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Determination of trapping parameters of the high temperature thermoluminescence peak in equilibrated ordinary chondrites

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Abstract

Most meteorites exhibit thermoluminescence (TL) that can be used to constrain their recent thermal and irradiation history, but quantitative conclusions require a knowledge of the detailed TL peak structure of the TL glow curve. We have determined TL peak parameters for the high temperature portion of the glow curve for six ordinary chondrites: Chicora (LL6); Innisfree (L5); Lost City (H5); Paragould (LL6); Pribram (H5); and Tilden (L6). The saturation dose for all these meteorites is approximately 3600 Gy. Published procedures were used to determine the number and temperatures of peaks in the high temperature (>570 K) portion of the glow curve and peak fitting was used to estimate TL trap parameters for each peak. These data were then tested and adjusted, if necessary, by comparing calculated decay results with TL glow curves for samples heated at ≈ 420 K for various times. We find evidence for four TL peaks in the high temperature portion of the glow curve, where trapping parameters vary slightly from meteorite to meteorite. For the Lost City meteorite, the TL peak temperatures (K), activation energies (E, eV), and Arrhenius factors (s, $\times\ 10^{-9}\ s^{-1}$) are: 325, 1.26, 4.8; 360, 1.33, 3.88; 401, 1.44, 5.8; and 455, 1.5, 2.25, respectively. These data could be used to estimate dose rates for meteorites; however, the albedo values required for the calculation are not yet sufficiently known. However, terrestrial ages, or surface exposure ages, for meteorite finds from hot deserts like those in Australia or North Africa, can be estimated from these data.

1. Introduction

Natural thermoluminescence (TL) has been used to study thermal and radiation history, terrestrial age, and surface exposure ages of meteorites (McKeever and Sears, 1979; Melcher 1981; Benoit et al., 1991; Benoit, 1995). It has also been used to help in the identification of paired meteorites and the specimens recovered some distance apart that were part of a single meteorite that fragmented during atmospheric passage or impact. These studies utilized the low temperature (150–300°C) region of the glow curve (Fig. 1). Christodoulides et al. (1971) suggested that the TL of meteorites could be used to study the cosmic ray dose rate that a meteorite is exposed to in space, in effect, using meteorites as dosimeters. However, because of the ease of drainage of natural TL in the low temperature region of the glow curve as a result of solar heating in space, it would be necessary to use the more

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stable, high temperature region of the glow curve for such studies. This requires the determination of the TL build-up and decay parameters of this region of the glow curve.

In the present study we examine only the equilibrated ordinary chondrites, the largest class of meteorites. The mineral responsible for most of the TL in these meteorites is sodic feldspar (Guimon et al., 1995). The TL of other meteorite classes is dominated by feldspar of different composition and structure and different TL properties (e.g., Hasan et al., 1986; Guimon et al., 1995). For example, like most terrestrial feldspar samples some meteorites exhibit "anomalous fading" (Wintle, 1973; Sears et al., 1991) but the equilibrated ordinary chondrites show no evidence for this process (Hasan et al., 1986).

2. Previous work concerning the TL parameters of meteorites

Most previous studies of the TL of meteorites have concentrated on specific applications, for example, the calculation of effective temperatures of irradiation in order to constrain orbital parameters (Prachabrued et al., 1971; McKeever and Durrani, 1977; Melcher, 1981). Due to problems in resolving individual TL peaks in the glow curves of meteorites (below), these studies assumed a simplified glow curve structure. Other studies have concentrated on determining TL parameters for the low temperature portion of the glow curve (e.g., Sears and Mills, 1974).

McKeever (1980) applied a least-squares peak fitting procedure to determine TL parameters for the entire glow curve for three ordinary chondrites. He showed that the glow curve of ordinary chondrites contains at least eight TL trap populations with significant peak overlap (e.g., Fig. 1). The high degree of overlap limited the accuracy of the parameters he determined, especially in the high temperature region of the glow curve. In this study, we seek to determine the TL parameters for the high temperature region of the glow curve of ordinary chondrites. Our results suggest that the parameters determined for the low temperature portion of the glow curve by McKeever (1980) are sufficient for most applications, but we have better constrained TL peak properties for the higher temperature portion of the glow curve, where peak overlap is a significant problem.

