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BEHAVIOR OF VARIOUS NIGERIAN QUARTZ SAMPLES TO REPEATED IRRADIATION AND HEATING

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Abstract: In the present work the sensitization of the entire glow-curve is studied in 6 different quartz samples of Nigerian origin. The investigation was applied to the un-fired "as is" samples as well as to samples fired at 900°C for 1 hour following cooling to room temperature. The results showed that in the case of "as is" glow-curve is sensitized as a whole. There is an abrupt transition from the "natural" sensitivity without any previous heating and the artificial sensitivity induced after the first heating. The sensitization is growing up strongly to the 10th heating but to a lower rate. The sensitization factor of the TL glow-peak at "110°C" was found to be linearly correlated to the higher temperature TL peaks. In the case of annealed samples there is an initial increase between the sensitivity immediately after the end of annealing and after the first heating. As the number of heating is increased up to the 10th heating the sensitization is stabilized at a constant value. The results are discussed in the framework of existing models and implications of the sensitization effect in various applications, while some explanations are attempted.

Keywords: Quartz, Thermoluminescence, 110°C TL glow peak, High Temperature TL peaks (HTTLPS), Pre-dose effect, Sensitization, luminescence centre, defects.

1. INTRODUCTION

Quartz thermoluminescence (TL) glow curve consists of a number of glow-peaks when heated from room temperature to 500°C at a constant heating rate subsequent to irradiation either in the laboratory or by dose acquired naturally (Wintle, 1997). Each one of these TL peaks corresponds to a specific trap. Franklin *et al.* (1995), have shown the presence of at least four of the quartz's TL peaks, namely at 95-110, 150-180, 200-220 as well as 305-325°C, depending on the heating rate. Similar results were also reported by other authors, such as Ogundare *et al.* (2006), Koul (2008) and Preusser *et al.* (2009). Additional glow peaks are also available in the literature for different quartz samples as well as different detection optics, such as at 250-270°C as well as the one at 350-375°C (Wintle, 1997; Thomsen, 2004). Recently, Subedi *et al.* (2011) presented the major TL peaks on various quartz types after deconvolution. A summary of all the TL peaks reported in the literature is tabulated by Preusser *et al.* (2009).

As high temperature TL peaks (denoted as HTTLPS hereafter) we consider those TL peaks that are yielded in the temperature region between 150°C and 500°C of the

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glow curve. While the 110°C glow-peak is employed in the pre-dose technique of dating (Bailiff, 1994; Koul *et al.*, 2010), the HTTLPS stand as the cornerstone of additive–dose dating procedure. Moreover, the fast component of Optically Stimulated Luminescence (OSL) has been confirmed to originate from a trap that is responsible for the TL peak around 325°C (Spooner 1994; Li and Chen, 2001; Jain *et al.*, 2003; Kitis *et al.*, 2010). Therefore, the HTTLPS are also somehow related to the single-aliquot regenerative-dose (SAR) method.

A very noticeable feature of quartz that has attracted the interest of many workers is its enhancement of luminescence sensitization as a result of previous irradiation and thermal activation, in both cases of TL and OSL (Bøtter-Jensen *et al.*, 1995; Bailiff, 1994; Chen and Li, 2000; Han *et al.*, 2000; Koul *et al.*, 2009; Kiyak *et al.*, 2007; Koul, *et al.*, 2010). Since SAR, pre-dose methods and many luminescence protocols involve a combination of heating (mainly in the form of pre-heating) and irradiation procedures, it is necessary to have adequate understanding of quartz sensitization so as to monitor sensitivity changes owing to the treatments the quartz samples receive in the course of measurement procedures. Such findings are desirable in order to avoid erroneous results in luminescence dating using quartz. There exists a significant body of literature with reports of sensitization study of the 110°C TL peak (Bailiff, 1994; Bøtter-Jensen *et al.*, 1995; Charitidis *et al.*, 1999; 2000; Polymeris *et al.*, 2006; Koul *et al.*, 2010). Furthermore, sensitivity changes of quartz OSL signals as a result of repeated preheating and irradiation have also been reported (Stokes, 1994; Wintle and Murray, 2000; Jain *et al.*, 2003; Wintle and Murray, 2006; Kiyak *et al.*, 2007). Nevertheless, to the best of the author's knowledge, sensitization investigations of HTTLPS are scanty in the literature (Liritzis, 1982; Han *et al.,* 2000; Afouxenidis *et al.*, 2007). Therefore, the aim of the present report is to study the sensitization that occurs in the HTTLPS signal of quartz after repeating measurements performed on the same aliquot.

