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In-homogeneity in the pre-dose sensitization of the 110 °C TL peak in various quartz samples: The influence of annealing

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ABSTRACT

The pre-dose sensitization effect of the 110 °C TL glow-peak of quartz is a basic tool in thermoluminescence and optically stimulated luminescence dating and retrospective dosimetry. In the present work, a homogeneity study was performed on pre-dose sensitization in grains obtained from large quartz crystals samples collected from 10 different origins. The aliquot – to – aliquot scatter of the pre-dose sensitization of the 110 °C TL peak within each quartz crystal was monitored. The influence of the annealing on this scattering was also studied. Therefore, the investigation was applied to the un-fired "as is" samples as well as to samples annealed at 900 °C for 1 h following cooling to room temperature in air. The results showed that in the case of "as is" quartz the sensitization effect vary strongly within each aliquot of the same quartz sample. This strong variation is removed by both the high temperature annealing as well as heating up to 500 °C, involved in the TL measurements. These results are generally discussed in the framework of existing models and applications of the effect.

1. Introduction

SiO_2 (silica) makes up 12.6% by weight of the Earth's crust as crystalline quartz and amorphous silica [1]. Quartz is the main material for retrospective dosimetry and for dating archeological pottery and geological sediments using both thermoluminescence (TL) as well as optically stimulated luminescence (OSL). Among the various TL glow peaks of quartz, the one which was found at just about 100 °C for a heating rate of 5 °C/s is known as the 110 °C TL peak [2]. This specific TL peak in quartz is known to decay at room temperature (RT) with a half-life of the order of an hour [3]. Therefore this peak can be seen in all quartz, whether natural or artificial [4] provided that it has been irradiated less than a few hours before measurement. Nevertheless, though unstable at RT, the 110 °C TL glow peak of quartz, has worked very efficiently due to its pre-dose sensitization property [5]. The pre-dose thermoluminescence technique of dating [6] is unique in its ability to measure radiation doses of as small as 10 mGy in contemporary ceramic materials such as bricks, tiles and porcelain plumbing fixtures [7]. Therefore, it is a well-established technique in both retrospective dosimetry and dating [8]. The main idea behind the use of both this technique and peak is to monitor the increase in the sensitivity rather than the, conventional, accumulation of TL [9], since the response of the 110 °C TL peak to a small test dose can be enhanced by heating to temperatures above 200 °C [10]. Utilizing this sensitization, the 110 °C TL glow peak of quartz has been attempted for the firing temperature assessment of ceramics also [11–13].

A workable TL peak at a temperature as low as 110 °C is a big advantage in TL measurements for a number of reasons, namely: (a) studying the TL resulting from charge release from this trap is not difficult and no instrumental limitation is implied, as the unwanted effects occurring due to heating at much higher temperatures can be avoided, (b) this TL peak is ubiquitously present in all quartz samples, natural or artificial, annealed and as-is, (c) several features of the 110 °C TL glow-peak were proved to be universal or prevalent, such as its main structure (shape, peak position and trapping parameters [14]) as well as its thermal quenching behavior [15], (d) due to its simplicity and non-composite nature, the 110 °C TL peak does not require de-convolution, (e) lastly, the 110 °C TL peak is sensitive and consequently, its study therefore does not require large doses of irradiation in the laboratory.

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Given all the aforementioned reasons, the pre-dose methods are basically centered on the 110 °C TL peak, thus, the pre-dose sensitization of the latter glow-peak has also become the subject of numerous studies in respect to:

(i) Its response to combined annealing and irradiation [4,5,16,17].
(ii) Its response to either irradiation [18] or thermal treatment [9,19].
(iii) The recombination center as well as the emission wavelength which are responsible for pre-dose effect, namely the emission at 370 nm which occurs due to $[H_3O_4]^0$ hole centers, silicon vacancies that are occupied by three hydrogen atoms and a trapped hole [20–22].
(iv) The sensitization and superlinearity in quartz, which are phenomena intimately related to each other [23–26].
(v) A comparative study of the pre-dose effect for different types, annealing temperatures and origins of quartz [26,27].
(vi) Explanation and modeling of the effect [28–32].
(vii) Its usefulness to monitor the sensitization of the OSL signal in various dating protocols [2,3,33–42].

