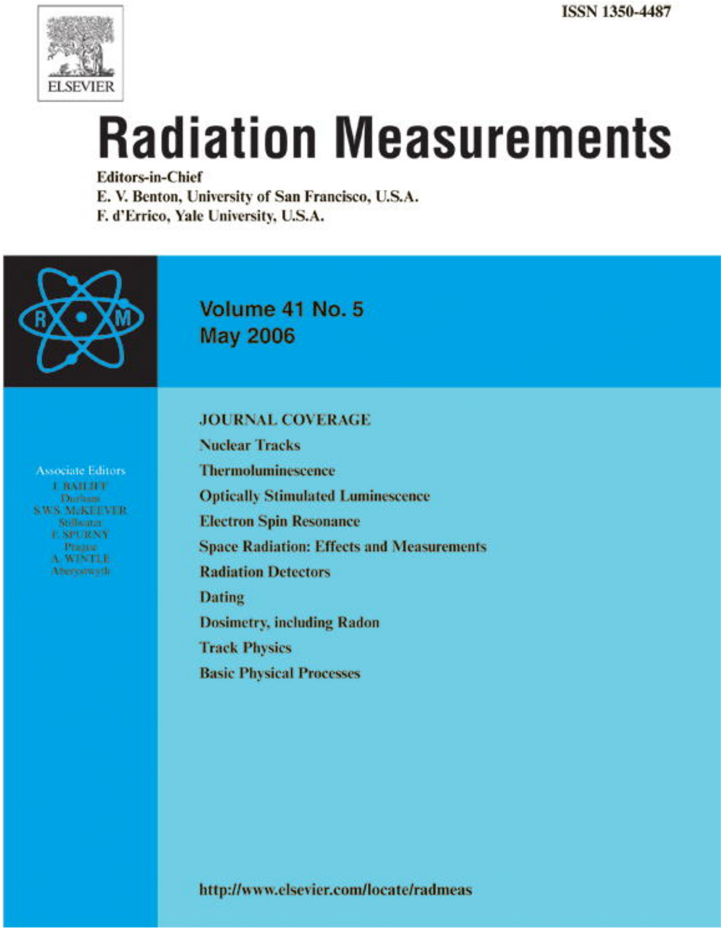

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The effects of annealing and irradiation on the sensitivity and superlinearity properties of the 110 °C thermoluminescence peak of quartz

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Abstract

Quartz samples which undergo heating and irradiation treatments exhibit changes in their sensitivity to irradiation, as well as in their TL dose response. These changes of thermoluminescence (TL) sensitivity and superlinearity of the 110 °C TL peak of quartz have been the subject of several experimental and theoretical studies, because they form the basis of the predose technique for dating ceramics and for accident dosimetry. In an effort to separate experimentally the effects of irradiation and annealing on the predose effect, three quartz samples of different origin were prepared under three different conditions: unannealed samples, samples annealed at 500 °C, and samples annealed at 900 °C. Complete TL versus dose and sensitivity S versus predose curves were obtained for the dose range of $0.1 < D < 400$ Gy. Additional complete sets of data were obtained for samples that underwent a combined predose irradiation and a heat treatment to 500 °C. Although the TL versus dose curves and the sensitivity versus predose data showed very different behaviors, preannealing the samples at 900 °C removed the observed differences that are due to the thermal or irradiation history of the quartz samples. The experimental data is consistent with the assumption that high-temperature anneals and/or high dose irradiation of the samples reduces the concentration of available competitor sites. The concentration of these competitor sites, as described by the Zimmerman model of quartz, is identified as the most important factor in causing the observed differences in predose behavior between quartz samples of different origin. Strong evidence in support of this competitor theory is provided by the estimated equivalent doses (EDs) for the three quartz samples.

Keywords: Thermoluminescence; Quartz; Annealed quartz; Predose technique; Sensitivity changes; Dose response

1. Introduction

The “110 °C” glow-peak of quartz is a glow-peak of unique interest in thermoluminescence (TL). The interest in this peak comes from the fact that quartz is the main material for dating archeological pottery and for retrospective dosimetry. This glow-peak has also become the subject of numerous studies and is of primary importance for the study of the basic mechanisms of TL. Some of the basic characteristics of quartz that make it invaluable for the understanding of TL mechanisms are:

1. This glow-peak appears in all types of synthetic and natural quartz.
2. Its main structure (shape, peak position and trapping parameters) varies within narrow limits even after the application of extreme conditions of temperature and irradiation (Pagonis et al., 2002)
3. This TL peak exhibits sensitization due to both irradiation and heat treatment, which is usually termed the predose effect (Bailiff, 1994).
4. Synthetic samples exhibit superlinearity properties in this peak, while such phenomena are mostly absent in geological samples (Bailey, 2001).
5. The peak undergoes large variations of its sensitivity as a function of the firing temperature and as a function of irradiation temperature (Charitidis et al., 1999, 2000).
6. Occasionally dose rate effects are present (Chen and McKeever, 1997, p. 182).

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Despite the large number of quartz studies found in the literature, there have been very few broad comparative studies of both the sensitization and superlinearity properties of quartz of different origins, under different predose irradiations and/or heating of the samples to various temperatures (Yang and McKeever, 1988; Bailiff, 1994, and references therein).

It is well known that the phenomena of sensitization and superlinearity in quartz are intimately related to each other (Chen and McKeever, 1997, p. 196). Charitidis et al. (1999, 2000) performed an extensive study of the superlinearity of synthetic quartz for heavily predosed samples, as well as for samples annealed between 500 and 900 °C. These authors found that preannealing of the quartz samples at high-temperatures ($> 500\,°\mathrm{C}$) caused a decrease in the superlinearity, as well as an increase in the sensitivity to a test dose. The same effect of decreased superlinearity and increased sensitivity could also be induced by heavy predose irradiation of the samples. These phenomena were explained by Pagonis et al. (2003) by using a modified version of the original Zimmerman (1971a,b) model of quartz.

The change of sensitization observed in the predose effect of quartz is caused by the combined effect of irradiating the sample (the predose), and by heating the sample to a high activation temperature, typically around 500 °C. The experimental protocol in the present paper is designed as an attempt to separate these combined effects on the sensitization and superlinearity of quartz, and to discriminate the individual influence of these two properties on the predose effect. To the best of our knowledge, this paper is the first comprehensive attempt in the literature to compare the predose sensitization and superlinearity phenomena in several different types of quartz.

In a recent paper, Kitis et al. (2005) presented a preliminary comparative study of the predose effect for three types of unfired quartz of different origin. Complete TL versus dose and sensitivity S versus predose curves were obtained for the dose range of $0.1 < D < 400\,\mathrm{Gy}$. Additional complete sensitivity versus predose curves were obtained for samples which underwent a combined predose irradiation and a subsequent heat treatment to 500 °C. Although the TL versus dose curves showed very different behaviors, the sensitivity versus predose curves showed several common characteristics. In particular, the sensitivity versus predose curves showed abrupt changes around 10 Gy, and the sensitivity after a combined predose irradiation and heat treatment to 500 °C showed a very gradual change in the whole dose range studied. These preliminary results were explained by using the modified Zimmerman model for quartz to simulate the complete experimental protocol in the study (Kitis et al., 2005).

This model is based on four energy levels consisting of the main TL trap at 110 °C (denoted as T), a deep thermally disconnected competitor trap S, a luminescence/recombination center L and a hole reservoir R. The model is a variation of a previous model developed by Chen and Leung (1999). This model has had several successes in describing a wide variety of phenomena in quartz. These phenomena include the thermal activation characteristics (Chen and Pagonis, 2004), as well as the superlinear characteristics of synthetic quartz as a function of both predose and annealing temperature (Pagonis et al., 2003). Within this model, the predose effect is explained by a twin mechanism involving irradiation of the sample in nature and thermal activation of the sample in the laboratory. During the natural irradiation of the sample holes accumulate in the thermally stable hole reservoir R over time. Heating of the sample in the laboratory (typically to a temperature of 500 °C), leads to the thermal activation of holes which are transferred from the reservoir to the luminescence center L. The increased concentration of holes in L results in an apparent increase of the TL sensitivity when a small test dose is administered to the sample.

