Study of the 110°C TL peak sensitivity in optical dating of quartz

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Abstract

As the 110°C TL emission in quartz uses the same luminescence centers as the OSL emission, the 110°C TL signal from a test dose may be used to monitor the OSL sensitivity change. It is thus important to study the relationship between the 110°C TL peak and the OSL sensitivity in studies related to optical dating from quartz. We have conducted a series of experiments using sedimentary quartz, where the annealing temperatures were varied between 260 and 1000°C before the measurement of OSL and 110°C TL sensitivities. Another series of experiments on two sedimentary quartz samples investigated the 110°C TL peak and OSL dose-dependent sensitivity change after different annealing temperatures. In these experiments, the 110°C TL and OSL signals from the test dose are shown to have similar sensitization characteristics: the 110°C TL sensitivity change is proportional to the OSL sensitivity change if the annealing temperature is lower than 500°C. It is concluded that the 110°C TL signal can be used to correct the OSL sensitivity change in the single- aliquot additive-dose protocol. © 2000 Elsevier Science Ltd. All rights reserved.

1. Introduction

Since the optical dating of quartz was first proposed (Huntley et al., 1985), many papers have been published on dating applications. Two basic methods, one using regeneration-doses and one using additive-doses have been employed, using both multiple- and single- aliquots. Duller (1991) first developed a viable method for the measurement of the equivalent dose using a single aliquot. He concluded that the additive method was more reliable than the regeneration method because of the possibility of sensitivity changes. Murray and Roberts (1998) later developed a single-aliquot regenerative-dose (SAR) protocol for OSL dating which used the response of the 110°C TL peak in quartz to correct for sensitivity changes in the OSL signal. Murray and Wintle (2000) described improvements to this procedure, which used the OSL signal from a test dose to replace the 110°C TL signal used by Murray and Roberts (1998). These two single-aliquot methods (additive and regenerative) are now widely employed in OSL dating.

Some form of heating of the sample (preheating) is always used before OSL measurement in quartz optical dating, and in this process the OSL sensitivity (light output per unit dose) of the quartz is changed. Aitken and Smith (1988) found that such OSL sensitivity change is paralleled by changes in the sensitivity of the 110°C TL peak and suggested that there might be a
common mechanism. Since then, the relationship between 110°C TL peak and OSL sensitivity has been studied by many authors (Stoneham and Stokes, 1991; Murray and Roberts, 1998; Wintle and Murray, 1998; Chen and Li, 2000). Franklin et al. (1995) used spectral measurements to argue that the 110°C TL emission used the same luminescence centers as the 325°C TL emission, and thus, the same as the OSL emission. Several workers have suggested that the OSL sensitivity change could be corrected using the 110°C TL peak sensitivity (e.g. using a small test dose given after each OSL measurement). Murray and Roberts (1998) demonstrated how this could be done in practice in a regenerative procedure. In the later form of the single-aliquot regenerative-dose protocol (Murray and Wintle, 2000), the sensitivity change in the OSL signal is measured directly by the OSL response to a subsequent test dose. But when using the single-aliquot additive-dose protocol, the OSL signal is not emptied during each measurement cycle, and the OSL test dose response cannot be used. However, the sensitivity of the 110°C TL peak can be applied to correct the OSL sensitivity change induced by the additive dose and preheat procedure. It is the potential of this correction procedure that has led to this study.

Bøtter-Jensen et al. (1995) have studied the OSL and photo-transferred TL (110°C peak) sensitivity changes with annealing temperature. In the present paper, we examine the relationship between the 110°C TL response and the OSL signal for different annealing temperatures and laboratory dosing. Our data are then discussed in terms of the deduced changes in hole population and numbers of hole centers.

2. Samples and analytical facilities

Two quartz sedimentary samples were used in this study. They are 970201 and WIDG8. Quartz (90–120 μm) was extracted from 970201, a marine sediment with an equivalent dose of about 54 Gy from Skørbok in north Denmark. Quartz (90–120 μm) was extracted (by R. G. Roberts) from WIDG8, an apron sediment about 30 ka old from Widgingarri in the Kimberley region of the northern part of Western Australia. Its OSL and TL behavior have been extensively studied in earlier papers (Wintle and Murray, 2000). The purity of the quartz grains was confirmed by the absence of infrared sensitivity to a regeneration dose (Aitken, 1998). All samples were mounted on 9.0 mm diameter nickel cups or on 9.8 mm diameter stainless-steel discs with silicone oil (Silkospray).

