

Relative features of the principal and secondary luminescence lifetimes in quartz

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The form of time-resolved luminescence spectra has been studied for quartz annealed at 800 °C and pulse-stimulated at 470 nm. Depending on the measurement temperature and the irradiation dose, the spectra may be de-convolved into two possible components to produce a principal and subsidiary lifetime component. Measurements between 20 and 200 °C show that both components are affected by thermal quenching of the associated luminescence, qualitatively so for the subsidiary one, but with an activation energy of 0.71 eV for the principal lifetime. Regarding influence of irradiation, time-resolved spectra consist of a single component below about 260 Gy but two components thereafter independent of irradiation. The irradiation-dependent features of the lifetimes are accounted for in terms of the relative importance of the various luminescence centers in quartz.

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1 Introduction Quartz is a common natural mineral with properties that make it amenable for application in radiation dosimetry using luminescence methods [1]. The luminescence properties of quartz including its sensitivity and luminescence lifetimes undergo notable changes when the quartz is annealed, and in particular, near its phase inversion temperatures of 573 and 867 °C [2, 3]. The physical processes leading up to the emission of luminescence in quartz may be investigated using time-resolved optical stimulation. The aim of this method is to separate in time the stimulation and emission of luminescence to enable measurement of time-resolved luminescence spectra which may be resolved into associated lifetimes, defined in this sense as the delay between stimulation and emission of luminescence [4, 5]. A number of such studies show that annealing defines the detailed distribution of lifetimes with measurement temperature as well as the irradiation-dependent characteristics of the lifetimes [3, 6]. In particular, it was noted [3] that spectra measured at certain temperatures could be accurately resolved into more than one component, a scenario that pointed to the possibility of involvement of multiple luminescence centers in the emission of luminescence from quartz.

The aim of this work is to build on the findings described, specifically to study the influence of measurement temperature and irradiation on the principal and subsidiary luminescence lifetimes in natural quartz.

2 Experimental details Measurements were done on samples of 90–500 µm quartz grains (BDH Ltd, UK) annealed at 800 °C for 10 minutes, the same quartz used in previous studies [3, 6]. The heating was intended to provide suitably annealed samples for the study and to enhance the sensitivity of the material to optical stimulation.

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Luminescence was measured using an LED based Pulsing System [4]. The pulses are generated from an integrated circuit multivibrator, a 2N74221, and used to drive a set of LEDs. The diodes thus pulse-stimulate luminescence from samples, which is detected by a photomultiplier (EMI 9635QA) and processed by a multichannel scaler (ORTEC MCS-plus). The MCS was used to simultaneously trigger the set of LEDs and initiate a signal measurement scan. The scaler acquires photon counts sequentially into its memory one channel at a time during a scan. Time-resolved luminescence spectra are then built up as a distribution of cumulative photon counts against time for the dynamic range selected.

A set of four 470 nm blue LEDs (Nichia NSPB-500) were used to stimulate luminescence at 11 μ s duration per scan. The luminescence was detected in the band 340 - 380 nm (FWHM) through filters Schott BG39 (transmission band 340–620 nm FWHM) and UG11 (transmission band 270–380 nm FWHM). A GG420 long pass filter was put in front of each LED to reduce scattered stimulating light. Lifetimes were evaluated from portions of the time-resolved luminescence spectra after the pulse by fitting exponential functions of the form $\sum A_i \exp(-t/\tau_i) + B$ where τ_i is the lifetime of the i th component, A_i is a scaling factor, t is time, and B a constant included to account for the background.

In terms of a model where for a short rectangular pulse of width t_1 , where λ is the probability of stimulation per unit time and $N(t_1)$ the number of stimulated electrons, the time-dependence $L(t)$ of the luminescence after the pulse is exponential, $L(t) = \lambda N(t_1) \exp(-(t - t_1)/\tau)$, where τ is the lifetime.

Figure 1 shows a time-resolved luminescence spectrum measured at 100 °C over 10^5 sweeps from a sample of quartz annealed at 800 °C for 10 minutes and beta-irradiated to 257 Gy. In this example, the signal after the pulse is better fitted by a sum of two exponentials rather than by a single exponential as evidenced by the corresponding residuals plots in the inset. The associated principal and secondary lifetimes in this case are $34.4 \pm 1.2 \mu$ s and $10.1 \pm 0.5 \mu$ s.

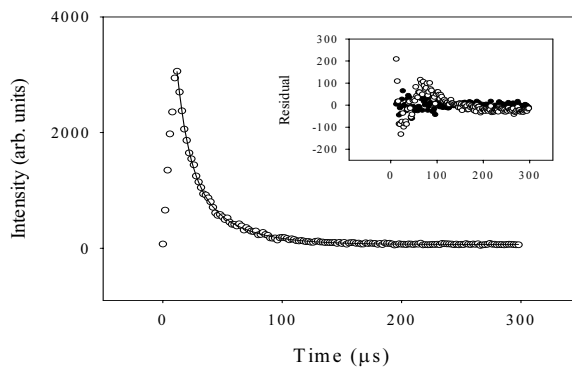


Fig. 1 A time-resolved luminescence spectrum from quartz measured at 100 °C. The solid line is the best fit of a sum of two exponentials, a better alternative (solid symbols, inset) over the use of a single exponential function (open symbols, inset).

