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Contributions of pre-exposure dose and thermal activation in pre-dose sensitizations of unfired and annealed quartz

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HIGHLIGHTS

- Thermal sensitization was the major mode of sensitization in unfired quartz samples.
- Pre-exposure dose played the chief role of sensitization in annealed samples.
- Pre-dose method of dating is only appropriate for annealed samples.
- Irradiation and TL histories are important in aliquot's sensitivity corrections.

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ABSTRACT

This work was undertaken to determine the separate contributions of pre-exposure dose and subsequent thermal activation in sensitisations of 110 °C Thermoluminescence (TL) peak in attempt to further unravel the complex nature of quartz luminescence properties. Two sets of quartz samples were used. Each of the two sets was divided into two parts; the first was unfired while the second part was annealed at 900 °C for 1 h. The TL measurements were carried out on each aliquot using an automated RISØ TL/OSL reader(model-TL/OSL–DA–15) utilizing different heating rates. Pre-dose sensitization in unfired samples was observed to be predominantly derived from thermal activation while it was pre-exposure dose that majorly contributed to that of annealed samples. This indicated opposite mode of pre-dose sensitizations in the two categories of samples. The findings in the present work revealed that pre-dose technique is not appropriate for unfired sample but rather ideal for fired/annealed quartz samples. All the observed modes of sensitization were explained under the framework of existing models. Effect of sensitization resulting from TL readout and radiation histories in respect of mass normalization in TL/OSL dating procedures was stressed.

1. Introduction

A very important feature of quartz's luminescence property is the enhancement in its sensitivity as a result of combined actions of pre-exposure dose of previously received irradiation and the subsequent thermal activation by thermoluminescence (TL) reading or annealing to an activation temperature. This characteristic, termed as pre-dose effect, is customarily mostly allied with the 110 °C TL peak of quartz and has been well studied and documented in the literature (Zimmerman, 1971; Chen, 1979; Bailiff, 1994; Koul et al., 1996; Bailey, 2001; Li and Yin, 2001; Adamiec et al., 2006; Galli et al., 2006). Apart from the application of 110 °C TL peak pre-dose method of dating in archeological dating and retrospective dosimetry, this technique has been effectively utilized in authenticity testing of artefacts and determination of firing temperature of pottery materials (Bailiff, 1994; Koul et al., 1996; Galli et al., 2006; Li and Yin, 2001).

Pre-dose effect was originally discovered by Fleming and Thompson (1970). A year later, Zimmerman (1971) proposed a

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model to explain the phenomenon. Her model is based on one electron trap responsible for the 110 °C TL peak, two hole-centers termed as non-luminescence reservoir centers (*R*) and luminescence recombination centers (*L*). During irradiation (i.e pre-exposure dose), newly created holes are preferentially captured by the *R* centers which resulted into low sensitivity. When this is followed by a TL reading to a selected temperature *T*, (typically above 200 °C) holes are transferred from *R* to *L*. This charge transfer therefore enhances the luminescence recombination rate due to increased population of holes trap in *L* and decreases competition from *R*. Thus, a subsequent administration of test dose (TD) and TL measurement produce increased sensitivity of 110 °C TL peak.

Apart from the pre-dose sensitization, annealing of quartz at temperatures higher than 200 °C has been observed to result in enhancement of the TL sensitivity of the 110 °C glow peak (Yang and McKeever, 1990; Chen et al., 1994; Rendell et al., 1994; Han et al., 2000; Schilles et al., 2001, Adamiec, 2005; Koul, et al., 2010). This sensitization, unlike pre-dose effect, is independent on pre-exposure dose but rather reliant on annealing temperature and duration. Therefore, this sensitization represents the sensitivity change induced by annealing process only. A viable proof of the existence of pure thermal sensitization was the enhancement in sensitivity after a high temperature annealing of a synthetic quartz that was not expected (Yang and McKeever, 1990;Bøtter-Jensen et al., 1999) in view of pre-dose senitization. This is because synthetic quartz, which is believed to have received insignificant level of irradiations in that past, is not supposed to display enhancement of sensitivity as a result of annealing to high temperature. In addition to their observations on synthetic quartz, and Lersen (1997) established the sole thermal senitization of sedimentary quartz samples also. This was observed in optically stimulated luminescence (OSL) and phototransferred TL (PTTL) of quartz to follow the same pattern of senitization after high temperature annealing.