The purpose of the present paper is to extend the experimental work by Kitis et al. (2005) to samples that were preannealed at 500 and 900 °C, prior to measuring their predose properties. The results of the present study show that the experimental curves for the three types of quartz studied show several common similarities along with a few major differences which disappear at high annealing temperatures. The observed behaviors of the TL versus dose and of the sensitivity versus predose curves are explained in a qualitative manner and are attributed to possible variations in the concentrations of competitor traps in the quartz samples.

2. Experimental procedure

The materials used in the present study were high purity synthetic quartz, natural Arkansas quartz of hydrothermal origin, and sedimentary quartz from the coast of the Chalkidiki region in Northern Greece. Three types of samples were prepared for each of these three kinds of quartz:

(a) Samples in their “as is” (or as received) state, which were not subjected to any irradiation and heat treatment.
(b) Samples preannealed for 1 h at 500 °C, and immediately cooled to room temperature.
(c) Samples preannealed for 1 h at 900 °C, and immediately cooled to room temperature.

These samples will be referred to in the rest of this paper as “as is”, “preannealed at 500 °C” and “preannealed at 900 °C” samples, correspondingly.

A preliminary account of the experimental procedure and sample preparation has been given in Kitis et al. (2005). The experimental protocol is as follows: the “as is” quartz sample is irradiated with a β predose, and its glow curve is measured by heating up to 150 °C. This yields the TL-response of the 110 °C glow-peak to the β predose. The sample is next irradiated with a β test dose and is heated to an activation temperature of 500 °C. This activation temperature is determined in a separate experiment by using a multiple aliquot thermal activation procedure. This step in the experimental protocol measures the sensitivity of the 110 °C glow-peak due to the β predose without any thermal activation. At the same time, the glow-peaks from 150 °C upwards are obtained and the sample is thermally activated up to the activation temperature of 500 °C. It is noted that this activation step is the basis of the predose technique.

The thermally activated sample is next irradiated by the same β test dose and is heated to 150 °C in order to obtain the sensitivity of the 110 °C glow-peak. This is the sensitivity due to *both* the predose β irradiation and to the thermal activation to 500 °C. The previous steps are repeated for a higher β predose administered in the first step, and by using a new aliquot of the sample each time.

This entire procedure was also repeated for samples which were preannealed at 500 °C, as well as for samples preannealed at 900 °C prior to any heat or irradiation treatment, and for all three types of quartz studied here.

In a separate experiment the thermal activation characteristics (TACs) of the three quartz samples were measured in detail, in order to determine the temperature necessary for thermal activation of the predose effect. These extensive TAC measurements were carried out using both a multiple aliquot and a single aliquot procedure, and were presented elsewhere (Kitis et al., 2006). It was found that a temperature of 450–500 °C was sufficient for thermal activation of the quartz samples, in agreement with previous studies (Aitken, 1985).

It is also noted that in addition to the data presented in this paper, which deals exclusively with the “110 °C” TL peak of quartz, the procedure outlined above also allows determination of the TL versus dose curves for the higher-temperature TL peaks of quartz. These results will be presented elsewhere.

3. Experimental results

3.1. The TL glow curves—estimation of the equivalent beta dose (ED) values

Examples of the TL glow curves at various beta doses for “as is” samples are shown in Fig. 1a–c for synthetic, Arkansas and sedimentary quartz, correspondingly. Fig. 2 shows the corresponding TL glow curves for samples preannealed at 500 °C prior to any treatment, and Fig. 3 shows the TL glow curves for samples preannealed at 900 °C.

It is possible to estimate the equivalent beta dose (ED) values for the Arkansas and sedimentary quartz, since these samples exhibit a geological TL signal with TL peaks above 200 °C. By applying the additive-dose method, the ED values for the Arkansas and sedimentary quartz were found to be ED $\geqslant$ 31 and 16 Gy, respectively. In the case of synthetic quartz the ED value was $ED \sim 0$ Gy for all practical purposes. These values represent a lower limit on the actual geological doses received by the samples, due to their unknown history of exposure to sunlight.

The difference in the ED values for the three quartz samples presents us with a unique experimental opportunity to test the effect of irradiation history on the predose properties of quartz. One of the fundamental assumptions of all quartz models is the existence of a competitor site, which competes for electrons with the “110 °C” TL trap. This competitor site has been previously identified as the E'-center of quartz (Chen et al., 1988). The competitor is assumed to be thermally disconnected, so that the concentration of available competitor sites decreases with time during the lifetime of the quartz sample, due to irradiation from natural sources.

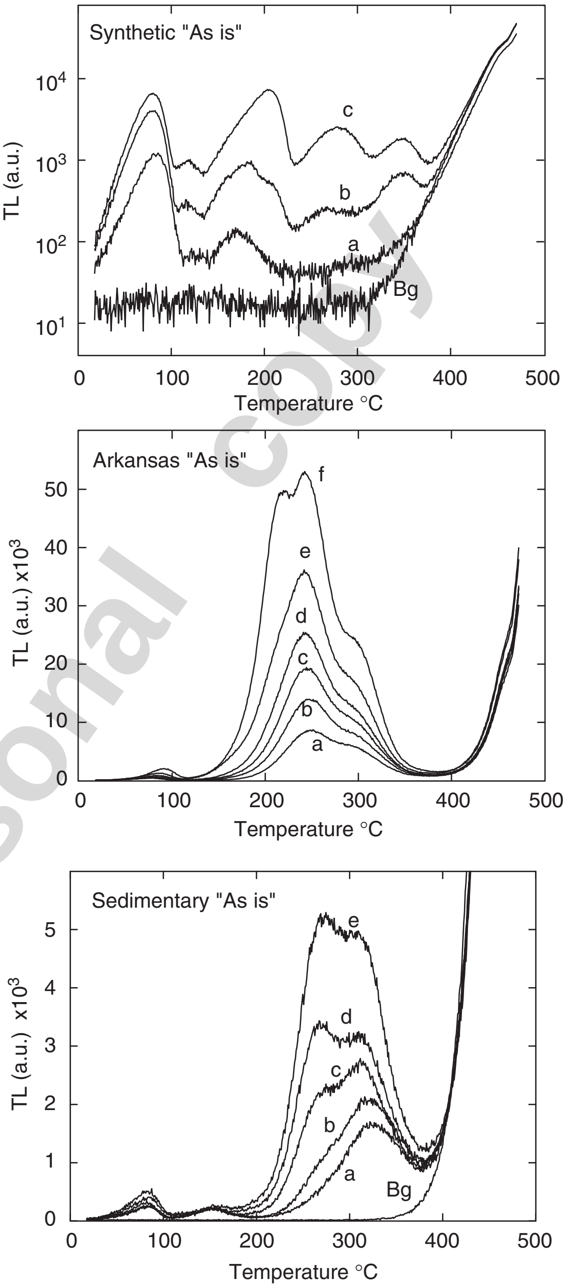

Fig. 1. The TL glow curves for there “as is” quartz samples. Top figure: synthetic quartz at doses (a) 22.4, (b) 89.6 and (c) 232 Gy. Middle figure: Arkansas quartz for (a) geological TL (G-TL) (b) G-TL plus 15.7 Gy (c) G-TL plus 31.3 Gy (d) G-TL plus 67.7 Gy (e) G-TL plus 119.6 Gy (f) G-TL plus 196 Gy. Bottom figure: Sedimentary quartz for (a) geological TL (G-TL) (b) G-TL plus 3.92 Gy (c) G-TL plus 15.7 Gy (d) G-TL plus 31.3 Gy (e) G-TL plus 125.5 Gy.

