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Dependence of Luminescence Lifetimes on Measurement Temperature and Thermal Activation Energy from Quartz using a Time-Resolved Pulsing System

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ABSTRACT

Luminescence is the light emitted from previously irradiated materials when exposed to light of certain wavelength or temperature. One important technique of luminescence is time-resolved optical stimulation of luminescence (TR-OSL). This technique is well known for the study of luminescence from quartz, feldspar, carbon doped aluminium oxide, etc. These materials are of interest in dosimetry. Time resolved optical stimulation is carried out by separating in time the stimulation and emission of luminescence. The luminescence is obtained from the sample using a short pulse of light and the emission of the luminescence is monitored. Here, a new pulsing system based on light emitting diode is designed and is used to study the dependence of luminescence lifetimes on measurement temperature in quartz. The Luminescence lifetimes measured decreased with measurement temperature from 40.4 ± 0.9 μs at 20 ^{0}C to 14.8 ± 1.8 μs at 200 ^{0}C. The dependence of luminescence lifetimes on measurement temperature in quartz was due to thermal quenching at high temperature. Thermal quenching is the decrease of luminescence because of increased non-radiative transitions at high temperature. The activation energy of thermal quenching was also investigated. The value of the activation energy for thermal quenching for quartz was evaluated as 0.67 ± 0.05 eV.

Keywords:
Pulsing system,
Quartz,
Time-resolved luminescence,
Temperature,
Thermal activation energy.

INTRODUCTION

Optically stimulated luminescence is a process whereby an irradiated material emits light when stimulated with light of different wavelength and absorb energy. The ability of quartz to record the amount ionizing radiation exposed to as a luminescence signal within its crystalline lattice makes it an important material for radiation dosimetry. However, for quartz to be used as a natural dosimeter, that part of the signal is removed by exposure to heat or light. In radiation dosimetry, quartz is used as a natural dosimeter to quantify the radiation history of materials.

Optically stimulated luminescence measurements are carried out by the stimulation of an irradiated sample with a light source emitting visible, ultra-violet or infrared light at a certain wavelength. A typical technique of optically stimulated luminescence is time-resolved optical stimulated luminescence (TR-OSL). This technique is frequently used for dating geological and archaeological materials deposited during the last half a million years of quaternary period (Preusser et al., 2009).

In TR-OSL, the luminescence is stimulated using a brief light pulse emitting visible, ultraviolet or infrared at a certain wavelength. The measured luminescence signal consists of linearly increasing luminescence component and scattered stimulating light. After the pulse, the luminescence and the scattered stimulation light are separated by the careful use of band pass filters and transmission filter. The band pass filters are used to transmit the luminescence and the transmission filters to attenuate the intensity of the scattered stimulation light (Botter-Jensen, 1997; Galloway et al., 1997). The measured intensity of the luminescence decreases exponentially in time to produce a decay curve. In time-resolved luminescence, the decay curves can be respectively deconvoluted by non-linear regression into three principal components; the fast, medium and slow component (Bailey et al., 1997; Chithambo & Galloway, 2001; Smith & Rhodes, 1994). Time-resolved luminescence technique gives high signal-to-noise ratio over extended measurement times (Galloway et al., 1997). The spectra obtained from time-resolved luminescence have the potential to provide information

concerning the dynamics of radiative recombination processes associated with specific bands and the nature of the defect where recombination occurs (Bailiff, 2000).

Measurement systems for time-resolved luminescence have been reported [Chithambo & Galloway, 2003; Chithambo, 2011; Sanderson & Clark, 1994; Markey et al., 1995]. Sanderson and Clark (Sanderso & Clark, 1994) make use of a 470 nm light from an N2 dye laser to pulse optically stimulated luminescence from alkali feldspar with a pulse width of the order of 10 ns. Chithambo and Galloway (Chithambo & Galloway, 2003) used a pulsed 525 nm green light-emitting-diode system to measure luminescence from feldspar and quartz. Markey et al. (Markey et al., 1995) carried-out time-resolved luminescence to study features of luminescence from α-Al2O3 in which the light from an Ar-ion laser was used to stimulate luminescence. The aim of this study is to develop a cheaper and easy to use light emitting diode pulsing system for time-resolved measurement and use the system to investigate the behaviour of luminescence lifetimes against measurement temperature in quartz.

