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Determination of the Trapping Parameters of Glow Peaks of CaF₂:Dy (TLD-200) by Using Computer Glow Curve Deconvolution Method

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Abstract

Computerized glow curve deconvolution method is used to determine the number of peaks, the order of kinetics b , the activation energy E_a , frequency factor s and number of the trapped electrons n_0 associated with the glow peaks in single crystal of CaF₂:Dy (TLD-200). Glow curve analysis indicate that the glow curve in the temperature region between room temperature and 250 °C can be best described as a superposition of six peaks. First five peaks are described by first-order kinetics and sixth peak general-order kinetic. The number of trapped electrons, activation energies and frequency factor for individual glow peaks are as follows: (Peak 1) 3×10^{13} , 1.27 and 2.13×10^{17} , (Peak 2) 6.3×10^{13} , 1.01 and 9.2×10^{12} , (Peak 3) 3.3×10^{14} , 0.7 and 6.12×10^7 , (Peak 4) 2.4×10^{14} , 1.14 and 3.28×10^{12} , (Peak 5) 1.5×10^{14} , 0.84 and 2.56×10^8 , (Peak 6) 2.4×10^{14} , 0.98 and 1.76×10^9 .

1. Introduction

Dysprosium-activated CaF₂ is widely used in the dosimetry of ionizing radiations using thermoluminescence. Many studies have been reported about the preparation of this material and its dosimetric properties [1-3]. However up to now no information has been found on the trapping parameters; namely, activation energy (trap depth) E_a (eV), frequency factor s (s⁻¹), number of the trapped electrons n_0 (m⁻³) and kinetic order b in literature. These data may provide a base for a more reliable analysis of the glow curve which is necessary to derive information on the radiation quality. There are many methods available to determine the trapping parameters from single glow peak [4-10]. For example, the trapping parameters can be determined by isothermal decay, peak shape, initial rise and heating rate methods. However in most TL samples (i.e.; LiF:Mg,Ti ,

CaF₂:Dy), the glow curves are obtained in the form of summation of many overlapping glow peaks. This type of glow curves are known as complex glow curves. In complex glow curves, separation of individual glow peaks from each other is very difficult. There are many methods to distinguish these peaks from each other. The most well known of them is a computerized curve fitting procedure which constructs a glow curve using the initial values of the parameters and compares the computed curve with the experimental curves.

In this study we determined the trapping parameters (E_a , s , n_0 , and b) of TLD-200 by using a computer glow curve deconvolution (CGCD) program.

2. Experiments and Methods

Standard TLD-200 loose chips (3.1 x 3.1 x 0.89 mm) purchased from Harshaw Chemical Company were employed for the experiment. The standard annealing was done at $410 \pm 10^\circ\text{C}$ for 10 min followed by a rapid cooling to room temperature (approximately $100^\circ\text{C}/\text{min}$) and then irradiated at room temperature immediately after quenching. All irradiations were carried out with a ^{90}Sr - ^{90}Y β -source. The irradiation equipment was an additional part of the 9010 Optical Dating System. Glow curves were obtained by using a Harshaw QS 3500 manual type reader interfaced to a PC where the TL signals were analyzed. Glow curve readout was carried out on a platinum planchet at a linear heating rate of $1^\circ\text{C}/\text{sec}$ up to 250°C . The time duration between irradiation and TL readout was always kept constant at about 1 min.

The glow curves were analyzed with a curve fitting computer program for thermally activated process using a PC. The program uses a linear least-squares minimization procedure to determine the peak area, peak temperature, peak intensity, activation energy, frequency factor and kinetic order. The computer program was developed at the Reactor Institute at Delft, The Netherlands 1993 [11]. Two different models were used in the computer program. In the first model, the glow curve is approximated from the first-order TL kinetics by the expression

$$I = n_0 s \exp\left(-\frac{E_a}{kT}\right) \exp\left[-\frac{s}{\beta} \frac{kT^2}{E_a} \exp\left(-\frac{E_a}{kT}\right) \left(0.9920 - 1.620 \frac{kT}{E_a}\right)\right]. \quad (1)$$