The reasons why HTTLPS have not been studied as much as the 110°C peak are (i) its complex composite nature, unlike the simplicity and non-composite nature of 110°C TL peak that does not require deconvolution (ii) the pre-dose methods incorporated the sensitization of 110°C, in particular and (iii) HTTLPS are not as sensitive as 110°C and, therefore, require large doses of irradiation in the laboratory for their adequate sensitization, resulting in time consuming irradiation in order to have good statistics.

A linear relationship between sensitivity changes in the 110°C TL peak of quartz and in its OSL emission was first reported by Stoneham and Stokes (1991), providing evidence for a strong link between the two luminescence signals. This is the reason why the applicability of the 110°C TL peak was suggested as a sensitivity monitor by Stokes (1994) in the case of single-aliquot additive dose protocol, as well as by Murray and Roberts (1998), while developing a single-aliquot regenerative-dose protocol. Therefore, they proposed the use of the former in order to monitor and correct for sensitivity changes while using the same sample for routine OSL measurements (Duller, 1991).

Due to luminescence characteristics of quartz that are generally complex and differ from sample to sample to some extent, the simultaneous investigation of sensitization of all TL glow-peaks of quartz is considered essential. One main difficulty that could be encountered in such concurrent study, as reported by Ogundare *et al.* (2006), is the overflowing of the PMT when detecting the 110°C TL peak at high doses that HTTLPS require to be well populated. A feasible TL protocol to overcome this hurdle has been employed in this work to systematically study the sensitization of all TL glow-peaks of quartz and to investigate the possible correlation between the sensitization of the 110°C TL glow-peak and the sensitization of the higher temperature TL peaks.

2. EXPERIMENTAL PROCEDURE

Materials and Apparatus

Six different quartz samples, collected from different locations all around Nigeria were analyzed in this work. They were given laboratory names S1, S2, S3, S4, S6 and S8. The first three of these samples were from central-western part of Nigeria, whereas S4, S6 and S8 were from western part of the country. The modus operandi behind sample selections in this study was to have quartz grains that possessed the same radiation, optical and thermal histories for each sample used, in order to rule out the possibility of grain to grain different sensitizations. Thus, instead of using sedimentary quartz samples, the original quartz samples were large crystals of hydrothermal and metamorphic origins which occurred in veins associated with metamorphic rocks. S1, S2, S3, S4 (clear rock crystal type) and S6 (rose-pink type) are hydrothermal while S8 (milky quartz species), in its own case, is of metamorphic origin. Grains of dimensions between 90 and 150 μm were obtained from each of the natural crystal quartz samples after, smashing in an agate mortar and sieving. Two types of measurements were performed on each of the 6 samples. The first sets are 'as is' (unfired) while the second sets have been annealed at 900°C for 1 hour and allowed to cool immediately to room temperature in the air afterwards in the laboratory. Each of the aliquots (sub-samples) was of equal mass of about 5 mg. 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 (Bøtter-Jensen *et al.*, 2000). The reader was fitted with a 9635QA photomultiplier tube. The detection optics consisted of a 7.5 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 dose administered to each of the 6 quartz samples (both 'as is' and annealed) is accordingly indicated in the experimental protocol presented bellow (case of 'as is' samples):

Protocol

Step 1: Give a dose (75 Gy) on a sample having its Natural TL (NTL).

Step 2: Pre-heat to 50°C at 1°C/s for 30 s meant to depopulate 110°C TL peak in order to avoid overflowing of PMT because of the high dose applied

Step 3: Measure TL up to 500°C

Step 4: Repeat Steps 1 to 3 for nine sequential times (i.e. perform 10 sequential TL measurements) on the same aliquot.

