Radiation Protection Dosimetry
Vol. 100, Nos 1–4, pp. 373–376 (2002)
Nuclear Technology Publishing

SEARCH FOR COMMON CHARACTERISTICS IN THE GLOW CURVES OF QUARTZ OF VARIOUS ORIGINS

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Abstract — The thermoluminescence (TL) glow curves of quartz of various origins were measured under two different conditions, (1) unannealed samples and (2) samples annealed at 500°C and 900°C. The different glow curves obtained were analysed using first order kinetics and glow curve deconvolution (GCD) analysis. The comparison of the glow curves obtained was mainly concentrated in studying the sensitivities of the glow peaks as a function of the annealing temperature, and in obtaining the kinetic parameters of the glow peak at '110°C'. Furthermore, in four samples the detailed comparison was extended to the trapping parameters of all existing glow peaks. It was found that despite their different origin and the different shapes of the glow curves, there are several basic characteristics that are common to all samples studied.

INTRODUCTION

The thermoluminescence (TL) properties of the '110°C' peak in quartz and in particular its sensitisation by heating and irradiation has been the subject of numerous studies because of its importance in dating and dosimetry (Reference 1 and references therein). Despite all this previous work, there have been few comparative studies of the sensitisation effect and of the TL activation energy E for quartz of various origins.

Gandhi *et al*[2] studied the effect of heating and irradiation on the TL glow curves for several synthetic forms of quartz. Benny *et al*[3] studied the sensitisation of the 110°C TL peak in quartz separated from sand. Specifically they studied the effect of annealing temperature, amount of pre-dose irradiation and annealing time on the sensitisation factor. They concluded that the radiation-induced sensitisation could be due to elimination of the competing traps. Rendell *et al*[4] studied the effects of high temperature annealing on the sensitisation as well as on the emission spectra of various types of quartz of different origins. They found that the TL signal was enhanced by factors up to 1000 times and that the emission spectra were influenced by the rate of cooling of the samples to room temperature. Kitis *et al*[5] in a series of papers studied the effect of annealing temperature and irradiation temperature on the TL properties of high purity synthetic quartz.

The purpose of this paper is to look for common characteristics in the TL glow curves of quartz of various origins, with a special emphasis on the kinetic parameters of the '110°C' peak in quartz. It was found that the temperature of maximum TL intensity as well as the activation energy of the '110°C' TL peak varies within a very narrow range of values for all quartz types studied. The T_m-T_{stop} method was found to be a reliable method of determining the number of peaks in the complex TL glow curves of both synthetic and geological quartz. In all cases the TL glow curves could be fitted with first order kinetics by using glow curve deconvolution (GCD) analysis. The sensitivity of the '110°C' TL peak caused by successive heating and irradiation cycles was also found to lie within a narrow range of values. Finally the 'plateau' test yielded very similar results for all quartz types studied.

The results point to a common type of TL centre being responsible for the '110°C' TL peak in quartz samples, regardless of their origin. Furthermore, it is concluded that TL studies of quartz samples extracted from sea sand could prove very useful in both dating and in retrospective dosimetry.

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EXPERIMENTAL

Table 1 shows the various quartz samples and the apparent location of their major TL peaks, as measured with a heating rate of 2°C.s^{-1}. There are two main types of quartz studied. Group 1 consists of natural crystals termed Arkansas, Norway, synthetic Q-quartz and Pink variety quartz, and Group 2 consists of 9 different types of quartz grains extracted from sea sand. The study included at least two different aliquots from each quartz sample.

TL measurements were performed with the 2000A-B TLD reader of the Harshaw Chemical Co. Light emission was detected by an EMI S-11 photomultiplier tube. The maximum readout temperature was 450°C and a heating rate of 2°C.s^{-1} was employed unless otherwise noted. The irradiations were performed with a ^{90}Sr beta source delivering about 0.59 Gy.min^{-1}. The black body radiation was digitally subtracted from all TL glow curves.

Annealing of the samples was performed in an oven at temperatures of 500°C and 900°C for 1 h in air. Immediately after the end of the annealing the samples were cooled quickly to room temperature by placing them on a copper block. After the high temperature annealing, two successive cycles of irradiation and readout were performed in order to see the variation of the sensitivity of the '110°C' TL peak relative to the unfired samples, and also to see the thermal activation of the sensitivity.

