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THERMOLUMINESCENCE GLOW CURVE PROPERTIES OF TLD-500 DOSIMETER

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Abstract

In this paper, TL characteristics and glow curves of α-Al_2O_3:C dosimeter, known as TLD-500, were analyzed using different methods and softwares. The effect of heating rate (HR) and low dose (from 10 cGy up to 50 cGy) on TL glow curves of α-Al_2O_3:C chips have been investigated after β-irradiation. TL kinetic parameters were also calculated by using computerized glow curve deconvolution (CGCD), peak shape (PS), various heating rate (VHR) and three points (TP) methods. Furthermore, using Mathematica software, all TL glow curves of TLD-500 were decomposed in order to compare with the results of other methods and simulated after exposed different beta doses.

Key words: *Thermoluminescence, TLD-500, dosimeter, glow curve, Mathematica.*

1. Introduction

Thermoluminescence (TL) detectors based on single crystal α-Al_2O_3:C (TLD-500) developed and produced at Urals Polytechnical Institute [1], have advantages comparing to well-known LiF TL detectors (TLD-100) since the sensitivity of TLD-500 was reported to be 30–40 times higher than TLD-100 which makes it possible to detect very low dose levels down to 300 nGy [2,3]. Also, TLD-500 is considered to be very good TL phosphors due to its high radiation sensitivity, simple glow curve, low background and dose threshold, low fading in the dark, good reproducibility, simple emission spectrum, wide dose range and relatively low atomic number [4,5]. TL intensity of α-Al_2O_3:C strongly depends on heating rate [3,6-8]. As the heating rate increases not only does the peak maximum shift to higher temperatures but the intensity falls off strongly on the high temperature side of the peak.

In studying the glow curve of α-Al_2O_3:C for increasing doses, the main TL glow peak shifts to lower temperatures as the dose increases, but not in a monotonic way. This behavior is consistent with an overlap of several first-order TL peaks. The shift of the peak maximum can be explained by the differential growth of the individual first-order peak components [9,10]. There are various methods to evaluate the trapping parameters from TL glow curves. If a glow peak is highly isolated from the others, variable heating rates (VHR), peak shape (PS), three points (TP), computerized glow curve deconvolution (CGCD) [11] and Mathematica software are suitable methods to determine them. Therefore, in the given study, these techniques were used to determine the kinetic parameters of α-Al_2O_3:C after different dose levels with beta (β) irradiation.

2. Materials and Methods

In this study, TLD-500 chips (α-Al_2O_3:C) with 1 mm thick and 5 mm in diameter were used. The chips based on anion-defective single crystal show more sensitivity to radiation and have simple TL curve [1]. It has useful dose range (0.05 μGy-10 Gy) and less thermal fading 3% per year [12,13]. All TL measurements were carried out using a Risø TL/OSL reader system (model TL/OSL-DA-20, Risø National Laboratory, Denmark) with ^{90}Sr-^{90}Y β irradiator source (dose rate≈6.689 Gy/min). UV transmitting filter (Hoya U-340, 7.5 mm thickness, ~290–370 nm transmission) was used in front of the EMI 9235 QA photomultiplier tube (PMT). TL glow curves of chips were recorded from room temperature (RT) up to 400°C with a linear heating rate of 2°C/s. N_2 gas was allowed to flow into the reader during all readout processes to avoid any spurious signals and background readouts were subtracted from the TL glow curves. The CGCD, PS, VHR, TP methods and Mathematica software were used to calculate TL kinetic parameters of TLD-500.

3. Results and discussion

3.1. Dose response and dose equation

The TL low-dose responses were measured over the β dose range from 10 cGy up to 50 cGy and TL glow curves were recorded (see Fig.1). As seen from Fig.1, TL glow curve of TLD-500 shows an intense and well defined peak with the maximum on ~204°C and this TL peak is shifting towards to lower temperature with increasing dose level. This peak shifting is within the error limits. According to TL theory, the peak temperatures are expected to change only with heating rate for first order kinetics (b=1). Hence for a constant heating rate the peak maximum should not be affected by other experimental parameters and should thus be fairly constant within the limit of experimental errors [14]. Therefore, if the TL peak temperature shifts to the lower side with increasing dose levels the peak is not first order kinetics ($b\neq 1$), so this peak should be first (b=1) order kinetics.