In the second model the glow curve is approximated with the general-order TL kinetics by using the expression

$$I = n_0 s \exp\left(-\frac{E_a}{kT}\right) \left[1 + \frac{(b-1)s}{\beta} \frac{kT^2}{E_a} \exp\left(-\frac{E_a}{kT}\right) \left(0.9920 - 1.620 \frac{kT}{E_a}\right)\right]^{\frac{b}{1-b}}, \quad (2)$$

where n_0 (m^{-3}) is the number of trapped electrons at $t=0$, s (sec^{-1}) the frequency factor, E_a (eV) the activation energy, T (K) the absolute temperature, k (eV K^{-1}) Boltzmanns constant, β ($^\circ\text{C s}^{-1}$) heating rate and b the kinetic order.

3. Results

A typical glow curve of a TLD-200 sample, which had been exposed to β -rays for 5 minutes after a standard annealing heat treatment at $400 \pm 10^\circ\text{C}$ for 10 minutes, is shown in Figure 1. As seen from this figure, there are at least three main glow peaks between room temperature and 250°C which they are appearing at about 115°C , 140°C and 200°C . In this study, they are labeled as peak 3, 4 and 6, respectively. In the analyses of the complex glow curves by CGCD to obtain best-fit results, it is very important to decide correctly how many glow peaks there are in the glow curve. In some cases, good fits can be obtained when different number of peaks is used in the CGCD analyses instead of the real number of the glow peaks to be in the glow curve. The values of the trapping parameters, which are obtained by using a wrong glow peaks number, do not reflect their real values even if good fits are obtained. Therefore, firstly one must decide the number of the peaks there must be in the complex glow curve of TLD-200 in the investigated region (between the room temperature and 250°C). In previous studies [1-3, 12], the reported number of glow peaks in the TLD-200 is 4 or 5 peaks. From Figure 1, it is seen that there are some shoulders at the low temperature side of peak 3. But it is not clear how many number of peaks there are in that region. To find the exact number of these peaks, we obtained the glow curve of this sample, as seen in Figure 2, from room temperature to 145°C . The two shoulders, which were not seen clearly in Figure 1, are seen obviously in this figure. It means that there are two glow peaks labeled as peak 1 and 2 at low-temperature side of peak 3. The low-temperature glow peaks 1 and 2 have high fading rates. We used this property to support the presence of peak 1 and 2. Therefore, we recorded the glow curve of TLD-200 after a few days and compared with the typical glow curve, as shown in Figure 3. In this figure, curves represented with the filled squares and triangles are typical glow curve and the glow curve of the sample TLD-200 obtained after 1 day β -irradiation, respectively. As seen from this Figure, while peak 2 still exists, peak 1 is completely destroyed. Under the light of the these two results, we decided that there are two-satellite peaks in the low temperature side of peak 3. After the determination of the number of glow peaks in the low-temperature side of glow peak 3, another question arises: whether there is any other glow peak or not between the glow peak 4 and 6. In the previous studies [1-3, 12], the existence of this kind of peak between these peaks had not been reported. In some cases, thermal cleaning method becomes a very useful technique to find the hidden glow peaks in the complex glow curves. Therefore, we applied this method to eliminate the intensity of the main peak 4 until the satellite peak, if there is one, appears on the high-temperature side of peak 4 by keeping the sample at an appropriate thermal cleaning temperature. In Figure 4, the glow curve of the sample subjected to the post-irradiation heat treatment at 115°C for 15 min is shown. A satellite peak on the high-temperature side of peak 4, which is labeled as peak 5, is clearly seen on the glow curve. After all of these investigations, we decided that there are six glow peaks between the room temperature and 250°C .