MATERIALS AND METHODS

The Pulsing System

Figure 1 shows a schematic diagram of the pulsing system. Figure 1(a) shows the schematic arrangement for detection and measurement of time-resolved luminescence spectra. Figure 1(b) is a schematic diagram showing the designed circuitry used to generates the brief pulse. The detecting system shown in Figure 1(a) was reported previously by Galloway (Galloway, 2002), Chithambo and Galloway (Chithambo & Galloway, 2003), and Chithambo (Chithambo, 2011). The pulsing circuit shown in Figure 1(b) is new and is discussed in detail. A monostable mul tivibrator based on the NE555N timer integrated circuit was used to generates pulses of various duration and the generated pulses are sent into a MOSFET transistor (2N700 MOSFET transistor). A set of 16 LEDs that are arranged in a dural holder were used to stimulate the luminescence, with a Schott GG-420 long-pass filter placed in front of each LED to prevent scattered stimulation light from entering the photomultiplier tube. A transmission filter (Schott BG39), with a transmission peak at 340 nm was placed in front of the photomultiplier tube to transmit the emitted luminescence to the photomultiplier tube. The detected luminescence from several scans is combined giving rise to a time-resolved luminescence spectrum. The spectrum is generated by timing the duration between a START and a STOP signal. A multichannel scaler produces a START signal that triggers the pulsing system to turn ON 16 LEDs for stimulation. The emitted luminescence is detected by a photomultiplier tube (EMI 9635QA) and the signal fed into the combination of a timing filter amplifier (Ortec 474) and a constant-fraction discriminator (Ortec 584). A valid STOP signal is provided by the first photon signal detected from the quartz sample under stimulation. The multichannel scaler then records the luminescence-photon counting rate until a STOP signal arrives. To prevent scattered stimulation light from reaching the photomultiplier tube, a long pass filter (Shott GG-420) was placed in front of the 16 LEDs. A transmission filter (Schott BG39) was then placed in front of the photomultiplier tube to transmit the emitted luminescence.

The signal detected by the photomultiplier tube is fed into the combination of a timing filter amplifier and a constant-fraction discriminator. The timing filter amplifier shape the pulses and optimize the signal-to-noise ratio for timing measurements. These are then furnished to a constant-fraction discriminator where they are counted. The multichannel scaler simultaneously triggers the pulsing circuitry and records the counting rate of events as a function of time.

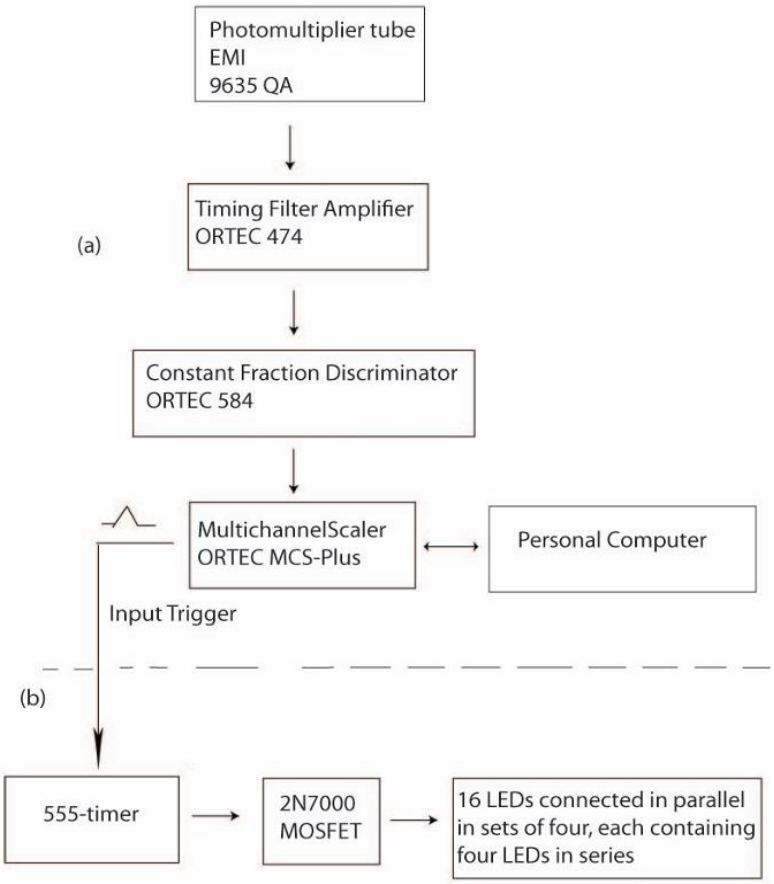

Figure 1: The new pulsing system showing the arrangement for detecting and recording time-resolved luminescence spectra (a) and the pulsing circuitry used to pulse the light emitting-diodes (b)