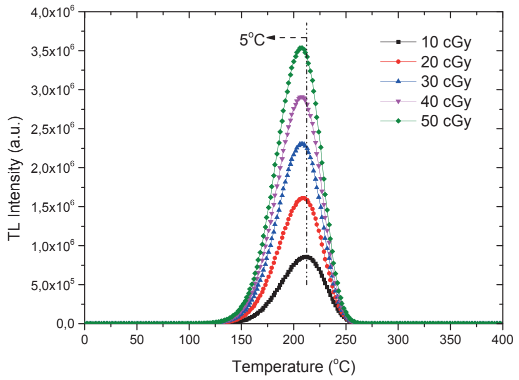

Fig. 1 TL low-dose glow curves of TLD-500 obtained with a linear heating rate of 2 °C s⁻¹.

TL response and low-dose relationship of TLD-500 is illustrated in Fig. 2. As seen in Fig. 2, TLD-500 has a linear dose range between 10 cGy and 50 cGy, and beta radiation dose can be calculated using this equation;

$$\beta\ Dose\ (cGy) \approx \frac{TL\ Intensity - 1.99x10^5}{6.69x10^4}$$

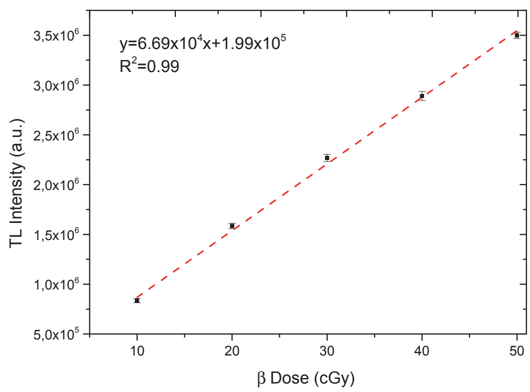

Fig. 2 Relationship between TL response and low-dose.

3.2. Heating rate effect

The heating rate effect on TL glow curves is a basic experimental variable in TL measurements [15]. The heating rate of the dosimetric materials influences the variation of their TL sensitivity and, hence, the trends of the dose curve [16]. In the application of TL dosimetry, TL glow peak (or curve) area and TL glow peak height are affected by changes in heating rate [17].

In order to determine the effect of heating rate on the TL glow curve and to calculate kinetic parameters (section 3.4.3) of TLD-500 dosimeter, three chips were irradiated with ~10 cGy β dose and then TL glow curves were recorded from RT to 400°C by using various heating rates of 1, 2, 3, 4, 5, 6, 7, 8, 9 and 10°C/s. TL glow curves of the chips are presented in Fig. 3a. As seen in Fig. 3a, the peak temperature of the glow peak is shifting to the higher temperature side as the heating rate rises and also peak intensity is continuously diminishing as expected in the TL theory. Results from the large number of studies showed a decrease in TL glow peak height (or area) intensity with increasing heating rate. This phenomenon has been explained to be due to thermal quenching, whose efficiency increases as temperature increases [18-23]. The maximum TL intensities of the glow peaks were normalized to lower heating rate (1°C/s) and it can be seen that the maximum TL intensity of peak decreases by about 65% (in Fig. 3b).

