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The relationship between thermal activation energy, infrared stimulated luminescence and anomalous fading of K-feldspars

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ABSTRACT

A strong dependence of thermal activation energy (TAE) on infrared (IR) stimulation time for the infrared stimulated luminescence (IRSL) signal was observed for K-feldspar grains extracted from several sediments and granites from China. A TAE value as low as ~0.1 eV was observed at the beginning of IR stimulation and increased to ~0.45 eV after 90 s. For a trap depth of ~2 eV below the conduction band for the IRSL traps, the TAE value of ~0.45 eV is consistent with the energy gap between the excited states (~0.5 eV below the conduction band) and conduction band. This phenomenon is explained as the result of the coexistence of thermally assisted recombination via conduction band or band-tail states hopping and athermal tunnelling recombination of electrons from the excited states under IR stimulation, leading to the observation of a higher anomalous fading rate in the initial part of the IRSL decay curve.

1. Introduction

The infrared stimulated luminescence (IRSL) from sedimentary feldspar has been used for optical dating of sediment for the last two decades since the first report of optical stimulation spectra of feldspar by Hütt et al. (1988). However, the application of optical dating using feldspar has been hampered due to anomalous fading effect, i.e. the reduction in IRSL signals from feldspar with storage at room temperature (Wintle, 1973; Spooner, 1992, 1994; Huntley and Lamothe, 2001; Huntley and Lian, 2006). The anomalous fading has been explained as the result of direct recombination of electron-hole pairs via tunnelling (Aitken, 1985; Visocekas, 1985; Visocekas et al., 1994). There have been many attempts to overcome the anomalous fading problem, e.g. making fading correction (Lamothe and Auclair, 1999; Huntley and Lamothe, 2001) and searching for stable (non-fading) signals (Tsukamoto et al., 2006; Li et al., 2008) or less fading signals (Thomsen et al., 2008).

It has been shown that the IRSL traps are associated with deep traps with a thermal depth of ~1.7 eV (Li and Tso, 1997) and an optical depth of ~2 eV below the conduction band (Hütt et al., 1988; Poolton et al., 2002a,b; Baril and Huntley, 2003). Therefore, it is impossible that the infrared (IR) photons (~1.4 eV) have enough energy to excite the trapped electrons into the conduction band and then recombine with holes to produce luminescence.

To explain the luminescence under IR stimulation, several possible recombination processes were considered (Fig. 1), i.e. (a) thermal assisted recombination from the excited state through the conduction band (Hütt et al., 1988) (Fig. 1a); (b) direct electron-hole tunnelling recombination from the excited state (Poolton et al., 2002b) (Fig. 1b); (c) band-tail states hopping from the excited state followed by tunnelling to a recombination centre (Poolton et al., 2002a) (Fig. 1c). The process of tunnelling recombination (Fig. 1b) requires no thermal assistance and thus is thermally independent, while the other two processes (Fig. 1a and c) are thermally dependent but in a different extent. Therefore, these processes can be marked by a different thermal dependency (thermal activation energy) of IRSL signal. In the model of Fig. 1a (Hütt et al., 1988), the electrons trapped in the defects in crystal lattice are ejected to an excited state by IR photons (~1.4 eV) and subsequently to the conduction band by thermal relaxation (or thermal assistance) (Fig. 1a). However, the thermal activation energy (TAE) measured for IRSL from feldspar is generally less than 0.2 eV (Bailiff and Poolton, 1991; Bailiff and Barnett, 1994; Clark and Sanderson, 1994; Rieser et al., 1997; Meisl and Huntley, 2005), which is far smaller than the expected energy gap between the excited state and the conduction band (e.g. 0.5 eV suggested by Poolton et al., 2002a,b). Alternatively, Poolton et al. (2002a,b) proposed an improved model for IRSL of feldspar, in which the luminescence can be a competitive process between direct electron-hole tunnelling recombination and non-tunnelling recombination through thermal hopping among the band-tail states (Fig. 1b and c). These models can explain why the thermal activation energy

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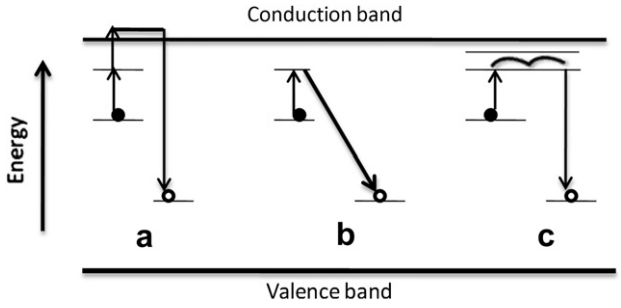

Fig. 1. Simplified bad-gap diagram (after Bøtter-Jensen et al., 2003) showing the processes involving an electron trap (filled circle) and a hole trap (open circle) that may give rise to IRSL from feldspar. (a) Thermal assistance: IR photons raise the electron to an excited state. The electron is then evicted into conduction band by thermal assistance (Hütt et al., 1988); (b) Direct recombination (tunnelling) from the excited state (Poolton et al., 2002b). (c) Band-tail states hopping (Poolton et al., 2002a).

measured for IRSL is far smaller than expected for a thermal assistance process (Fig. 1a) (Hütt et al., 1988) because the tunnelling process has little thermal assistance effect. Based on their model, Poolton et al. (2002a) suggested that the extent of anomalous fading can be predicted by measuring the thermal activation energy, although this was subsequently shown to be not applicable (Meisl and Huntley, 2005). In a recent study, Thomsen et al. (2008) observed a lower fading rate for the later part of IRSL signal when compared with that from the initial part. This led them to conclude that the initial part of IRSL signal is mainly from tunnelling between spatially close donor–acceptor pairs while the latter part are results of tunnelling between distant donor–acceptor pairs and non-tunnelling hopping among band-tail states. However, previous studies of the luminescence characteristics of IRSL from feldspar were mainly concentrated on the initial part of the signal, which might lead to incomplete consideration and inappropriate explanations for the process of IRSL from feldspar. In this study we investigated the time-dependent properties of the IRSL signal from K-feldspar grains extracted from sediments.

