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TEMPERATURE DEPENDENCE OF LUMINESCENCE LIFETIMES OF A-AL$_2$O$_3$:C USING TIME-RESOLVED OPTICAL STIMULATION

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ABSTRACT

This study reported the performance of a newly developed time-resolved pulsing system by investigating the components of time-resolved optically stimulated luminescence (TR-OSL) signal from carbon-doped aluminium oxide, α-Al$_2$O$_3$:C, a material of interest in dosimetry. The study demonstrates time-resolved optical stimulation (TR-OSL) measurements under a brief light-pulse LEDs stimulation. TR-OSL is a technique that separates in time the stimulation and emission of luminescence. In this work, we report the influence of measurement temperature on luminescence lifetimes in α-Al$_2$O$_3$:C obtained using a new pulsing system. Luminescence lifetimes were measured from 20°C to 140°C using a pulsed LED system with a 17 ms stimulation duration. Despite extensive studies on the luminescence properties of α-Al$_2$O$_3$:C, the precise influence of temperature on time-resolved optical stimulation luminescence lifetimes remains insufficiently characterized. Here, we designed and developed a new cost-effective time-resolved pulsing system based on blue LEDs. The measured luminescence lifetimes decreased from 36.8 ± 0.1 ms at 20 ^{0}C to 28.0 ± 0.6 ms at 140 ^{0}C. Luminescence lifetimes in α-Al$_2$O$_3$:C exhibited thermal quenching at elevated temperatures, attributed to increased non-radiative transitions. The value of the activation energy for thermal quenching for α-Al$_2$O$_3$:C was evaluated as 1.02 ± 0.01 eV. The measured activation energy of 1.02 ± 0.01 eV agrees with earlier findings 1.075 ± 1.0 eV reported by Pagonis et al. (Pagonis et al., 2013), 0.96 ± 0.005 eV reported by Ogundare et al. (Ogundare, 2012) and 0.95 ± 0.04 eV by Chithambo et al. (Chithambo, 2014), confirming the thermal quenching model.

Keywords: Time-resolved luminescence, α-Al$_2$O$_3$:C, Temperature, Thermal activation energy, Pulsing system, LEDs

INTRODUCTION

Optically stimulated luminescence (OSL) is the emitted light at certain wavelength that arises from previously irradiated minerals or materials when exposed to light at different wavelength. During exposure, the absorbed energy by the ionizing radiation by the mineral generates free electrons and holes withing the volume of the detectors and some of which gets trapped at point defect within the mineral (Uriri, 2016). During optical stimulation with photons, the trapped charges (usually electrons) are released and some of these charges recombine with charge carriers of opposite sign (usually holes) and release energy that consist of luminescence light because of the electronic transition at the luminescence centers. The population of electrons in the traps is due to the irradiation of the material, and therefore the OSL intensity is related to the absorbed radiation dose (Denis et al., 2011). There are three basic modes for optical stimulation of luminescence. These are continuous-wave OSL (CW-OSL), linear-modulation OSL (LM-OSL), and time-resolved OSL (TR-OSL).

Time-resolved optical stimulation (TR-OSL) is an important technique for measurement of OSL whereby light pulses of constant intensity are used to separate in time the stimulation and emission of luminescence (Uriri, 2016). The luminescence is stimulated using a short light pulse emitting visible, infrared light or ultra-violet at a certain wavelength which is detected by a photomultiplier tube. The measured 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 luminescence intensity decreases exponentially in time, resulting to 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 and Galloway, 2001; Smith and Rhodes, 1994). Time-resolved luminescence technique gives high signal-to-noise ratio over extended measurement times. 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 (Baliff, 2000).