One among the uncertainties encountered on the extensively studied phenomenon of sensitization of 110 °C TL peak is the degree of sensitization that changes from sample to sample. The complexity of this peak, just like other quartz luminescence properties, is always attributed to the different crystallization environments during formation of respective sedimentary quartz samples that varies from location to location [43,44]. Nevertheless, even within the same quartz sample, 'grain to grain' variation has been also reported for the case of some sedimentary quartz samples [44–46], leading to the devising of a 'Single grain OSL attachment'. Grain to grain variation has been reported to happen as a result of different grain to grain origin, and various irradiations, thermal and optical/bleaching histories that each grain might have possessed during transportation. Each one of these factors is widely known to be highly influential on pre-dose sensitization [4,5,9,16–19]. Ideally, the same degree of pre-dose sensitization is expected from all the grains of any crystalline quartz. This assumption results from the fact that all the grains are (i) from a common origin, (ii) and possess the same irradiations, thermal and optical/bleaching histories. However averaging effect over many grains is generated by using an aliquot with a layer of grains of quartz. Still disc to disc variations are expected due to statistical reasons but in that case these variations are expected to be very low. Therefore, large variation in sensitization that is exhibited by different grains of a crystalline quartz sample should arise from the intrinsic nature of the crystal and not from prevailing external factors. Should such characteristic exist, it is expected to shed more light on the complexity nature of quartz luminescence characteristics; and subsequently be of high assistance in all luminescence area of applications. Based on the above, the aim of this work was to study the homogeneity of the pre-dose sensitization of the 110 °C TL peak within the same quartz crystal for several quartz samples of various origins by assessing the aliquot – to – aliquot scattering. The influence of the annealing to this scattering was also studied. To the best of the author's knowledge, no similar study is reported in literature, despite the voluminous literature dealing with the 110 °C TL peak in quartz.

2. Experimental procedure

The original quartz samples were large crystals of hydrothermal and metamorphic origins which occurred in vein-associated metamorphic rocks. In order to make the study to be universal, 10 different quartz samples were collected from different origins spanning through Africa, Europe and Asia. The laboratory code names, locations and types of each of the 10 quartz samples are presented in Table 1. Each natural crystal quartz sample was crushed and smashed in an agate mortar. Grains of dimensions between 90 and 150 μm were obtained by sieving, cleaned in acetone and dried in the air. Two types of measurements were performed on each of the ten samples. The first sets were performed to 'as is' (unfired) material while for the second sets the material was previously annealed at 900 °C for 1 h and allowed to cool to room temperature in the air afterwards. For each set, 10 aliquots of equal mass of about 5 mg were analysed in this work with aim of observing variation in the sensitization among ten aliquots of the same sample.

All the TL measurements on the quartz samples were carried out using a RISØ TL/OSL reader (model TL/OSL–DA–15) equipped with a 0.075 Gy/s $^{90}Sr/^{90}Y$ β ray source [46]. The reader was fitted with a 9635QA photomultiplier tube. The detection optics consisted of a 2.5 mm Hoya U-340 ($\lambda_p \sim 340$ nm, FWHM 80 nm) filter. All measurements were performed in a nitrogen atmosphere with a constant heating rate of 1 °C/s in order to avoid significant temperature lag, up to a maximum heat temperature of 500 °C. The experimental protocol used was the following:

Step 1: Give a TD to a natural sample having its Natural TL as NTL.
Step 2: Measure TL up to 180 °C. This step measures the sensitivity S_{01} of the TL glow-peak at "110 °C" acting additionally as a mass normalization.
Step 3: Give the same TD and measure TL up to 500 °C. This step (a) measures the sensitivity S_{02} of the TL glow-peak at 110 °C and (b) acts as a thermal activation the sample.
Step 4: Give the same TD and measure TL up to 500 °C. This step (a) measures the sensitivity S_1 of the TL glow-peak at 110 °C and (b) acts as a thermal activation the sample again.

Table 1
Laboratory code name, origin and test dose administered for both 'as is' and annealed samples.

Sample's laboratory name	Sample origin	TD administered, 'as is' (Gy)	TD administered, annealed (Gy)
A1	Nepal, Asia	5	2
B2	Nepal, Asia	2	0.5
Kilkis	Greece, Europe	5	2
Koupa	Greece, Europe	5	2
S1	Nigeria, Africa	5	0.5
S2	Nigeria, Africa	2	1
S3	Nigeria, Africa	5	0.5
S4	Nigeria, Africa	2	0.5
S6	Nigeria, Africa	5	0.5
S8	Nigeria, Africa	5	0.5