EXPERIMENTAL RESULTS

The T_m-T_{stop} method

The well-known T_m-T_{stop} method was applied to both the natural Arkansas quartz and the high purity Q-quartz. Figure 1 shows the results for both quartz samples. The synthetic sample exhibits 13 well-defined TL peaks at T_{max} = 90,108, 124, 148,161, 172, 192, 212, 231, 257, 288, 322 and 360°C (as measured with a heating rate 4°C.s^{-1}). The initial rise method provides a first estimate of the activation energies and the corresponding energies are determined by GCD analysis of 24 glow curves, as described elsewhere in these proceedings[6]. The activation energies found are E = 0.85, 1.00, 1.10, 1.10, 1.20, 1.22, 1.20, 1.20, 1.20, 1.25, 1.30, 1.33, 1.40 eV.

In the geological quartz sample the TL peaks at 108°C and 124°C are much smaller than in the synthetic sample, and the dominant TL peaks are located at 90, 147 and 254°C. Apart from the size of these two smaller peaks, the glow curves of synthetic and geological samples are very similar. The results indicate that the number and location of the TL peaks are the same for the synthetic and geological quartz of the Arkansas variety, at least within the accuracy of the T_m-T_{stop} data.

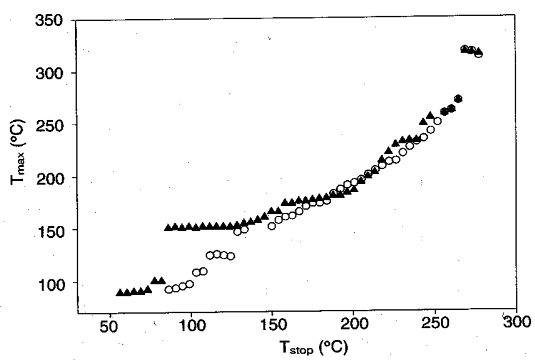

Figure 1. The T_m-T_{stop} method yields several distinct TL peaks for (○) synthetic quartz and (▲) Arkansas quartz.

GCD results

A GCD analysis was performed for the 110°C peak of all 13 samples studied. Figure 2(a) shows the distribution of the activation energy E and Figure 2(b) shows the similar distribution for the temperature of maximum TL intensity T_{max}. The horizontal dashed lines indicate the 67% confidence intervals for the data. It is concluded that both E and T_{max} lie within narrow ranges given by E = 0.86 ± 0.12 eV and T_{max} = 82.1 ± 3.1°C. By using these values of E and T_{max}, the frequency factor is found to be s = (2.5 ± 0.8) × 10^{11} s^{-1}.

Sensitisation results

The TL glow curves for four samples, namely the Arkansas and synthetic quartz annealed at 500 and

Table 1. Summary of TL characteristics of quartz samples of various origins

Index no	Quartz variety	Apparent colouration	Apparent major TL peaks (β = 2°C.s^{-1})
Group 1: Crystalline quartz			
1	Arkansas (Source: Carolina Scientific)	Clear	90, 160, 230, 260, 320
2	Pink variety (Source: Carolina Scientific)	Pink	90, 180, 330
3	Norwegian	Clear	90, 130, 160, 200, 220, 320
4	Synthetic Q-quartz (Source: Sawyers Co.)	Clear	90, 130, 180, 240, 320
Group 2: Quartz from sea sand			
5	Sea sand	Milky white	90, 130, 170, 200, 280, 320
6	Sea sand	Grey	90, 130, 170, 200, 270, 320
7	Sea sand	Milky white	90, 130, 180, 220, 280, 330, 370
8	Sea sand	Red	90, 130, 180, 200, 280, 330, 360
9	Sea sand	Light pink	90, 160, 200, 320, 360
10	Sea sand	Milky brown	90, 150, 170, 230, 280, 350
11	Sea sand	Milky red	90, 130, 180, 220, 270, 320, 350
12	Sea sand	Milky white	90, 140, 170, 220, 320
13	Sea sand	Brown	90, 120, 160, 200, 260, 320, 370

900°C, were studied in detail using standard GCD analysis[7]. The TL glow curves for these four samples were fitted using the same set of kinetic parameters obtained from the T_m-T_{stop} method. The only adjustments necessary for a successful fit were the shifting of the '110°C' TL peak to lower temperatures as the annealing temperature is increased. This shifting in temperature has been reported previously by several researchers and has been explained using theoretical models (Reference 8 and references therein). The successful fitting of these four glow curves indicates that the annealing process most likely does not change the kinetic parameters E and s, but rather changes the number of available recombination centres.