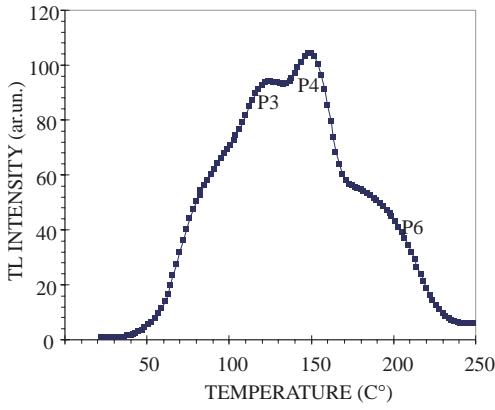


Figure 1. A typical glow curve of CaF₂:Dy (TLD-200) after a standard annealing procedure at 410°C for 10 min. $\beta=1^\circ\text{C}/\text{sec}$. The filled squares represent experimental points.

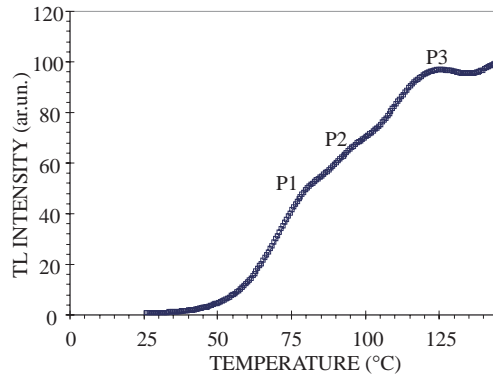


Figure 2. The glow curve of TLD-200 recorded up to 145°C with a heating rate $\beta=1^\circ\text{C}/\text{sec}$. The filled squares represent experimental points.

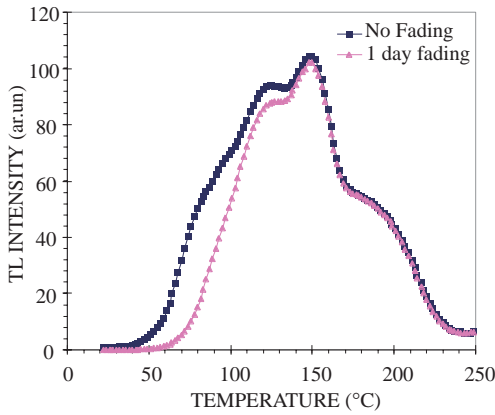


Figure 3. A set of glow curves for CaF₂:Dy crystals. The filled squares represent the typical glow curve (no fading), the filled triangles represent the glow curve after 1 day fading.

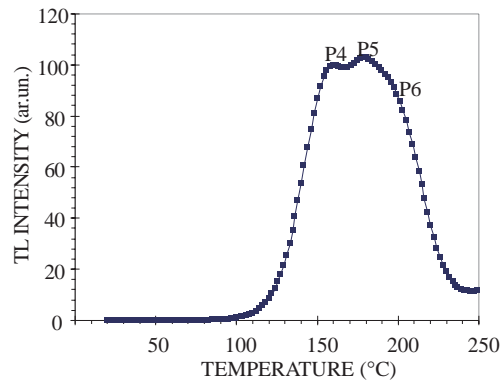


Figure 4. The glow curve of TLD-200 after 15 min. post-annealing at 115°C. $\beta=1^\circ\text{C}/\text{sec}$. The filled squares represent experimental points.

The second important point is to decide whether glow peaks are first- or general-order kinetics. Our investigations have represented that best-fit results were obtained for first five peaks due to first-order kinetics and sixth peak due to general-order kinetic. The determination of the kinetic order of individual glow peaks in the complex glow curve is very difficult. Especially, thermal cleaning method was used to isolate the glow peaks

from each other and then the kinetic order of isolated glow peak can be determined easily by using peak shape method. According to this method, there is a high relationship between the shape factor ($\mu_g = \frac{\delta}{\omega} = \frac{T_2 - T_m}{T_2 - T_1}$) and the kinetic order b . The values of the kinetic order as a function of μ_g can be found elsewhere [1].

If we use these methods to determine the kinetic order of the sixth peak of TLD-200, firstly the samples were subjected to the post-irradiation annealing temperature at 145°C for 10 min to isolate peak 6 from its low-temperature glow peaks. The isolated peak 6 is shown in Figure 5. After the isolation of glow peak 6, the kinetic order of this peak is determined by employing peak shape method. T_1 , T_m and T_2 are found approximately as 176°C, 201°C, and 222°C, respectively, from Figure 5. The shape factor is determined as ≈ 0.46 by using the values of T_1 , T_m and T_2 . The kinetic order of peak 6 corresponding to this value was found on the order of ≈ 1.3 .