Since ED $\sim$ 0 Gy for the young synthetic quartz samples, one may reasonably assume that the available competitor sites in this material are empty. On the other hand, since ED $\geqslant$ 31 and 16 Gy for the Arkansas and sedimentary quartz samples, correspondingly, one may reasonably assume that at least part of the available competitor sites in these geological quartz samples are filled. This observation was useful in explaining the TL dose response differences between that “as is” synthetic

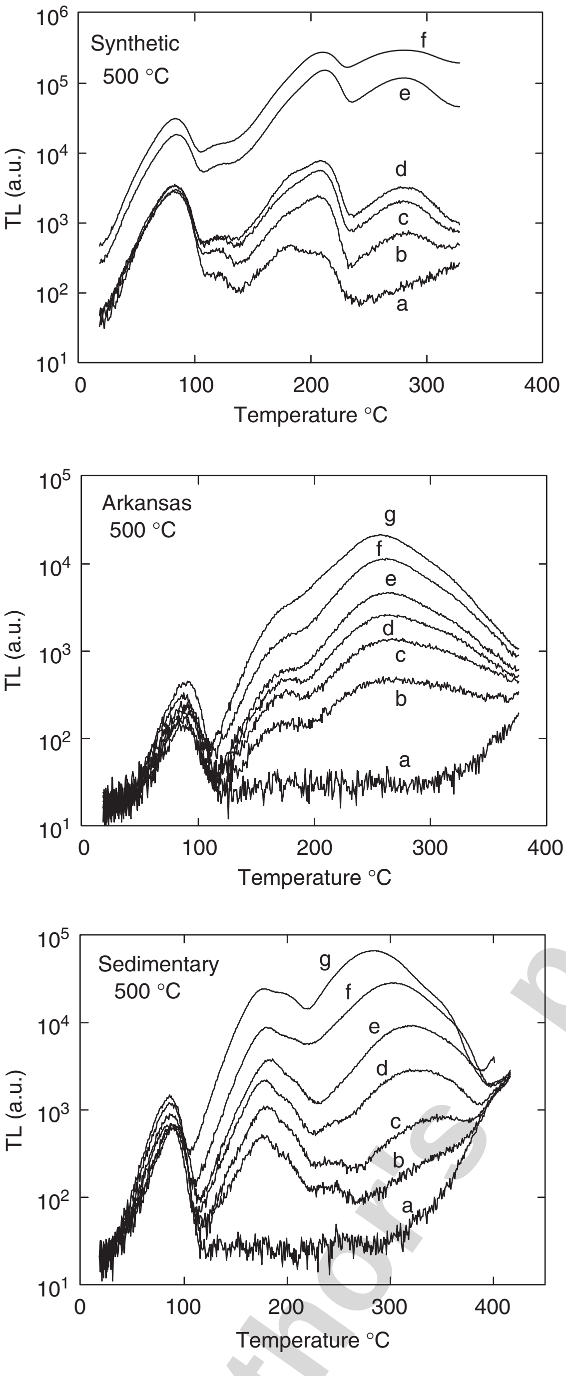

Fig. 2. The TL glow curves for quartz samples preannealed for 1 h at 500 °C and irradiated at different doses. Top figure: synthetic quartz (a) 18 Gy (b) 36 Gy (c) 50 Gy (d) 67 Gy (e) 190 Gy (f) 403 Gy. Middle figure: Arkansas quartz (a) 0.09 Gy (b) 2.8 Gy (c) 6 Gy (d) 12 Gy (e) 24 Gy (f) 48 Gy (g) 96 Gy. Bottom figure: Sedimentary quartz (a) 0.0036 Gy (b) 1.12 Gy (c) 2.8 Gy (d) 6 Gy (e) 12 Gy (f) 24 Gy (g) 48 Gy.

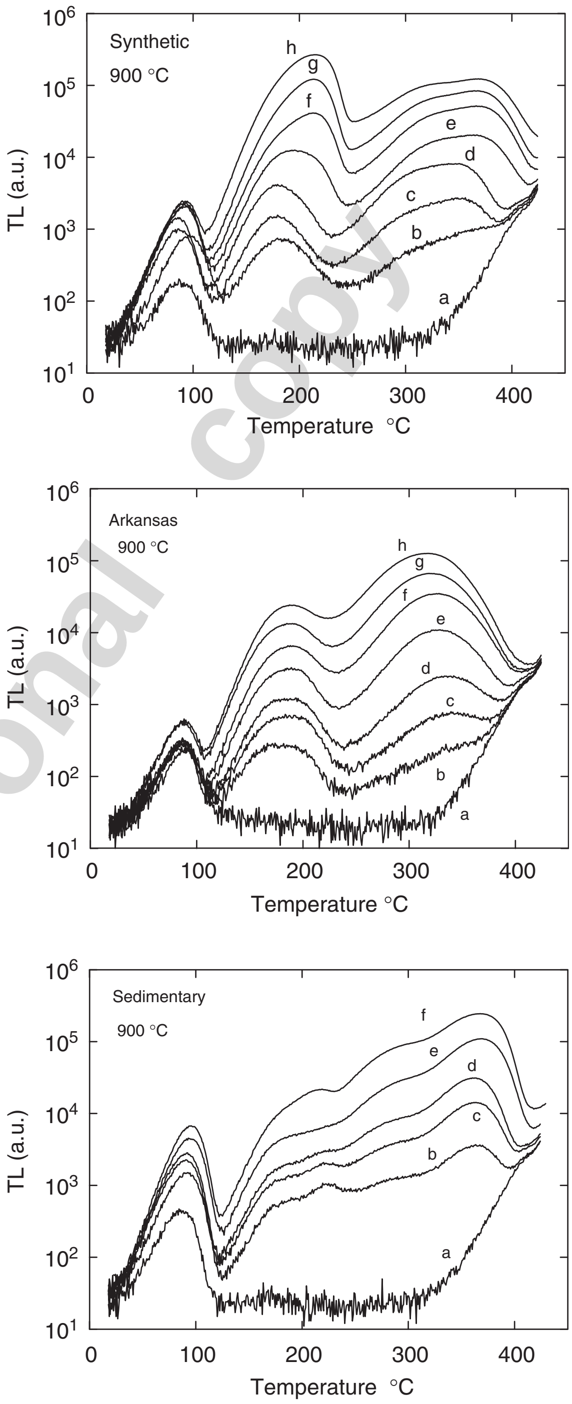

Fig. 3. The TL glow curves for quartz samples preannealed for 1 h at 900 °C and irradiated at different beta doses. Top figure: synthetic quartz (a) 0.0036 Gy (b) 1.68 Gy (c) 3.36 Gy (d) 6 Gy (e) 12 Gy (f) 24 Gy (g) 40 Gy (h) 60 Gy. Middle figure: Arkansas quartz (a) 0.0036 Gy (b) 0.75 Gy (c) 1.68 Gy (d) 3.36 Gy (e) 8 Gy (f) 18 Gy (g) 32 Gy (h) 56 Gy. Bottom figure: Sedimentary quartz (a) 0.0036 Gy (b) 1.68 Gy (c) 3.36 Gy (d) 4 Gy (e) 8 Gy (f) 16 Gy.

and “as is” Arkansas and sedimentary quartz samples (see next section).

3.2. The TL dose response curves

Fig. 4 shows the TL versus dose response curves of the 110 °C glow-peak for the three “as is” quartz samples: (a) synthetic (b) Arkansas and (c) sedimentary quartz. Figs. 5 and 6 show the corresponding results for quartz samples that were preannealed for 1 h at 500 and 900 °C, correspondingly. It is noted that the data in Figs. 4 and 7 have been previously presented in Kitis et al. (2005), but they are included here for the

