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Development of a Thermoluminescence Parameter Calculator Program

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***Abstract*— In the literature many attempts have been made to calculate the related phenomena of thermoluminescence (TL) from a kinetic model primarily in order to study crystal defect properties and to have a tool for determining trapping parameters such as activation energies (or trap depths, E), frequency factors (s) and order of kinetics (b). Determining the kinetic model of TL glow curves reveals the dosimetric properties of the experienced material. Thus, the subject of TL kinetic parameters calculation is very significant in TL dosimetry studies. In this study, "Thermoluminescence Parameter Calculator (TLPC)" application was developed with C# programming language on Microsoft Visual Studio .Net platform. The developed TLPC software can calculate E, s and b values easily, practically and correctly by using various heating rate (VHR), peak shape (PS) and three points (TP) methods. In order to determine these values, non-overlapping TL glow peaks of any dosimetric material must be recorded from TL measurement and maximum peak temperatures (T_M), heating rate values (β), maximum intensities (I_M) and peak areas (maximum, half and quarter) of these TL peaks must be obtained. Using the first, second and general order isolated TL glow peaks, the reliability of TLPC software were checked with obtained data from the Mathematica.**

***Keywords*—TLPC software, thermoluminescence, kinetic parameters, programming, dosimeter.**

I. Introduction

When radiation is incident on a material, some of its energy may be absorbed and re-emitted as light of longer wavelength (Stoke's Law). This is the process of luminescence. The wavelength of the emitted light is characteristic of the luminescent substance and not of the incident radiation. The various luminescence phenomena are given names, which reflect the type of radiation used to excite the emission. Thus, we have thermoluminescence (TL), photoluminescence (PL), cathodoluminescence (CL) and etc [1, 2].

The term thermoluminescence (TL) consist of two words: thermo, meaning heat and luminescence, meaning emission of light. These words may apper to mean that the emission of luminescence is caused by heat. However, this is not totally true since heat has only a secondary role in this case [3]. In TL, an insulator or semiconductor sample firstly excited by irradiation and then heated at a linear heating rate. During the heating, some of the energy absorbed by the sample emitted as light in the form of a glow curve, which contains one or more glow peaks. The shape, position and intensity of the glow peaks are related to various trapping parameters of the trapping states responsible for the TL emission [4]. The complete description of the defects in solids and TL characteristics require the knowledge of kinetic parameters [5]. These parameters include the order of kinetics (b), the activation energy/trap depth (E), the frequency factor (s) or the pre-exponential factor (s″) and the relative value of the initial concentration of trapped electrons (n_0). The knowledge of the trapping parameters is important in many fields of studies such as understanding of the TL phenomenon; in the studies on the stability of the stored energy in the trap level against the environmental condition, in the practical use of the thermoluminescent material as dosimeters and it can provide information on the types of defects in solids [4].

In this work, "Thermoluminescence Parameter Calculator (TLPC)" named a novel application was developed with C# programming language on Microsoft Visual Studio .Net platform.

II. Theoretical Background and Methods

A. Thermoluminescence Kinetics

Thermoluminescence processes have been given by Randall-Wilkins [6], Garlick-Gibson [7] and May-Partridge [8] equations for first, second and general orders, respectively:

$$I(t) = -\frac{dn}{dt} = nse^{-E/kT} \quad (b = 1)$$

$$I(t) = -\frac{dn}{dt} = \frac{n^2}{N} se^{-E/kT} \quad (b = 2)$$

$$I(t) = -\frac{dn}{dt} = n^b s' e^{-E/kT} \quad (1 < b < 2)$$

where I(t): TL intensity, n: concentration of trapped electrons at time t, s: frequency factor, N: the total trap concentration, E: activation energy or trap depth, k: Boltzmann's constant, T: the absolute temperature and s': the so-called effective pre-exponential factor for general order kinetics [9].

