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STRUCTURAL STATE AND ANOMALOUS FADING OF THERMOLUMINESCENCE OF OLIGOCLASE

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Abstract—Anomalous fading of thermoluminescence was measured in four samples of oligoclase from Muskwa Lake, Canada. Two samples were annealed below the order/disorder transformation temperature and two were annealed at higher temperatures; X-ray diffraction measurements confirmed that the first two samples were fully ordered, while the others underwent appreciable, but not complete, disordering. After β-irradiation and a 15-day storage at room temperature, all samples showed anomalous fading in the high temperature regions (>400°C) of the glow curve, but the amount of fading was less for the partially disordered samples than for the fully ordered samples. We suggest that these data are consistent with the suggestion that the structural state of feldspars plays an important part in determining their anomalous fading properties.

1. INTRODUCTION

THE APPLICATION of natural thermoluminescence (TL) measurements to archaeological and extraterrestrial problems normally assumes that the TL in the higher temperature regions of the glow curve is thermally stable over the time-scales of interest (Aitken, 1985; Sears, 1988). Often this is so, but in many samples, especially those containing feldspar, the TL decays more rapidly than widely accepted models predict, and the high temperature regions of the glow curve may show appreciable loss of TL signal in days or weeks (Hoyt *et al.*, 1972; Wintle, 1973, 1977). In efforts to circumvent this difficulty, it has been found that it is sometimes possible to measure the TL of a stable component in the presence of a component showing such anomalous fading (Wintle, 1977).

The mechanism for anomalous fading is unknown. Garlick and Robinson (1972) and Visocekas (1985) have explored the possibility that quantum mechanical tunnelling between excited and ground states may be responsible, while Templer (1986) has suggested that transition between localized trapping sites may be involved. In both cases, the need for promotion of electrons to the conduction band is avoided, so that it is possible for very deep traps, which produce peaks at high glow curve temperatures, to have low stability. Other mechanisms involving the destruction of traps or luminescence centers have also been discussed (Wintle, 1977; Moharil *et al.*, 1989).

Based on the anomalous fading characteristics and annealing experiments on various meteorite classes (Guimon *et al.*, 1985), and the relationship between TL peak temperature and structural state of Amelia albite (Pasternak, 1978), Hasan *et al.* (1986) suggested that it was feldspar in the low-temperature (ordered) form that was showing anomalous fading while feldspar in the high-temperature (disordered) form was not. However, Tyler and McKeever (1988) measured the anomalous fading of samples annealed below and above the order/disorder transformation temperature and found no difference in their anomalous fading properties.

In the present paper, we report anomalous fading measurements similar to those of Tyler and McKeever, but X-ray diffraction techniques were also used to determine the degree of disordering in the samples. We conclude that Tyler and McKeever's samples were not fully disorderd, and hence still showed anomalous fading. In our experiments the amount of anomalous fading was less for partially disordered samples than for the fully ordered samples, suggesting that fully disordered samples would not show anomalous fading.

2. EXPERIMENTAL

Approximately 400 mg of terrestrial oligoclase ($Or_{2.3}$, $Ab_{78.0}$, $An_{19.7}$) from Muskwa Lake in Canada, obtained from B. Robertson of the Canadian Department of Energy, Mines and Resources, was crushed, and four 100 mg aliquots placed in quartz vials in an inert atmosphere (N_2) for annealing for 100 h at 300, 400, 900 and 1000°C in wire-wound tube furnaces, the order/disorder transition temperature being

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500–600°C (Smith, 1974, p. 6). After annealing, two or three 4 mg portions from each vial were irradiated to about 20 Gy using a 200 mCi Sr-90 source and stored for 15 days in the dark, in an air-conditioned room (20–23°C). The TL signal was then recorded, the samples re-irradiated with the same dose and the TL signal measured once more. Another aliquot of the annealed powder was spread on a glass slide and the powder X-ray diffraction pattern recorded 3–5 times over the 2Θ interval 20–30° using a Diano 8500 diffractometer. Checks were also made of the entire X-ray diffraction pattern to check for mineral impurities or chemical changes, but none were present. Further details of the annealing and X-ray diffraction measurements may be found in Hartmetz (1988), whose results wee briefly discussed by Hartmetz and Sears (1987).

3. RESULTS

Glow curves for the four samples after the 15-day delay period ($t = 15$) and without a delay period ($t = 0$, are shown in Fig. 1; Figs 1(a), (b), (c) and (d) show the curves for the samples annealed at 300, 400, 900 and 1000°C, respectively). In each case, the glow curves shown are an average of the three replicates. As discussed by Hartmetz and Sears (1987), the annealing caused the temperature of the major TL peak to move from approximately 120°C in curves for the samples annealed at 300 and 400°C to approximately 250°C in the curves for the samples annealed at 900 and 1000°C. The same effect has been observed with other ordered feldspars and certain meteorites (Hartmetz and Sears, 1987; Guimon *et al.*, 1985).