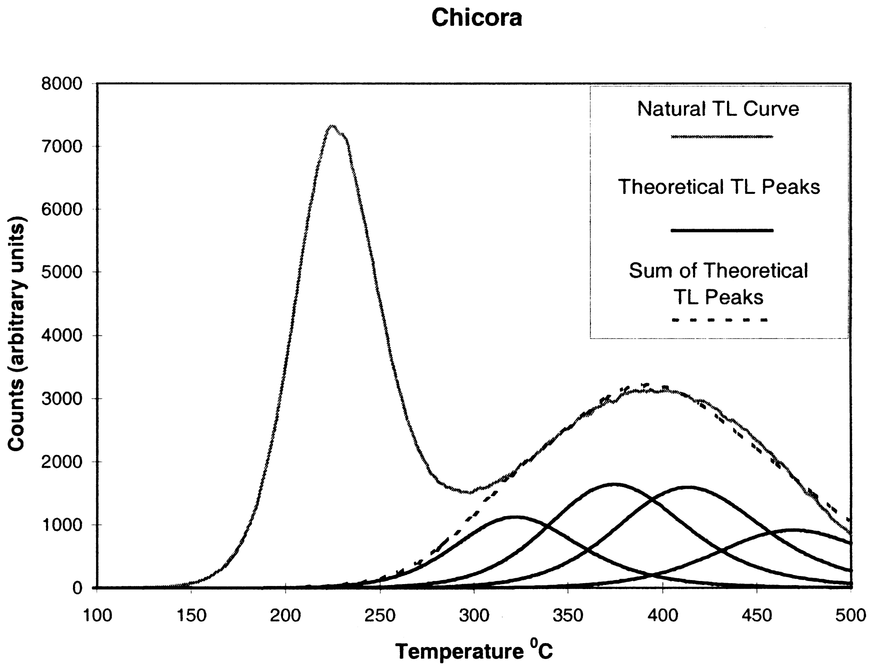

Fig. 1. The natural TL glow curve of the Chicora meteorite. The glow curve contains two apparent peaks one at about 220°C and the second at about 370°C. Peak fitting suggests that there are four overlapping peaks in the high temperature (> 300°C) portion of the glow curve.

3. Theory

3.1. TL equation

The TL decay and build-up for a single population of TL traps for meteorites can be described in terms of an equilibrium between second-order decay and first-order buildup (Garlick and Gibson, 1948; Christodoulides et al., 1971; McKeever, 1980, 1985):

$$\frac{dn}{d} = \frac{r(N-n)}{R_0} \text{-} s'n^2 \exp\left(\frac{-N}{kT}\right) \quad (1)$$

where s' is a second-order frequency factor, equal to s/n_0, where s is the first-order frequency factor, t = time, n is the number of trapped electrons, N is proportional to the total number of traps, R_0 = the dose necessary to fill 63.2% of the total available traps, and r = dose rate, n_0 = initial number of trapped electrons, E = energy depth of the trap, k = Boltzmann's constant, and T = storage temperature. The evidence for second-order kinetics for the TL of ordinary chondrites was summarized by McKeever (1980) and our observations are in accord with his. The parameters N (Gy), R_0 (Gy), E (eV), and s (s^{-1}) describe a single trap population and are determined by the material. For a given sample the value of n (Gy) is dependent upon two environmental variables, the temperature, T (K), and the dose rate, r (Gy/s). While Eq. (1) seems to describe the physical behavior of TL in many phosphors, the detailed physics of the TL mechanism is not well known (Chen and McKeever, 1997).