RESULTS AND DISCUSSION

A typical example of a time-resolved optical stimulation spectrum obtained from a sample of quartz irradiated to 5 Gy and annealed at 500 ^{0}C is shown in Figure 2. A pulse width of 11 s was used to stimulate the luminescence. The solid squares represent the background counts, and the open circles represent the TR-OSL spectrum. The inset shows a portion of the TR-OSL spectrum after the light pulse. The measurements results reported here were obtained using the new designed pulsing system as described above. Each run of measurement was repeated five times, and the mean lifetime is obtained. After the light pulse, Equation 1 was used to fit the TR-OSL spectrum obtained.

$$I(t) = A\,exp\left(-\frac{t}{\tau}\right) + \beta\,. \qquad (1)$$

Here, A is a scaling parameter, τ is the luminescence lifetime, l(t) is the time dependence of luminescence after the light-pulse, t is time and β a constant added to account for the background signal. The fit yielded a luminescence lifetime of 40.2 ± 2.0 s. This value is consistent with ones reported for quartz annealed at 500 ^{0}C. For example, Chithambo and Galloway [Galloway and Chithambo, 2000] reported a luminescence lifetime of 40.0 ± 0.3 s for quartz annealed at 500 ^{0}C, Galloway (Galloway, 2002) reported a lifetime of 39.9 ± 0.4 s for a sample of quartz annealed at 500 ^{0}C, and a lifetime of 41.8 ± 0.3 s for quartz annealed at 500 ^{0}C was reported by Chithambo (Chithambo, 2006). Clearly, the lifetimes obtained using the new pulsing system are in good agreement with those reported in the literature.

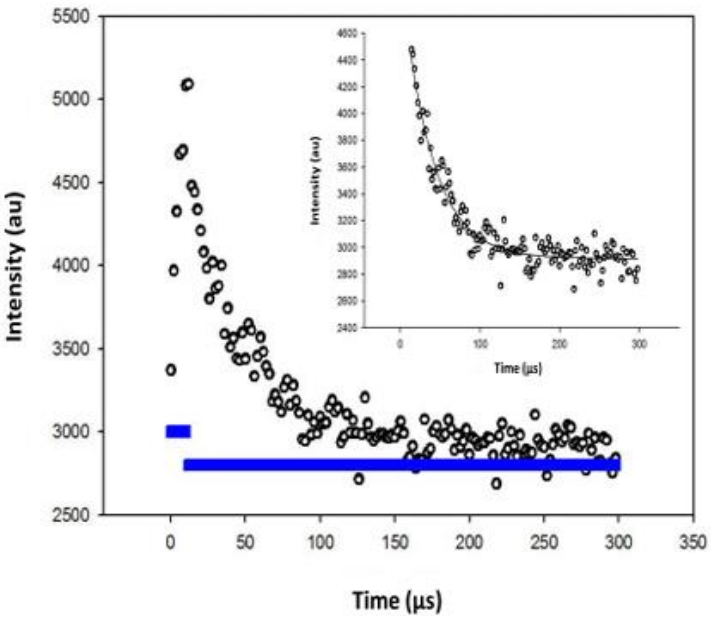

Figure 2: TR-OSL spectrum from a quartz sample irradiated to 5 Gy and annealed at 500 ^{0}C before 470 nm pulsed stimulation. Background counts (solid squares) are shown for comparison. The inset shows the luminescence after the light-pulse

Dependence of luminescence lifetimes on measurement temperature

The effect of measurement temperature on luminescence lifetimes was investigated in quartz annealed at 500 ^{0}C. All measurements were made at 11 s pulse-width, 90 mA pulse-current and a 300 s dynamic range. Note that all data is described in degrees Celsius unless otherwise specified. Figure 4 shows the dependence of luminescence lifetimes on measurement temperature from 20 to 200 ^{0}C represented in the figure as (heating) and from 200 to 20 ^{0}C denoted as (cooling) in steps of 20 ^{0}C, respectively. Luminescence lifetimes obtained from 20 to 100 ^{0}C were constant at about 40.4 ± 0.9 s. Thereafter, the values decreased to a minimum of 14.8 ± 1.8 s at 200 ^{0}C. The decrease of the luminescence lifetimes with temperature is due to increased probability of non-radiative transitions at high temperature known as thermal quenching. Thermal quenching is the loss of luminescence efficiency with increasing temperature (Botter-Jensen et al., 2003). The change in luminescence lifetimes with increasing temperature as shown in Figure 2 is described by and was fitted with a thermal quenching equation of the form:

