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Quartz luminescence response to a mixed alpha-beta field: Investigations on Romanian loess

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HIGHLIGHTS

- Prior alpha irradiation history does not influence the laboratory beta growth curves.
- Alpha and beta induced photon emissions in quartz follow the same recombination path.
- Laboratory alpha and beta growth curves overlap up to total alpha doses of ~1250 Gy.
- Mixed alpha-beta growth curves reproduce the beta dose response curves up to ~800 Gy.
- Mixed radiation field is not causing the deviation of natural OSL signal growth curves from laboratory growth curves.

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ABSTRACT

Previous SAR-OSL dating studies using quartz extracted from Romanian and Serbian loess samples report SAR-OSL dose–response curves on fine grained (4–11 μm) quartz that grow to much higher doses compared to those of coarse-grained (63–90, 90–125, 125–180 μm) quartz. Furthermore, quartz SAR-OSL laboratory dose response curves do not reflect the growth of the OSL signal in nature. A main difference in coarse- and fine-grained quartz dating lies in the alpha irradiation history, but the effect of mixed alpha-beta fields has so far received little attention. In the present study we investigate whether the alpha dose experienced by fine grains over geological cycles of irradiation and bleaching may have an effect on the saturation characteristics of the laboratory dose response. By applying time resolved optically stimulated luminescence we confirm that the OSL signals induced in quartz by alpha and beta radiation follow the same recombination path. We also show that a mixed alpha-beta dose response reproduces the beta dose response only up to about 800 Gy. Assuming an a-value of 0.04 we have shown that laboratory alpha and beta dose response curves overlap up to effective alpha doses of ~50 Gy. Based on these results, we conclude that exposure of fine grains to alpha radiation during burial and transport cycles prior to deposition, as well exposure to the mixed radiation field experienced during burial are not responsible for the age discrepancies previously reported on fine and coarse grained quartz extracted from Romanian and Serbian loess.

1. Introduction

Heavy charged particles such as alpha particles (helium ions) deposit a large amount of energy per unit track length when interacting with matter and cause dense ionization and, in the case of luminescence dosimeters, local charge saturation effects in the vicinity of their tracks. Because of these saturation effects, the OSL output per unit dose from alpha particles is lower than that from high-energy electrons (beta irradiation) or photons (gamma irradiation or X rays). This difference in OSL response is quantified by the alpha efficiency parameter relative to beta irradiation (a-value). The lower luminescence response to alpha irradiation with respect

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to that of beta and gamma radiation in quartz, both in the case of TL and OSL has been known for a long time (Zimmerman, 1972; Aitken and Bowman, 1975). This effect becomes especially important in ‘fine-grain’ dating, because external alpha particles can irradiate the entire volume of the grain. Although, there has been an increasing focus on dating fine grained quartz in the last decade (e.g. Lowick et al. (2010); Sugisaki et al. (2010); Timar et al. (2010)) using the single aliquot regenerative dose protocol (Murray and Wintle, 2000), the subject of alpha efficiency has received little attention (Tribolo et al., 2001; Galloway, 2002; Burbidge et al., 2009; Polymeris et al., 2011). Reported estimates of alpha efficiency for fine grained quartz extracts is known to be around 0.1 in the case of thermoluminescence (Zimmerman (1972); Aitken (1985 appendix K)), while in the case of OSL reported values vary between 0.03 and 0.04 (Rees-Jones, 1995; Mauz et al., 2006; Lai et al., 2008).

The range of alpha particles is ~20 μm in quartz (Aitken, 1985). Thus, they deposit energy in the entire volume of fine (4–11 μm) grains, while only in an outer shell in coarse (63–90 μm) grains. In the latter case the alpha-irradiated shell is etched away before measurement. The question we ask here is whether this difference in alpha affected volume can explain the differences in the OSL behaviour of the two size fractions from the loess in the Lower-as well as the Middle Carpathian-Danube basin (Timar-Gabor et al., 2011, 2012; Timar-Gabor and Wintle, 2013; Constantin et al., 2014a). These differences in OSL behaviour are observed in two ways: 1) on the one hand as observed in the Romanian loess, the laboratory dose response curves of fine quartz (D_0 of ~175 Gy and ~1800 Gy) saturate much later than the etched coarse quartz (D_0 of ~55 Gy and ~600 Gy), (see Timar-Gabor et al. (2012)); this behaviour has also been observed in other sites (e.g. Kreutzer et al. (2012); (Lomax et al., 2014; Timar-Gabor et al., 2014). 2) there is a general disagreement between fine and coarse quartz ages (Constantin et al., 2014a,b; Timar-Gabor et al., 2011, 2012) which is likely to be due to the difference between the naturally and laboratory generated dose response curves (Timar-Gabor et al., 2011, 2012; Timar-Gabor and Wintle, 2013; Constantin et al., 2014a, 2014b). The divergence observed between the natural and the laboratory dose responses is observed for both grain fractions but is more pronounced in the case of fine grains (see Timar-Gabor and Wintle (2013), Fig. 6 and Fig. S1a).

