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Radioluminescence of annealed synthetic quartz

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H I G H L I G H T S

- Radioluminescence of synthetic quartz is reported.
- The emission band amplitude increases with annealing temperature.
- Beyond 700 °C, the intensity scales up with annealing temperature.
- The emission spectra can be deconvoluted to 7 emission bands.
- The change in emission bands is consistent with change in lifetimes.

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The radioluminescence of synthetic quartz annealed at various temperatures up to 1000 °C is reported. The amplitude of the emission bands increases with annealing temperature. In addition, when samples are annealed at temperatures exceeding 700 °C, the intensity of the radioluminescence increases with duration of annealing. The corresponding emission spectra show seven emission bands at 2.04, 2.54, 2.77, 3.04, 3.40, 3.75 and 3.91 eV. The change in dominant emission band with annealing is consistent with annealing-induced variations in lifetimes determined previously from time-resolved optically stimulated luminescence spectra in the same samples.

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1. Introduction

It is well known that the sensitivity of optically stimulated luminescence (OSL) from quartz is enhanced with annealing to temperatures above its first phase inversion temperature (Bøtter-Jensen et al., 2003; Preusser et al., 2009). A number of other studies (Chithambo and Ogundare, 2009; Chithambo et al., 2011; Galloway, 2002) show that annealing also affects lifetimes evaluated from time-resolved optically stimulated luminescence in natural and synthetic quartz. This change in lifetimes is associated with change in the principal luminescent centres which implies that emission bands should also change with annealing temperature.

The luminescence lifetime (henceforth referred to only as lifetime) described here denotes the delay between optical stimulation and emission of luminescence and consist of the time to evict an

electron from a trap, transit time through the conduction band, and the lifetime of the excited state at the luminescence centre with the latter being the dominant term (Chithambo et al., 2016). Since the lifetimes are closely linked with transitions at luminescence centres, their use aids investigation of dynamics of OSL.

Using the same natural quartz as used previously by Chithambo and Ogundare (2009) and Galloway (2002), Pagonis et al. (2014), reported radioluminescence (RL) measurements where a total of seven emission bands between 1.5 and 4.5 eV were observed and their behaviour studied as a function of annealing temperature. Radioluminescence, which is emitted during exposure of a sample to ionizing radiation, is measured as a function of the emission wavelength and was thus a suitable means to study the influence of annealing on emission bands. In this regard, an emission band at ~3.44 eV (360 nm) was found to be strongly enhanced when the annealing temperature was increased to 500 °C. It was also noted that a new emission band at about 3.73 eV (330 nm) appeared for annealing in the range 600–700 °C. Although the question of whether the recombination centres associated with lifetimes are the same ones involved in RL was not settled, the study confirmed

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in outline that annealing changes luminescence emission bands.

It has previously been shown, using a specific type of synthetic quartz manufactured by Sawyer Research Products (Ohio, USA), that lifetimes in synthetic quartz are also affected by annealing (Chithambo, 2004; Chithambo et al., 2011). In particular, it was noted that whereas lifetimes in natural quartz decrease when the annealing temperature is increased beyond 600 °C (Galloway, 2002), the opposite is observed in synthetic quartz (Chithambo et al., 2011). In view of this, the aim of this work is to report radioluminescence carried out to examine the influence of annealing on luminescence emission bands in the same synthetic quartz, that is, with an identical thermal history and specific distributions of defects and impurities. The purpose of this brief report is to examine whether the change of lifetimes in synthetic quartz can be also be associated with alterations in luminescence emission bands.

2. Experimental details

Granular samples of size between 90 and 500 μm were prepared from a high purity synthetic α -quartz block supplied by Sawyer Research Products (Ohio, USA). This is the same material used previously e.g. (Chithambo et al., 2011; Pagonis et al., 2014). Identical aliquots for use were monolayered on stainless steel sample discs. Measurements were made on both unannealed and annealed synthetic quartz. For the latter, samples were annealed in air at 500, 600, 700, 800, 900 and 1000 °C for 10, 30 and 60 min respectively in quartz boats in a furnace (Galenkamp muffle furnace) and cooled in air thereafter. The RL was stimulated using X-rays from a Philips 2274 X-ray tube with a tungsten target. The operating current and voltage were 20 mA and 20 kV respectively. For each sample, 40 consecutive spectra were measured with each spectrum corresponding to a dose of 10 Gy. The RL was detected using an apparatus build in-house at the University of Milano-Bicocca, Italy, featuring a charge coupled device (Jobin-Yvon Spectrum One 3000) coupled to a spectrograph operating in the wavelength range 200–1100 (Martini et al., 2012a,b).