The same protocol was also applied in the case of annealed samples; however a dose of 37.5 Gy was attributed instead of 75 Gy during step 1. The criterion for the selection of the dose was to achieve a good statistics for the high temperature TL glow-peaks, especially, in case of the first measurement. Sensitization was studied in terms of the maximum peak intensity for each TL peak. Therefore, deconvolution was not applied.

3. EXPERIMENTAL RESULTS

"As is" quartz

The results for all "as is" quartz samples studied are shown in **Fig. 1**. Plots A, C, E, G, I and K present the 10 successive glow curves for each quartz sample under study. In all cases, despite the depopulation of 110°C TL peak by pre-heating to 50°C, the TL peak still remained quite strong in case of all the samples. Furthermore, the sensitivity of the partially depleted 110°C TL glow-peak is much higher than the sensitivities of the HTTLPS. Therefore, in order to present the entire glow-curves in one figure, the Y-axis of all left hand-sided panels is presented in a way so that the details for the higher temperature TL peaks become very clear.

Besides the 110°C which is common for all quartz samples (Pagonis *et al.*, 2002) (in fact the mean value of the peak maximum temperature of this glow-peak for the quartz samples studied and for a heating rate of 1°C/s used in the present work appears at about 83±6°C), it is clearly seen that the glow-curve shapes of all samples are different. This observed variability should be attributed to the different crystal origin and formation, as well as to different conditions for processes that are taking place subsequent to the crystal formation, such as heating, bleaching and irradiation. Nevertheless, sensitization occurs along the entire glow-curve up to the temperature of 400°C in the case of 5 quartz samples and only in the case of sample S8 the HTTLPS are not sensitized. It should be recalled here that S8 is the only sample that is metamorphic while the remaining 5 samples are of hydrothermal origin.

Plots B, D, F, H, J and L in **Fig. 1** present the normalized TL intensity for HTTLPS with T_{max} (temperature at peak maximum intensity) located in the temperature region between 200°C and 320°C. Normalization of all peaks was carried with the corresponding peaks only. In all these plots of **Fig. 1** the first point of all the curves (a-d) corresponds to the sensitivity of each sample, which was not previously heated. So, this sensitivity represents the sensitivity of the samples without any previous heating (thermal activation), which was affected only by irradiation. The corresponding "thermally in-activated" glow curve of each sample is labelled as curve (1) in plots A, C, E, G, I and K.

However, in order to obtain the first thermally activated (artificial sensitivity i.e. curve (2) in plots A, C, E, G, I and K of **Fig. 1**) sensitivity the un-heated sample is irradiated and subsequently heated. Therefore, the new sensitivity is artificially affected by both heating and irradiation. Similarly, all sensitivities up to the 10^{th} heating are similarly affected. In the present work, all artificial sensitivities are normalized over the "thermally in-activated" sensitivity; this ratio of pre-dosed sensitivities over the "thermally in-activated" sensitivity (i.e. normalization) will be called sensitization factor hereafter.

As glow curves (1) and (2) of all plots in **Fig. 1** will further reveal, there is an abrupt change on the sensitivity of all samples between the first and the second heating. This abrupt transition from the "thermally in-activated" sensitivity to the first pre-dosed one is clearly shown in all plots of **Fig. 1**. This transition is more intense in the cases of the TL peak at 110°C. The sensitization factor of this glow-peak, represented by curve (a), is sample-dependent, varying between 10 and 800 from sample to sample. Nevertheless, sensitization is also observed in the case of HTTLPS, generally, possessing two peaks in the temperature region between 200°C and 320°C, which are represented by curves (b) and (d) correspondingly. Sample S2 presents a third peak also which is represented by cases (c) in the figure. The sensitization factor of these peaks is much lower compared to that of the TL peak at 110°C, varying between 2 to 10. However, in that case, the maximum value of sensitization factor seems to be of prevalent nature for all the quartz samples that were subjected to study.