Randall and Wilkins were the first to suggest a theoretical model for a well isolated TL glow peaks, and they assumed that the retrapping may be negligible. The TL intensity for first order kinetics is given by [6];

$$I(T) = n_0 \times s \times \exp\left(-\frac{E}{kT}\right) \times \exp\left[-\frac{s}{\beta} \times \int_{T_0}^{T} \exp\left(-\frac{E}{kT'}\right) dT'\right]$$

Garlick and Gibson considered the case where electrons may be retrapped and they modified the first order kinetics model. The TL intensity for second order kinetics is given by [7];

$$I(T) = \frac{n_0^2 \times s \times exp\left(-\frac{E}{kT}\right)}{\mathrm{N} \times [1 + [\frac{n_0 \times \mathrm{s}}{\beta N}] \times \int_{T_0}^{T} \exp(-\frac{E}{kT'})\, dT']^2}$$

May and Partridge suggested an empirical equation has been proposed to describe the TL glow peak when conditions for neither first-order nor second order are satisfied. The TL intensity for general order kinetics is given by [8];

$$I(T) = \frac{n_0 \times s'' \times exp\left(-\frac{E}{kT}\right)}{[1 + [(\mathrm{b} - 1) \times \frac{s''}{\beta}] \times \int_{T_0}^{T} \exp(-\frac{E}{kT'})\, dT']^{\frac{b}{b-1}}}$$

where $s'' = s \times (\frac{n_0}{N})^{b-1}$ is known as the pre-exponential factor and b the order of kinetics.

B. *Determination of the Kinetic Parameters*

There are several methods in order to determine the trap parameters of TL glow peaks such as initial rise (IR), various heating rates (VHR), isothermal decay, peak shape (PS), three points (TP), curve fitting (or glow curve deconvolution, CGCD) methods and etc. In this paper, only three of these methods, VHR, PS and TP, are selected to implementation.

Various heating rate (VHR) method depends on the relation between the heating rate (β) and the temperature (T_M) corresponding to the maximum intensity (I_M) to obtain the kinetic parameters [9]. From the Booth–Bohun method [10, 11], for different sets of heating rates and their corresponding T_M, the activation energy (E) and frequency factor (s) can be calculated by the following expressions:

$$E = k\left(\frac{T_{M_1} \times T_{M_2}}{T_{M_1} - T_{M_2}}\right) ln\left[\left(\frac{\beta_1}{\beta_2}\right)\left(\frac{T_{M_2}}{T_{M_1}}\right)^2\right]$$

$$\frac{\beta_1 \times E}{k \times T_{M_1}^2} = s \times exp\left(-\frac{E}{k \times T_{M_1}}\right)$$

$$\frac{\beta_2 \times E}{k \times T_{M_2}^2} = s \times exp\left(-\frac{E}{k \times T_{M_2}}\right)$$

or

$$s = \frac{E \times \beta_1}{k \times (T_{M_1})^2} \times \left[\left[\left(\frac{\beta_1}{\beta_2} \times \left(\frac{T_{M_2}}{T_{M_1}}\right)^2\right)\right]^{\frac{T_{M_2}}{T_{M_1} - T_{M_2}}}\right] \quad [5]$$

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) [12] 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 [13, 14].

$$E = 1.51 \times \left[\frac{T_M \times T_1}{T_M - T_1}\right]$$

$$s = \frac{\beta \times \mathrm{E}}{k \times T_M^2} \times \exp\left(\frac{E}{k \times T_M}\right)$$

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

Rasheedy [15] has developed a technique (Three Points Method) for obtaining the trap parameters which includes the order of kinetics (b), the activation energy (E), the frequency factor (s) or the pre-exponential factor (s'') and the relative value of the initial concentration of trapped electrons (n_0). The activation energy, frequency factor and order of kinetics are given by

$$E = \frac{\left[ln2 - b \times ln\left(\frac{A_M}{A_2}\right)\right] \times k \times T_M \times T_2}{T_M - T_2}$$

$$E = \frac{\left[ln2 - b \times ln\left(\frac{A_M}{A_4}\right)\right] \times k \times T_M \times T_4}{T_M - T_4}$$

$$b = \frac{T_2 \times (T_M - T_4) \times ln2 - T_4 \times (T_M - T_2) \times ln4}{T_2 \times (T_M - T_4) \times ln\left(\frac{A_M}{A_2}\right) - T_4 \times (T_M - T_2) \times ln\left(\frac{A_M}{A_4}\right)}$$

$$s'' = s \times \left(\frac{n_0}{N}\right)^{b-1}$$

III. DEVELOPMENT OF THE PROGRAM

Thermoluminescence Parameter Calculator (TLPC) program is developed with C# programming language on Microsoft Visual Studio .Net 2015 platform. The developed program requires .NET Framework 4.5 installed on the Microsoft Windows based operating systems in order to run properly. This program is a Microsoft Windows based application which provides the following methods for the determination of the TL kinetic parameters:

- Various Heating Rate,
- Peak Shape,
- Three Points.

