Radiation Protection Dosimetry (2006), Vol. 119, No. 1–4, pp. 438–441
doi:10.1093/rpd/nci548 Advance Access published on May 22, 2006

A COMPREHENSIVE COMPARATIVE STUDY OF THE PREDOSE EFFECT FOR THREE QUARTZ CRYSTALS OF DIFFERENT ORIGIN

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The study of the thermoluminiscence (TL) sensitivity of quartz due to heat and irradiation treatments is of importance in dating and retrospective dosimetry. A comprehensive comparative study of the predose effect was carried out for three types of quartz of different origin. Complete TL vs. dose and sensitivity *S* vs. predose curves were obtained for the dose range of $0.1 < D < 400$ Gy. Additional complete sensitivity vs. predose curves were obtained for samples which underwent a combined predose irradiation and a subsequent heat treatment to 500°C. Although the TL vs. dose curves showed very different behaviours, the sensitivity vs. predose curves showed several common characteristics. The sensitivity vs. predose curves showed abrupt changes ~10 Gy. 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 results are explained qualitatively by using the modified Zimmerman model for quartz.

INTRODUCTION

The study of the changes in the thermoluminiscence (TL) sensitivity of quartz due to heat and irradiation treatments is of fundamental importance in dating and retrospective dosimetry applications. In particular, the change in sensitivity due to thermal activation forms the basis of the well-established predose technique for retrospective dosimetry. The phenomena of non-linear response to administered dose, thermal sensitisation and sensitisation owing irradiation treatments, have been studied extensively for both synthetic and natural quartz [Ref. (1) and references therein].

There have been very few comparative studies of the sensitisation properties of different types of quartz, under different 'pre-dose' irradiations and/or heating of the samples to various temperatures. The purpose of the present study was to carry out a complete characterisation of the TL and sensitisation characteristics for three types of quartz of very different origin.

The complete experimental protocol employed in this study was simulated using a kinetic model introduced by Chen and Leung[(2)]. It is noted that the model of Chen and Leung[(2)] has had several successes in describing several phenomena in quartz. These phenomena include the thermal activation characteristics[(3)], as well as the superlinear characteristics of synthetic quartz as a function of both predose and annealing temperature[(4)].

The new experiments presented here show that not only the TL vs. dose curves, but also the sensitivity vs. predose curves (before and after a thermal treatment to 500°C) can also be simulated using essentially the same kinetic parameters previously employed to explain the superlinearity properties of quartz[(4)].

MATERIALS AND METHODS

The materials used in the present study were high purity synthetic quartz, natural Arkansas quartz of hydrothermal origin and sedimentary quartz. The three kinds of quartz used in the present study were in their 'as is' (or as received) state, so that they were not subjected to any irradiation and heat treatment.

All quartz samples were used in powder form of dimension 80–200 μm, which were obtained by crushing and sieving large quartz portions. The sample was prepared using a stable volume dispenser, which provides a sample of 2.5 mg in weight. The powder flows from the dispenser on a brass disc of an area 1 cm^2, which was previously covered by a very thin layer of non-luminescent silicon oil. The reproducibility of the dispenser is better than 5%.

The experimental procedure was performed according to the following steps:

Step 1. The 'as is' quartz sample was irradiated with a beta predose.

Step 2. The irradiated 'as is' quartz sample was readout up to 150°C. This obtains the TL-response of the 110°C glow-peak to the beta dose used in Step 1 above.

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Step 3. The sample was irradiated with the beta test dose and heated to an activation temperature of 500°C. This temperature is determined in a separate experiment by using a Multiple Aliquot Thermal Activation procedure. The aim of this step in the experimental protocol is three-fold as follows:

(1) The sensitivity of the 110°C glow-peak, owing to the predose of Step 1 without any thermal activation, is obtained.
(2) The glow-peaks from 150°C upwards are obtained.
(3) The sample is thermally activated up to the activation temperature of 500°C. It is noted that this step is the basis of the predose technique.

Step 4. The thermally activated sample was irradiated by a beta test dose and heated to 150°C to obtain the sensitivity of the 110°C glow-peak. This is the sensitivity due to both the predose irradiation (administered in Step 1) and the thermal activation to 500°C (administered in Step 3).