2. Samples and analytical facilities

Five Aeolian sedimentary samples (Sm1, Sm7, SY3, HLD3 and WG3), and three granites (CKD, JK3 and ZGC) from different regions of China were used. The sedimentary samples Sm1 and Sm7 were taken from Shimao section at the transition zone between the Mu Us Desert and the Loess plateau in central China. The detailed description of the stratigraphy of the section has been given by Sun et al. (1999). The age of Sm1 (~10 ka) has been determined using OSL dating of quartz and isochron IRSL (iIRSL) dating of K-feldspar (Li et al., 2008). The age of Sm7 (~440 ka) was estimated by correlation of the stratigraphy and OIS stages (Li and Li, 2008). Previous determinations of the equivalent dose for the samples from the section suggest that the IRSL signal for sample Sm7 has reached an equilibrium state between electron filling and escaping (or fading) (Li and Li, 2008), referred to as "field saturation" (Lamothe et al., 2003; Huntley and Lian, 2006). The sand samples WG3 and HLD3 are both from the Hulun Buir Desert and SY3 is from the Hunshandake Desert (Li et al., 2002; Li and Sun, 2006) with quartz OSL ages of 10–13 ka. Granite samples CKD, JK3 and ZGC are from Xinjiang, Tibet and Northern China, respectively, and were crystallised in the Paleozoic, Neogene and Precambrian, respectively.

The sedimentary samples were treated with HCl and H_2O_2 to remove carbonate and organic matter in subdued red safe-light conditions. After drying, 150–180 μm grains were obtained by sieving. The granites were first ground into sand sizes and then sieved to obtain 90–150 μm grains. The K-feldspar grains from all samples were separated using heavy liquids. The extracted K-feldspar grains were cleaned using 10% HF for 5 min. Aliquots containing several hundred grains were prepared by mounting the grains in a monolayer, on a 9.8 mm diameter aluminum disc coated with "Silkospay" silicone oil. The K-feldspar IRSL measurements were made using an automated Risø TL-DA-20 reader equipped with IR diodes (870Δ40 nm) for stimulation with a total power at the sample position of ~40 mW/cm^2 (Bøtter-Jensen et al., 2000). Irradiations were carried out within the reader using a ^{90}Sr/^{90}Y beta source. The IRSL signals were detected using a photomultiplier tube after passing through Schott BG-39 and Corning 7–59 filters, which allows blue transmission (320–480 nm).

3. Results and discussion

3.1. Thermal activation energy

The TAE for the IRSL signal from K-feldspar as a function of IR stimulation time was measured using the following procedure. Aliquots from different samples containing natural signals or regenerative signals were first cut-heated to 260 °C. This was followed by an IRSL measurement using a reduced stimulation intensity (10% of the maximum power) for a short period (0.1 s), at progressively higher sample temperatures (50, 60, 70, 80, 90 and 100 °C). These measurements made negligible depletion (<1%) of the trapped charge can be used for estimating the TAE value directly. Following these measurements, the aliquots were optically bleached for different periods (up to 90 s) using IR (90% power at 60 °C) and the above measurement sequence repeated.

Fig. 2a shows the natural IRSL intensity from sample Sm1 as a function of stimulation temperature from 50 to 100 °C (upper x-axis in Fig. 2a). Similar results were observed for other samples. This was expected because of the predominantly thermally assisted escape of charge which is well described by the Arrhenius equation (Bøtter-Jensen et al., 2003),

$$I(T) = I_0 \exp\left(-\frac{E}{kT}\right), \quad (1)$$

where $I(T)$ is the IRSL signal at measurement temperature T, I_0 is the IRSL signal at infinitely large temperature, k is the Boltzmann's constant and E is the thermal activation energy. The latter can be calculated by taking the logarithm of Eq. (1); Fig. 2a shows the Arrhenius plot of ln(I) against 1/kT (lower x-axis) within the initial part of the natural IRSL decay curve for Sm1 which shows excellent linearity. The TAE results for the natural IRSL from Sm1 (Fig. 2b, LH scale; 90% power, 60 °C) as a function of IR stimulation time are shown in Fig. 2b (RH scale). The TAE value starts at a value of 0.12 eV and rises to ~0.45 eV at $t = 90$ s. The TAE was also measured using a bleached and irradiated (regenerative) aliquot of Sm1. A similar dependence of TAE on IR stimulation time was observed (red squares in Fig. 2b, rising from 0.11 eV to ~0.47 eV at $t = 90$ s.

It should be noted that the increase of IRSL with stimulation temperature is not an apparent result of the photo-transfer effect, in which the electrons from the IRSL traps were transferred into empty shallow TL traps associated with peaks in the range 50–150 °C. To check this effect, the luminescence was monitored at the elevated temperatures (50–150 °C). The luminescence instantaneously reduce to a negligible level following IR stimulation, suggesting that any photo-transferred luminescence is negligible compared with the observed IRSL signal.

Fig. 3 shows the values of TAE as a function of IR stimulation time for the IRSL signal from all samples used in the study. It is