A model proposed by Bøtter-Jensen et al., (1995) and Lersen (1997) to explain sole thermal sensitization caused by thermal activation is based on removal of competitors at high temperature annealing and alterations to the recombination centers as earlier presumed by McKeever et al. (1983) and Hashimoto et al. (1994). The model includes three trapping states. As Fig. 1 reveals, a shallow electron trap T_s represents 110 °C TL peak and all those shallow trap centers in the real material. The electron trap center T_t represents all the optically active trap centers that are depopulated during OSL measurements and thermal stable traps in quartz (e.g 325 °C TL peak). T_c represents the thermally deep electron traps that are not optically or thermally emptied during TL or OSL measurement. The model has two recombination centers, *L*-center and *K*-center, in which *L*-center is radiative and *K*-center non-radiative. Based on the assumption that annealing at high temperature alters recombination centers, the concentration of *L*-center is increased relatively to that of the *K*-center for annealed case in this model. This is achieved by either increasing *L* concentration or reducing *K*-center concentration. Also, to model the removal of competitors at high temperature annealing, the concentration of T_c was reduced. By applying all these in the model, the following as observed experimentally were simulated (Bøtter-Jensen et al., 1995; Lersen, 1997:

i. OSL and PTTL sensitivity changes due to annealing temperature.
ii. Temperature shift of the photo-transferred 110 °C TL peak.
iii. Dose response of OSL in sedimentary quartz.

The agreement between the simulated and experimental results supports the hypothesis that thermal senitization observed in

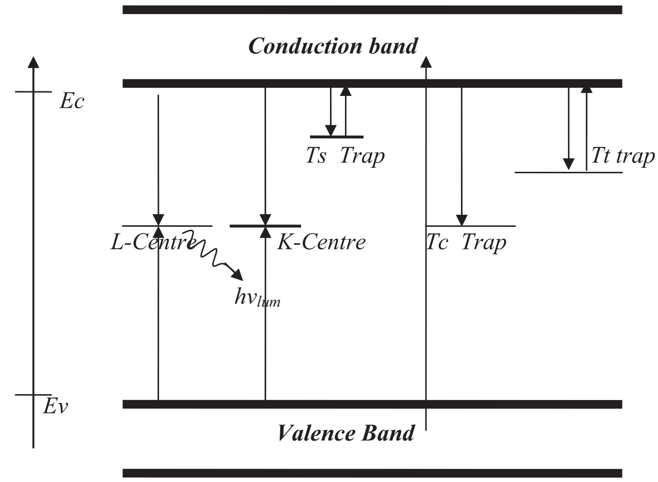

Fig. 1. Thermal sensitization schematic band model after Bøtter-Jensen et al., (1995) and Lersen (1997).

quartz is due to alterations to the concentrations of the recombination centers and trap centers and different from pre-dose effect.

Han et al., (2000) was the first to correlate the annealing duration with the sensitization magnitude. In another direction of thought, Chen and Li, 2000 who reported a similar observation have proposed a modified Zimmerman's model with multiple R centers (each with different life time leading to a more complex function of time and temperature) to explain their findings. tried to identify the possibility of pure thermal sensitization in the pre-dose mechanism of the 110 °C TL peak of quartz by way of using different annealing temperatures and heating rates for thermal activation process. The basis of incorporating different heating rates is on the premise of different time of heating that is associated with each heating rate (Koul et al., 2010). These authors confirmed the sole thermal sensitizations and pre-dose sensitization in their study on three natural quartz samples and one synthetic. More recently, investigated a component resolved study towards the contribution of the thermal activation in case of linear modulated (LM) OSL pre-dose sensitization signals measured at room temperature (RT) and 125 °C among other findings. These authors equally ascribed the reliance of pre-dose sensitization of 110 °C TL peak and RT LM-OSL on heating rates of thermal activation to the pure thermal sensitization.

Despite the recent few findings throughout the literature (Koul et al., 2010; Oniya et al., 2012) coupled with other initial reports on the existence of pure thermal sensitization of quartz, the conditions leading to this phenomenon of quartz sensitization is yet to be established in the literature. Identification of conditions leading to pure thermal sensitization of quartz is important in the pre-dose dating method. This is because there is always the possibility of erroneous age estimation in dating if pure thermal sensitization happens to be the most prevalent mode of sensitization in a sample to be dated using pre-dose dating technique.

Thus, distinction of factors or conditions responsible for both pre-dose and pure thermal sensitizations of quartz is essential in order to establish a standard criterion for selection of suitable materials for pre-dose dating technique and probe more into quartz luminescence features. This investigation is an extension of the report of Oniya et al., (2012) with a view to identifying the modes of luminescence sensitizations in quartz by employing systematic luminescence sequences that were designed to distinguish the two modes of sensitizations in question. This was achieved by going further than employing only different heating rates for thermal activation TL that is associated with intervals of