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PARAMETERS OF OSL TRAPS DETERMINED WITH VARIOUS LINEAR HEATING RATES

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Abstract—An analysis method is introduced for traps which give rise to optically stimulated luminescence (OSL). For pulse-annealing experiments, the relationship between the temperature at which maximum OSL depletion occurs and the linear heating rate used to deplete the trapped charge is established, and this allows trap parameters to be determined. The lifetime at ambient temperature can be obtained from these parameters assuming first-order kinetics. Experimental data for a sedimentary K-feldspar separate are presented. © 1997 Elsevier Science Ltd

1. INTRODUCTION

Dating with the optically stimulated luminescence (OSL) signal from natural minerals has been widely applied to Quaternary sediments and archaeological materials (Wintle, 1993) and ages from a few decades to half a million years have been obtained (e.g. Huntley *et al.*, 1993). As with thermoluminescence (TL) dating methods (Aitken, 1985), one of the fundamental requirements of OSL dating is that the lifetime of the trapped charges responsible for the OSL signal must be substantially greater than the age of the material being dated. Despite several advantages, such as being more sensitive to light and having no unbleachable component, the measured OSL signal, unlike TL, yields no information about its thermal stability or lifetime. For minerals governed by first-order kinetics, the lifetime and the traps parameters are related by the equation $\tau = S^{-1}\exp(E/kT')$, where τ is the trap lifetime (s), S is the frequency factor (s^{-1}), E is the thermal activation energy (eV), k is Boltzmann's constant and T' is the ambient temperature (K) at which the mineral is stored.

Although various analysis methods have been introduced for TL traps (Chen and Kirsh, 1981), these methods may not be applied to OSL traps directly because of the different stimulation processes involved. It is reported that the OSL traps are different from the TL traps for signals from K-feldspar samples (Duller and Bøtter-Jensen, 1993; Duller, 1995). Hence, methods of directly determining the parameters of OSL traps must be developed.

In a recent publication, Short and Tso (1994) proposed methods for thermal activation energy analysis, including one which involved a hyperbolic heating strategy, which cannot be achieved in normal luminescence dating apparatus. In this paper, we describe a method of direct measurement of OSL traps, which involves linear heating profiles with different heating rates.

2. THEORETICAL BASIS

In order to simplify the problem, general one-trap kinetics are assumed (Randall and Wilkins, 1945; Levy, 1983). When measuring the OSL signal at a temperature T, thermal release and optical stimulation are both involved in the draining of charge from the OSL trap. For an OSL signal measured at a fixed and low temperature T_0 (e.g. < 50°C), we assume the OSL traps are deep enough that there is no effect of thermal release on the concentration of trapped charge. Within the confines of this model, we also assume that the intensity of the OSL signal is proportional to the concentration of the trapped charge. Hence, the OSL intensity measured at temperature T_0 can be expressed as

$$I = Cn \tag{1}$$

where C is a constant and n is the concentration of charge in the OSL trap. The OSL measurement can be made with a short light exposure and the loss of signal resulting from this measurement (δn) can be neglected, i.e. $\delta n \ll n$.

When heating the sample to a temperature T higher than T_0 without optical stimulation, thermal release of the trapped charges takes place. The probability of trapped charge escaping from the trap can be expressed by the following equation

$$-\frac{dn}{dt} = S\exp\left(-\frac{E}{kT}\right)f(n) \tag{2}$$

where $f(n)$ is a function of n. For first-order kinetics, i.e. no retrapping, $f(n) = n$ and equation (2) becomes

$$- \frac{dn}{dt} = Sn \exp\left(- \frac{E}{kT} \right). \quad (3)$$

For second order kinetics, i.e. with retrapping, $f(n) = n^2$ and equation (2) becomes

$$- \frac{dn}{dt} = Sn^2 \exp\left(- \frac{E}{kT} \right) \quad (4)$$

where $S = S/N$, and N is the total trap concentration.