Step 5. Steps 1–4 were repeated for a higher beta predose (as administered in Step 1), using a new aliquot of the sample.

For the TL measurements a TL analyser type 711 of the Littlemore Co was used. The light emission was detected by an EMI 9635 QA photomultiplier tube and the glow-curves were stored in a computer via a 1024-channel ADC card operating in the MCS mode. The heating strip was nichrome of thickness 0.8 mm, with a Cr–Al thermocouple fixed on it. The heating rate used was 2°C s^{-1}. The irradiations were performed with a ^{90}Sr–^{90}Y beta-ray source delivering 0.56 mGy min^{-1} in a sample distance of 15 mm. The time elapsed between the end of irradiation and the readout was in all cases <1 min.

RESULTS AND DISCUSSION

The TL dose–response curves of the 110°C glow-peak of the three quartz samples are shown in Figure 1. It is noted that the synthetic quartz has no initial linear region, but rather shows superlinearity from the lowest dose. The saturation region for the quartz samples starts at ~100 Gy. The TL dose–response of the sedimentary quartz is linear up to 10 Gy and then becomes slightly superlinear. Finally, the Arkansas quartz is linear up to 4 Gy and then becomes clearly sublinear.

The sensitivity of the 110°C peak as a function of predose only (obtained in Step 3 of the experimental protocol) is shown in Figure 2, normalised over the sensitivity at the lowest predose. The qualitative behaviour shown in Figure 2 is the same for all three types of quartz, i.e. after a value of the predose ~10 Gy the sensitivity increases abruptly as a function of predose. The test doses used were as follows: synthetic 4 Gy, Arkansas 0.56 Gy and sedimentary 0.56 Gy.

The sensitivity of the 110°C peak for samples that underwent both a predose and thermal activation up to 500°C (as obtained in Step 4 of the experimental protocol) is shown in Figure 3. The data of Figure 3 are also normalised over the sensitivity at the lowest predose for each of the three types of quartz. The qualitative behaviour is shown to be similar for the Arkansas and sedimentary quartz, and slightly different for synthetic quartz. Nevertheless, all three curves in Figure 3 show the same general behaviour, a slowly varying function of the sensitivity as a function of the predose. The test doses used were 4 Gy

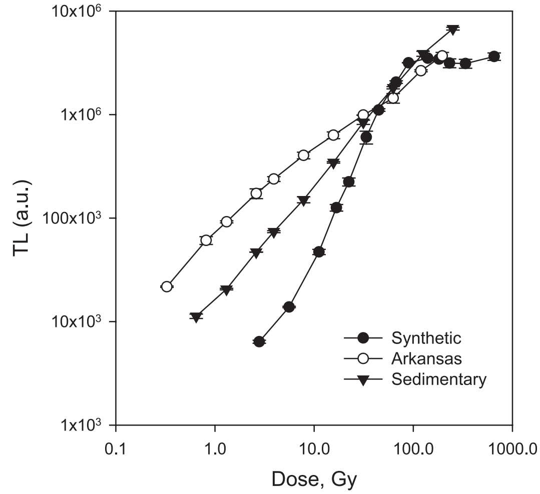

Figure 1. The TL vs. dose-response of the ‘110°C’ glow-peak of the three quartz types as a function of the dose.

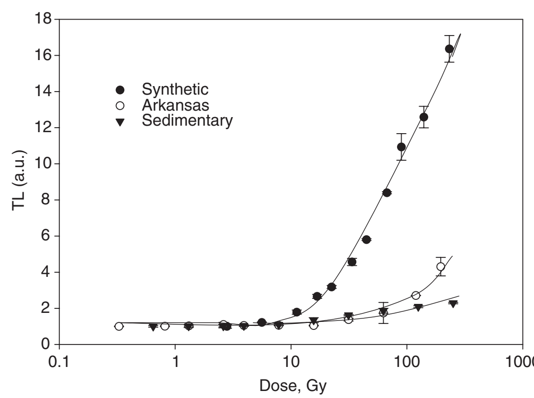

Figure 2. The sensitivity of the ‘110°C’ glow-peak as a function of the predose only, normalised to the sensitivity at the lowest predose.