The TLPC program needs three steps to calculate kinetic parameters with the selected methods. In the first step, TLPC request user to select one of the two options to calculate order

of kinetics which are calculate or input order of kinetics value manually. The result of choices made in the first step, in the second step, if the user selects the calculate option, the program takes the T_1, T_2 and T_M parameters from user to calculate order of kinetics (b) and geometric factor (μ) (Fig 1a). On the other hand, if the user selects manual option, the application takes the order of kinetics value from user manually (Fig. 1b).

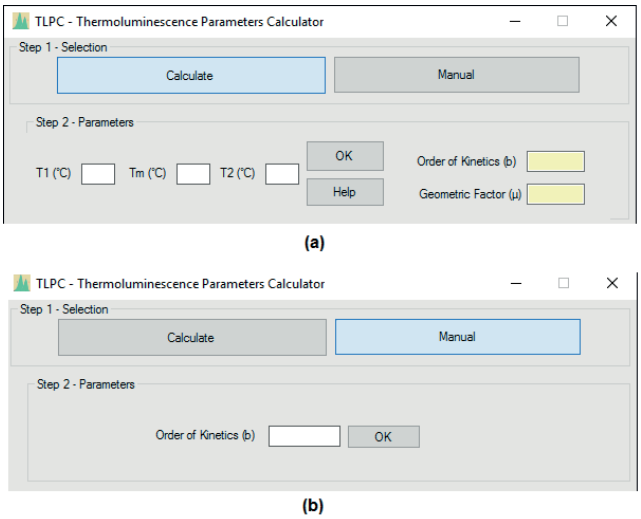

Fig. 1 Determining order of kinetics a) calculate option, b) input manually.

In the third and last step, the TLPC program calculates the activation energy (E) and frequency factor (s) parameters from the selected method.

A. *Various Heating Rate Method*

The VHR method takes six parameters from the user in order to calculate activation energy (E) and frequency factor (s) of the given thermoluminescence glow peak (Fig. 2). The heating rate 1 (β_1) should be smaller than heating rate 2 (β_2) for this method. The application also provides a help section to inform users with a graphical explanation of the method.

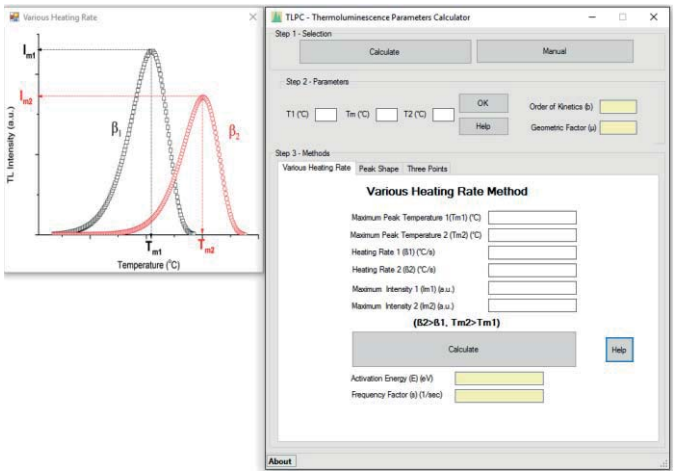

Fig. 2 Various Heating Rate (VHR) method screen.

B. *Peak Shape Method*

The PS method takes four parameters from the user in order to calculate activation energy (E) and frequency factor (s) of the given thermoluminescence glow peak (Fig. 3). The results of this method is listed at the bottom of the application. Explanation of this method is given in a graphical help section.