Measurement systems for time-resolved luminescence based on LED systems and lasers have been reported (Kang et al., 2024; Haggar et al., 2022; Kang et al., 2020; Chithambo and Galloway, 2003; Chithambo, 2011; Sanderson and Clark, 1994; Markey et al., 1995). Sanderson and Clark (Sanderson & 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. (1995) carried-out time-resolved luminescence to study features of luminescence from α-Al$_2$O$_3$ in which the light from an Ar-ion laser was used to stimulate luminescence. α-Al$_2$O$_3$ is a crystalline phase of trigonal symmetry with a rhombohedral lattice (Chithambo & Gostin, 2016). Research interest in aluminium oxide, particularly in its polymorphic phase is rapidly growing due to its unique physical and chemical properties which generate point-defects, such as oxygen vacancies, that can be used for a different purpose including luminescence-based applications (Agullo-Lopez, 1988; Evans, 1995). α-Al_2O_3 is relevance in dosimetry due to its sensitivity to radiation and it is mostly used as a sensitive synthetic dosimeter. Although time-resolved optical stimulation techniques have been applied to various luminescent materials, a systematic study on the temperature dependence of luminescence lifetimes in α-Al_2O_3:C remains limited. Understanding these variations is crucial for improving dosimetric accuracy and refining TR-OSL methodologies.

In this study, a newly designed pulsing system that uses blue light emitting diodes is used to study the luminescence lifetimes from α-Al_2O_3. The pulsing system has been developed to enable the automatic control of pulse width and stimulation power and to drive a high pulse-current of up to 90 mA per light emitting-diode.

MATERIALS AND METHODS

The new pulsing system

A new light-emitting diode based pulsing system is developed that is capable for time-resolved optically stimulated luminescence. 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 for generating the pulse. The detecting system shown in Figure 1(a) was reported previously by Galloway (2003) and Chithambo (2011). The pulsing circuit shown in Figure 1(b) is new and is discussed in detail.

A monostable multivibrator 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. In order to measure emission spectra of the light-emitting-diodes, calibration of the spectrophotometer was necessary. An experiment was conducted to calibrate the spectrophotometer. The spectrophotometer was calibrated using a mercury lamp as a reference source. Spectrum of this lamp was first obtained via measurement using the spectrophotometer software. As the spectrum from the lamp was displayed, the spectrophotometer was then calibrated using the calibration button in the software.

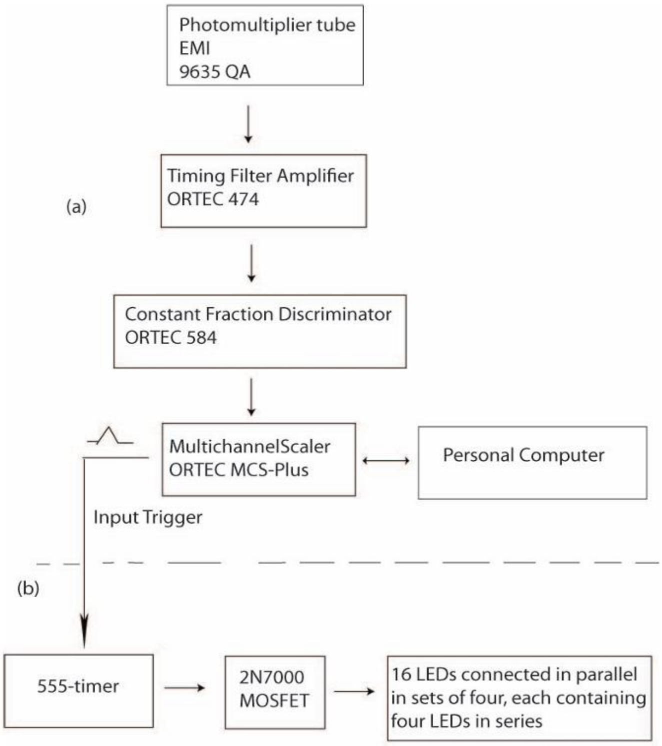

Figure 1: Schematic diagram of the pulsing system showing the arrangement for detecting and recording time-resolved optical luminescence spectra (a) and the pulsing circuitry used to pulse the light emitting-diodes (b)

RESULTS AND DISCUSSION

A new LED based pulsing system was designed to study the dependence of luminescence lifetimes on measurement temperature in α-Al_2O_3, a highly sensitive luminescence dosimeter. The sample used was irradiated to 1.0 Gy before 470 nm pulsed stimulation. The luminescence was stimulated at a 17 ms pulse-width using 150 ms dynamic range. TR-OSL spectra were measured from 20 to 140 ^{0}C in steps of 20 ^{0}C. The irradiation was carried out in a RISO optical stimulated luminescence reader prior to the LEDs stimulation.