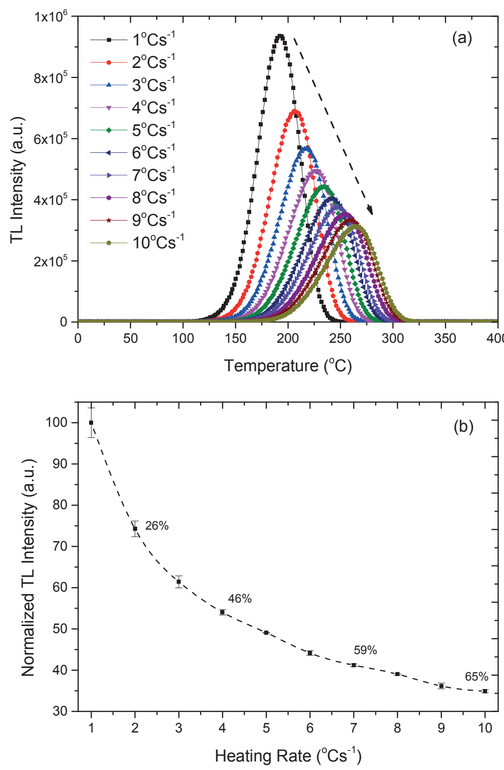

Fig. 3 (a) The measured and (b) normalized TL glow curves after different heating rates (D≈10 cGy).

3.3. Reusability

Reusability is one of the most important properties for any dosimetric material [24] and TL sensitivity of a good dosimeter should not change with repeated TL readout cycle. In order to test reusability of TLD-500 dosimeter without annealing, it was irradiated with 10 cGy by β from ^{90}Sr-^{90}Y source and the TL glow curve was recorded. The cycle of irradiate-readout process was repeated ten times and TL measurement was carried out before irradiation to see that no TL signal was left after per readout. Normalized peak height intensities were obtained from maximum peak intensity. Fig. 4a shows the TL glow peaks of TLD-500 dosimeter for per readout cycle and the TL sensitivity of the TLD-500 after the different cycles of usage are shown in Fig 4b. As seen in Fig. 4a and Fig. 4b, TL sensitivity is decreasing per readout from 1 to 10 cycle and this decreasing is about 11% at tenth TL readout. Therefore, the best combination of time and temperature for an optimum annealing procedure should be determined and applied to obtain the highest reproducibility for TL signals. In previous studies, this procedure is defined as 1 hour at 400°C and then 16 hours at 80°C for TLD-500 [25].

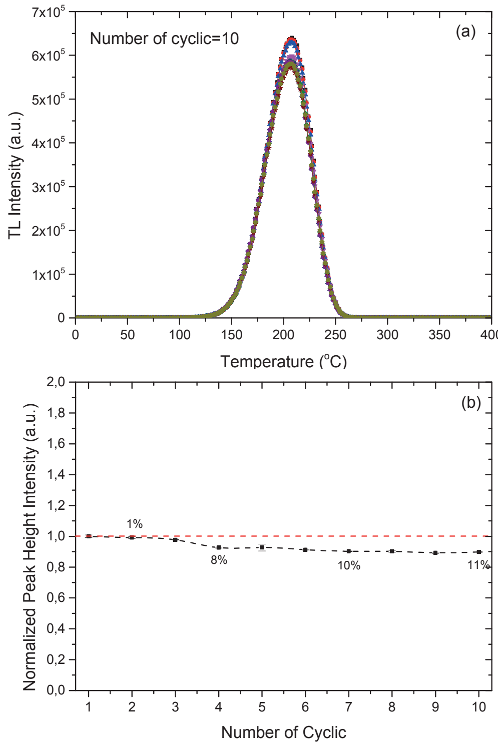

Fig. 4 (a) TL glow curves and (b) TL sensitivity change of TLD-500 at repeated β-irradiation.

3.4. Kinetic parameters

The shape of a TL glow peak takes an important role in basic research and TL applications. In TL research, it is the basis of important and suitable methods for calculating the trapping parameters of different energy levels within the crystal. The complete description of the TL characteristics of TL material requires the knowledge of these parameters. The TL glow curves are characterised by determining various factors like the activation energy (E), order of kinetics (b), and frequency factor (s) [26].

In this study, kinetic parameters of the main TL glow peaks of TLD-500 chips (i.e. order of kinetics (b), activation energies (E) and frequency factors (s)) were determined and evaluated by the method of computerized glow curve deconvolution (*CGCD*), peak shape (PS), various heating rate (VHR) and three points (TP) using the glow curve data. Furthermore, using Mathematica software, all TL glow curves of TLD-500 were decomposed in order to compare with the results of other methods.