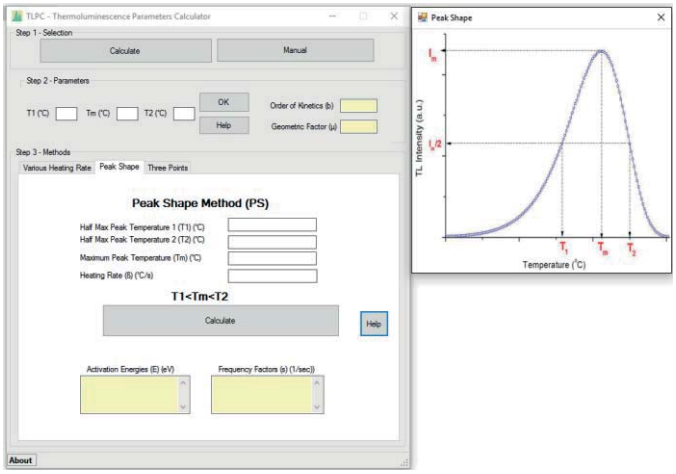

Fig. 3 Peak Shape (PS) method screen.

C. *Three Points Method*

The TP method takes seven parameters from the user in order to calculate activation energy (E) and frequency factor (s) of the given thermoluminescence glow peak (Fig. 4). This method calculates order of kinetics (b), two different activation energies (E_1, E_2) and two different frequency factors (s_1, s_2). The usage of this method is explained with a graphical help section of TP method tab.

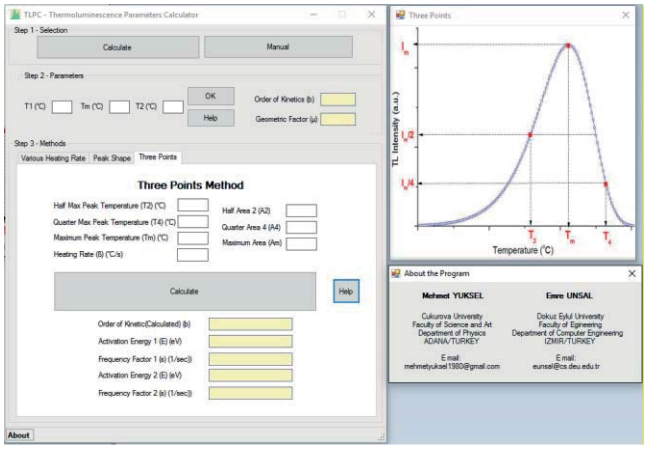

Fig. 4 Three Points (TP) method screen.

IV. Results and Discussion

In this study, during the development of TLPC program, the equations given in section two and the following data of TL glow peaks (Fig. 5, Fig. 6 and Fig. 7) are used simultaneously. The data of all glow peaks were obtained using Mathematica software in order to compare results.

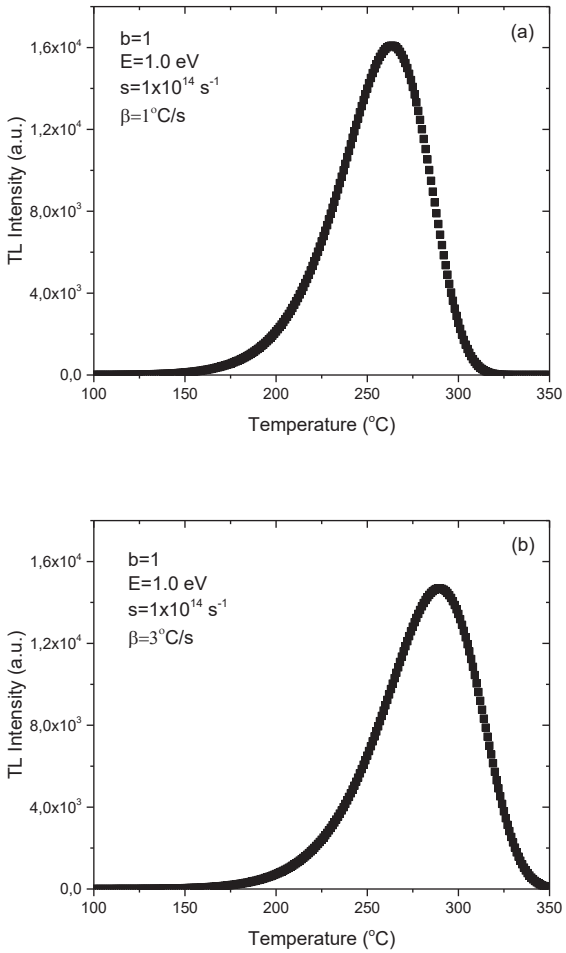

Fig. 5 Thermoluminescence peaks which have first order kinetics from obtained Mathematica (a) β=1°C/s (b) β=3°C/s.

