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The Effect of Experimental Cycles on the Traps Depths of Dosimetric Traps of Natural Calcite Minerals

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ABSTRACT

A trap found in a solid state radiation dosimetry is characterized by kinetic parameters such as trap depth (E_a), frequency factor (s), kinetic order (b) and carrier concentration (n_o). Trap depth (Activation energy) is the required energy to release carriers in the trap. In this study, it is investigated that how the dosimetric trap depths of the traps found in the four natural calcite minerals are affected by reusable of them as a dosimeter. All samples were irradiated about 36 Gy beta dose and read out by a thermoluminescence dosimeter (TLD) reader. A computer glow curve deconvulation program (CGCD) was used to get the kinetic parameters. And the results are compared for the four calcite samples.

Keywords: Thermoluminescence, Dosimetry, Radiation, Calcite.

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Introduction

Some insulators and semiconductors emit light when they are stimulated by heat after absorbing radiation dose from some radiation source such as beta source. This special emitting light is called as thermoluminescence (TL). The energy of the emitted TL light is different from the absorbed energy due to radiative transition between localized and delocalized band in the structure. In the TL process, an insulator absorbs radiation energy then electron-hole pairs are generated in conduction and valance band. The charge carriers (electrons and holes) are trapped by electron and hole traps (also known as localized energy levels) found in forbidden bandgap. The traps are metastable states in the forbidden bandgap. These are defects such as vacancy, dislocations and impurity atoms. Without applying thermal energy, the charge carries stay in the traps. When enough thermal energy is given to the solid, these charge carriers release from traps and moves into the delocalized bands (conduction and valance band). Finally they recombine radiatively with their opposite charges in the recombination center and thermoluminescence light emits. In the TL studies, the variation of TL light intensity as a function of temperature is obtained to know informations about structure of the material. The graph given in figure1 is called as TL glow curve. And also, in dosimetric studies, the absorbed radiation dose is calculated by calculating area under the TL glow curve.

In the Figure1, each peak generally represents a trap. The TL glow curve has four peaks in the Figure1. Each trap is evicted completely at specific peak temperature.

A trap found in the sample is characterized by kinetic parameters such as activation energy (E_a), frequency factor (s), kinetic order (b) and carrier concentration (n_o). Activation energy, also known as trap depth, is the required energy to release carriers in the trap. Kinetic order gives the relation between the rate of retraping and recombination. Frequency factor is the number of attempt of carrier to escape from the trap. The carrier concentration gives the number of trapped carrier and peak integral generally gives it. Trap depth or activation energy (E_a) is required energy, expressed in electronvolt, to release charge carrier (electron or hole) from trap to conduction or valance band of a crystal. These traps are metastable state or level within the forbidden bandgap.

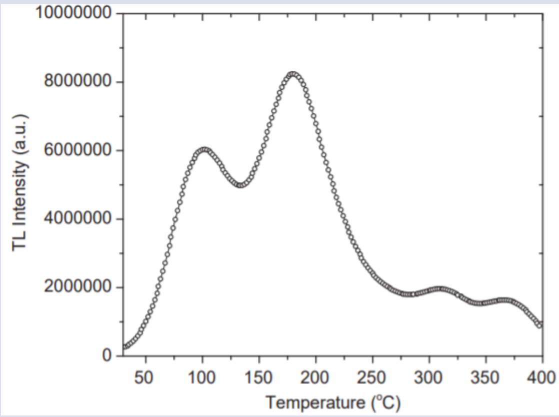

Figure 1. An example of TL glow curve [1]

The metastable state can be electon trap near to the conduction band or hole trap near to the valance band, or a luminescence center (recombination center) near to the middle of bandgap [2-5]. The trap depth for electron trap is measured from the trap to the bottom of conduction band and for the hole trap from trap to top of the valance band.

In the dosimetric studies, reusability (also known as reproducibilty) effect plays important role. At every usage of radiation dosimeter, different TL glow curve may be obtained and this is undesired result. For a good dosimeter, same TL glow curve is desired at the end of each reading.