The abrupt transition of the sensitivity is a clear pre-dose sensitization effect. The pre-dose in "as is" quartz consists of the natural dose plus the test dose of 75 Gy. The subsequent readout heating to obtain the "thermally in-activated" sensitivity activates the pre-dose effect causing the strong increase of the sensitivity. This is common result for the TL peak at 110°C. However, it is seen that the pre-dose sensitization effect concerns the entire TL glow-curve in "as is" quartz. In both cases of the 110°C TL peak as well as HTTLPS, the sensitization

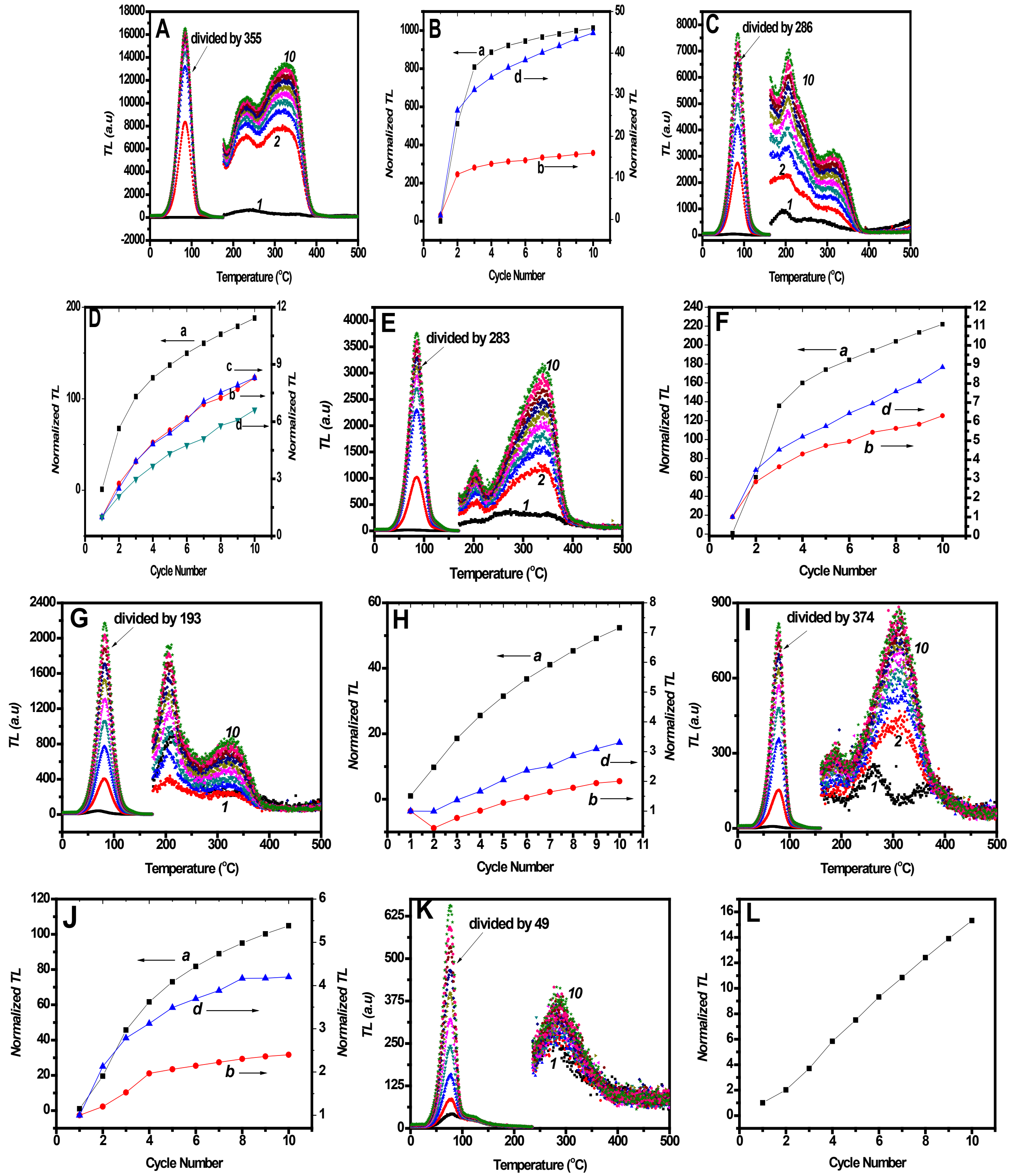

Fig. 1. *Results of the "as is" quartz samples. Plots A, C, E, G, I and K present TL glow-curves as a function of the thermal activation cycle number for all quartz samples under study. In all cases curve 1 corresponds to the TL sensitivity when the test dose is given to the sample having its natural dose, i.e. it represents the "thermally in-activated" sensitivity of all glow-peaks studied here. Curve 10 corresponds to the 10th thermal activation cycle. The plots B, D, F, H, J, and L present the normalized TL, of the glow-curves on the left hand side, normalized by the corresponding peak heights of the 1st run. In all cases curve (a) corresponds to the TL glow-peak at "110°C" whereas curves (b), (c) and (d) to glow-peaks centred around 200, 250 and 320°C correspondingly.*