When the sample is heated to a temperature T linearly, i.e. $T = T_0 + \beta t$, where T_0 is the starting temperature and β is the heating rate (K/s), the reduction rate of the trapped charge will be

$$- \frac{dn}{dT} = - \frac{1}{\beta} \frac{dn}{dt} = \frac{S}{\beta} \exp\left(- \frac{E}{kT} \right) f(n). \quad (5)$$

Once temperature T has been reached, the sample is cooled to T_0 very quickly. It is assumed that the cooling of the sample happens so fast that the reduction of trapped charges during this cooling period is insignificant. The loss of trapped charges $- \Delta n$ will be shown by the reduction of the OSL signal measured at temperature T_0, $I = C(n_0 - \Delta n) = I_0 - C\Delta n$, where n_0 is the original charge concentration. The reduction rate of the OSL signal to the original signal I_0 will be

$$- \frac{dI}{dT} = - \frac{C}{I_0} \frac{dn}{dT} = \frac{C}{I_0} \frac{S}{\beta} \exp\left(- \frac{E}{kT} \right) f(n). \quad (6)$$

The reduction rate of the OSL signal will reach its maximum at a temperature T_m if the derivative of equation (6) is set to zero, i.e.

$$\frac{d\left(\frac{dI}{dT} \right)}{dT} = C \frac{d\left(\frac{dn}{dT} \right)}{dT} = 0. \quad (7)$$

Substituting equations (3) – (5) into equation (7) and rearranging the terms, we have

$$\ln\left(\frac{T_m^2}{\beta} \right) = \frac{E}{kT_m} + \ln\left(\frac{E}{Sk} \right) \quad (8)$$

for first-order kinetics and

$$\ln\left(\frac{T_m^2}{\beta} \right) = \frac{E}{kT_m} + \ln\left(\frac{E}{2nSk} \right) \quad (9)$$

for second order-kinetics. As can be seen from equation (8), the temperature T_m is simply related to the heating rate and the parameters of the OSL trap for first-order kinetics. For various heating rates, trap parameters can be determined from a plot of $\ln(T_m^2/\beta)$ vs $1/T_m$ which has a slope of E/k and an intercept of $\ln(E/Sk)$. Therefore, the lifetime at ambient temperature can be obtained. For second-order kinetics the value of T_m also relates to the concentration of the trapped charges, as shown in equation (9).

3. EXPERIMENTAL PROCEDURES AND RESULTS

The necessary data can be obtained using Pulse Annealing experiments (Duller, 1994). Experimentally, this can be achieved in two ways, using multiple aliquots and a single aliquot. For the multiple aliquots approach, a series of sample aliquots is prepared. Different aliquots are used for each heating, each starting from the same temperature, but heated to a different final temperature in certain temperature intervals at a heating rate β. The final temperatures ranged from T_1 to T_x (e.g. from 100 to 500°C). Once the final temperatures are reached, the aliquots are quickly cooled to temperature T_0. The OSL signals are measured at T_0 using a short stimulation time. Other groups of aliquots are used for different heating rates. The reduction rate of OSL signal vs temperature can be plotted and the temperature at which the reduction rate reaches a maximum value can be obtained for each heating rate. Hence, the parameters (E and S) of the light sensitive trap can be obtained using equation (8). In this way each aliquot is heated only once, but a large amount of sample aliquots is required and normalization is necessary.

On the assumption that significant eviction of trapped charge occurs as a result of heating, a single aliquot may be used for measurements at each heating rate. The single aliquot is heated, with the same temperature increment, to successively higher temperatures at a heating rate β, and quickly cooled to temperature T_0. The OSL signals are measured at temperature T_0 using a short stimulation time after each heating. This single aliquot approach has advantages of improving reproducibility, as no normalization is required, and working with small quantities of sample. However, each aliquot must be heated a number of times. We have compared the results using both procedures with a K-feldspar sample. The same OSL reduction was found for heating rate of 2°C/s with an increment of 10°C.

As a supplement to the basis of the method, we have applied the single aliquot procedure, because of limited sample availability, to a sedimentary K-feldspar separate from Hong Kong. The natural sample was preheated first at 220°C for 10 min to isolate thermally stable OSL signals (Li, 1991). For each heating rate, aliquot was heated from 100°C to 500°C with 10°C increment. After heating to each appropriate temperature, aliquots were quickly cooled to 50°C. OSL signals were measured on Risø TL/OSL reader at 50°C with a 0.1 s exposure of infrared from an array of light emitting diodes with

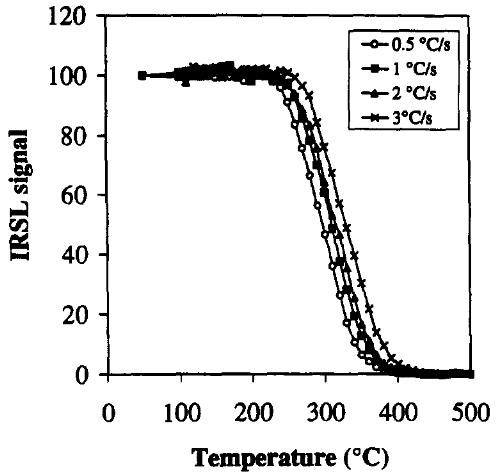

Fig. 1. Pulse annealing curves of natural K-feldspar after preheating at 220°C for 10 min. Data have been plotted as the percentage of OSL signal remaining after annealing at a given temperature.