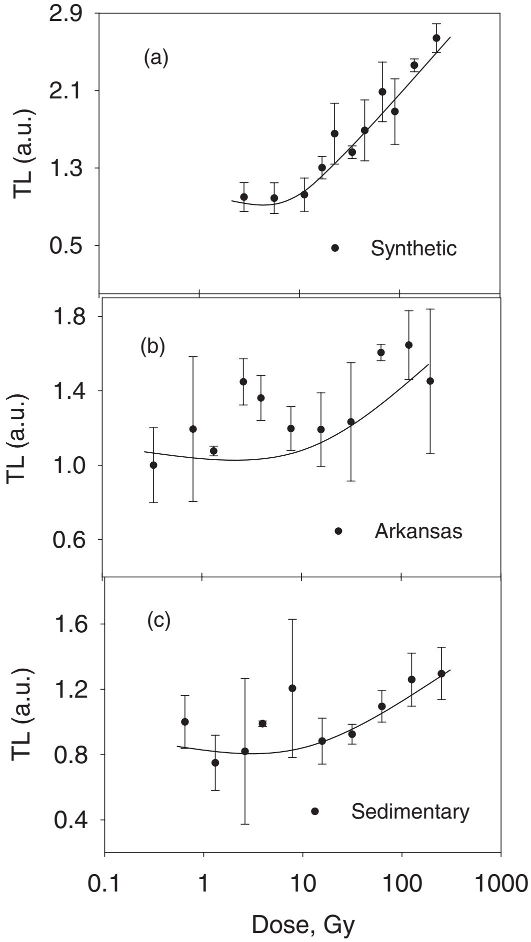

Figure 3. The sensitivity of the '110°C' glow-peak for samples that underwent both a predose irradiation and a thermal activation up to 500°C, normalised to the sensitivity at the lowest predose. (a) synthetic (b) Arkansas (c) sedimentary quartz.

for synthetic, 0.28 Gy for Arkansas and 0.19 Gy for sedimentary quartz. It must be noted that the behaviour shown in Figures 2 and 3 is of fundamental importance in applying the predose dating technique.

The sensitivities at the lowest predose in Figure 2 (sensitisation owing to predose only), are increased relative to the respective initial sensitivities of Figure 1 by no more than 15%. However, the sensitivities at the lowest predose in Figure 3 (sensitisation owing to both predose and thermal activation) are increased relative to the respective normalisation values of Figure 2 as follows. The increase for synthetic quartz is by a factor of 6, in Arkansas quartz by a factor of 10 and in sedimentary quartz by a factor of 60. 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 >200°C. By applying the additive-dose method, the ED values for the Arkansas and sedimentary quartz were found to be 31 and 16 Gy, respectively. In the case of synthetic quartz the ED value was zero for all practical purposes. These values represent a low limit on the actual geological doses received by the samples, owing to their unknown history of exposure to sunlight.

Another result, which deserves attention, is the large experimental errors existing in Figure 3. Each point in Figures 1–3 is the mean value of several runs made with the same sample. In the cases of Figures 1 and 2 the relative errors for the most part are <5%, whereas in the case of Figure 3 the error bars are much higher—of the order of 25%. The only difference between the results of Figures 1 and 2, and the results of Figure 3 is the thermal activation up to 500°C in Figure 3. It is possible that the thermal activation process perturbs the high temperature region of the glow-curve, where the unknown optical, irradiation and grinding history of the samples are accumulated. The large errors in Figure 3 could be attributed to these unknown histories of the three quartz samples.

SIMULATION OF THE EXPERIMENTAL RESULTS

Chen and Leung[2], used a mathematical model based on a thermally connected trapping state T, a thermally disconnected trapping state S, a hole reservoir R and a luminescence centre L to provide a mathematical description of the predose effect in quartz. This model and the kinetic equations associated with it have been presented in detail elsewhere[4] and will not be repeated here.