In this study, it is investigated that how the dosimetric trap depths of the traps found in the four natural calcite minerals are affected by reusable of them as a dosimeter.

Samples and Methods

Four different natural calcite minerals were used in this work. They are tagged as sample1, sample2, sample3 and sample4. The TL glow curves of the four samples were obtained after irradiation sample by a beta source and read out by a TLD reader. All examples were irradiated about 36 Gy at room temperature with a point beta source (90Sr-90Y) which delivers 0.040 Gy/s. Its activity is about 3.7 GBq (100 mCi). Glow curve measurements were made using a Harshaw TLD System 3500 Manual TLD Reader at 1 oC/s heating rate experiment. The irradiator and the TLD Reader were shown in Figure 2. The kinetic parameters of the traps found in the samples were obtained by a computerised glow curve deconvulation (CGCD) method developed by Afouxenidis et al [6]

Figure 2. (a) Irradiator and (b) TLD Reader

When the glow curve has overlapping peaks it is suitable to use Computer Glow Curve Deconvolution Method program, which become popular way to study the trapping parameters parameters such as trap depth (Ea), frequency factor (s), and kinetic order (b) from thermoluminescence glow curves in recent years [7]. In this method, the TL intensity-Temperature data is imported to the program and several satellite peaks are simulated by entering kinetic parameters. Due to the suitable simulated satellite peaks, a simulated TL glow curve is created. The main aim in the program that superimpose experimental TL glow curve and simulated TL glow curve on each other. Then this simulated curve is fitted to experimental curve.

Results and Discussion

In the results part the fitted glow curves of the four samples and their variation of trap depth as a function of reusability were given.

Calcite sample1

In the Figure 3 (a) the fitted TL glow curve of one of the repeat experiment for the sample1 is given. The TL glow curve is fitted with six satellite peaks which locate at different temperatures. The Figure of merite (FOM) for this cycle of measurement was found 0.947%. The variation of the activation energies of two main satellite peaks located at ~140 oC and at ~320 oC are given in the Figure 3(b). The activation energy of the peak at ~140 oC does not affected by the repeat of experiment and found 0.6 eV but there is a change for the activation energy of the peak at ~320 oC. Its value fluctuates from 1.8 eV to 2. 1eV. Besides these, the FOM values for the fitted TL glow curves of each cycle is given Table 1. The standard deviation for the peak at ~140 oC and at ~320 oC are 1.57% and 7.07%, respectively.

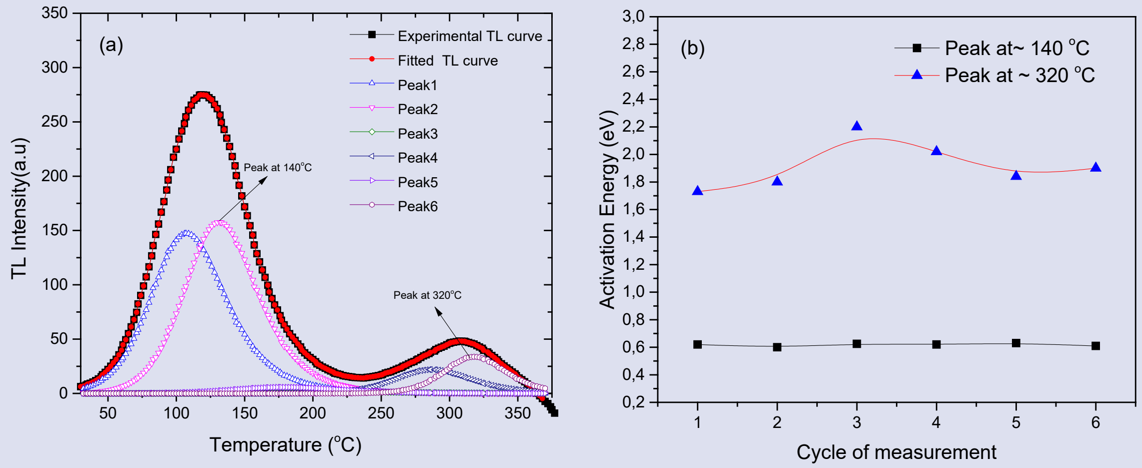

Figure 3. For the sample1: (a) The fitted TL glow curve of one of the cycle of measurement, (b) variation of activation energy of two main peaks.