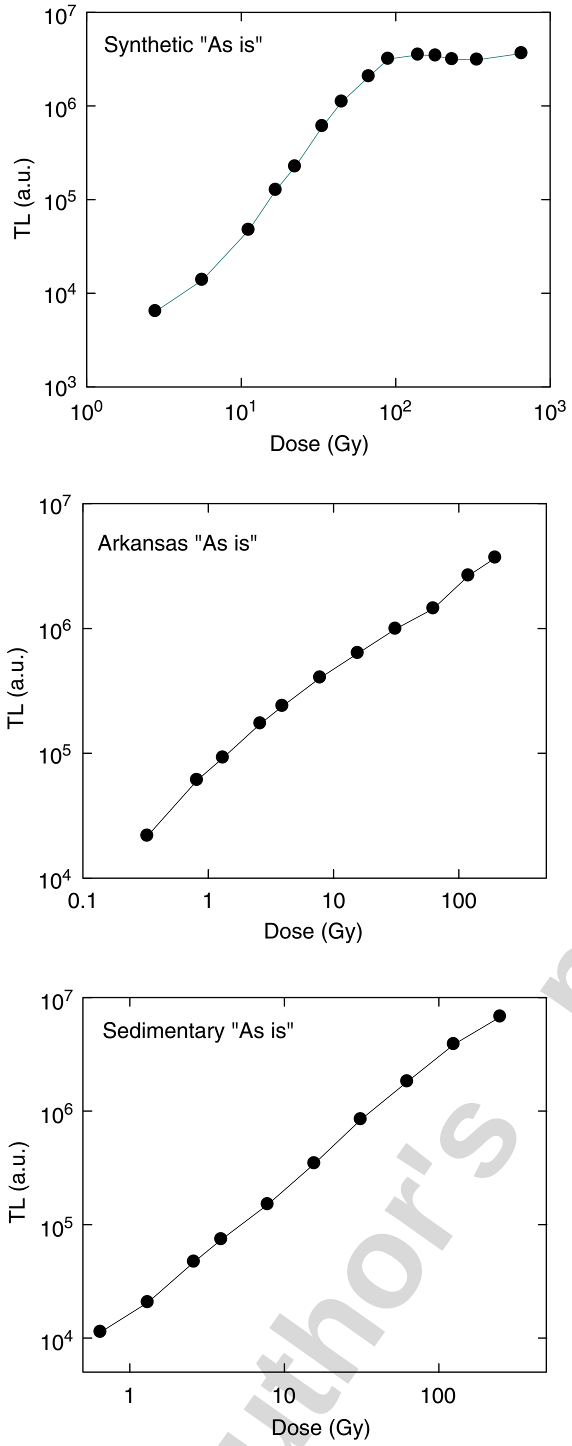

Fig. 4. The TL versus dose response of the "110 °C" glow-peak of the three "as is" quartz samples: (a) synthetic quartz (b) Arkansas quartz (c) sedimentary quartz.

sake of completeness of the presentation and for comparison purposes.

The TL dose response graphs shown in Fig. 4 are obviously very different for each kind of unfired quartz. The unfired synthetic quartz data in Fig. 4a shows no initial linear region, but rather shows superlinearity from the lowest dose. This is consistent with the published data of Charitidis et al. (1999) and Chen et al. (1988) for high purity synthetic quartz. These previous studies found that synthetic quartz exhibits two distinct superlinearity regions, and that high-temperature annealing and/or heavy predose irradiation removed the superlinearity effect.

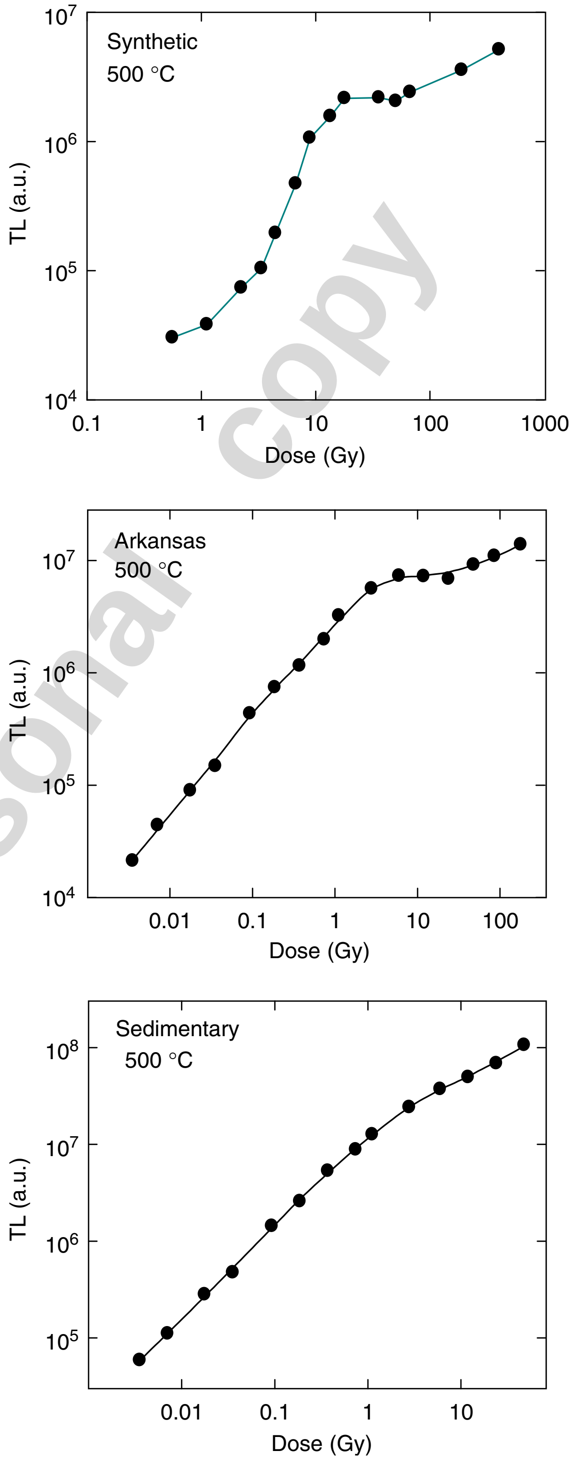

Fig. 5. The TL versus dose response of the "110 °C" glow-peak for quartz samples preannealed for 1 h at 500 °C: (a) synthetic quartz (b) Arkansas quartz (c) sedimentary quartz.

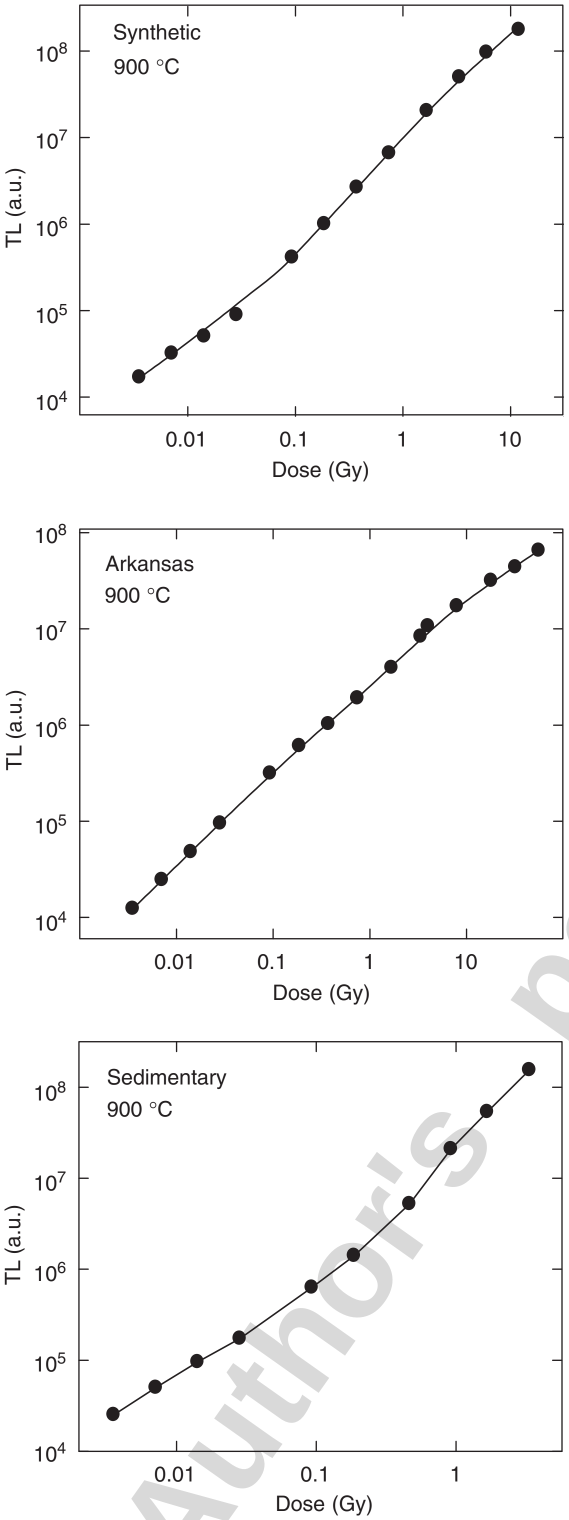

Fig. 6. The TL versus dose response of the “110 °C” glow-peak for quartz samples preannealed for 1 h at 900 °C: (a) synthetic quartz (b) Arkansas quartz (c) sedimentary quartz.