For each sample, the glow curve obtained after the 15-day storage was divided by the curve obtained without the 15-day storage. This ratio is also plotted in Fig. 1 ($t = 15/t = 0$). The noise on the curves provides a good indication of the uncertainty in the data. For the samples annealed at 300 and 400°C, the ratio of the two curves begins to rise at about 100°C and levels off at about 200°C to values between 0.4 and 0.7. There is a step in the curve for the sample annealed at 300°C, but for the 400°C sample the curve is fairly flat. Considerable fading has occurred in both samples. The TL in the 400–500°C region of the glow curves, where no thermal fading is expected

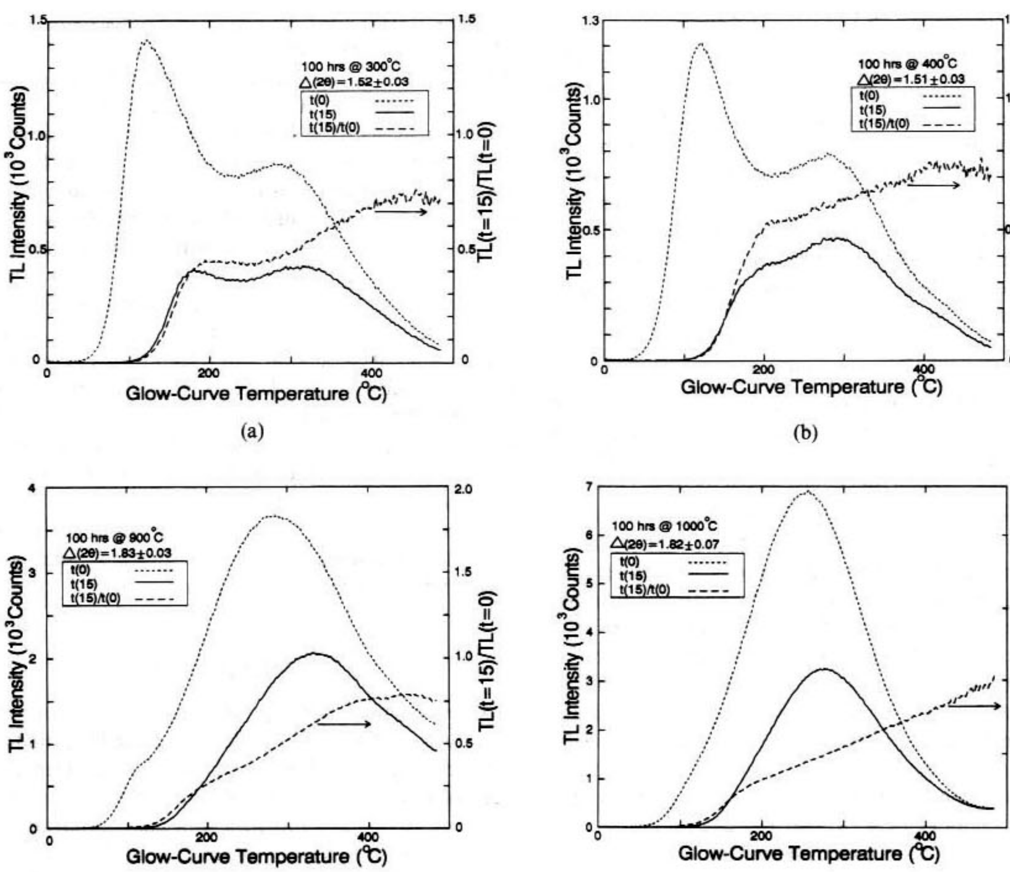

FIG. 1. Glow curves for four samples of oligoclase annealed for 100 h at (a) 300°C; (b) 400°C; (c) 900°C and (d) 1000°C. Curves run immediately [$t(0)$], and after a 15-day delay [$t(15)$] are shown. Also indicated is the ratio of the glow curve obtained after a 15-day delay to that obtained immediately, [$t(15)/t(0)$].

the ages being determined. Even empirical methods which have been applied successfully to historically dated samples could presumably be improved by a knowledge of the mechanisms responsible.

Hasan *et al.* (1986) also argued that a relationship between degree of ordering in feldspar and anomalous fading might be understandable in terms of a tunnelling mechanism, since it increases the bond distance between the postulated Mn luminescent centers and the nearest oxygens. It remains to be seen whether other plausible mechanisms for anomalous fading might also be affected by major changes in structural state. Tyler and McKeever (1988) argued that the kinetics of fading and phosphorescence at 280°C were more consistent with the localized transition mechanism than with anomalous fading. However, Tyler and McKeever (1988) point out that the trap distribution is unknown and, as is evident from their Figs 4 and 5, the models make very similar predictions for the decay kinetics of TL and phosphorescence.

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