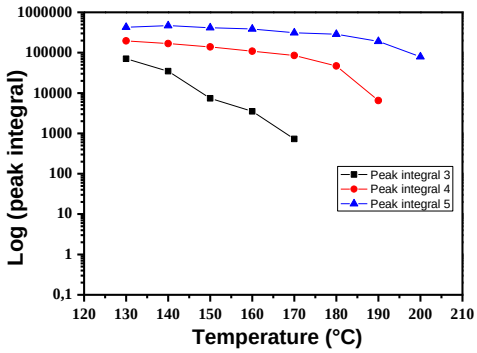


Fig.3 *Peak integral dependence on pre-heat temperature.*

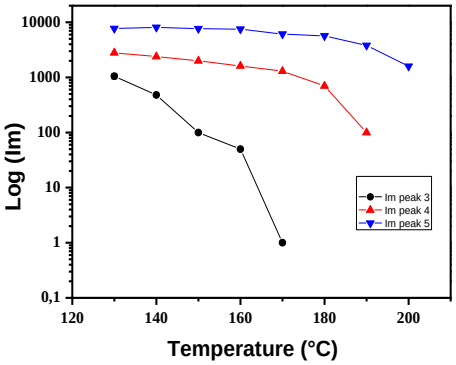


Fig.4 *Maximum TL intensity dependence on pre-heat temperature.*

3. Numerical modeling

The model consists of four electron traps and one recombination center to investigate whether the empirical results presented above could be qualitatively reproduced by means of numerical simulations.

3.1 Rate equations

A program within the Wolfram Mathematica was written, with the following parameters: the concentration of electron traps or hole centers N_i (cm^{-3}); the concentration of trapped electrons or holes n_i (cm^{-3}); the frequency factor s (s^{-1}); the electron trap depth below the conduction band or hole center energy levels above the valence band E (eV); the instantaneous concentration of holes in centers m (cm^{-3}); the instantaneous concentration of electrons in the valence band n_v (cm^{-3}); the instantaneous concentration of electrons in the conduction band n_c (cm^{-3}); A_n and A_m ($cm^{-3}.s^{-1}$) are the trapping and recombination probability coefficients respectively; the capture coefficient of the competitor trap A_d (cm^3/s); the rate of production of electron-hole pairs R and Bk=0.00008617 is the Boltzmann constants.

The equations used in this study are as follows:

$$\beta = \frac{dT}{dt} \qquad (5.1.1)$$

$$\frac{dn_i}{dt} = n_i[t]\, s_i\, exp\left(\frac{-Ei}{(Bk\, T[t])}\right) + n_c[t]\,(N_i - n_i[t])A_{\mathrm{ni}} \qquad (5.1.2)$$

(i=3-5)

$$\frac{dn_6}{dt} = n_c[t]\,(N_6 - n_6[t])A_d \qquad (5.1.3)$$

$$\frac{dm}{dt} = -A_m\, m[t]\, n_c[t] \qquad (5.1.4)$$

$$\frac{\mathrm{dn_c}}{\mathrm{dt}} = \mathrm{n_3'[t] + n_4'[t] + n_5'[t] + n_6'[t] + A_m\, m[t]\, n_c[t]} \qquad (5.1.5)$$

The values of i=3 to 5 stand for the TL glow-peaks 3-5, whereas the sixth electron trapping level corresponds to thermally disconnected traps (TDT). These TDT are considered to be thermally stable during the TL experiment.

The concentration of electron traps are: $N_5=n_5$, $N_4= n_4$, $N_3=n_3$ because most algorithms describing thermoluminescence consider $n_i=Ni$ [10, 12].