Figure 2 shows an example of a TR-OSL spectrum obtained from α-Al_2O_3:C following beta irradiation to 1.0 Gy. Measurements were made at 40 and 140 ^{0}C. The solid squares in the figure show the background counts. The inset shows the portion after the light-pulse. The portion after the light-pulse was analysed using Equation 1 to give the associated luminescence lifetime at a particular temperature. A luminescence lifetime of 35.5 ± 0.5 ms was obtained from the fit of Figure 2 (inset). All data were analysed using the powerful scientific data analysis software called sigmaplot.

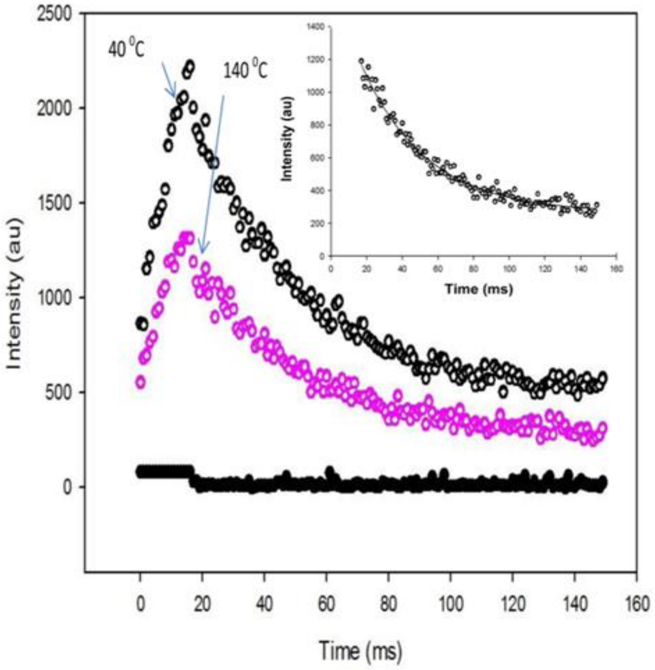

Figure 2: A TR-OSL spectrum from a sample of α-Al_2O_3:C following beta irradiation to 1 Gy. Measurements were made at 40 and 140 ^{0}C. Background counts (solid squares) are also shown. The inset shows the portion after the light-pulse.

Figure 3 shows the dependence of luminescence lifetimes on measurement temperature. The experiment was conducted at measurement temperatures from 20 to 140 ^{0}C in steps of 20 ^{0}C. Luminescence lifetimes from 20 to 60 ^{0}C were constant at about 36.8 ± 0.1 ms. Thereafter, the values decreased to a minimum of 28.0 ± 0.6 ms at 140 ^{0}C. This decrease in luminescence lifetime is evidence of thermal quenching. The luminescence lifetime decreases as a function of temperature as described before as

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

where all parameters are as described previously. The data in Figure 3 was fitted with Equation 5.4, and the values of ΔE and C obtained from the fit are 1.02 ± 0.01 eV and 9 x 10^{11} respectively. The value of ΔE is in good agreement with 1.08 ± 0.03 eV reported by Akselrod et al. (1998), 1.075 ± 1.0 eV reported by Pagonis et al. (2013), 0.96 ± 0.005 eV reported by Ogundare et al. (2012) and 0.95 ± 0.04 eV by Chithambo et al. (2014). Thermal quenching is a process caused by loss of luminescence efficiency. Nikiforov et al. (2001) developed a model that describes thermal quenching in α-Al_2O_3:C based on thermal and optical ionization of F-centres. However, in this work, thermal quenching in α-Al_2O_3:C is described in terms of the model of Pagonis et al. (2011). Figure 4 shows the model of Pagonis et al. (2011) for thermal quenching in α-Al_2O_3:C. This model consists of an electron trap, two deep traps, and excited levels of the F-centre. The model allows non radiative transition into the ground state with thermal activation energy of quenching, ΔE. Pagonis et al (2011) described thermal quenching based on competition between radiative and non-radiative electronic transitions occurring within the recombination centre. The electrons from the trap are stimulated by optical or thermal stimulation into the conduction band. The electrons then transit 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. During this non radiative transition into the ground state of the recombination centre, the absorbed energy is released as phonons.