factor of the second data point depends on the sum of 75 Gy plus the naturally accumulated dose. Consequently, this sensitization factor varies according to the dose received in case of each sample. In the subsequent irradiation-heating cycle the pre-dose is equal to the test dose received in the previous iteration. Therefore, in the ten runs carried in this study the pre-dose is decided by the accumulated dose experienced by the sample till the current read out.

Annealed quartz

The results for all annealed quartz samples are shown in **Fig. 2**. The reading of the figure is exactly similar to the **Fig. 1** for "as is" quartzes. In the case of annealed quartz, instead of the "natural" sensitivity of the "as is" quartz, we have the sensitivity of the samples immediately after the end of the high temperature annealing. The high temperature annealing removes all traces of the natural dose. Therefore, this sensitivity corresponds to a zero pre-dose. Once the sample is heated to obtain the zero pre-dose sensitivity and then subsequently irradiated and heated again, the new sensitivity is due to a thermal activation and a pre-dose equal to the test dose. The same holds true in case of all subsequent irradiation-heating cycles.

The transition from the zero pre-dose sensitivity to the respective pre-dosed is steep, but not as abrupt as in the case of "as is" quartz, while the sensitization factor is much lower. Namely, for the case of the TL glow-peak at 110°C it varies between 2 to 10. Except for the sample S6, in all other cases the system of the TL glow-peaks centred at about 200°C is the one that is sensitized the most. A high sensitization factor is also observed for the system of TL glow-peak centred at about 200-250°C. In fact, this TL peak appears to be the most sensitized one. Nevertheless, the sensitization factor of the system of TL glow-peaks centred at about 320°C is the lowest observed, once again except in the case of S6 quartz.

A general characteristic for the behaviour of the sensitization factor is that it attains a stable value. The number of repeated cycles of irradiations-TL measurements required for that stability to be reached depends strongly on both sample and TL peak. In most cases the TL glow-peak centred at about 320°C is just sensitized only after the first cycle of measurements, while the stability is starting even from the second cycle, which is the first pre-dose cycle respectively. Generally, following 4 or 5 cycles of irradiation-TL measurement, the sensitivity of the entire glow curve of all annealed quartz samples is stabilized.

Correlation

Franklin *et al*. (1995) have concluded that, for their Australian sedimentary quartz sample, four of the quartz's TL peaks, namely at 95-110, 150-180, 200-220 as well as 305-325°C (depending on the heating rate) use the same luminescence centre, accessing it via the conduction band. Other luminescence features such as bleaching rates, thermal quenching and the inter-relation between these peaks through optical stimulation, the so-called photo transferred TL (PTTL) (Kaylor *et al*., 1995) support the connection between them throughout the use of the same luminescence centre (Bailey, 2001; Itoh, 2002; Guzzo *et al*., 2006; Khoury *et al*., 2007). In the present study, another luminescence feature, namely the correlation of the TL output between the HTTLPS and the corresponding of the 110°C TL peak in the case of annealed quartz samples, further supports this aforementioned connection.

The comparison of glow curves between the 'as is' and annealed samples permits easy observation of modifications made to the 'as is' samples as a result of high temperature annealing. These changes in the nature of the glow-curves after annealing are attributed to the alterations made to the recombination pathways and competitions during irradiation and heating. These are thought to result from the introduction of more traps and recombination centres as Bøtter-Jensen *et al*. (1995) and Lima *et al*. (2002) suggested.