All experiments were undertaken using two automated Risø TL/OSL readers equipped with broadband green light (420–550 nm) or high-power blue diodes (470 ± 30 nm; Bøtter-Jensen, 1997) respectively, and U-340 detection filters. Software-controlled beta irradiators equipped with a 90Sr/90Y sources delivering 0.090 Gy/s and 0.232 Gy/s were attached.

Two heating ovens were used to anneal the samples at the selected temperature: a VULCAN oven (maximum temperature 1100°C) and a Carbolite oven (maximum temperature 1000°C).

3. Dependence of 110°C TL peak and OSL sensitivity on annealing temperature

This section examines the effect of different initial thermal treatments on the way in which the 110°C TL and OSL sensitivities change with heating.

3.1. Sample pretreatment

One sedimentary quartz sample, 970201, was used for the annealing experiment. Four groups of samples were prepared (each of three aliquots): (i) natural, and groups annealed at (ii) 500; (iii) 800 and (iv) 1000°C. All annealing was for 10 min at the selected temperature. The annealed samples were then given a labora-

Fig. 1. Variation of test dose response for four initial annealing conditions (natural, 500, 800 and 1000°C) using sample 970201. (a1–a4) The 110°C TL peak and OSL sensitivity change with annealing temperature. (b1–b4) The ratio of OSL sensitivity to 110°C TL sensitivity change with annealing temperature.
tory dose of 54 Gy, equal to the equivalent dose of the sample. A 100 s green light exposure was then applied to all samples at 160°C to eliminate the 110°C TL and OSL signal produced by the natural dose and the laboratory dose of 54 Gy.

3.2. Experimental sequence

Each of the pretreated aliquots was then subjected to multiple cycles of OSL/TL sensitivity measurement and thermal treatment. Ten annealing temperatures were chosen for this experiment between 160 and 1000°C. The annealing time was 10 min. After each annealing treatment, the same experimental sequence was applied to all the aliquots — A test dose of 0.9 Gy was given, and the aliquot was heated to 160°C at 5°C/s, during which the 110°C TL peak sensitivity to the test dose was measured. The OSL sensitivity to the test dose was then measured for 100 s of green light stimulation (at 160°C to prevent re-trapping of charges in the traps corresponding to the 110°C TL peak). This cycle of test dose for 110°C TL and OSL measurement was repeated three times on each aliquot before annealing at the next annealing temperature and repeating the measurement sequence until completing the 1000°C cycle.

3.3. Results

The 110°C TL and OSL sensitivity changes are plotted as a function of pre-annealing temperature in Fig. 1(a1)-(a4), and the ratios of OSL sensitivity to 110°C TL sensitivity in Fig. 1(b1)-(b4). The 110°C TL sensitivity change with annealing temperature is broadly similar to the OSL sensitivity change in both the natural and annealed. However, the ratio of the OSL sensitivity to 110°C TL sensitivity dose change with annealing temperature, especially in the cases of natural and 500°C annealed samples. The curve of OSL sensitivity change with the annealing temperature in Fig. 1(a1) is very similar to that shown by Bøtter-Jensen et al. (1995). The absolute sensitivity changes for the sample annealed at 500°C show similar trends to those of the natural sample (Fig. 1(a1) and (a2). In contrast, the curves from 800 and 1000°C annealed aliquots show a completely different trend. At the lower annealing temperature (natural and 500°C), the OSL increases more than the 110°C TL sensitivity. At the two higher annealing temperatures (800 and 1000°C), the opposite is true.

4. Dependence of 110°C TL peak and OSL sensitivity on prior laboratory dose

Six discs of sample 970201 were annealed at 800°C for 10 min. After cooling to room temperature, they were given different laboratory doses of 0, 5, 10, 20, 40 and 80 Gy. All aliquots were then exposed to green light for 100 s at 160°C to reset the 110°C TL and OSL signal to zero. Ten annealing temperatures were used in this annealing experiment between 160 and 1000°C; each disc was successively annealed at each of the temperatures for 10 min. After each annealing, the aliquots were measured using the same experimental sequence as in Section 3.2, and the results are shown in Fig. 2 (1–6). The changes in the 110°C TL and OSL sensitivity with annealing temperature are almost completely independent of dose, at least over the dose range examined here. The OSL to 110°C TL ratio is mainly less than 1 in all aliquots, as was seen earlier in the aliquots heated to 800 and 1000°C (Fig. 1(a3), (a4), (b3), (b4)).