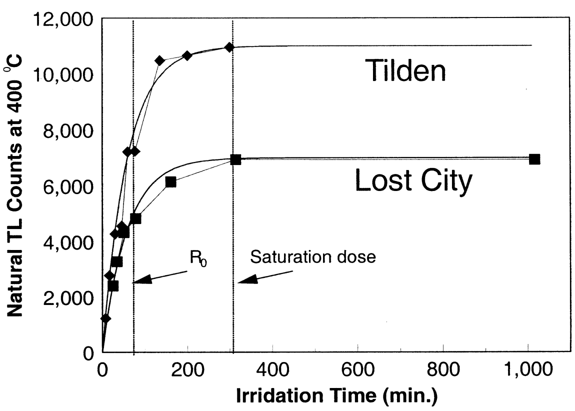

Fig. 2. Experimental dose response (points) and the calculated dose response (solid lines) at 400°C in the glow curve. The dashed lines correspond to the time at which saturation is reached, N; and the time where 63.2% of the total counts are reached, R_0.

3.2. Determination of build-up parameters

The trapping parameters for the build-up of TL, N and R_0, can be determined experimentally by subjecting samples to known doses of radiation at very high dose rates (Fig. 2), so that thermal decay is negligible (Christodoulides et al., 1971). An estimate of the saturation level, at which traps are filled and further dose causes no increases in TL signal, is shown in Fig. 2, at saturation $n = N$.

3.3. Determination of decay parameters

A summary of the techniques to determine s and E values can be found in Chen and McKeever (1997). The technique that gives the most accurate values for overlapping peaks is the peak fitting procedure (McKeever, 1980). This method fits a series of theoretical peaks to a glow curve by a least-squares regression, assuming which each peak can be described by:

$$I(T) = n_0^2 s' \exp\left(\frac{-E}{kT}\right)\left[1 + \left(\frac{n_0 s'}{\beta}\right)\int_{\tau_0}^{T} \exp\left(\frac{-E}{kT}\right) dT\right]^2 \quad (2)$$

where β = the heating rate (K/s) and the other parameters are as described above (Garlick and Gibson, 1948). However, this method requires an initial estimate of the number and approximate locations of each TL peak.

The most common method for determining the number and location of TL peaks is to read out a number of glow curves from a single sample obtained by interrupting and restarting the heating ramp a number of times, each time at a higher temperature. Apparent peak temperature (T_m) can then be determined as a function of stopping temperature (T_{stop}). This method is best suited to well-resolved peaks or peaks that overlap only slightly, but even peaks with significant overlap can be observed, although estimates of their positions in the glow curve will carry significant uncertainties (Chen and McKeever, 1997). The individual peaks appear on a T_m versus T_{stop} plot as a series of steps.

4. Experimental

4.1. Sample preparation

Chips of the Paragould (LL6), Lost City (H5), Tilden (L6), Chicora (LL6), Innisfree (L5) and Pribram (H5) meteorites, each weighing about 150–300 mg, were separately ground in a mortar and pestle and non-luminescent metal removed with a hand magnet. Four-mg portions of each sample were placed in copper pans with 0.02 in. thick bases. The TL measurements were performed with Daybreak Nuclear and Medical Systems' TL apparatus fitted with blue band-pass and IR filters (Corning 7–59 and 4–69), with a heating rate of 3°C/s and maximum temperature of 500°C. Luminescence intensity at 1°C increments was automatically recorded in electronic form for later peak fitting.

4.2. Build-up parameters

Dose response curves were measured for meteorites using a ^{90}Y–^{90}Sr irradiation cell with a dose rate of ≈ 12 Gy/min. Each sample was irradiated multiple times to produce a graph of TL intensity versus dose.

4.3. Decay parameters

T_m–T_{stop} curves were produced using 4 mg samples and T_{stop} increments of 4°C for Lost City and 10°C for Paragould. Due to the amount of material necessary for these experiments, T_m–T_{stop} data were not collected for the other four meteorites.

Samples of all six meteorites were heated for 2, 25, 67 and 166 h at a constant temperature of 154 ± 2°C and the natural TL measured in order to determine curves that could be used to test and further refine the parameters obtained by curve fitting. TL parameters were estimated using least-squares refinement of the 300–450°C portion of the glow curves, modeling TL peaks using Eq. (2), using the PC-based PEAKFIT program. This program enables simultaneous refinement of multiple overlapping peaks.