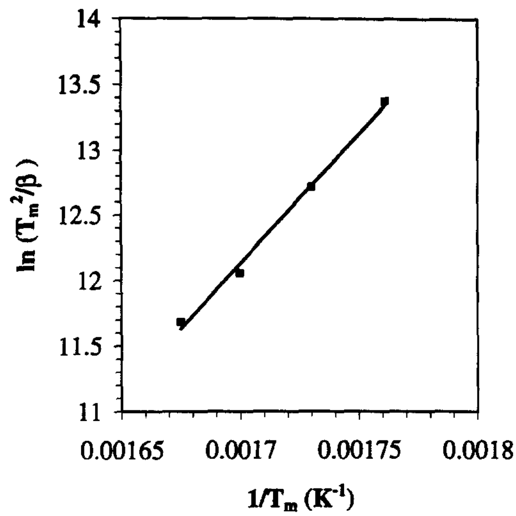

Fig. 3. Plot of $\ln(T_m^2/\beta)$ vs $1/T_m$ of pulse annealing results from K-feldspar.

emitting wavelengths peaked at 880 nm. The OSL was measured by an EMI 9236QA photomultiplier with one 2 mm thick BG 39 filter. At least four repetitive measurements were made for each heating rate. The heating rates were 0.5, 1, 2, 3°C/s.

Pulse annealing curves of OSL signals vs annealing temperature were obtained. Figures 1 and 2 show the fraction of remaining OSL signal and the percentage of OSL reduction per °C vs annealing temperature respectively. Each curve represents the average of four aliquots. The original OSL signal was taken as 100%. The reduction rate of OSL signal was calculated as percentage to the original signal. Because a 10°C increment was applied to each heating phase in the experiments, and the temperatures in Fig. 2 is the temperature reached, the average temperature for each heating phase would be a minus 5°C of the temperature reached since the loss of

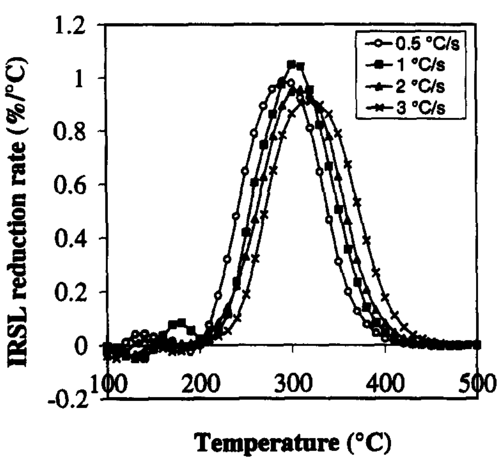

Fig. 2. The same pulse annealing results as plotted in Fig. 1, but now plotted as the percentage of the OSL signal reduction per °C. The data have been smoothed using three-point running mean.

charges can be any temperature within the 10°C. Hence, a − 5°C was justified in choosing the maximum temperature (T_m) from Fig. 2. A plot of $\ln(T_m^2/\beta)$ vs $1/T_m$ is shown in Fig. 3. Trap parameters were calculated from the slope and intercept of the plot. For the K-feldspar sample, the thermal activation energy (E) is 1.72 eV and the frequency factor is 6.1×10^{13} Hz. Hence the lifetime of the OSL signal is 1.0×10^9 y at an ambient temperature of 10°C for first-order kinetics.

4. DISCUSSION

The lifetime of the OSL signals from the K-feldspar is similar to the lifetime of 1×10^9 y of the TL peak of temperature around 350°C with $E = 1.68$ eV and $S = 2.8 \times 10^{12}$ Hz (Aitken, 1985). Because a preheating procedure, at 220°C for 10 min, has been applied to the natural sample and resulted in only high temperature TL remaining, the similarity in the trap parameters between the OSL signals obtained here and the high temperature TL peaks obtained independently suggests that the thermal stabilities of both OSL and TL traps are the same for the preheated samples. It is suggested further that the preheating procedure is suitable for isolating the thermally stable component in dating of Quaternary sediments.