Table 1. The values of Figure of merit (FOM) for the six cycle of measurements

	Cycle 1	Cycle 2	Cycle 3	Cycle 4	Cycle 5	Cycle 6
FOM (%)	1.29	1.06	1.2	1.01	1.26	1.05

Calcite sample2

The fitted TL glow curve and their five satellite peaks for the second calcite sample are shown in Figure 4 (a). The reusability of this sample is investigated via three trap depths located at ~90 oC, ~140 oC and ~245 oC shown in Figure 4(b). It is seen that the peak at ~90 oC is more stable than others at ~140 oC and ~245 oC. The standard deviation for the peak at ~90 oC, ~140 oC and ~245 oC are 5%, 6.2%, and 5.2%, respectively. For the second calcite sample, the FOM values for the fitted TL glow curves of each cycle is given Table 2

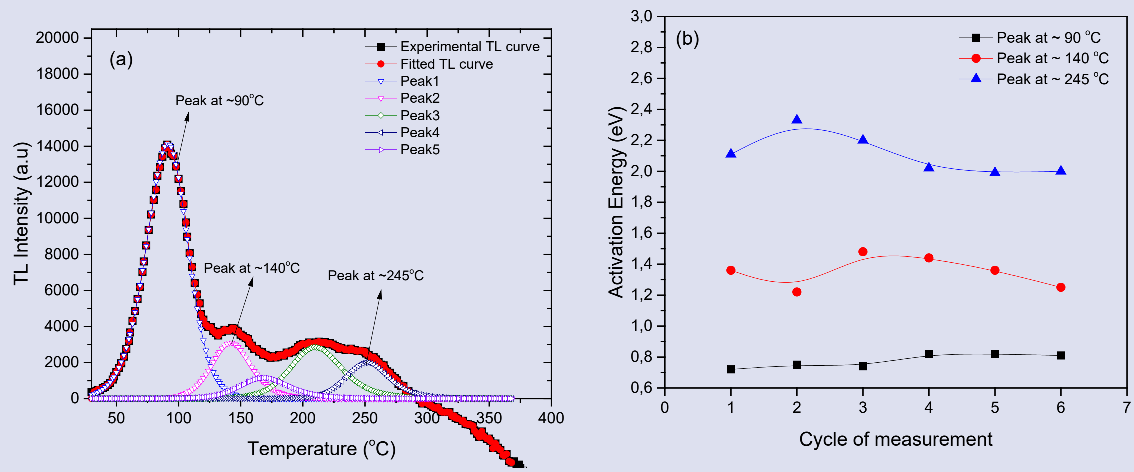

Figure 4. For the sample2: (a) The fitted TL glow curve of one of the cycle of measurement, (b) variation of activation energy of three main peaks.

Table 2. The values of Figure of merit (FOM) for the six cycle of measurements

	Cycle 1	Cycle 2	Cycle 3	Cycle 4	Cycle 5	Cycle 6
FOM (%)	1.74	2.23	2.38	2.2	1.35	1.55

Calcite sample3

The TL glow curve of the third calcite sample is fitted by six satellite peaks shown in Figure 5(a). The FOM values of the each cycle is given in the Table 3. The variation of the traph depths of three main peaks at ~115 oC, ~245 oC and ~290 oC as a function of repeat of experiment is drawn in Figure 5(b). The stabilities of the peak at ~115 oC and ~245 oC are better than the peak at ~290 oC. The standard deviation for the peak at ~115 oC, ~245 oC and ~290 oC are 0%, 3.8%, and 12.5%, respectively.