The TL dose response of unfired Arkansas quartz in Fig. 4b is linear in a rather narrow dose region up to 4 Gy, and then becomes clearly sublinear. Finally, the TL dose response of the unfired sedimentary quartz in Fig. 4c is linear over a rather broad range up to 10 Gy, and then becomes slightly superlinear.

The absence of substantial supralinearity in unfired Arkansas and sedimentary quartz samples indicates also the absence of an appreciable number of available competitors, “because no competitors no supralinearity” (Mische and McKeever, 1989). This means that the estimated ED values must be in fact much larger, so that the major part of the competitors in these materials is filled, leaving empty only a small percentage which accounts for the slight supralinearity observed.

Similarly, the TL dose responses shown in Figs. 5a–c for samples preannealed at 500 °C show that synthetic quartz exhibits a markedly different behavior than the Arkansas and sedimentary quartz samples. The data in Fig. 5a for synthetic quartz show that the initial region of superlinearity is now smaller than in Fig. 4a, and is extending up to about 20 Gy. The corresponding data for Arkansas and sedimentary quartz in Figs. 5b and c show a linear behavior through most of the dose region.

The differences shown in Fig. 5 between the three samples that were preannealed at 500 °C, can be explained, again, on the basis of the different occupational state of deep competitors in each material. In the case of Arkansas and sedimentary quartz samples the transition from the slight supralinearity of Figs. 4b and c, to the observed linearity of Figs. 5b and c is consistent with the assumption that preannealing at 500 °C is sufficient to remove (destroy) the small number of available competitors sites in these geological quartz samples. On the other hand, Fig. 5a is consistent with the assumption that preannealing at 500 °C is insufficient to remove all competitor sites in synthetic quartz, because of the much larger concentration of competitor traps available in this sample.

This trend is further supported even more convincingly by the data of Fig. 6, in which samples preannealed at 900 °C exhibit a linear behavior almost throughout the whole dose region, and the TL versus dose behavior of the three quartz samples is essentially the same. This is consistent with the data by Charitidis et al. (1999, 2000), who found that a 900 °C annealing removed almost completely the superlinearity effect in synthetic quartz. Annealing of the samples destroys the competitors leaving only a small percentage which accounts for the remaining small degree of superlinearity, even after annealing at 900 °C (Charitidis et al., 1999; Chen et al., 1988; Yang and McKeever, 1988).

3.3. The sensitization curves due to irradiation only—radiation quenching

Fig. 7 shows the sensitivity of the “110 °C” peak as a function of predose only, normalized over the sensitivity at the lowest predose. The data shown in Fig. 7 is for the three “as is” quartz samples: (a) synthetic (b) Arkansas and (c) sedimentary quartz. Figs. 8 and 9 show the corresponding results for quartz samples that were preannealed at 500 and 900 °C, correspondingly.

The qualitative behavior shown in Figs. 7a–c is the same for all three types of unfired quartz, i.e. after a value of the predose around 10 Gy the sensitivity increases abruptly as a function of predose. The test doses used were: synthetic 4.5 Gy, Arkansas 0.56 Gy and sedimentary 0.56 Gy. A rather high test dose of

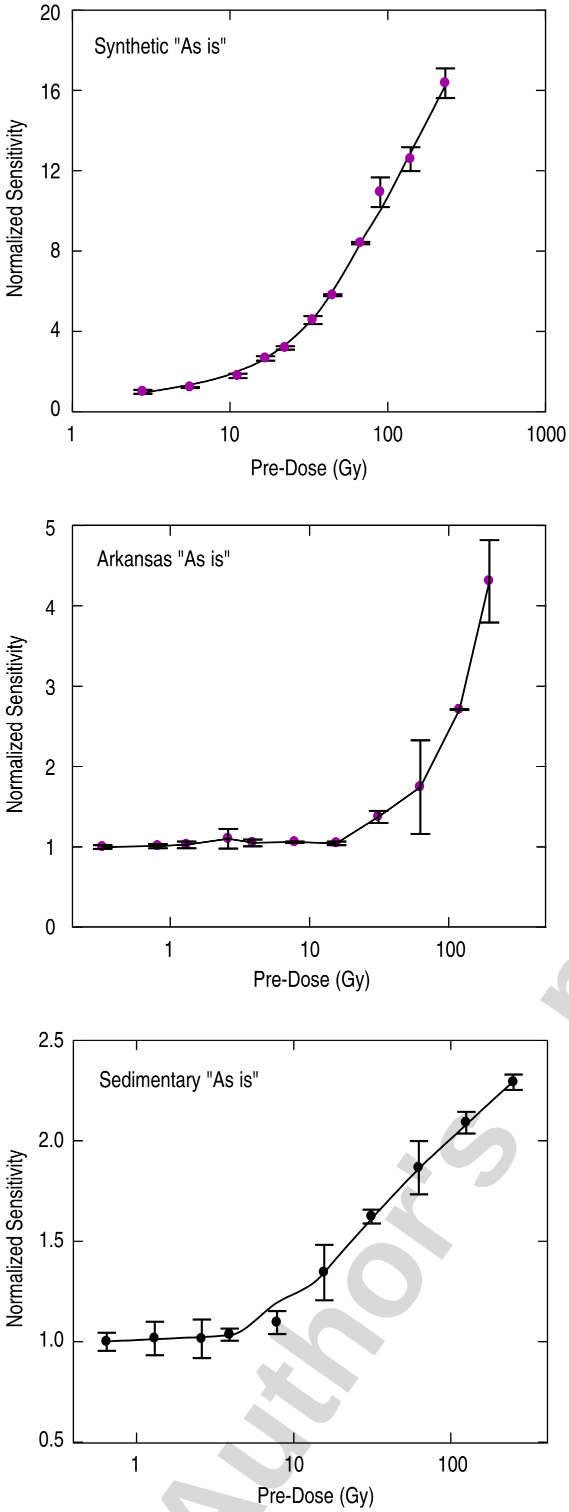

Fig. 7. The sensitivity of the "110 °C" glow-peak as a function of the predose only, normalized to the sensitivity at the lowest predose, for "as is" samples: (a) synthetic quartz (b) Arkansas quartz (c) sedimentary quartz.