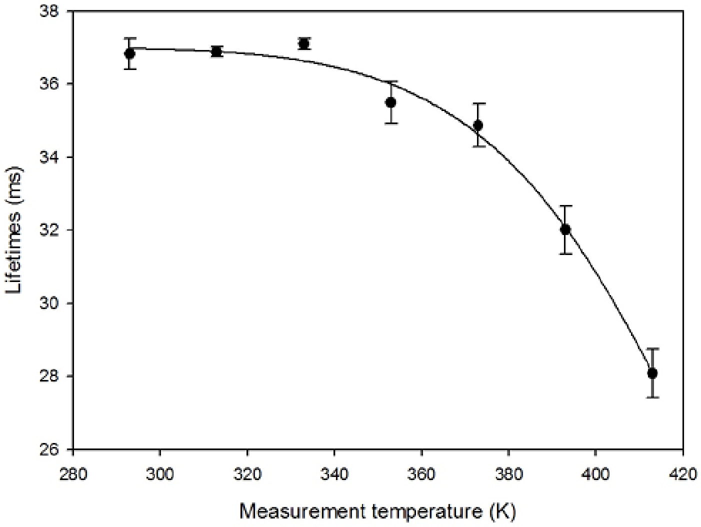

Figure 3: Dependence of luminescence lifetimes on measurement temperature in α-Al_2O_3:C following beta irradiation to 1 Gy. All measurements were made from 20 to 140 ^{0}C in steps of 20 ^{0}C.

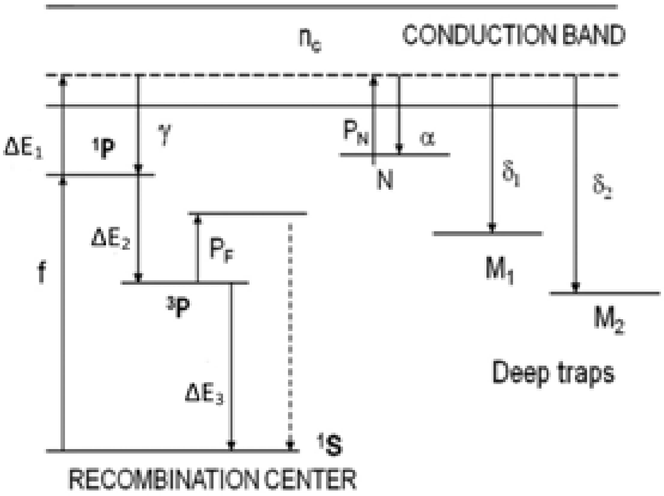

Figure 4: An energy band model used to describe thermal quenching based on competition between radiative and non-radiative electronic transitions occurring within the recombination centre in α-Al_2O_3:C **(**Pagonis et al., 2011)

CONCLUSION

Time-resolved optical stimulation of luminescence spectra from α-Al_2O_3:C were obtained using a newly designed TR-OSL pulsing system based on light-emitting diodes (LEDs). The LEDs in the pulsing system are pulsed at various durations by a 555-timer integrated circuit operated as a monostable multivibrator. The output signal from the pulsing system was detected by a photomultiplier tube and sent to a computer screen for recording. The time-resolved optical stimulation spectra from α-Al_2O_3:C were measured to demonstrate the system performance. A sample of α-Al_2O_3:C was irradiated to a beta dose of 1 Gy and temperature dependence measurements were made from 20 to 140 ^{0}C in steps of 20 ^{0}C to investigates the dependence of luminescence lifetime on temperature. The luminescence lifetime decreases with an increased in temperature. The luminescence lifetimes show evidence of thermal quenching as the measurement temperature increased. The activation energy of thermal quenching ΔE measured for α-Al_2O_3:C was 1.02 ± 0.01 eV. The results of this study contribute to the optimization of luminescence-based dosimeters, where precise temperature-dependent lifetime measurements are crucial for accurate dose assessments. The study was conducted within a limited temperature range (20–140°C), and future research could explore luminescence behavior at lower and higher temperatures to determine if additional quenching mechanisms exist. Further work could investigate the influence of different doping concentrations in α-Al_2O_3:C on thermal quenching behavior, as well as the applicability of the new pulsing system to other luminescent materials. The performance of the new pulsing system had shown to be consistent with the results of the dependence of lifetimes on measurement temperature in α-Al_2O_3:C obtained with others measurement systems.

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