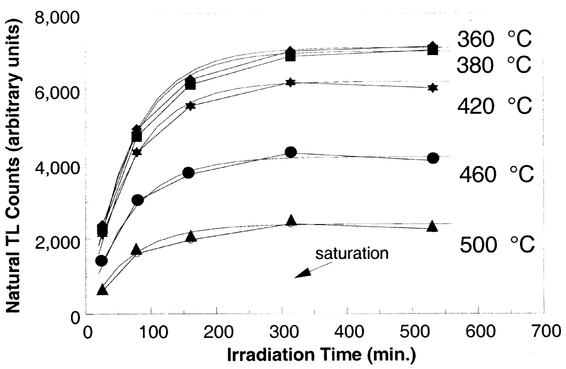

Fig. 3. Experimental and theoretical dose response curves for the Lost City meteorite taken at several temperatures in the glow curve. The time at which saturation is reached, N, does not change throughout this portion of the glow curve.

5. Results

5.1. Build-up parameters

The dose response curves for the TL of Lost City and Tilden at 400°C in the glow curve are shown in Fig. 2. The dose response curves for the remaining meteorites are similar. The samples reach saturation at 3600 ± 100 Gy and have a R_0 value of 800 ± 70 Gy. The saturation dose is approximately constant throughout the high temperature portion of the glow curve (Fig. 3).

5.2. Decay parameters

The glow curve for Paragould appears to contain three well-defined peaks in the high temperature portion of the glow curve at ≈ 360, 370 and 400°C (Fig. 4), with a possible fourth peak at > 435°C, but this is not well-resolved because it is obscured by blackbody radiation. These peak temperatures were used as initial estimates for peak fitting. The least-squares refinement of the TL glow curves resulted in slight modifications of TL peak temperatures, E and s. The E and s combinations calculated from this initial peak fitting were used to predict the amount of TL decay expected during the heating experiments using the relationship:

$$I(t) = \frac{I_0}{1 + sn_0 t \exp\left(\frac{-E}{kT}\right)} \tag{3}$$

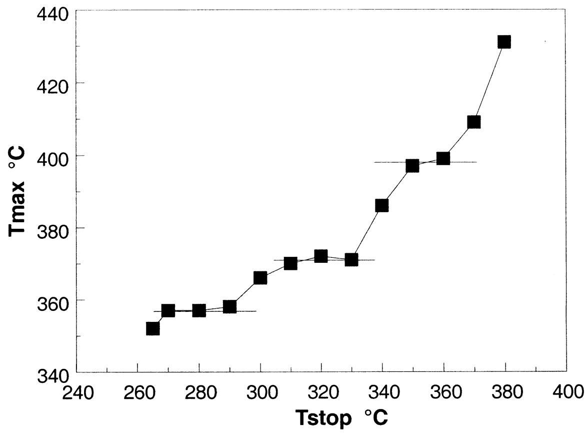

Fig. 4. Apparent peak temperature (T_m) for glow curves obtained for the high temperature region of the glow curve for the Paragould meteorite by heating to intermediate temperatures (T_{stop}) and heating again to a higher temperature. There are three well-defined "steps" at about 355, 370 and 400 °C, corresponding to three discrete peaks, with perhaps a fourth at ≈ 435°C.

where I_0 is the initial TL intensity and $I(t)$ is the TL intensity at a given time, t (Garlick and Gibson, 1948). *For a given meteorite, a range of E and s combinations was found to produce matches to the TL data. However, E and s combinations that did not result in TL intensities similar to those observed in the laboratory heating data were eliminated and the peak fitting done again with slightly revised initial estimates of the peak temperatures, E and s. Uncertainties* (Table 1) *reflect the range in E and s combinations producing matches to the heating experiments rather than statistical uncertainties.*

Fig. 5 shows a comparison between experimental and theoretical decay glow curves using the parameters reported in Table 1. The calculated peak positions of Paragould, Lost City, Tilden, Chicora, Innisfree and Pribram match well with the approximate positions predicted by the $T_m - T_{stop}$ curve of Paragould (Table 1, Fig. 4).