Furthermore, quantitatively, this correlation is further established from the data presented in **Fig. 3**. For the case of quartz samples S2 and S4, the intensity of the HTTLPS is plotted versus the intensity of 110°C TL peak. As the figure is going to further reveal, the TL intensity of the HTTLPS is linearly increasing with the intensity of the 110°C TL peak. This linear behaviour between these signals stands at least up to first four data points for all quartz samples that were subjected to the present study. Especially in the case of the 325°C TL peak (curves d), the linearity that is beyond the first four data points further supports the interconnection of these two peaks through PTTL as was suggested by Kaylor *et al.,* (1995). These experimental features provide another indication towards the use of the same radiative recombination centre for both emissions. These are $[H_3O_4]^\circ$ hole centres, silicon vacancies that are occupied by three hydrogen atoms and a trapped hole (Wintle and Murray, 1999; Preusser *et al*., 2009).

4. DISCUSSION

In the case of the 110°C TL peak, the pre-dose sensitization is explained by models involving a thermal transfer of holes from reservoirs to luminescence centres (Zimmerman, 1971; Bailiff, 1994). A reservoir could be any hole trap which is thermally excited and releases holes in valence band when the temperature exceeds 200°C. Based on the results of the present study, it is reasonable to assume that the same model and mechanism could be also responsible for the sensitization of the HTTLPS. Especially in the case of the "as is" quartz samples, which were not previously heated, the first TL measurement up to 500°C thermally activates the mecha-

