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General features and kinetic analysis of thermoluminescence from annealed natural quartz

Sunil Thomas, M.L. Chithambo*

Department of Physics and Electronics, Rhodes University, Grahamstown 6140, South Africa

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ABSTRACT

The aim of this study is to explore the thermoluminescence properties of beta irradiated natural quartz annealed at 900 °C, that is, beyond its second phase inversion temperature of 867 °C. The sample was annealed to improve its sensitivity and to deplete any residual charge from electron traps. The glow-curve corresponding to 10 Gy shows three peaks when measured at 1 °C/s; a dominant one at 71 °C, the subject of this report, and two other ones at 125 and 177 °C. The dose response of the main peak was studied in the range 1–300 Gy. The activation energy of the main glow-peak was evaluated as ~1 eV. Kinetic analyses using various methods show that the main glow-peak follows first order kinetics.

1. Introduction

Quartz is one of the most abundant natural minerals used in retrospective dosimetry and is composed of silicon dioxide (SiO_2). Since quartz has in most cases well-defined thermoluminescence (TL) glow-peaks, it is suitable for use in radiation dosimetry [1]. Quartz is also used in a range of luminescence based applications such as testing the authenticity of art objects, nuclear accident dosimetry, food irradiation control, and dating of geological and archaeological materials [2,3]. The luminescence properties of quartz have been investigated widely in both its natural and synthetic forms [4–11].

The TL sensitivity of quartz is affected by annealing as has been shown in a number of reports e.g. [4,5,7,11–16]. The influence of annealing on the sensitivity of TL of high purity synthetic quartz, natural quartz of hydrothermal origin from Arkansas, USA, and sedimentary quartz from Chalkidiki in Northern Greece has been reported by Polymeris et al. [7]. The conclusion of Polymeris et al. [7] was that sensitization increases with annealing temperature due to the combined influence of the pre-dose effect and thermal activation. The effect of heat treatment on the TL intensity and emission spectra of quartz of various origin such as hydrothermal, synthetic and volcanic was reported by Rendell et al. [4] who noted that annealing to 1000 °C enhanced the TL intensity of quartz samples by factors of up to 1000 times.

The annealing-induced change in TL sensitivity of quartz in early studies was attributed to increased recombination probability and modifications in the physical processes of luminescence [14]. The difference between TL emission bands of annealed and unannealed quartz reflects changes in the physical processes of luminescence [17]. The sensitivity of optically stimulated luminescence increases for quartz annealed near its first phase inversion point of 573 °C [17] with further enhancements noted near 870 °C [11]. Studies of time-resolved luminescence spectra of quartz show that changes in lifetimes and sensitivity are due to the transfer of holes between luminescence centres caused by annealing [9,11]. In another work on synthetic quartz [18], it was determined that annealing did not create additional defects. The change in the emission wavelength and the lifetimes of quartz annealed beyond 500 °C was also discussed by Pagonis et al. [19]. There are other relevant studies on, for example, TL of unannealed quartz [20], quartz annealed at 460 °C [21] or between 350 and 700 °C [22] or at 800 °C [23].

The luminescence lifetimes of quartz are independent of annealing up to 500 °C but decrease thereafter and a small increase in lifetimes with annealing temperature is observed between 800 and 1000 °C [8,11]. In addition, lifetimes decrease monotonically with measurement temperature and analysis of the change produces unusually low values of the activation energy of thermal quenching [8]. This unusual response of lifetimes to annealing between 800 and 1000 °C was one of the motivations for choosing the quartz annealed at 900 °C for this work.

The primary aim of this work is to study thermoluminescence of quartz annealed at 900 °C, that is beyond its second phase inversion temperature, to assess any thermal effects such as thermal quenching, and to compare the results with the literature. In particular, we investigate the kinetic parameters such as activation energy, frequency factor and order of kinetics of the main TL peak using various methods.

* Corresponding author.
E-mail address: m.chithambo@ru.ac.za (M.L. Chithambo).

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Supplementary studies namely reproducibility, fading and dose response have been made on the main glow-peak. Identifying the point defects responsible for the luminescence in the quartz was outside the scope of this work.

The reasons for choosing the main glow-peak (the "110 °C" peak) for the present study are as follows. (1) This peak shows sensitization owing to both irradiation and heat treatment [7,24] (2) Its kinetic parameters change only slightly even after extreme conditions of irradiation and temperature [25] (3) This peak appears in almost all varieties of natural and synthetic quartz (4) The sensitivity of this peak varies either with irradiation temperature [15] or with firing temperature [16].

2. Experimental

Granular natural quartz of size 90–500 μm (BDH Ltd., UK) was used. The sample was annealed at 900 °C for 1 h before use to remove any residual signal and to enhance its sensitivity. The TL measurements were carried out using a RISØ-TL/OSL/DA-20 Luminescence Reader. Samples were irradiated at ambient temperature using a $^{90}Sr/^{90}Y$ beta source at a nominal rate of 0.1028 Gy/s. Luminescence was detected using an EMI 9235QB photomultiplier tube through a 7 mm thick Hoya U-340 filter (transmission band 250–390 nm FWHM).