6. Discussion

Our $T_m - T_{stop}$ data agree with those of McKeever (1980) in the approximate position of three major peaks in the high temperature portion of the TL glow curve. We found peaks at ≈360, 380 and 400°C for our samples. In comparison, McKeever (1980) identified peaks at ≈360, 380 and 420°C using samples of the Soko Banja meteorite and a heating rate of 1°C/s. However, the TL parameters determined by McKeever (1980) by curve fitting to the natural glow curve for Lost City did not reproduce the decay seen in our laboratory heating experiments (Fig. 6) whereas our parameters were refined to agree with the experimental data (Fig. 5). *Variation of E and s values outside the uncertainty estimates* (Table 1) *results in significantly different TL decay behavior and thus do not reproduce our experimental data.*

The E and s values of each peak for different meteorites are very similar but not identical (Table 1). Thus, while these ordinary chondrites have similar TL properties, for highly precise applications, like accurate dosimetry, individual E and s values will need to be determined. However, other factors are also important, and may involve higher degrees of uncertainty than that imposed by the TL parameters. For example, Cristodoulides et al. (1971) and McKeever (1980) have suggested using meteorites for radiation dosimetry. They could be used to evaluate radiation gradients in the inner solar system, presently not known in detail (e.g., McKibben, 1975; Potgeiter et al., 1989; McDonald et al., 1997), and, given the long integration time for TL, they might be used for evaluation of changes in the radiation environment on the 10^5–10^6 year time-

Table 1
Thermoluminescence parameters calculated for the four high temperature peaks

	Peak #1	Peak #2	Peak #3	Peak #4
Lost City (H5)				
E (eV)	1.26 ± 0.01	1.33 ± 0.01	1.44 ± 0.01	1.50 ± 0.01
s ($\times 10^9$ s^{-1})	4.80 ± 0.5	3.88 ± 0.5	5.80 ± 0.5	2.25 ± 0.5
T (°C)	325 ± 1	360 ± 1	401 ± 1	455 ± 1
Innisfree (L5)				
E (eV)	1.26 ± 0.01	1.37 ± 0.01	1.46 ± 0.01	1.50 ± 0.01
s ($\times 10^9$ s^{-1})	8.55 ± 0.5	9.17 ± 0.5	8.64 ± 0.5	1.75 ± 0.5
T (°C)	312 ± 1	360 ± 1	401 ± 1	462 ± 1
Chicora (LL6)				
E (eV)	1.28 ± 0.01	1.38 ± 0.01	1.45 ± 0.01	1.52 ± 0.01
s ($\times 10^9$ s^{-1})	8.25 ± 0.5	5.76 ± 0.5	4.35 ± 0.5	1.91 ± 0.5
T (°C)	325 ± 1	374 ± 1	413 ± 1	469 ± 1
Pribram (H5)				
E (eV)	1.25 ± 0.01	1.36 ± 0.01	1.44 ± 0.01	1.50 ± 0.01
s ($\times 10^9$ s^{-1})	7.50 ± 0.5	5.10 ± 0.5	4.89 ± 0.5	1.37 ± 0.5
T (°C)	312 ± 1	369 ± 1	406 ± 1	469 ± 1
Tilden (L6)				
E (eV)	1.28 ± 0.01	1.41 ± 0.01	1.44 ± 0.01	1.52 ± 0.01
s ($\times 10^9$ s^{-1})	7.60 ± 0.5	11.90 ± 0.5	4.83 ± 0.5	2.75 ± 0.5
T (°C)	324 ± 1	371 ± 1	407 ± 1	459 ± 1
Paragould (LL6)				
E (eV)	1.31 ± 0.01	1.44 ± 0.01	1.47 ± 0.01	1.50 ± 0.01
s ($\times 10^9$ s^{-1})	10.50 ± 0.5	23.50 ± 0.5	7.20 ± 0.5	2.00 ± 0.5
T (°C)	329 ± 1	368 ± 1	410 ± 1	458 ± 1