Figure 3 shows the results of studying the sensitisation effect in the quartz samples extracted from sea sand. The samples underwent the following treatment. The existing TL was not erased by a measurement like the zero dose reading. Instead, the test dose was given directly on the as-received samples in order to see the clear response of the '110°C' TL peak. Next two successive cycles of irradiation and readout were performed in order to see the variation of the '110° C' TL peak. The test doses administered were in the range of 0.05–0.3 Gy.

Figure 3 shows the ratio of the TL peak heights for two successive irradiation/heating cycles for both annealed and unannealed samples. It can be seen that the sensitisation factor lies within a very narrow range of values of 1.22 ± 0.24.

The plateau tests

The well-known plateau test[9] was applied to all samples and it was found that the quartz samples extracted from sand (Group II) displayed clear plateaus at distinct temperatures of 90, 148, 172, 231 and 322°C. The quartz samples of Group 1 showed clear plateaus for the '110° C' TL peak, while less distinct plateaus were found for the rest of the TL peaks.

DISCUSSION AND CONCLUSIONS

From the results presented in this paper some very general conclusions can be drawn as follows:

(a) The '110°C' TL peak exhibits a universality in its properties, independent of the origin of the quartz samples. These properties are the location of maximum TL intensity T_{max}, the activation energy E and frequency factor s, the kinetic order and the sensitisation factor to small doses.

(b) The nine random samples of quartz extracted from sea sand exhibit a consistent common behaviour in their TL properties, within very narrow statistical limits.

(c) The results presented here demonstrate that studies of the basic TL mechanism (at least for the '110°C' TL peak) in quartz could be carried out in such samples too, and not only on the commonly used samples of sedimentary or synthetic origin.

(d) As a result of the above observations, it can be concluded that TL studies of quartz samples extracted from sea sand could prove very useful in both dating and in retrospective dosimetry.

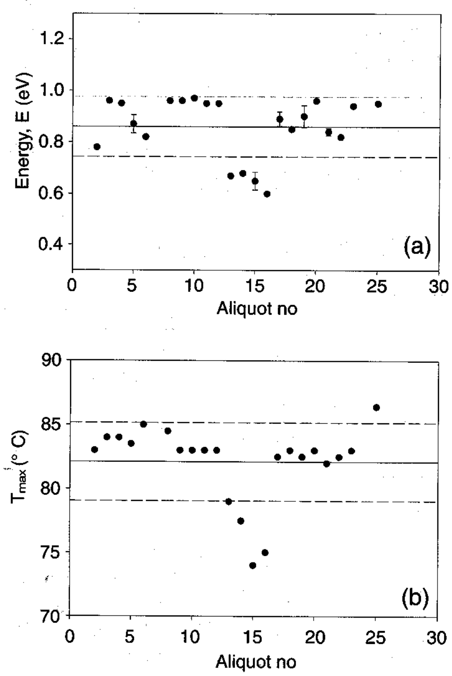

Figure 2. Distribution of (a) the activation energy E obtained using GCD analysis and (b) the temperature of maximum TL intensity T_{max} for unannealed samples. The dashed lines indicate the 67% confidence intervals.

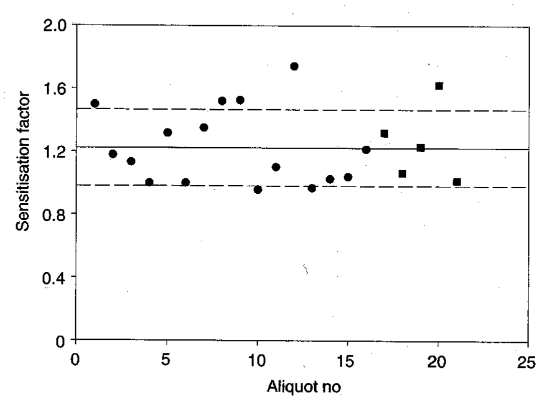

Figure 3. Distribution of the sensitisation factor following two successive cycles of irradiation and heating. Both annealed (■) and unannealed (●) samples are shown.

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