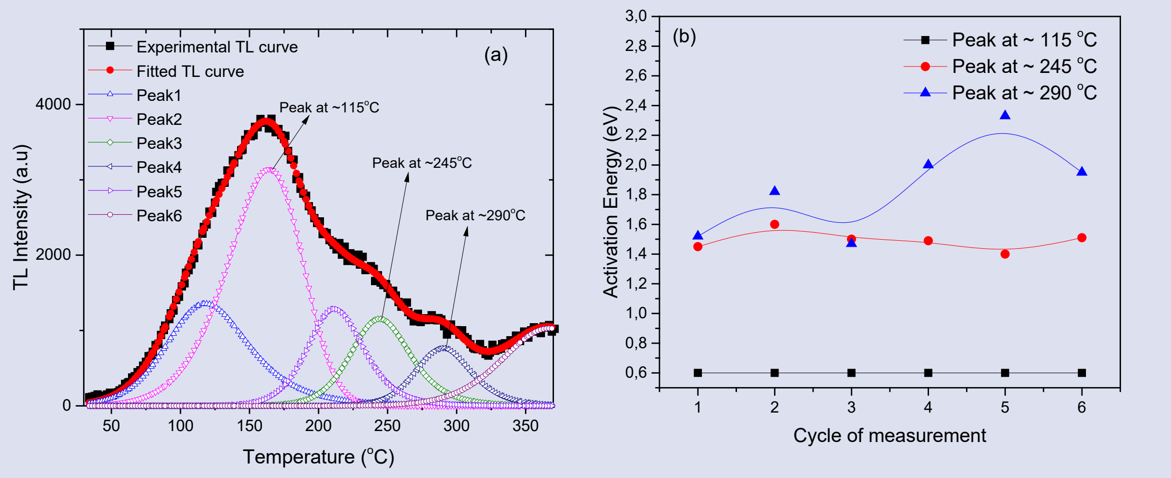

Figure 5. For the sample3: (a) The fitted TL glow curve of one of the cycle of measurement, (b) variation of activation energy of three main peaks.

Table 3. The values of Figure of merit (FOM) for the six cycle of measurements

	Cycle 1	Cycle 2	Cycle 3	Cycle 4	Cycle 5	Cycle 6
FOM (%)	3.03	2.91	2.89	2.74	2.49	2.32

Calcite sample4

For the last calcite sample, the fitted TL glow curve of one of the cycles is obtained by using five satellite peaks shown in Figure 6(a). In the Table 4, all FOM values for each cycle of the fourth calcite sample are tabulated. Like other samples, the stabilities of the traph depths of three main traps located at ~115 °C, ~225 °C and ~340 °C are investigated in Figure 6(b). It is seen that the trap depth of the trap at ~115 °C is more stable than of the traps at ~225 °C and ~340 °C. The standard deviation for the peak at ~115 °C, ~225 °C and ~340 °C are 0.6%, 12.7%, and 6.3%, respectively.