3 Results and discussion

3.1 Temperature dependence of lifetimes Figure 2 shows the change of the principal and subsidiary lifetime with measurement temperature for a quartz irradiated to 257 Gy beta dose. In both cases, lifetimes decrease with temperature between 30 and 200 °C. The principal lifetime is about 33 μ s below 120 °C and decreases rapidly thereafter down to about 7 μ s at 200 °C. In comparison, the decrease in the subsidiary lifetime with temperature is continuous from about 15 μ s at 30 °C to 1.5 μ s at 180 °C. The temperature dependence of the principal lifetimes in Fig. 2 has been reported before [3, 6] and is due to increased thermal effect on lifetimes described as $\tau(T) = \tau_{\text{rad}}/(1 + C \exp(-\Delta E/kT))$ where τ_{rad} is the radiative lifetime at absolute zero of temperature, ΔE is the activation energy of thermal quenching, k is Boltzmann's constant, T is absolute temperature in K and C is a constant [3, 6]. The activation energy of thermal quenching was evaluated as 0.71 ± 0.16 eV, in good agreement with typical values in quartz [3,

6]. On the other hand, subsidiary lifetimes cannot be accurately described similarly but do qualitatively reflect the thermal quenching effect.

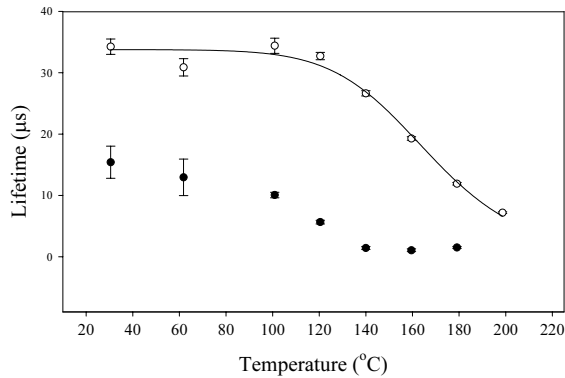


Fig. 2 Temperature dependence of the principal (open symbols) and subsidiary lifetimes (solid symbols). The solid line is the best fit of the thermal quenching equation described in the text.

3.2 Dose dependence of lifetimes Figure 3 shows the dependence of lifetimes on irradiation dose. Time-resolved luminescence spectra corresponding to irradiation doses below 260 Gy could not be reliably resolved into two components and so only one lifetime is shown. The initial increase in the lifetime is consistent with previous observations [3]. However, beyond 260 Gy, a principal lifetime of about 33 μs and a secondary component of about 10 μs , inset, are apparent.

The dose dependence of lifetimes may be explained with reference to the energy band scheme proposed by Galloway [3]. On the basis of the model, the principal lifetimes may be associated with luminescence emitted from one set of centers, L_H and L_L , and the subsidiary lifetimes from another set, L_S , whose corresponding lifetimes are manifested only at certain combinations of annealing and measurement temperature.

The initial increase of lifetimes with dose in Fig. 3 can then be explained by the increase in lifetime from τ_L to τ_H because of the preferential trapping at the L_H center of holes produced during irradiation. The L_H center eventually becomes dominant as seen by the attainment of a stable value of the τ_H lifetime. On the other hand, we suppose that the appearance of the short lifetime τ_S with increase in dose is evidence of hole trapping at the L_S center.

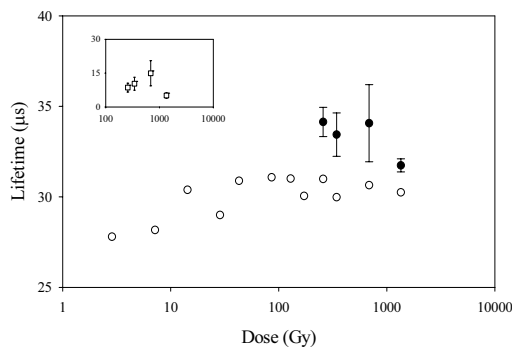


Fig. 3 The dependence of luminescence lifetime on irradiation for the principal lifetime (solid circles) and subsidiary lifetimes (inset) compared to values evaluated using a single exponential.

4 Conclusion The temperature and dose dependent features of luminescence lifetimes from quartz annealed at 800 °C for 10 minutes have been investigated. We have observed that the lifetimes can be resolved into a principal and secondary component, particularly so for samples beta irradiated to greater than 260 Gy. The distribution of principal lifetimes with measurement temperature is affected by thermal quenching of luminescence with an activation energy of about 0.71 eV. On the other hand, the subsidiary lifetimes only qualitatively display this effect. Regarding the effect of irradiation, time-resolved spectra consist of essentially a single lifetime at low irradiation doses up to about 260 Gy. As the dose is increased from then on, the principal and subsidiary lifetimes could be distinguished. The properties of the lifetimes in this case can be explained by the relative dominance of luminescence centres involved in the luminescence emission process [2, 3].

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