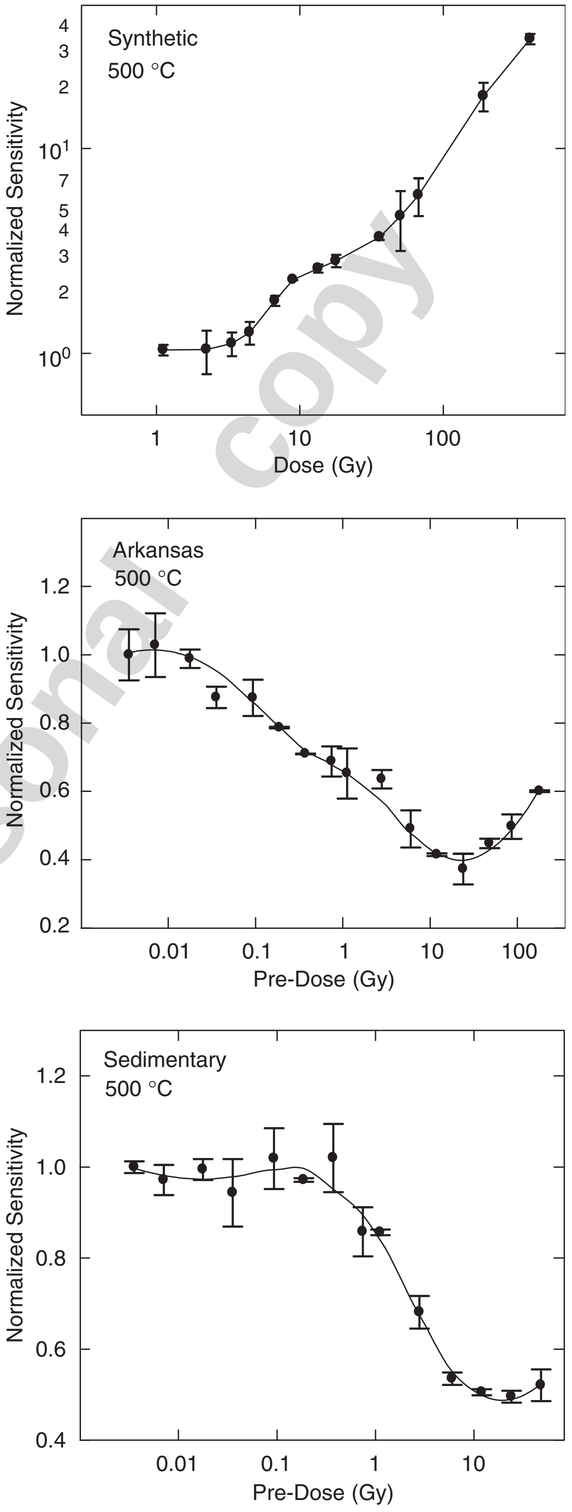

Fig. 8. The sensitivity of the "110 °C" glow-peak as a function of the predose only, normalized to the sensitivity at the lowest predose, for samples preannealed for 1 h at 500 °C: (a) synthetic quartz (b) Arkansas quartz (c) sedimentary quartz. The data in (b) and (c) shows the phenomena of radiation quenching.

4.5 Gy was used in the case of synthetic quartz because of its extreme insensitivity at low doses.

The data in Fig. 8a–c shows a dramatically different behavior between the synthetic, Arkansas and sedimentary quartz samples which were preannealed at 500 °C. Synthetic quartz in Fig. 8a shows a constant sensitivity at low doses followed by an almost continuous increase in sensitivity at higher doses. On the

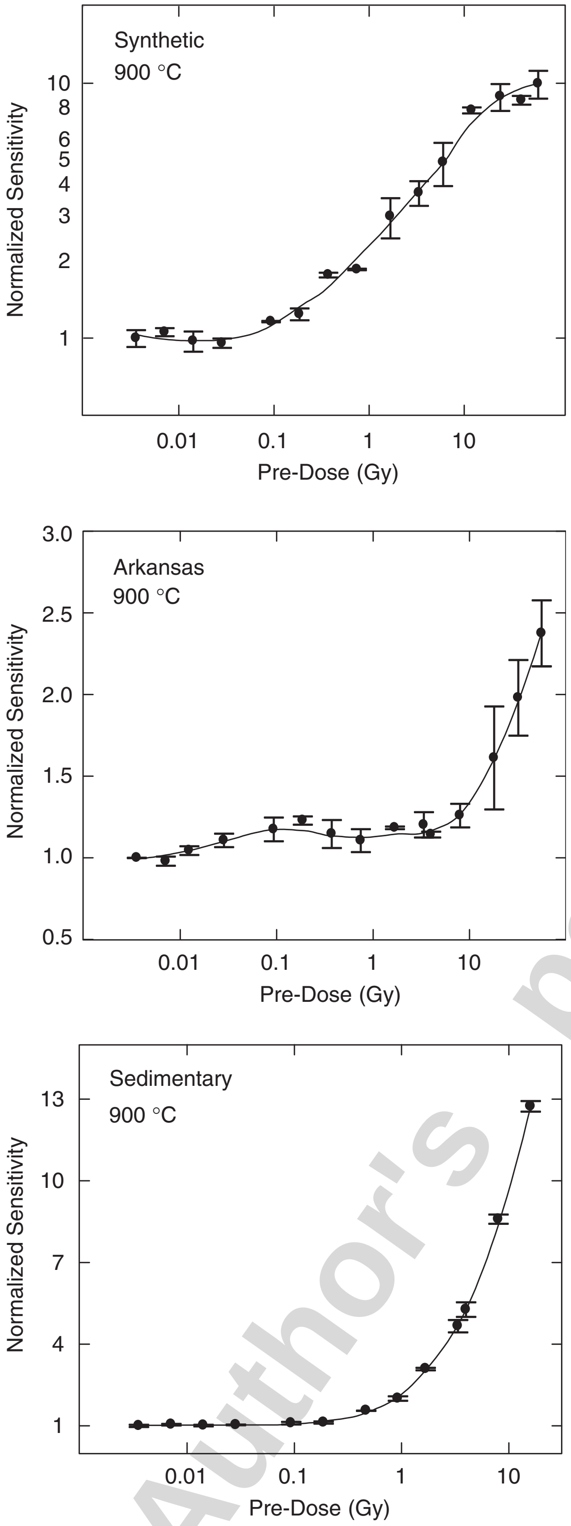

Fig. 9. The sensitivity of the "110 °C" glow-peak as a function of the predose only, normalized to the sensitivity at the lowest predose, for samples preannealed for 1 h at 900 °C: (a) synthetic quartz (b) Arkansas quartz (c) sedimentary quartz.

other hand, the data for Arkansas quartz in Fig. 8b shows an initial dramatic drop in the sensitivity of the order of 50%, before the sensitivity starts recovering at a dose of about 20 Gy. The same behavior is shown in Fig. 8c by the sedimentary quartz sample. At the highest dose available in the data the decrease in sensitivity ceases and a trend for recovery becomes apparent.

The data shown in Fig. 9 for samples preannealed at 900 °C shows that the three quartz samples exhibit again very similar behavior, with an initial region of constant sensitivity up to a dose of 0.1–1 Gy, followed by an abrupt increase in sensitivity at higher doses.

The differences exhibited in Figs. 8a–c for the three quartz samples can be explained qualitatively as follows. The drop of sensitivity due to predose irradiation is a common phenomenon termed "radiation quenching" (Aitken, 1985; Bailey, 2001). According to a previously proposed mechanism for radiation quenching by Aitken (1985), during the heating of the quartz sample to 150 °C in order to empty the main traps *T* (before one measures the TL sensitivity of the "110 °C" TL peak), electrons are released from the main trap *T* and can recombine directly with holes at the luminescence centers *L*. This recombination reduces the concentration of available holes in *L*, and leads to a measured reduced sensitivity to the TL test dose in the next step of the experiment.

According to this proposed mechanism, one would expect that radiation quenching would be more likely to occur in samples with fewer available competitor sites, as in the case of Arkansas and sedimentary quartz. In the case of synthetic quartz many more competitor sites are available for the electrons released from *T*, and these act in competition to the direct recombination process in the center *L*, and thus one is much less likely to observe the radiation quenching effect.

The quenching phenomenon is not present in Figs. 9a–c because preannealing at 900 °C removes the competitor sites and once more the three quartz samples exhibit the same monotonic behavior as a function of the predose.

3.4. *The sensitization curves due to both irradiation and annealing to* 500 °C

Fig. 10 shows the sensitivity of the 110 °C peak for samples that underwent both a predose and thermal activation up to 500 °C. The data in Fig. 10 are also normalized over the sensitivity at the lowest predose for each of the three types of quartz: (a) synthetic (b) Arkansas and (c) sedimentary quartz. The test doses used were 4 Gy for synthetic, 0.28 Gy for Arkansas and 0.19 Gy for sedimentary quartz.

Figs. 11 and 12 show the corresponding results for quartz samples that were preannealed at 500 and 900 °C, correspondingly. The test doses for these samples were 1.2 Gy for synthetic quartz, and 0.0036 Gy for Arkansas and sedimentary quartz samples.

The qualitative behavior for unfired quartz shown in Figs. 10a–c is seen to be very similar for the three quartz samples, exhibiting a slowly varying function of the sensitivity as a function of the predose. This very gradual change in sensitivity for samples that underwent both a predose and a 500 °C annealing, was also previously explained within the modified Zimmerman model (Kitis et al., 2005), by simulating the complete experimental protocol used in this experiment.