The four-level model produces a broad qualitative agreement with the above experimental results. This is shown in Figure 4, where we simulate the exact experimental procedure described in the Materials and Methods section (Steps 1–5). The kinetic parameters used are the same as those used by Pagonis *et al.*[4], with the only exception of the activation energy E_R of the reservoir R. The value of E_R was increased from $E_R = 1.4$ eV to $E_R = 1.6$ eV in order to obtain better agreement with the experimental thermal activation curves (TACs) of the three quartz samples. The TACs were measured in a separate detailed experiment, to be presented elsewhere.

Figure 4a shows the sensitivity vs. predose curves calculated by the model of Chen and Leung, and it shows the same abrupt change in the sensitivity as seen in the experimental data of Figure 2. The results of the computer simulation show that this abrupt change in the TL sensitivity observed experimentally

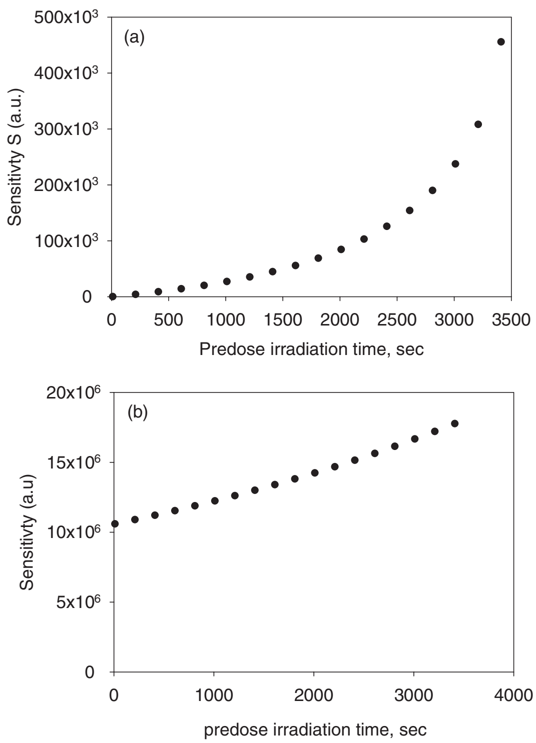

Figure 4. The results of the simulation of the experiments (a) Sensitivity vs. predose only and (b) Sensitivity vs. predose after heating to 500°C. These are to be compared with the experimental data shown in Figures 2 and 3 above.

for all three types of quartz in Figure 2, may be caused by the competitor becoming filled at a dose of ~10 Gy.

Figure 4b shows the sensitivity vs. predose curves for samples which underwent both a predose and heating treatment. The results from the model demonstrate the same very gradual change in the sensitivity as seen in the experimental data of Figure 3, for all three types of quartz.

CONCLUSIONS

This paper presents the results of a comprehensive comparative study of the predose effect in three different types of quartz of different origin. Although the TL vs. dose curves showed very different behaviours containing linear, sublinear and superlinear regions, the sensitivity vs. predose curves for the three quartz types showed several common characteristics. These changes include abrupt changes ~10 Gy in the sensitivity vs. predose curves. 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 common experimental sensitisation characteristics have been explained qualitatively by using the four-level modified Zimmerman model introduced by Chen and Leung. The complete experimental protocol used in this study was simulated and the abrupt changes in the sensitivity vs. predose curves seen at ~10 Gy were explained by the saturation of the competitor trap within the model of Chen and Leung.

REFERENCES

1. McKeever, S. W. S. *Mechanisms of thermoluminescence production: some problems and a few answers?* Nucl. Tracks Radiat. Meas. **18**, 5–12 (1991).
2. Chen, R. and Leung, P. L. *Modeling the pre-dose effect in thermoluminescence*. Radiat. Prot. Dosim. **84**, 43–46 (1999).
3. Chen, R. and Pagonis, V. *Modelling thermal activation characteristics of the sensitization of thermoluminescence in quartz*. J. Phys. D Appl. Phys. **37**, 159–164 (2004).
4. Pagonis, V., Kitis, G. and Chen, R. *Applicability of the Zimmerman predose model in the thermoluminescence of predosed and annealed synthetic quartz samples*. Radiat. Meas. **37**, 267–274 (2003).