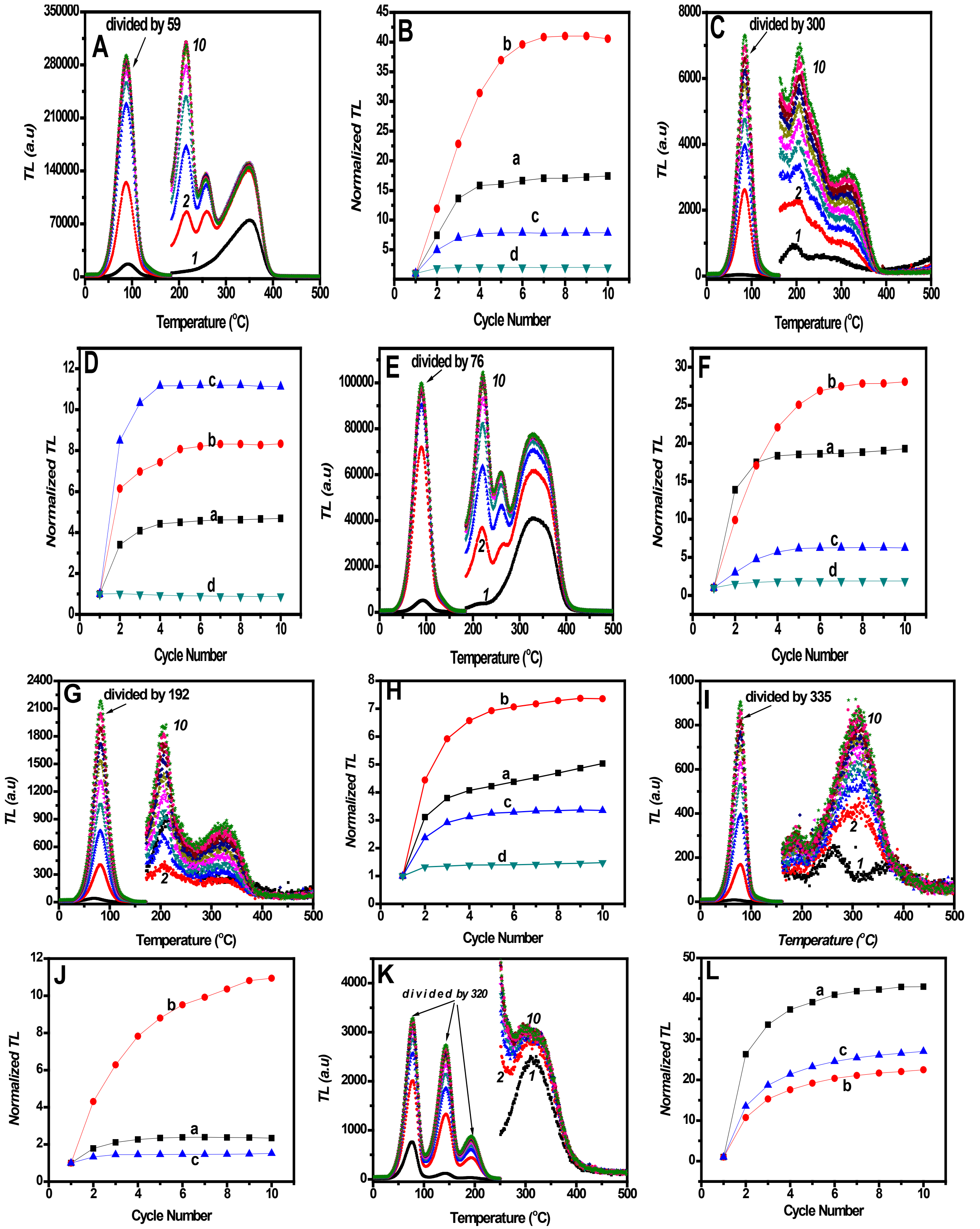

Fig. 2. *Results of the annealed quartz. Plots A, C, E, G, I and K present TL glow-curves as a function of the thermal activation cycle number. In all cases curve 1 corresponds to the TL sensitivity measured immediately after the end of the annealing. Curve 10 corresponds to the 10th thermal activation cycle. The plots B, D, F, H, J, and L present the normalized TL, from the glow-curves of left hand side, normalized by the respective peak heights recorded immediately after annealing. In all cases curve (a) corresponds to the TL glow-peak at "110°C" whereas curves (b), (c) and (d) to glow-peaks centered around 200, 250 and 320°C correspondingly.*

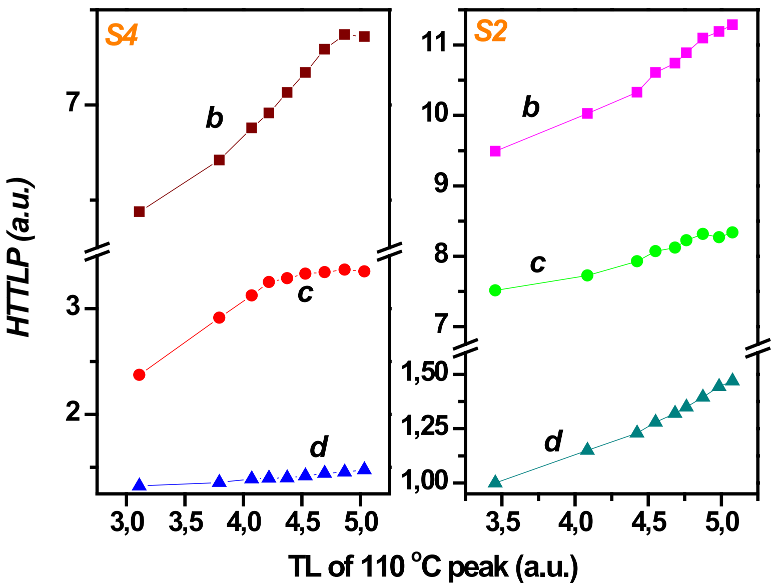

Fig. 3. *Plot of the HTTLPS intensity versus the corresponding intensity of the TL peak at "110°C" in case of S2 and S4 annealed samples. In all cases curves (b), (c) and (d) correspond to TL glow-peaks centred around 200, 250 and 320°C respectively.*

nism resulting in a great enhancement of TL sensitivity as it was already observed experimentally.

Nevertheless, the TL emission of quartz is also affected by phase, chemical and rheological changes occurred after crystal formation (Sawakuchi *et al*., 2010). The 340 nm TL band emission of many silicate minerals, including quartz, is strongly correlated to silicon-oxygen bonding defects produced when the crystal lattice is stressed (Yang and McKeever, 1990; Krbetschek *et al*., 1997; Garcia-Guinea *et al*., 2007). In consequence, heating up to the deformation of quartz crystal lattice, would have an effect on the TL emission of quartz.