3.4.1. Computerized Glow Curve Deconvolution (CGCD) Method

CGCD analysis has become very popular during the last two decades with the development of sophisticated glow curve deconvolution (GCD) techniques [27, 28]. The glow curves of dosimetric materials are in most cases complex curves consisting of many overlapping glow peaks. Hence, the deconvolution of complex glow curves into their individual components is widely applied for dosimetric purposes and for evaluating the kinetic parameters using curve fitting methods [28]. In addition, *CGCD* method is very important to decide correctly how many glow peaks there are in the complex glow curve and which of them have first, second or general order kinetics to obtain correct results [14].

There are many *CGCD* programs used by researchers, one of these programs have been TL Glow Curve Analyzer (TLanal) [29, 30]. TLanal is written in C# of the Visual Studio .Net and has been tested in the Microsoft Windows-based operating systems. It is possible to convert this program to an ActiveX control which can run in a web browser. In this program, a new method based on the general one-trap TL equation was adopted to analyze the TL glow curve with the traditional first-order, second-order and general-order kinetics model. The method described here, general approximation, generates TL glow peaks and interpolates the relevant TL parameters from the glow data [29, 30]. The accuracy of decomposing was tested with calculating the value of figure of merit (FOM) for all of the TL glow peaks.

In this study, the TL glow curve of TLD-500 dosimeter for the selected 50 cGy beta radiation dose was decomposed with TLanal software. Fig. 5 shows the analyzed TL glow curve of dosimeter. The analyzed TL glow curve as shown in the figure consist of two glow peaks at around 195 and 213°C (Fig. 5). The accuracy of these presented values is supportable with figure of merit (FOM) values. FOM values are 1.35% for TLD-500 dosimeter. Therefore, the TL kinetic parameters (E (eV), s (s^{-1}) and b) associated with the glow peaks were obtained by *CGCD* method. The obtained kinetic parameters and peak temperatures at the maximum (T_m) were given in Table 1. As seen in Table 1, all of the samples have general order kinetic (b=1.30 for P1 and b=1.44 for P2 by calculated CGCD method).

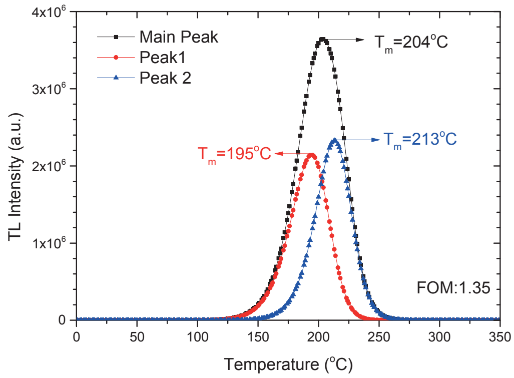

Fig. 5 Decomposed TL glow curves of TLD-500 (with TLanal software).

The TL glow curve of TLD-500 dosimeter was also decomposed for each heating rate values from 1 °C/s to 10 °C using TLanal software and different heating rate values versus activation energies were plotted for all peaks (Fig. 6). As seen in Fig. 6, average activation energy is 1.26±0.02 eV for peak 1 and 1.44±0.05 eV for peak 2. These results show that the obtained activation energies using CGCD and Mathematica software are compatible within the limit of experimental errors. The accuracy of these presented values is supportable with figure of merit (FOM) values. Many studies have shown that if the FOM value is between 0.0% and 2.5%, the fit is good [31-33]. In this study, average FOM values were calculated as 1.45% for TLD-500 dosimeter.