Table 2
Dose rates calculated from natural TL levels for samples of three meteorites with known orbits. Apparent storage temperatures were calculated by integrating temperature over the entire orbit of each meteorite, for two albedo values. *Uncertainties in calculated dose rates reflect uncertainties in E and s parameters* (Table 1)

Meteorite	Assumed albedo	Calculated temperature of irradiation (K)	Calculated dose rate (Gy/year)
Lost City	0.1	285	0.010 ± 0.002
	0.3	270	0.0009 ± 0.0002
Innisfree	0.1	276	0.0027 ± 0.0006
	0.3	269	0.0007 ± 0.0002
Pribram	0.1	301	0.23 ± 0.05
	0.3	284	0.014 ± 0.002

scale (e.g., Nishiizumi et al., 1980). TL is highly suited for such studies, because "shielding" or depth effects appear to have minor influence on TL levels in typical meteoroid-sized bodies (Benoit and Chen, 1996). However, this application requires knowledge of storage temperature for individual meteoroid bodies. The primary heat source for meteoroid is solar heating, and thus storage temperature largely reflects distance from the Sun during the meteorite's orbit (Benoit et al., 1991). If the orbit of the body is known from visual observations (Simonenko, 1975; Wetherill and Chapman, 1988), an effective storage temperature integrated over a full orbit can be estimated (Table 2). *This temperature is not the average temperature, but is rather weighted towards temperatures experienced during closest approach to the sun, when TL decay rates are great-*