After the determination of the number and kinetic order of all the glow peaks in the glow curve of the TLD-200, the typical glow curve is analyzed by applying first-order kinetics for the first five glow peaks and general-order kinetic for the sixth glow peak in the CGCD. The fitted glow curves corresponding to the six peaks, the glow curve represented by solid line obtained from the superposition of six glow curves, and typical glow curve are seen in Figure 6. The peak number is also illustrated on the figure 6. The evaluated activation energies E_a , frequency factor s , number of trapped electrons n_0 and kinetic order b given in Table 1.

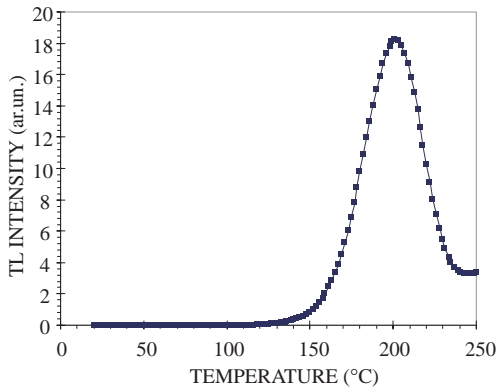


Figure 5. The glow curve of TLD-200 after 10 min. post-annealing at 145°C. $\beta=1^\circ\text{C}/\text{sec}$. The filled squares represent experimental points.

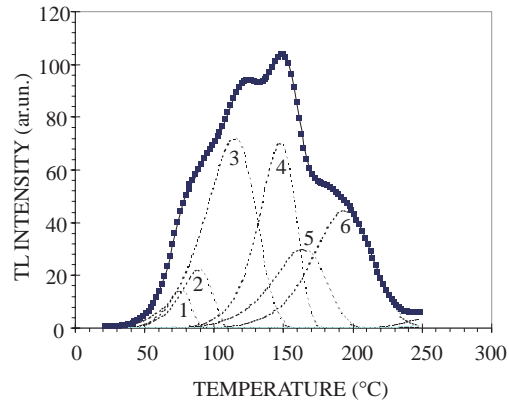


Figure 6. Fitted glow curve of TLD-200 measured after a standard annealing procedure. In the figure filled squares represent experimental points, the global fitting is shown as a full curve and the fitted peaks are represented by broken curves. $\beta=1^\circ\text{C}/\text{sec}$.

Table 1. Evaluated trapping parameters of glow peaks of TLD-200 after a standard annealing procedure of 10 min at $410 \pm 1^\circ\text{C}$ and readout at a linear heating rate $\beta=1^\circ\text{Cs}^{-1}$. Obtained from computer curve fitting program.

Peak Number	n_0 (m^{-3})	E_a (eV)	$\ln(s)$	b
1	3.06×10^{13}	1.27	39.90	1.00
2	6.28×10^{13}	1.00	29.85	1.00
3	3.32×10^{14}	0.70	17.93	1.00
4	2.41×10^{14}	1.14	28.82	1.00
5	1.46×10^{14}	0.84	19.36	1.00
6	2.42×10^{14}	0.98	21.29	1.25

4. Conclusion

In the last years CGCD program has become very popular method for the determination of the trapping parameters from TL glow curves [13-18]. When the initial parameters (number of glow peaks, kinetic order of each glow peak) were entered with the appropriate values to the CGCD program, a best-fit and real trapping parameters can always be obtained. If the trapping parameters are determined correctly, a lot of information on the radiation quantity can be obtained. For example, the mean-life time and fading quantity of any dosimetric peak in the TLD can be determined easily by using these parameters. Finally, the determination of the trapping parameters of all of the glow peaks in the TLD-200 is not possible by using heating rate, initial rise, peak shape and isothermal methods. CGCD is a more valuable method, which can be used instead of all of these methods, for the determination of trapping parameters of the samples having complex glow curves.

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