The assumptions of a single trap and first-order kinetics, made in describing the theoretical basis, are satisfied for the sample after the preheating. This suggests that the traps responding to OSL at 50°C can be represented by a single trap as far as the thermal stability is concerned. In other words, if there are several traps causing OSL the difference in the thermal stability for the OSL traps is insignificant after the preheating. Because the cooling rate after reaching the designated temperature is far greater than the heating rates used, the assumption of eviction of trapped charges occurring during the heating phase only has been met. We have tested a similar K-feldspar sample and found no differences between single aliquot and multiple aliquots (unpublished data). Hence, single aliquots can be used for the relatively slow heating rates used in this study.

The analysis method is quite similar to the Hoogenstraaten method applied in TL studies (Hoogenstraaten, 1958). However, there are significant differences between the two applications. The prerequisites of sufficient luminescence centres and radiative recombinations existing at all recorded temperatures in the TL glow curve are not required in this method because thermally stimulated charge recombination is not used for the analysis. Only the thermal removal of trapped charge, reflected by the reduction of OSL signal, is relevant. Similarly, the temperature of the maximum reduction rate T_m will be independent of the detection wavelength region, provided that there is no change in luminescence centres at the measurement temperature T_0. For TL, however, the value of T_m can change with the detection window used as a result of the change in availability of the related luminescence centres at higher temperatures. In particular there is less effect due to thermal quenching, because the OSL measurements are made at a lower temperature. Thermal quenching effects have been found for TL in quartz (Wintle, 1975) and for OSL signals measured at higher temperatures in feldspar (Poolton *et al.*, 1995).

In theory, this method can be applied to materials which have more than one OSL trap, if there is no charge transfer between these traps, and no transfer of charge between OSL traps and non-OSL traps, i.e. the charge concentration in each trap has to be independent of any other during heating and the OSL measurement. This implies that even traps which contribute only a small portion of the total OSL signal can still be analysed with this method. Traps with different parameters may result in data sets which overlap at one heating rate, but can be separated by using various different heating rates.

Despite differences between T_m for TL and T_m for the OSL reduction rate, some of the other techniques for analysing the TL signal with different heating rates might also be applied to the OSL signal (see Chen and Winer, 1970 for details). The advantages and disadvantages of the heating rate analysis in studies of TL may also be applicable (Chen and Kirsh, 1981).

In the single aliquot pulse annealing experiment, the TL signal can be recorded for each heating phase, if the annealing is performed in a TL oven. The TL thus recorded would be similar to the results of Initial Rise measurements (Aitken, 1985), except for the negligible loss of TL due to the OSL measurement at temperature T_0. This would provide extra information

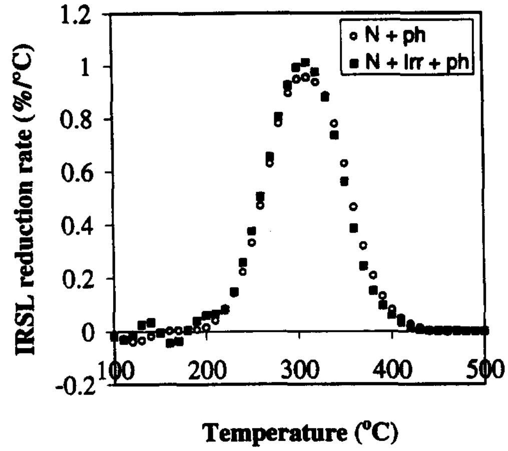

Fig. 4. Percentage reductions of the OSL signal per °C from natural and natural plus beta dose (79 Gy) samples. The natural dose of the sample is 80 Gy measured with K-feldspar OSL additive dose method.

on the trap parameters and the mechanism involved for both TL and OSL signals.

The kinetic order of the thermal drainage of trapped charge may be indicated by the shape and the maximum of the reduction rate curve, because there is a significant difference between the behaviour for first- and second-order kinetics. As in TL, a kinetic order check can be performed by seeing whether T_m changes when the charge concentration is altered, e.g. by changing the radiation dose. The same T_m has been found for both natural and natural plus beta dose aliquots of the K-feldspar sample after the preheating (Fig. 4, Tso *et al.*, 1996).

This method can also be applied to electron spin resonance (ESR) signal analysis where there is no signal loss caused by the ESR measurements.

5. CONCLUSION

The theoretical basis of a new OSL trap analysis method has been proposed here. Encouraging results have been obtained for OSL signals stimulated with infrared from a potassium feldspar separate.

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