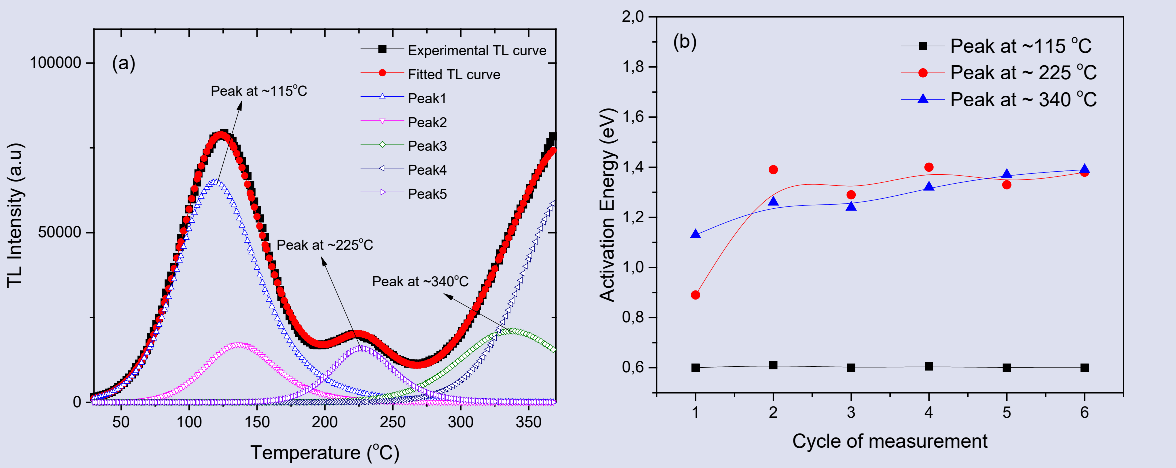

Figure 6. For the sample4: (a) The fitted TL glow curve of one of the cycle of measurement, (b) variation of activation energy of three main peaks.

Table 4. The values of Figure of merit (FOM) for the six cycle of measurements

	Cycle 1	Cycle 2	Cycle 3	Cycle 4	Cycle 5	Cycle 6
FOM (%)	1.23	1.86	1.31	1.63	1.74	1.24

Conclusion

Trap depth (activation energy) is one of the kinetic parameters of a trap. It is the required stimulation energy to evict all trapped charge carriers from the trap. The kinetic parameters are found by several methods. Computer glow curve deconvulation (CGCD) or computerised curve deconvolution (CCD) is a analysis program to get kinetic parameters of traps. The computerised curve deconvolution (CCD) analysis of thermoluminescence (TL) glow curves and optically stimulated luminescence (OSL) decay curves into their individual glow peaks and components respectively have been recognised over the last 30 years to be of major importance[6, 8-12]. The main aim in the program that superimpose experimental TL glow curve and simulated TL glow curve on each other. Then this simulated curve is fitted to experimental curve. The goodness of fit is tested by using the figure of merit (FOM) [13,14]. Generally, in the cases of experimental TL glow-curves the fit is acceptable for FOM% values less than 2%.

The FOM is a very convenient expression to take as ameasure of goodness of fit:

$$FOM[\%] = \sum_{J_i}^{J_f} \frac{100|y_j - y(x_j)|}{A} x100\% \qquad (1)$$

where J_i= first channel in the region of interest, J_f= last channel in the region of interest, y_j = information content j, $y(x_j)$ = value of the fitting function in channel j, A= integral of the fitted glow curve in the region of interest. It can be said that if the values of the FOM are between 0.0% and 2.5% the fit is good, 2.5 % and 3.5% the fit is fair, and > 3.5% it is bad fit [1,3]. In this study, the effects of repeat of experiment on the trap depth of the main traps found in four different natural calcite ($CaCO_3$) samples were investigated. For the four samples, a simulated TL glow curve with several satellite peaks is fitted to the experimental TL glow curve. Their FOM values are given in table 1, 2, 3 and 4. The FOM values of all cycles (repeat) for the sample1 and 4 are in the range of acceptable value (< 2%). Although some of cycles for the sample2 and all of the cycles for the sample3 have bigger FOM values but they are very close to 2%. Generally, the trap depth of the trap found at ~100°C is not affected by the cycle of measurement (repeat of experiment) for all samples. Its value is about 0.6eV with very small standard deviation. However, the trap depths of the traps found at higher temperature region between 200°C and ~350°C fluctuate between 1.4eV and 2.1eV. It means repeat of experiment with higher temperature changes the structure of the traps. In the other words, there is a trap conversion between too close traps with repeat of experiment due to the higher temperature [2,15]. The stabilizing the trap structure can be achieved by annealing preparation. In order to prepare a thermoluminescent material for use, it is needed to perform a thermal treatment, usually called annealing, carriedout in oven or/and furnace, which consists of heating up the TL samples to a predetermined temperature, keeping them at that temperature for a predetermined period of time and then cooling down the samples to room temperature [16,17].

In conclusion, the trap depths of shallow traps are almost unaffected by the repeat of experiment but the reusage of the sample changes the traph depths of deeper traps.

Conflicts of interest

The authors state that did not have conflict of interests.

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