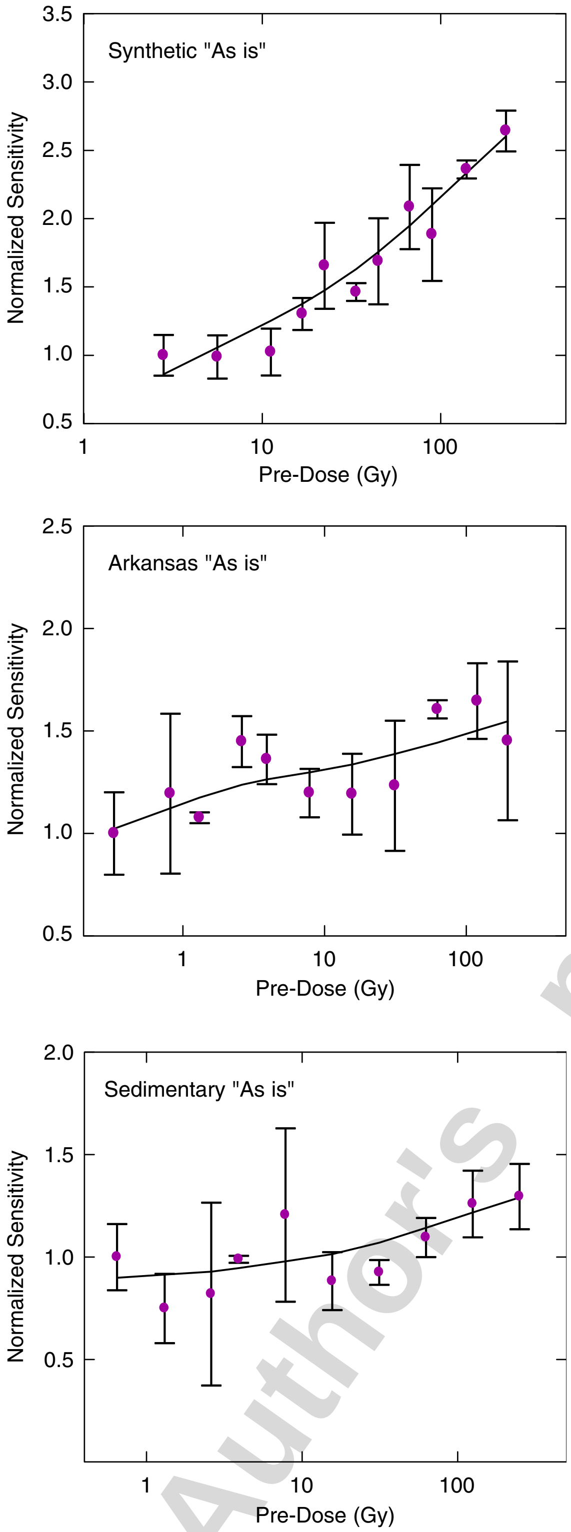

Fig. 10. The sensitivity of the “110 °C” glow-peak for samples that underwent both a predose irradiation and a thermal activation up to 500 °C, normalized to the sensitivity at the lowest predose. The samples were “as is”: (a) synthetic quartz (b) Arkansas quartz (c) sedimentary quartz.

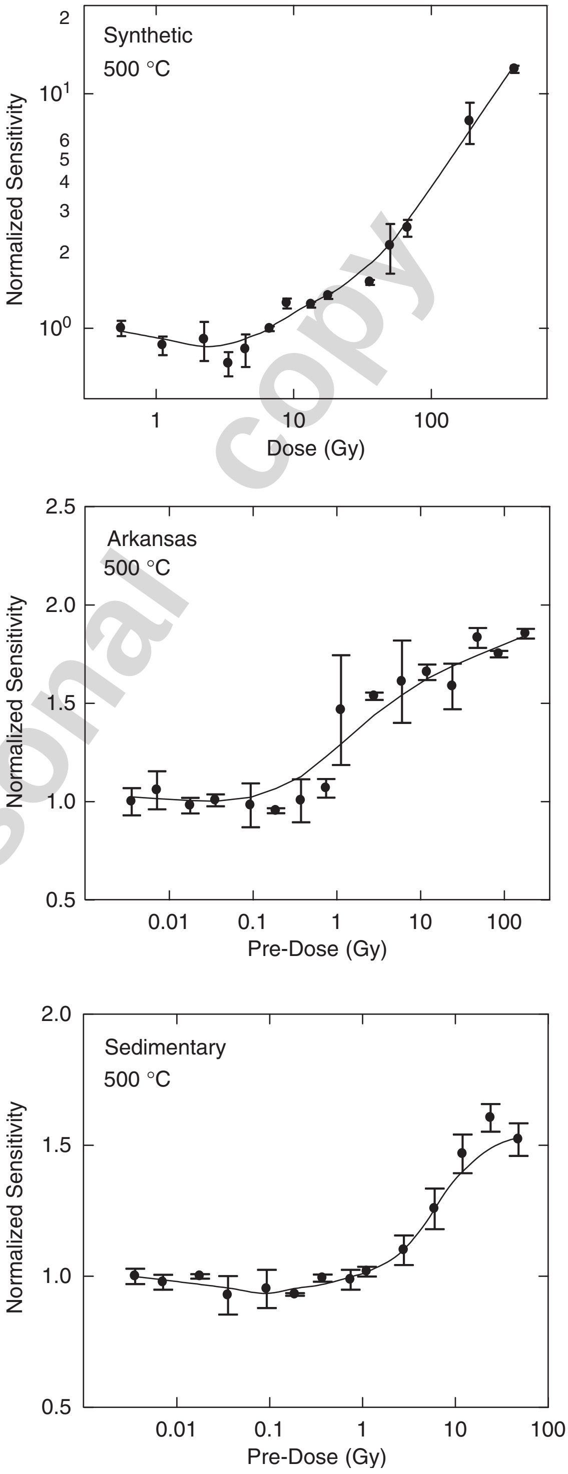

Fig. 11. The sensitivity of the “110 °C” glow-peak for samples that underwent both a predose irradiation and a thermal activation up to 500 °C, normalized to the sensitivity at the lowest predose. The samples were preannealed for 1 h at 500 °C: (a) synthetic quartz (b) Arkansas quartz (c) sedimentary quartz.

The synthetic quartz data in Fig. 10a shows a total increase of the sensitivity by a factor of about 3 over the dose range $1\,\mathrm{Gy} < D < 40\,\mathrm{Gy}$, while the corresponding graphs in Fig. 10b–c for Arkansas and sedimentary quartz show an increase of 50% and 20%, correspondingly. Similar behaviors are shown in Figs. 11a–c, with the synthetic quartz showing an increase of sensitivity by a factor of 10, while the Arkansas and sedimentary quartz samples show an increase by a factor of 2 and 1.5, correspondingly.