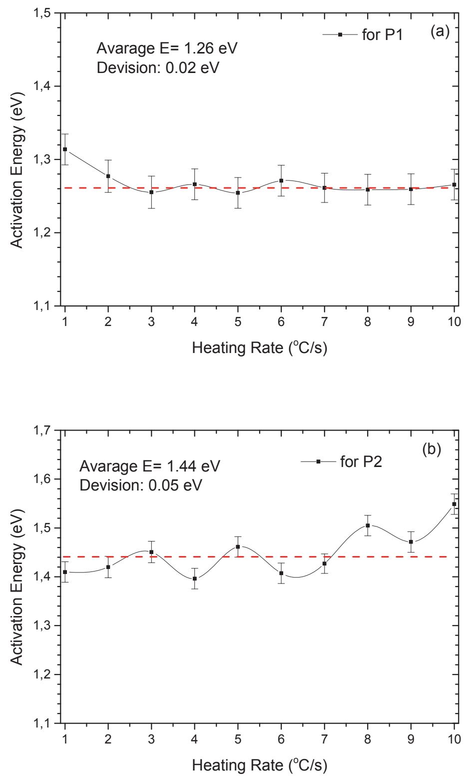

Fig. 6 Graphs of different heating rate values versus activation energies (a) for peak 1 and (b) for peak 2.

3.4.2. Peak Shape (PS) Method

One way of analyzing a TL glow peak, obtained using a linear heating rate, is by considering its geometrical properties with peak shape method. *PS* method is generally called as Chen's (1969) [34] method, which is used to determine the kinetic parameters of the main glow peak of the TL materials. This method is mainly based on the analysis of the glow-curve shape by using temperatures (T_m, T_1 and T_2). The second-order peaks are characterized by a symmetrical shape whereas the first-order peaks are asymmetrical in this method [35].

Geometrical shape parameters of TLD-500 were firstly determined with the help of TL glow curves readings for the selected 50 cGy beta radiation dose. Then *PS* method was applied to the main TL glow peaks at around 204°C. E, *s* and *b* (μ_g = 0.42*)* values were calculated (See Table 1). The kinetic parameters were calculated by using following expressions:

$$E = 1.51k\left[\frac{T_m T_1}{T_m - T_1}\right]$$

$$s = \frac{\beta E}{kT_m^2} exp\left(\frac{E}{kT_m}\right)$$

$$\mu_g = \frac{\delta}{\omega} = \frac{T_2 - T_m}{T_2 - T_1}$$

3.4.3. Various Heating Rate (VHR) Method

This method essentially makes use of change in glow peak temperature T_m with respect to heating rate. When the heating rate changes the maximum temperature (T_m) of the TL glow peak also changes: at faster heating rates corresponds a shift temperature towards higher values of T_m. (see Fig. 3a) [35]. The kinetic parameters of dosimeters can be calculated by the following expressions for different sets of heating rates and their corresponding T_m, E and s [26]:

$$E = \left[\frac{kT_{m1}T_{m2}}{T_{m1} - T_{m2}}\right]\left[ln\left(\frac{\beta_1}{\beta_2}\left(\frac{T_{m2}}{T_{m1}}\right)^2\right)\right]$$

$$s = \frac{E\beta_1}{k(T_{m1})^2}\left[\left[\left(\frac{\beta_1}{\beta_2}\left(\frac{T_{m2}}{T_{m1}}\right)^2\right)\right]^{\frac{T_{m2}}{(T_{m1}-T_{m2})}}\right]$$

In this study, E and s were calculated by using above equations. The values of T_{m1} and T_{m2} are selected 227°C and 264°C and the values of β_1 and β_2 are 4°C/s and 10°C/s, respectively. The calculated values are mentioned in Table 1.

3.4.4. Three Points (TP) Method

The trapping parameters of TLD-500 chip were calculated by the Rasheedy's [36] three points method (TPM) by using TL Parameters Calculator (TLPC) (Fig. 7) software that it is written in C# of the Visual Studio .Net [37]. The average values of the trapping parameters are given by E= 1.41±0.07 eV, s= 9.72x10¹³ s⁻¹ and b= 1.03 (See Table 1).