Assuming that the quartz has the same origin and physical characteristics irrespective of grain size, an important dosimetric difference between natural and laboratory irradiations lies in the dose rate and radiation field in the sediment matrix. The dose rate is ~11 orders of magnitude lower in nature compared to that in the laboratory. Similarly natural irradiations occur in a mixed radiation field (alpha, beta, gamma, x-rays) while laboratory irradiations usually consist entirely of either beta particles or photons (gamma or X rays). These differences may be reflected in calibration of luminescence response to dose and may arise from the different interaction mechanisms of alphas and betas, and the possibility of additional defect creation due to alphas.

In the present study we compare the dose response curves and sensitivity changes measured using continuous wave OSL (CW-OSL), under different combinations of alpha and beta irradiations designed to simulate the effect of a natural radiation field. We use time-resolved optically stimulated luminescence (TR-OSL) as a tool to gain insights into recombination pathways leading to the emissions generated by the two different types of radiations. These results are reported below.

2. Samples and measurement facilities

Analyses have been performed on 4–11 μm and 63–90 μm quartz grains of sample CST 3 (Timar-Gabor and Wintle, 2013; Constantin et al., 2014a). Additional measurements have been carried out on 4–11 μm quartz from sample LCA 26 (Constantin et al., 2014b). Relevant information on the samples is presented in Table S1. All investigations have been performed using large (9-mm diameter) aliquots that have been tested for purity by OSL IR depletion ratio (Duller, 2003; Wintle and Murray, 2006). The natural OSL signal was bleached by exposure to blue LEDs for 200 s at 20 °C prior to experiencing two cycles of regenerative beta dose of 350 Gy (preheat at 220 °C for 10 s), test dose of 17 Gy (cutheat 180 °C) and elevated temperature bleach (280 °C for 40 s). In the second cycle, an IR diode exposure for 100 s at 125 °C preceded reading of the OSL signal induced by the regenerative dose.

Experiments have been carried out at DTU Nutech (DTU Risø Campus, Denmark) on a standard TL/OSL-DA-20 reader (Thomsen et al., 2006) using an in-built $^{90}Sr/^{90}Y$ beta source (dose rate of 0.172 Gy s^{-1} to 4–11 μm grains deposited on aluminium disks and 0.219 Gy s^{-1} to 180–225 μm quartz grains mounted on stainless steel disks) and an uncalibrated ^{241}Am source under vacuum conditions. The OSL signal induced by 5000 s of alpha irradiation was found to be 2.22 times higher than that resulting from ~8.5 Gy of beta irradiation, giving an effective alpha dose rate of 0.095 Gy s^{-1}. This can be converted to a tentative absolute alpha dose rate of 2.370 Gy s^{-1} assuming an *a*-value of 0.04 for fine grained quartz (Rees-Jones, 1995).

All investigations into the optical emission characteristics of fine (4–11 μm) and coarse (63–90 μm) quartz used the SAR protocol (Murray and Wintle, 2003). The luminescence signals induced by alpha, beta and mixed alpha-beta radiation were stimulated with blue LEDs in continuous-wave (CW) mode for 40 s at 125 °C and the net CW-OSL signal was determined from the initial 0.32 s of the decay curve, less a background integrated between 1.76 s and 2.40 s (Ballarini et al., 2007; Cunningham and Wallinga, 2010). For the sake of consistency with previously reported data (Timar-Gabor et al., 2011, 2012; Timar-Gabor and Wintle, 2013) a preheat at 220 °C for 10 s and a cutheat to 180 °C was used, together with stimulation at 280 °C for 40 s at the end of each SAR cycle. Sensitivity changes induced by alpha and beta irradiation were monitored using the response to a constant 17 Gy beta test dose.

Time-resolved optically stimulated luminescence (TR-OSL) was measured on a Risø TL/OSL-DA-20 reader equipped with an integrated pulsing option to control the stimulation LEDs and a photon timer attachment with a detection resolution (bin-width) of 100 ps (Lapp et al., 2009). TR-OSL experiments have been carried out using the same parameters of the SAR protocol as in the CW-OSL measurements. The luminescence signals were stimulated and recorded at 125 °C for 100 s using pulsed optical stimulation (POSL). Each pulse consisted of an *on*-time (the duration of each LED stimulation pulse) of 50 μs and an *off*-time time (the following period when the LEDs are not illuminated) of 450 μs. The photon counter was set to count photons during both *on* and *off-time*. The dose rates for beta irradiation on this particular reader were 0.099 Gy s^{-1} to 4–11 μm quartz and 0.113 Gy s^{-1} to 180–225 μm quartz.

3. Results and discussions

3.1. The effect of alpha irradiation history on the beta dose–response curve

One major difference when applying OSL dating to the fine and coarse quartz extracts is the fact that for the coarse fraction the outer alpha irradiated rim is etched away using HF. Thus, in the case of coarse quartz the material used for dating has not experienced alpha irradiation. Due to the different interaction mechanisms of alpha and beta particles with matter, the multiple cycles of irradiation and light exposure in the geological past of the fine grains and