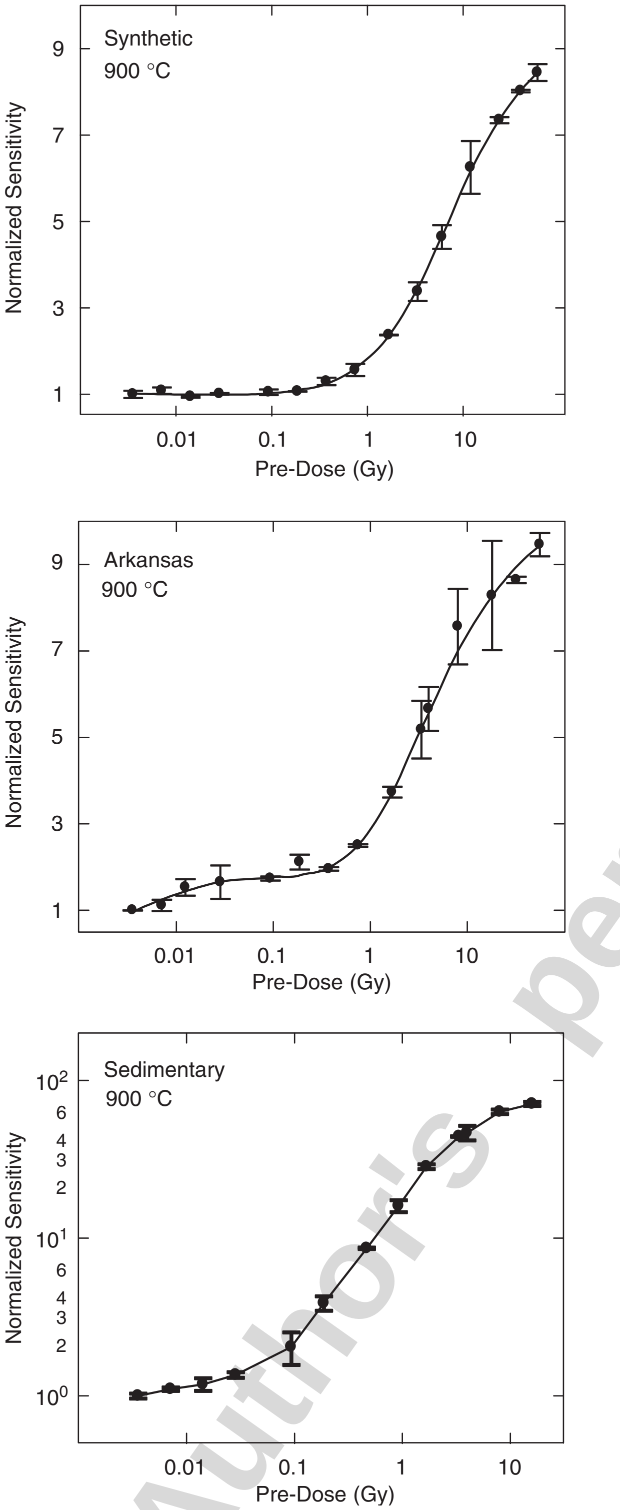

Fig. 12. The sensitivity of the "110 °C" glow-peak for samples that underwent both a predose irradiation and a thermal activation up to 500 °C, normalized to the sensitivity at the lowest predose. The samples were preannealed for 1 h at 900 °C: (a) synthetic quartz (b) Arkansas quartz (c) sedimentary quartz.

The data in Fig. 12a–c show that the three quartz samples preannealed at 900 °C exhibit almost identical behaviors, with an abrupt change in sensitivity at about 0.1 Gy.

In summary the data in Figs. 10–12 lead to two very important conclusions: (a) the sensitization due to the combined predose and thermal activation, increases with the annealing temperature and (b) the sensitization due to the combined predose and thermal activation is very different from the sensitization induced by predose only.

4. Discussion

The explanation of the results of the present work requires careful consideration of the various steps in the experimental procedure. According to the steps followed in this study, the TL dose response, the sensitization due to predose only, and the sensitization due to the combined predose and thermal activation, are all obtained from measurements on the same sample. Taking, for example, the case of "As is" quartz, every experimental point of Fig. 4 (the TL dose response) is measured before the experimental points of Fig. 7 (the sensitization due to predose only), and subsequently the data of Fig. 10 is obtained on the same sample (the sensitization due to combined predose and thermal activation). Therefore, one has to explain how the same samples which give the significant sensitization of Fig. 7, give subsequently the negligible sensitization of Fig. 10. Similarly, in the case of the annealing at 500 °C it is impressive how the radiation quenching behavior of Figs. 8b and c for Arkansas and sedimentary quartz changes to the sensitization of Fig. 11. Finally, in the case of annealing at 900 °C the strong sensitization of Fig. 9 changes to the larger sensitization of Fig. 12, i.e. a behavior opposite to that observed in the case of the "As is" samples.

It is crucial to observe here that the only procedure interposed between the sensitization due to predose only data of Figs. 7–9 and the very different sensitization due to both predose and thermal activation in Figs. 10–12, is the thermal activation step. Therefore, the effects of thermal activation can be summarized as follows. The thermal activation up to 500 °C removes the sensitization of Fig. 7, restores the radiation quenching of Fig. 8 and enhances even more the sensitivity of Fig. 9. Furthermore, the thermal transfer of holes from the reservoirs *R* to luminescence centers *L* gives rise to the results of Figs. 10 and 11 and to a further enhancement of the sensitization of Fig. 12.

An extra factor that must be taken into consideration here is that in addition to the very deep competitor electron traps considered above, the electron traps responsible for the glow curve in the region 150–500 °C also must play a role in the effects observed, since they can also act as competitors to the trap responsible for the 110 °C glow-peak. Under this assumption, the sensitization of Fig. 7 exists because as the dose increases, the traps responsible for the peaks between 150–500 °C are filled and do not compete effectively with the trap responsible for the 110 °C glow-peak. As a result, the sensitivity of the 110 °C glow-peak increases. Subsequently, the thermal activation up to 500 °C empties the high-temperature traps, which will then compete with the 110 °C glow-peak more effectively, removing thus its sensitization. The thermal transfer of holes from the reservoirs *R* to luminescence centers *L* gives rise to the slight increase of the sensitivity observed in Fig. 10.

Table 1
Changes in the sensitization of the three quartz samples (synthetic, Arkansas and sedimentary), for "as is" samples, samples preannealed at 500 and 900 °C

	S_{pa}/S_p for "as is" samples	S_{pa}/S_p for samples preannealed at 500 °C	S_{pa}/S_p for samples preannealed at 900 °C
Synthetic quartz	6.0	2.8	1.27 ± 0.10
Arkansas quartz	5.1	1.02	1.12 ± 0.10
Sedimentary quartz	20.4	1.04	1.25 ± 0.10

The symbols shown are: $S_{\text{as is}}$, Sensitivity to a test dose for "as is" samples. The sensitivity is measured at the lowest available experimental predose; $S_p = S_{\text{predose}}$, Sensitivity to the same test dose for samples that only received a predose irradiation. The sensitivity is measured at the lowest available experimental predose; $S_{pa} = S_{\text{predose+500−Anneal}}$, Sensitivity to the same test dose for samples that received a combined predose irradiation and an 1-h anneal at 500 °C. The sensitivity is measured at the lowest available experimental predose.

In the case of the Arkansas and sedimentary quartz samples (the synthetic quartz shows its own behavior related to the fact that a small percentage of the deep electron competitors still exists), the thermal activation empties again all the high-temperature traps, so that many competitors are now available and the radiation quenching effect is restored into a sensitization behavior. Again this can be attributed to the thermal transfer of holes from the reservoirs R to luminescence centers L giving rise to the significant increase of the sensitivity in Fig. 11.

Finally, in the case of the samples annealed at 900 °C the sensitization effects become similar to all quartz samples for both predose only sensitization and predose plus thermal activation sensitization. Moreover, in all cases the sensitization graphs are shifted to much lower doses. The deep electron competitors do not play any major role here, since they were destroyed by the high-temperature annealing.

5. Concluding remarks

The experimental results presented in the previous sections are consistent with the measured low limits of the equivalent doses (EDs) for the three types of quartz used in this study. Specifically, the differences in behavior between the synthetic quartz and the geological Arkansas and sedimentary samples were explained in a qualitative manner by the assumption that a large concentration of competitor traps is available in synthetic quartz, while a much reduced concentration of competitors is available in the geological Arkansas and sedimentary quartz samples. When the three quartz samples were preannealed at 900 °C, these competitor sites become unavailable and the three quartz samples show an almost identical predose behavior in terms of their sensitivity and superlinearity properties.

This elimination of the differences between the three quartz samples by high-temperature preannealing is shown in a quantitative manner in Table 1. In this table the changes in sensitivity at the lowest available doses are tabulated for all three quartzes and for different annealing treatments. The data in Table 1 shows that as the preannealing temperature increases, the differences in the sensitivity changes disappear and the sensitivity ratios become identical, at least within the accuracy of the experimental data.

In conclusion, this paper presents the results of a comprehensive comparative study of the predose effect in three types of quartz of different origin. The concentration of available competitor sites in quartz emerges as the possible fundamental factor causing the observed differences in predose behavior between the three quartz samples.

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