In order to explain the experimental fact that the TL glow-peak at about 200-250°C appears to be the most sensitized one, one has to refer to Correcher *et al*. (2009). These authors have studied the TL emission at approximately 220°C from cristobalite, which is a SiO_2 polymorph. They have concluded that this emission is linked to aluminium-related defects which are altered due to the phase transition from alpha to beta structure. This phase transition provokes changes in the TL emission through the movement of alkali ions away from aluminium sites, enhancing thus both sensitivity and sensitization. Nevertheless, Correcher *et al*. (2009) put forward that the alpha to beta phase transition affects not only the 220°C but the entire TL emission of quartz in the same way, despite the fact that the α to β-quartz transition occurs at 573°C, a temperature well beyond the maximum stimulation temperature generally used in TL measurements, being 500°C.

The case of sample S8 is extremely interesting. Prior annealing, the HTTLPS signal of this specific quartz is not sensitized. Furthermore, its intensity is relatively low according to **Fig. 1**. However, the annealing resulted in both increasing the HTTLPS signal intensity as well as inducing sensitization with successive cycles of irradiations – TL measurements. In the case of as is sample, the signal corresponding to HTTLPS results from recombination at the $(AlO_4)^0$ luminescence centre of quartz, emitting at 470 nm, in which an aluminium atom replaces a silicon one (Martini *et al.*, 1995). Quartz TL peaks that are associated with the latter have been found to remain almost unaffected by the pre-dose effect (Yang and McKeever, 1990; Koul and Chougaonkar, 2007). However, annealing changes the emission spectra since interstitial ions merge in interstitial sites of the crystal. These ions are mostly monovalent alkali ions as well as H^+ which are linked mainly to Al ions. These ions are important for charge compensation of substitutional trivalent impurities. Therefore, subsequent annealing the HTTLPS signal results from recombination at the (AlO_4/M^+) luminescence center (M^+=Na^+, H^+). According to Krbetschek *et al*. (1997) and references therein, the emission wavelength of this specific luminescence centre has a peak between 330 and 340 nm. Therefore, the sensitization that is monitored after annealing could be attributed to the change of the emission spectra towards the blue spectral region.

The results in the case of annealed quartz samples may have implications to dating of ceramics as well as to the retrospective dosimetry using modern bricks. In most cases the signal resulting from HTTLPS is sensitized only after the first two or three heatings. Especially, the signal resulting from the peak of 305-325°C is sensitized after the first TL readout only. Furthermore, this sensitization is not so abrupt but very much milder. Consequently, in the case of heated quartz samples, the application of a single aliquot equivalent dose estimation protocol may be feasible, provided that the sensitivity changes might be monitored as well as corrected. Therefore, the present study strongly suggests the application of 10 successive cycles of irradiations and TL readouts prior equivalent dose estimation to any heated quartz sample to be dated. A linear relationship between TL output of peaks 305-325°C and 110°C would provide a way of both monitoring and correcting sensitivity changes; therefore a single aliquot TL regenerative protocol could be effectively applied. In the case of unheated quartz samples, due to the lack of previous heating, the entire glow curve is sensitized following successive irradiations and TL readouts. Furthermore, the sensitivity did not stabilize following a small number of irradiation-TL readout cycles. Moreover, the sensitivity change after the first TL readout is much steeper in the case of the 110°C TL than the signal resulting from HTTLPS.

5. SUMMARY

The conclusions of the present study can be summarized as follows:

1) All quartz samples, regardless of annealed or unannealed, exhibit different glow-curve shapes.
2) All HTTLPS monitored up to the temperature of 400°C are sensitized (except sample S8). In the case of annealed samples, the pre-dosed sensitivities of all TL glow-peaks reach some stable value.
3) In the case of "as is" quartz samples there is an abrupt transition from the "thermally in-activated" sensitivity to the first artificial one, which is attributed to pure pre-dose effect. Correspondingly, in the case of annealed samples, there is an abrupt transition from the zero pre-dose sensitivity to the first pre-dosed one, which however, is much lower than that of the "as is" quartz.
4) The pre-dose sensitization effect appears to present in all TL glow-peaks of both "as is" and annealed quartz samples.
5) There is a very good correlation between the sensitization factors of the HTTLPS with the sensitization factor of the TL glow-peak at 110°C.
6) Although the pre-dose sensitization does not modify the glow curve structure of any of the annealed samples, annealing above 500°C introduced some alteration to the glow curve structure of the annealed samples as compared to the "as is".

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