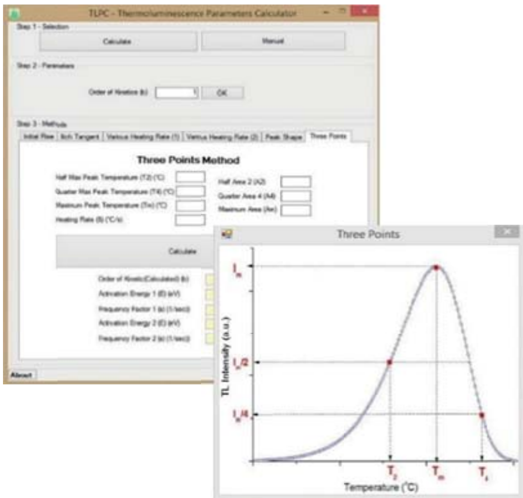

Fig. 7 Thermoluminescence Parameters Calculator (TLPC) software.

3.4.5. Mathematica Software

Mathematica is a computational software program used in many scientific, engineering, mathematical and computing fields, based on symbolic mathematics. It was conceived by Stephen Wolfram and is developed by Wolfram Research of Champaign, Illinois [38]. In this study, kinetic parameters of TLD-500 dosimeter were calculated by using Mathematica software. For this calculation, TL glow peaks of TLD-500 were simulated the Mathematica software entering numerical values of E, s and b. Then the X-Y data were plotted in Mathematica software (See Fig. 8 and Table 1).

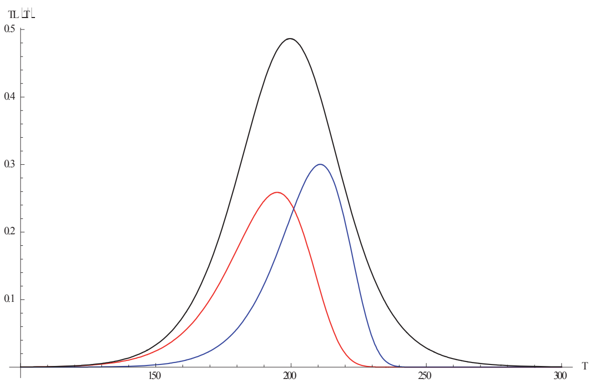

Fig. 8 Simulated TL glow curves of TLD-500 (with Mathematica software).

Table 1 Values of kinetic parameters.

Methods	Peak	T_M (°C)	E (eV)	s (s^{-1})	b
CGCD (TLanal)	P1	195	1.33±0.07	$1.98x10^{13}$	1.30
	P2	213	1.66±0.08	$1.64x10^{16}$	1.44
Peak Shape (PS)	Peak	204	1.12±0.06	$7.58x10^{11}$	1.00
Various Heating Rate (VHR)	Peak	204	1.31±0.06	$1.83x10^{10}$	-
Three Points (TP)	Peak	204	1.41±0.07	$9.72x10^{13}$	1.03
Mathematica	P1	195	1.34±0.06	$1.98x10^{13}$	1.37
	P2	214	1.67±0.08	$1.63x10^{16}$	1.45

4. Conclusions

For a dosimeter, it is important to be characterized in terms of dosimetric properties such as dose response, heating rate effect and reusability. Given study, TL characteristics and glow curves of α-Al_2O_3:C dosimeter were analyzed using different methods and softwares. The results of dose response experiment indicate that the glow curve of TLD-500 shows an intense and well defined peak with the maximum on approximately 204°C and this TL peak is shifting towards to lower temperature with increasing dose level within the error limits. The effect of heating rate (HR) experiment results are coherent expected in the TL theory in this study. According to reusability experiments, TL sensitivity of TLD-500 is decreasing about 11% at tenth TL readout. TL kinetic parameters were calculated by using CGCD, PS, VHR and TP methods. Furthermore, using Mathematica software, all TL glow curves of TLD-500 were simulated in order to compare with the results of CGCD. Both CGCD and Mathematica simulation results are compatible each other within the error limits. We believe in Mathematica software is also a very useful tool to obtain TL kinetic parameters and should be used for other dosimeters to see its efficiency.

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