The intensity of the emitted light is assumed to be the result of recombination of free electrons with trapped holes in the luminescence center, therefore it is given by:

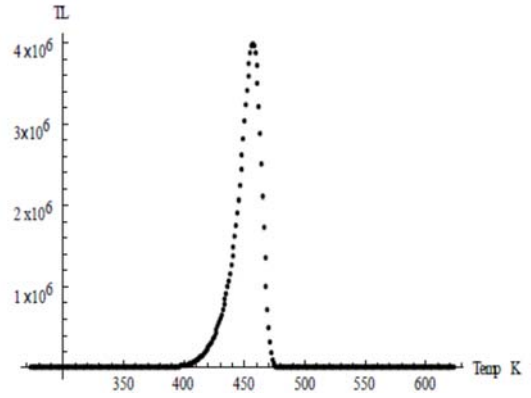
$$I(t) = \mathrm{A_m \times m \times n_c} \qquad (5.1.6)$$

3.2 Numerical results:

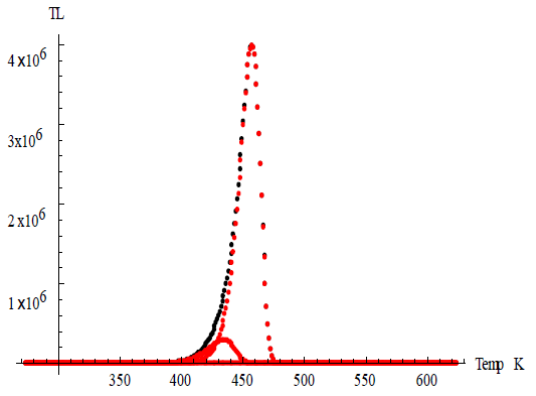
In order to obtain the numerical results, trapping parameter sets are selected and relevant sets of equations are solved numerically. The Mathematica solver has been used.

Table 1. *Kinetic parameters were obtained by deconvolution for the TL glow curves of LiF: Mg, Ti (0.5 at %).*

Peak	E (eV)	T_m (K)	b	s (s^{-1})	Peak integral	Normalization
3	1.4±0.05	409±1.8	1.0001±0.00002	7.1059E+14	34903±1577	0.05
4	1.5±0.05	434 ±2.1	1.0001±0.00003	2.0004E+16	168615±15352	0.25
5	2.15±0.06	457±2.7	1.00001±0.00005	3.6514E+21	467046±17353	1



(a)



(b)

Fig. 5 *Simulation results of the thermal fading at140°C with: E_3=1.4 eV; T_{m3}=409K; E_4=1.5eV; T_{m4}=434K; E_5=2.15eV; Tm_5=457K respectively.*

All input the trapping parameters for the simulation are of two types:

(*) experimentally evaluated (listed in Table1)

E_3=1.4 eV; T_{m3}=409 K.

E_4=1.5 eV; T_{m4}=434 K.

E_5=2.15 eV; T_{m5}=457 K.

(**) evaluated experimentally relative values:

$N_5=n_5=1.0\times10^{10}$ cm^{-3};

$N_4= n_4=0.277\times10^{10}$ cm^{-3};

$N_3= n_3 =0.0893\times10^{10}$ cm^{-3};

With $A_d=10^{-7}$ ($cm^{-3}.s^{-1}$), $A_5=10^{-8}$ (cm^{-3}), $A_4=10^{-8}$ (cm^{-3}), $A_3=10^{-8}$ (cm^{-3}) and $A_m =10^{-7}$ ($cm^{-3}.s^{-1}$).

The simulated TL glow curves shown in Fig.5a were obtained by the resolution of the sets of Eqs. (5.1.1-7). These glow curves consist of three peaks centered at 409, 434 and 457 K as calculated with a heating rate of 2 K/s. The results obtained by this model were in excellent agreement with experimental results at the pre heat temperature of 140°C.

The Fig. 5b evaluates each individual TL peak separately.

4. Conclusion

In the present work, the experimental results not only show the behavior of main TL peaks of LiF:Mg,Ti after special treatment thermal but explain lower order kinetics b and small changes in T_m. Moreover it can be said that the model for a post-irradiation fading (LiF: Mg,Ti) includes four trapping states and one recombination center. By solving the appropriate sets of coupled differential equations in sequence describe fairly well the post-irradiation fading.

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Conflict of Interest

The authors have no conflict of interest.

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