3. Results and discussion

3.1. General features of the TL glow-curve

A glow-curve measured from the sample at 1 °C/s after irradiation to 10 Gy is shown in Fig. 1. The glow-curve shows three peaks, an intense peak at 71 °C, labelled I, and two other ones at 125 and 177 °C designated as II and III. The analysis of TL presented in this work was carried out on the prominent peak (I) only.

3.2. Reproducibility

In order to examine the reproducibility of peak I, the TL was measured consecutively at 1 °C/s from a sample dosed to 10 Gy. The maximum measurement temperature was 500 °C. Fig. 2 shows the intensity (I_m) against number of repeat. The coefficient of variation for the ten measurements was calculated as 0.95%. For dosimetric application, reproducibility is considered good if the coefficient of variation in the TL intensity after 5–10 cycles is less than 5% [26]. Therefore, the reproducibility of peak I is acceptable. This may be a consequence of annealing above the second phase inversion temperature, as it is well known that the main glow-peak ("110 °C") of quartz is thermally sensitized under repeated cycles of irradiation and measurement.

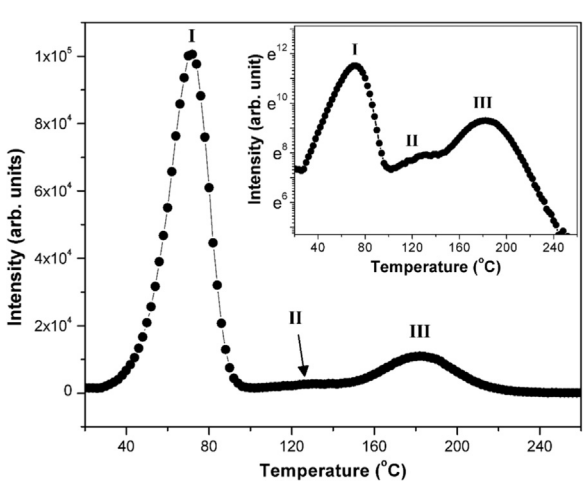

Fig. 1. A TL glow-curve measured at 1 °C/s after irradiation to 10 Gy. The inset shows the same data on a semi-logarithmic scale.

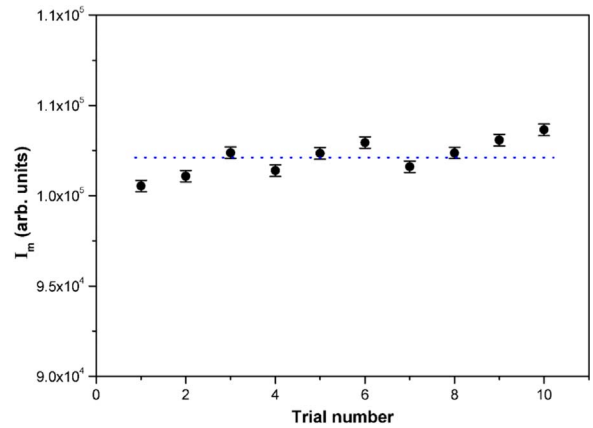

Fig. 2. The intensity of peak I against repeated measurement. The dotted line is only a visual guide.

3.3. Fading

There is a possibility that electrons can escape from electron traps between irradiation and measurement resulting in fading of the TL signal. The fading of peak I was studied by monitoring the change in its intensity at different times between irradiation and measurement. Delays of up to 4 h between beta irradiation and TL measurement were used. Fig. 3 shows the fading of peak I for TL measurements corresponding to 10 Gy. The peak intensity (I_m), noted as peak height, decreased with a mean lifetime (τ) of 2110 s at ambient temperature as deduced by fitting the experimental data with the function $I_m = I_0 exp(-t/\tau)$, where t is time and I_0 is the initial intensity.

3.4. Dose response

The dose response was measured for doses from 1 to 300 Gy. Fig. 4 shows the dose response curve. The TL intensity increases monotonically between 1 and 40 Gy and decreases thereafter when the dose is increased further to 130 Gy with little change in intensity from then on. The figure revealed that the intensity saturates beyond 130 Gy. The reason for this saturation is as follows. The half life of the energy level responsible for the peak (1200 s) is less than the irradiation time above 130 Gy. Therefore, this peak is an example for which simultaneous trap filling due to irradiation and trap emptying due to fading is taking place. This may be the reason for the saturation of the TL intensity at

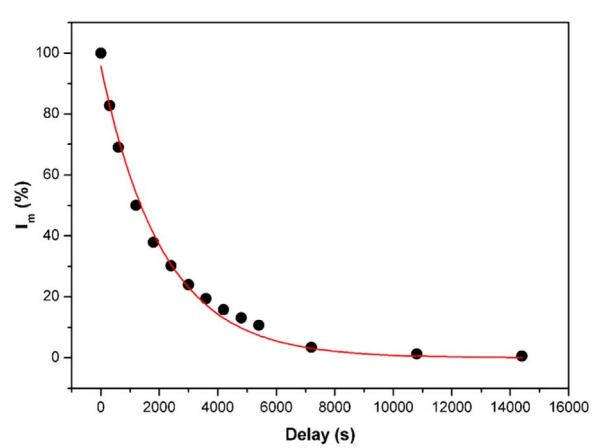

Fig. 3. The change in intensity of peak I with delay between irradiation and measurement. The solid line is a fit of the function $I_m = I_0 exp(-t/\tau)$.

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