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Assessing the impact of pulsed-irradiation procedures on the thermally transferred OSL signal in quartz

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HIGHLIGHTS

- The difference between constant- and pulsed-irradiation TT-OSL protocols is studied.
- The extent to which the TT-OSL signal is annealed by heat treatments is assessed.
- Partial annealing accounts for the difference in outcome observed between protocols.
- Pulsed-irradiation may not be appropriate for dating using the TT-OSL signal.
- Future studies using pulsed-irradiation should test for partial annealing.

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ABSTRACT

Thermally transferred optically stimulated luminescence (TT-OSL) dating protocols have been suggested as a means of extending the age range of luminescence dating. Several studies demonstrate that TT-OSL signals increase with large radiation doses (>2000 Gy) and yet, few studies report older TT-OSL ages (>400 Gy) in agreement with independent absolute age control. In one such study, agreement with independent chronology was only achieved for the old samples by implementing a pulsed-irradiation procedure. Pulsed-irradiation is suggested to compensate for dose rate dependent competition effects by dividing the laboratory irradiation into discrete irradiation steps interspersed with heat treatments. However, every inter-step heat treatment has the potential to anneal part of the TT-OSL dating signal. This study compares constant- and pulsed-irradiation TT-OSL protocols and investigates the degree of partial thermal annealing. The results suggest that almost all of the difference in outcome between constant- and pulsed-irradiation protocols can be explained by partial annealing of the TT-OSL signal rather than by competition effects. Partial annealing distorts the laboratory dose response curve but has no impact on the natural signal, resulting in unreliable equivalent dose estimates. This means that pulsed-irradiation procedures may not be viable for TT-OSL dating measurements. Future studies implementing pulsed-irradiation procedures should carefully consider the extent to which inter-step thermal treatments partially anneal the signal.

1. Introduction

The thermally transferred optically stimulated luminescence signal (TT-OSL) from quartz has been observed to grow to larger laboratory radiation doses than the more commonly used optically stimulated luminescence (OSL) signal (Wang et al., 2006b), thus giving the TT-OSL signal the potential to extend the age range of luminescence dating. The first study to use the TT-OSL signal for dating (Wang et al., 2006a) demonstrated agreement between TT-OSL and OSL equivalent doses (D_e) up to ~400 Gy. However, sample IEE424 from near the Brunhes-Matuyama (B/M) palaeomagnetic reversal obtained a D_e value of 1460 ± 62 Gy resulting in an age estimate of only 474 ± 27 ka (Wang et al., 2006a), far from the age obtained using Ar–Ar dating (775.6 ± 1.9 ka; Coe et al., 2004).

The severe age underestimation of this sample led Wang et al. (2006a) to investigate pulsed-irradiation protocols. Using a protocol that divides laboratory irradiation into ~150 Gy irradiation steps separated by a 240 °C cut heat, Wang et al. (2006a) were able to obtain equivalent doses that resulted in ages agreeing with the B/M age for sample IEE424 and for three other samples taken from near the B/M boundary. Although this suggested that a pulsed-

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irradiation protocol was necessary for the older samples in that study, subsequent investigations and applications of the TT-OSL signal have used only constant-irradiation protocols. In agreement with Wang et al. (2006a), most of these subsequent investigations have been unable to successfully date an older sample (>400 Gy) with independent age control (Duller and Wintle, 2012; Thiel et al., 2012). However, one recent investigation using constant irradiation has reported two older ages (~700 and ~1000 ka) in agreement with independent U–Pb ages and palaeomagnetic reversals (Pickering et al., 2013).

Pulsed-irradiation was initially proposed by Bailey (2004) as a means of compensating for differences between dose rates in nature and in the laboratory. This work was on simulations of modelled OSL data, but there has been limited application to real samples. The assumption that natural and laboratory radiation result in the same trapped charge distribution is fundamental to luminescence dating, but the two radiation sources have known differences. Natural dose rates are typically 1–4 Gy/ka and are comprised of a mixed radiation field including alpha, beta, and gamma radiation, whereas laboratory dose rates are typically of the order of 10^9 Gy/ka and are comprised of a single radiation type (most commonly beta radiation).

Due to the complexity of the charge distribution in quartz grains, the difference in magnitude between natural and laboratory dose rates could result in either age underestimation or overestimation (Qin and Zhou, 2009). For example, if the quartz grain has an abundance of temporally unstable electron traps (such as those associated with the 110 °C thermoluminescence peak), constant- laboratory irradiation could potentially underestimate the natural signal. In nature, these shallow traps capture electrons slowly due to the low dose rates and these electrons are then released due to the long duration of the irradiation and the short lifetime of charge in this trap at room temperature. The result is that these traps are effectively empty such that they can compete for each free electron in the conduction band. During the short, high dose rate laboratory irradiations these shallow traps collect electrons more rapidly than they escape, resulting in a higher equilibrium concentration than in nature and therefore reducing the competition for electrons with the deeper traps used for dating. This results in signals from laboratory doses that are brighter than signals from equivalent natural doses, and therefore results in age underestimation.