$$\tau = \frac{\tau_0}{1+C\exp\left(-\frac{\Delta E}{KT}\right)}, \qquad (2)$$

Where τ is the luminescence lifetime at any temperature T, τ_0 the luminescence lifetime at 0 K, C a constant, ΔE the activation energy of thermal quenching and k Boltzmanns constant [Akselrod et al., 1998]. From the fit, ΔE = 0.67 ± 0.05 eV; C = 2 x 10^7 for measurements from 20 to 200 ^{0}C and ΔE = 0.72 ± 0.03 eV; C = 1 x 10^8 for measurements from 200 to 20 ^{0}C. The two experimental values of ΔE agree within experimental error as expected since they refer to the same process. These results are also consistent with values of reported for samples of quartz annealed at 500 ^{0}C as reported in the literature for example, 0.66 ± 0.04 eV by Chithambo and Galloway (Chithambo & Galloway, 2001) and 0.77 ± 0.06 eV by Galloway (Galloway, 2002). Values of ΔE are compared in Table 5.1. Figure 5.5 shows a model used to explain thermal quenching in quartz. The model consists of one radiative centre and one non-radiative centre within the recombination centre. The model allows for non-radiative transitions into the ground state with thermal activation energy of quenching, ΔE (Pagonis et al., 2011, Pagonis etal., 2010). In this model electrons from a trap are raised by optical or thermal stimulation into the conduction band, followed by an electronic transition from the conduction band into an excited state of the recombination centre. Subsequently electrons in this excited state undergo either a direct radiative transition into a recombination centre, or a competing thermally assisted non-radiative process into the ground state of the recombination centre

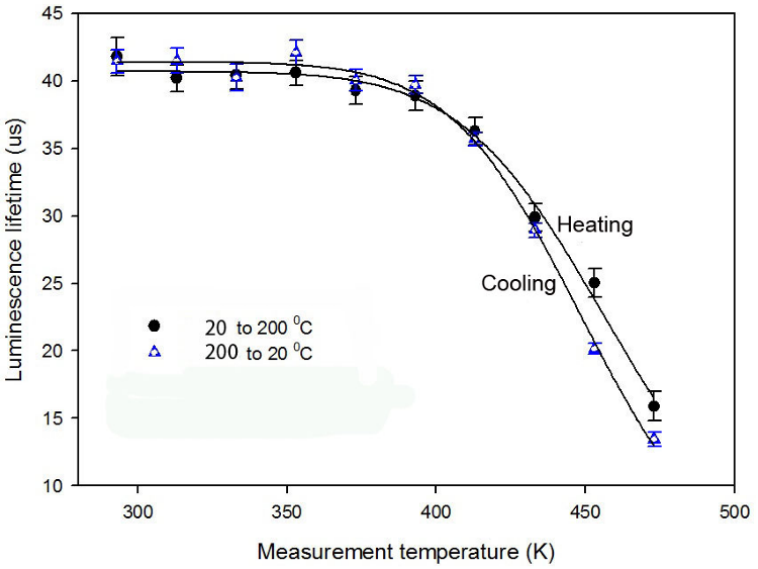

Figure 3: Dependence of luminescence lifetime on measurement temperature in quartz. The TR-OSL spectra were measured from 20 to 200 ^{0}C (a) and from 200 to 20 ^{0}C (b), in steps of 20 ^{0}C

CONCLUSION

A time-resolved optical stimulation spectra obtained from quartz using a new pulsing system that was developed was reported. The pulsing system was used to study the dependence of luminescence lifetime on measurement temperature. Results obtained using the pulsing system to measure the dependence of luminescence lifetime on measurement temperature were presented and discussed. The time-resolved optical stimulation spectra from quartz were measured to demonstrate the system performance. The time-resolved optical luminescence was measured at various temperatures from 20 to 200 ^{0}C and from 200 to 20 ^{0}C in steps of 20 ^{0}C, respectively for a sample of quartz annealed at 500 ^{0}C. The luminescence lifetimes decrease as the temperature increases. The decrease of the lifetimes is due to thermal quenching. For measurements from 20 to 200 ^{0}C, the activation energy of thermal quenching ΔE of 0.67 ± 0.05 eV was obtained for the quartz. When the temperature was decreased from 200 to 20 ^{0}C, we obtained 0.72 ± 0.03 eV activation energy of thermal quenching. These values of ΔE agree with each other and with others reported in the literature. The performance of the new pulsing system had shown to be consistent with the results of luminescence lifetimes dependence on measurement temperature of quartz.

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