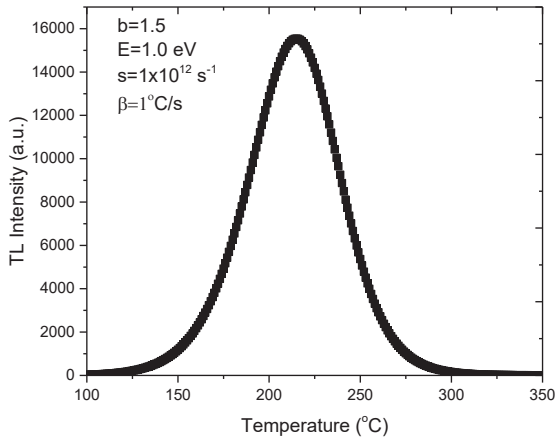

Fig. 6 Thermoluminescence peak which has general order kinetics from obtained Mathematica.

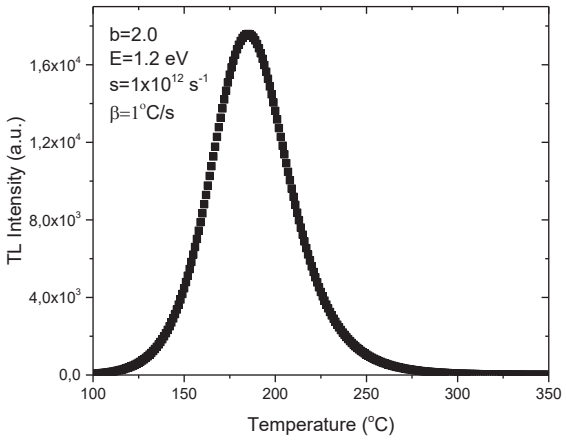

Fig. 7 Thermoluminescence peak which has second order kinetics from obtained Mathematica.

The determined activation energies (E) as 1.00 eV (for b=1), 1.00 eV (for b=1.5) and 1.20 eV (for b=2) and frequency factors (s) as 1.00×10^{14} s^{-1} (for b=1), 1.00×10^{12} s^{-1} (for b=1.5) and 1.00×10^{12} s^{-1} (for b=2) in Mathematica software were calculated using TLPC program. The activation energies according to VHR, PS and TP methods were calculated as 1.05 eV, 1.01 eV and 1.03 eV for b=1, 1.03 eV, 1.02 eV and 1.04 eV for b=1.5 and 1.25 eV, 1.21 eV and 1.23 eV for b=2, and the frequency factors (s) were calculated as 1.02×10^{14} s^{-1}, 1.03×10^{14} s^{-1} and 9.97×10^{13} s^{-1} for b=1, 9.95×10^{11} s^{-1}, 9.80×10^{11} s^{-1} and 1.09×10^{12} s^{-1} for b=1.5 and 1.22×10^{12} s^{-1}, 1.25×10^{12} s^{-1} and 1.21×10^{12} s^{-1} for b=2, respectively (Table 1).

TABLE I
OBTAINED TL KINETIC PARAMETERS USING DIFFERENT METHODS.

Methods	Order of Kinetics, b	Activation Energy, E (eV)	Frequency factor, s (s^{-1})
Mathematica	1.0	1.00	1.00×10^{14}
	1.5	1.00	1.00×10^{12}
	2.0	1.20	1.00×10^{12}
TLPC (VHR)	1.0	1.05	1.02×10^{14}
	1.5	1.03	9.95×10^{11}
	2.0	1.25	1.22×10^{12}
TLPC (PS)	1.0	1.01	1.03×10^{14}
	1.5	1.02	9.80×10^{11}
	2.0	1.21	1.25×10^{12}
TLPC (TP)	1.0	1.03	9.97×10^{13}
	1.5	1.04	1.09×10^{12}
	2.0	1.23	1.21×10^{12}

V. CONCLUSIONS

In this study, the named Thermoluminescence Parameters Calculator (TLPC) software was developed by using C# programming language at Visual Studio.Net platform and

TLPC has been tested in the Microsoft Windows-based operating systems. In order to verify the calculated values and reliability of TLPC program, the results are compared with the first, second and general order TL glow peaks obtained from the Mathematica. As a result of the comparisons, it can be said that the TL kinetic parameters of the studied material can be calculated easily, practically and correctly by using the developed TLPC program.

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