5. Dependence of 110°C TL peak and OSL sensitivity on regeneration dose

Two samples, 970201 and WIDG8, were used in this experiment, in which the relationship of 110°C TL peak and OSL sensitivity was examined as a function of regeneration dose. The experimental sequence is based on the single-aliquot regenerative-dose protocol.
and WIDG8 after different prior treatments (natural, 500 and 900°C). Each aliquot was given a regeneration dose and preheated at 260°C for 10 s. After OSL measurement of the regeneration dose at 125°C, the sample disc was given a 0.93 Gy test dose. The 110°C TL peak and OSL resulting from this test dose were then measured. This sequence was then repeated using the next regeneration dose. The regeneration doses were selected as 0, 23, 37, 60, 98, 160, 260, 410 and 670 Gy. The OSL measurement of regeneration dose and test dose was carried out by exposing the sample for 40 s to blue light at 125°C. All OSL measurements reduced the OSL count rate to a negligible fraction of its original value.

The results are shown in Fig. 3 (1) (970201) and (2) (WIDG8). From Fig. 3(1b) and (2b) the ratios of OSL sensitivity to 110°C TL sensitivity for the natural and 500°C annealed aliquots remain almost constant as a function of regeneration dose (the initial ratios were normalized to 1). This demonstrates that 110°C TL sensitivity change is almost directly proportional to OSL sensitivity change if the annealing temperature is lower than 500°C for these two samples. However, for the aliquots annealed at 900°C, this proportional relationship is destroyed, and the ratio changes by more than a factor of two in both curves.

6. Discussion

In Figs. 1 and 2, we see broadly similar changes in the sensitivities of the 110°C TL and OSL signals from two distinct electron traps with very different stabilities. It is thought that both these traps access the same luminescence centers, and so because of similarities in behavior, we assume that sensitivity changes (Aitken, 1998; Wintle and Murray, 2000) relate primarily to the recombination process, rather than electron trapping.

However, these sensitivity changes, although similar, are not identical. In Fig. 1 they vary in ratio by several times, depending on initial annealing temperature. From Figs. 1 and 3, if the sample is annealed at higher temperatures (above about 600°C) either before a regenerative sequence (Fig. 3) or during such a sequence (Fig. 1), the relationship becomes particularly variable. Zimmerman (1971) introduced the concept of the sensitivity of the 110°C TL peak being controlled by hole transfer between non-radiative Reservoir (R) centers and Luminescence (L) centers. Her model was developed for archaeological (heated) quartz and low annealing temperatures (up to 500°C). If we accept that the same luminescence centers are used by both the 110°C TL and OSL traps, then we must presume that the ratio of these L centers to non-radiative (R) centers is different for the two traps. This in turn implies two different recombination pathways i.e., electrons released from the 110°C TL trap are not indistinguishable from those released from the OSL trap (the 325°C TL trap). It is already known that a true delocalised conduction band cannot exist in quartz (the 375°C TL peak uses a different center from the 325°C OSL/TL trap). Based on Zimmerman’s original model, Chen and Li (2000) have proposed the existence of multiple levels of holes (R centers). Wintle and Murray (1997) and Murray and Roberts (1998) have also deduced that the 110°C TL trap and the 325°C OSL/TL trap must access different suites of R centers. We are forced to conclude that at least some of the charge derived from these two traps cannot recombine via the conduction band.

The mechanism for sensitivity change remains to be discussed. Chen et al. (2001) suggest that either R centers are preferentially destroyed (with respect to L centers) by heating to above 700°C, or L centers are created. Below this temperature sensitivity, changes are probably related to the differential filling and emptying of L and R centers such as that postulated by Zimmerman (1971). From the work described here, we conclude that it is more likely that at high temperatures R centers are destroyed, because the 110°C TL and OSL sensitivity changes are not proportional. Since the 110°C TL sensitivity increases most, we deduce that
the R centers preferentially accessed by the electrons from the 110°C trap are preferentially destroyed.

We conclude that if the 110°C TL trap is to be used to monitor sensitivity changes in the OSL trap, samples should not be heated above 600°C. At temperatures below this, differential filling and emptying of R and L centers appears to be the dominant process giving rise to sensitivity change. Even at these lower temperatures, however, the changes are not always directly proportion (Figs. 2 and 3, and Murray and Roberts, 1998), probably because different R centers are available to electrons, depending on their source.

7. Conclusion

We have shown that the ratio of OSL to 110°C TL response to a test dose changes significantly with the annealing temperature. We deduce that electrons related from the 110°C TL and OSL traps probably use the same type of L center, and different types of R centers. It is concluded that low annealing temperatures (below 500°C) do not greatly influence the relative hole population in the different R centers, in contrast to high annealing temperatures. High temperatures appear to preferentially destroy those R centers available to the 110°C TL trap, increasing its relative sensitivity. However, the observation that low temperature annealing does not significantly affect the relationship may indicate that the 110°C TL signal can be used to correct sensitivity changes in the single-aliquot additive-dose protocol.

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