The opposite effect can occur if the quartz grain has an abundance of temporally unstable electronic hole traps. In this instance, competition for electrons during recombination would be greater for laboratory irradiation due to a higher concentration of electronic holes in alternative recombination centres, resulting in the natural signal being brighter than an equivalent laboratory signal and therefore leading to age overestimation (Bailey, 2004). Pulsed-irradiation divides the laboratory irradiation dose into smaller irradiation steps and heats the sample between steps. The thermal treatments between steps are designed to deplete the temporally unstable traps and thus make laboratory competition effects in the quartz crystal similar to those which occur due to the low radiation dose rates received in nature.

Although pulsed-irradiation has the potential to address charge competition, an additional, unwanted effect of the protocol may be to partially anneal the target signal, if the thermal treatments are carried out at temperatures which are too high. Every inter-step thermal treatment during the protocol has the potential to remove a certain percentage of the signal used for dating. This additional effect could result in undesirable changes to the age estimate, unless the partial annealing is negligibly small.

This study explores the use of pulsed-irradiation for TT-OSL protocols by observing the difference in outcome between constant- and pulsed-irradiation protocols, and then investigates the relative influence of any partial annealing caused by the heat treatments between irradiation steps.

2. Sample description and instrumentation

Sample PT2, collected from the top of L3 at the Luochuan section of the Chinese Loess Plateau, was used for all of the measurements reported in this paper. This section is the same as that analysed by Wang et al. (2006a). The sample was collected as a small block wrapped in black plastic. Once in the darkroom conditions of the laboratory, the exterior of the block was removed and the remaining material was treated with 10% dilution by volume of concentrated HCl and 20 vols H_2O_2 until no continued reaction could be identified. The material was then settled in sodium oxalate using Stokes Law to obtain the 4–11 μm fraction, treated with H_2SiF_6 for 14 days to remove feldspar (Roberts, 2007), and subsequently re-settled as a further quartz purification step. All of the measurements were undertaken on a single aliquot that was pre-sensitized by numerous cycles of irradiation, heating, and optical stimulation, in order to reduce the trap type variability and minimize sensitivity changes.

Luminescence measurements were performed on a Risø TL-DA-20 reader incorporating blue LEDs emitting at 470 nm and delivering 50 mW/cm^2 (Bøtter-Jensen et al., 2003). The luminescence signal was recorded using an EMI9635QA photomultiplier tube equipped with 7.5 mm of U-340 filter, and a convex quartz lens to improve signal collection efficiency (giving ~75% brighter signal). A strontium/yttrium beta source with a dose rate of 0.083 Gy/s was used for laboratory irradiation, and a preheat of 260 °C for 10 s was applied to all samples before the OSL stimulations to yield the natural (L_n), regenerative (L_x) and test (T_x) dose signals. Integration intervals included the first two channels (2 s) of measurement minus a background of the last ten channels (10 s) of measurement.

3. Advances in TT-OSL measurement protocols

The basic steps of TT-OSL measurement involve an optical stimulation to empty electron traps associated with the fast component of the OSL signal, and a heat treatment to thermally transfer electrons from other less light-sensitive traps into the fast component traps. A second optical stimulation then gives the TT-OSL signal. The original sequence proposed by Wang et al. (2006a) has been thoroughly investigated in later studies, and the measurement protocol underwent several alterations, including the transition from a multiple aliquot protocol to a single aliquot protocol within a year of publication (Wang et al., 2007).

Wang et al. (2006a) described two parts to the TT-OSL signal, the BT-OSL signal and the recuperated OSL (Re-OSL) signal. The Re-OSL signal was the target signal for dating and could be obtained by subtracting the BT-OSL signal from the entire TT-OSL signal. Division of the TT-OSL signal into two parts was based on the theoretical understanding that Re-OSL originated from a double transfer process while BT-OSL originated from a single transfer process. In a double transfer process, a proportion of the electrons in the traps associated with the OSL fast component are temporarily stored in alternative traps after eviction by the initial blue light stimulation; the subsequent thermal transfer preheat then transfers these electrons from the alternative trap back into the OSL fast component traps where they can then be measured during the second light stimulation. In a single transfer process, the thermal transfer preheat accesses electrons in alternative traps that were not originally stored in the OSL fast component traps and transfers them into those traps.