3. Results and discussion

3.1. General observations

Fig. 1 shows the RL emission spectra measured from the unannealed sample. Two broad emission regions are apparent between

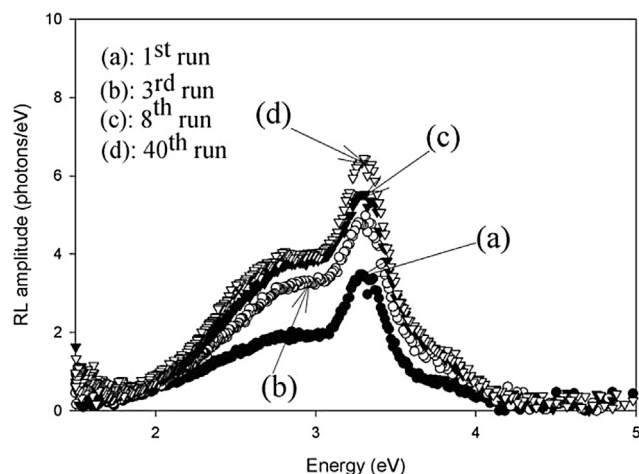


Fig. 1. Radioluminescence spectra of unannealed synthetic quartz. Each run corresponds to an X-ray dose of 10 Gy.

2 and 3 eV and between 3 and 4 eV. The emission bands between 3 and 4 eV are dominant and consistently increase with irradiation as compared to the emission bands between 2 and 3 eV. Fig. 2 shows the RL emission spectra for the sample annealed at 700 °C for 60 min. In this case, the RL emission bands between 2 and 3 eV are more intense and increase faster with irradiation than is observed for the emission bands between 3 and 4 eV. The effect exemplified in Fig. 2 for the sample annealed at 700 °C was also observed in samples annealed at 500 and 600 °C for 10, 30 and 60 min. Fig. 3 shows the spectra for a sample annealed at 1000 °C for 60 min. In this sample, there are at least three distinct bands apparent near 2 eV, between 2 and 3 eV and between 3 and 4 eV. The emission at about 2 eV and the one between 2 and 3 eV initially increases with irradiation and by the 8th run has decreased to a lower intensity. In contrast, the emission between 3 and 4 eV increases to a maximum with irradiation.

3.2. Effect of continuous irradiation on radioluminescence intensity

The main effect of continuous irradiation in the unannealed sample is to increase the RL intensity of all bands in the emission spectrum as shown in Fig. 1. This increase is faster for low doses and eventually shows evidence of saturation at 80 Gy. This slow increase implies that the recombination centre involved eventually saturates. The feature shown in Fig. 3 for the sample annealed at 1000 °C for 60 min is instructive. For this sample, the 2.7 eV emission band starts by increasing with irradiation, gets to a maximum at a dose of 30 Gy i.e. by the third run and then starts to decrease as dose is further increased. Indeed, even the emission band at 2.04 eV is a further illustration of how the emission bands decrease with irradiation after an initial enhancement. In comparison, the two other bands at 3.4 and 3.9 eV increase with dose initially but then tend to saturate with further irradiation. It is not clear what is responsible for these changes. However, one can speculate that radiation-induced effects as radiation damage, trapping and detrapping could cause the decrease or increase of RL signals.

3.3. Effect of duration of annealing on radioluminescence

Fig. 4(a) and (b) compares the RL emission spectra for measurements on the unannealed sample and ones annealed at 700 °C and 800 °C for 10, 30 and 60 min respectively. Fig. 4(a) shows that

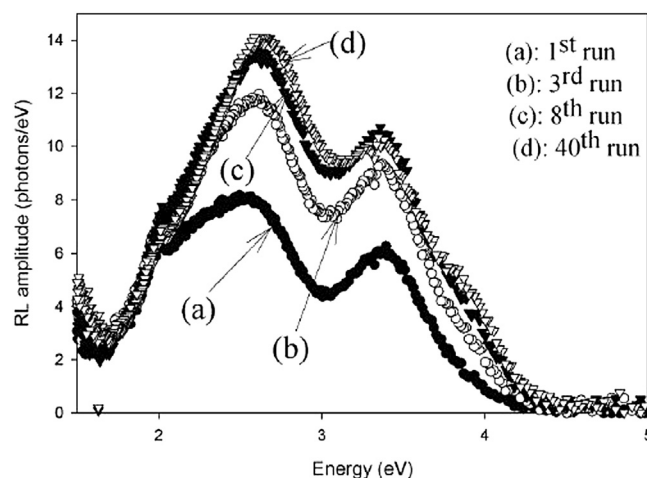


Fig. 2. Radioluminescence spectra of synthetic quartz annealed at 700 °C for 60 min. For each run, a dose of 10 Gy is given to the sample.

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