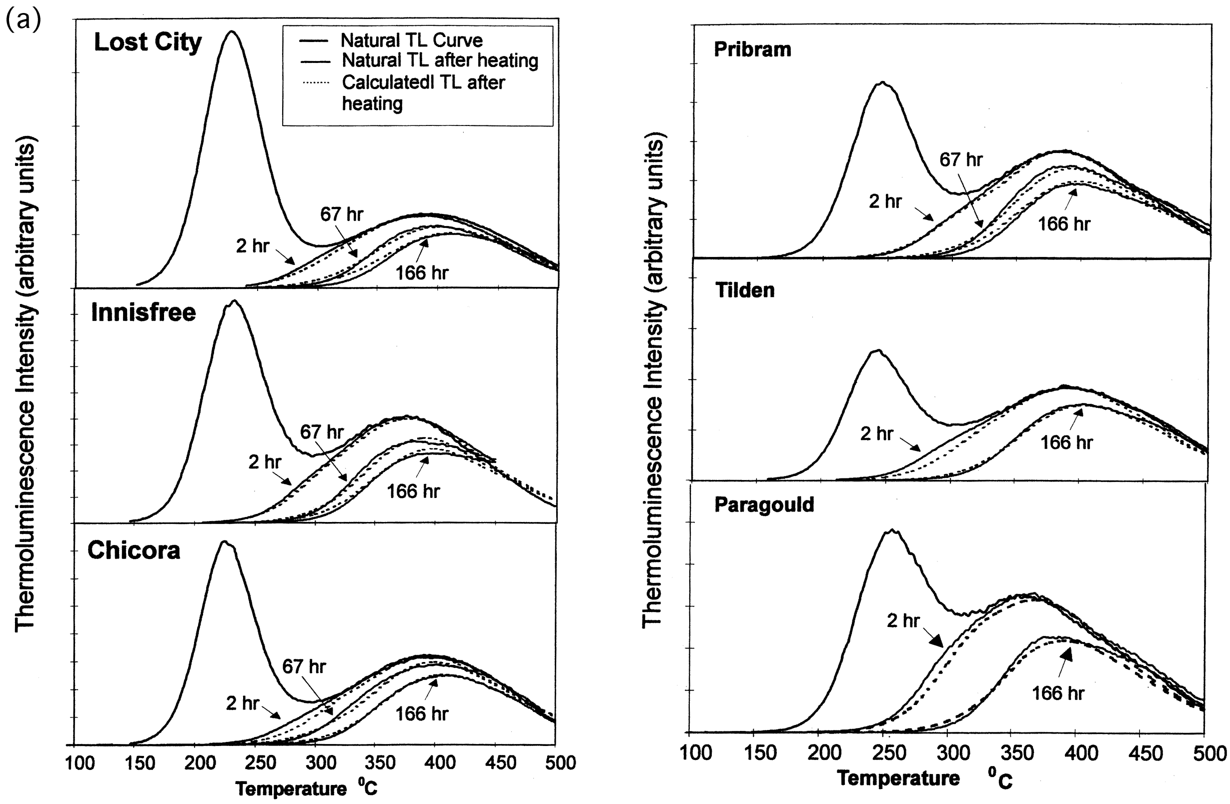

Fig. 5. The natural TL glow curves for the meteorites in this study (solid black lines) are shown along with the curves measured after 2, 67 and 166 h of heating at 155°C (solid gray lines). The experimental curves can be compared to the theoretical curves (dashed lines) calculated using the decay parameters given in Table 1. The E and s values have been adjusted to most closely fit the experimental heating data.

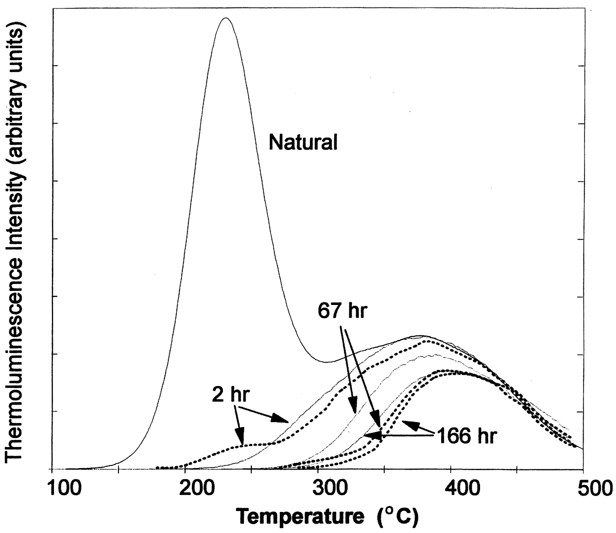

Fig. 6. Calculated glow curves for heated samples of Lost City, using the TL peak parameters determined by McKeever (1980) using peak fitting, compared against samples heated at 150°C in the glow curve. In general, these parameters poorly fit the experimental results.

est, using TL kinetics (Eq. (1)). However, the temperature of the body is significantly influenced by the albedo of the meteoroid in space. For example, Table 2 gives our calculated dose rates for three meteorites with precisely known orbits (Ceplecha, 1961; McCrosky et al., 1971; Halliday et al., 1978), based on observed TL levels and the parameters in Table 1, using the $\approx$300°C portion of the glow curve. We have shown calculations for albedos of 0.1 and 0.3, approximately bracketing the range observed in laboratory studies of ordinary chondrites (Chapman and Salisbury, 1973). It is apparent that uncertainty in albedo by a factor of three results in uncertainty in dose rate by about an order of magnitude. While the albedo of the meteorite sample could be used to delineate albedo, it is not clear that sample albedo is strongly related to the albedo of the surface of the parent meteoroid, which influences the degree of solar heating (Chapman and Salisbury, 1973; Clark et al., 1992).

7. Conclusions

The build-up parameters in the high temperature portion of the glow curve were found to be very similar in our suite of ordinary chondrites, the most common type of meteorite. The TL decay parameters differ from the preliminary values reported by McKeever (1980) reflecting more refined peak fitting and, in particular, the addition of laboratory heating experiments to eliminate incorrect E and s pairs.

Application of these data for radiation dosimetry using meteorites is hampered by the significant uncertainties in the degree of heating experienced by the samples. However, these data may be useful in another application, namely estimating terrestrial ages of meteorites found in hot deserts. Hundreds of meteorites have been recovered in the deserts of Australia,

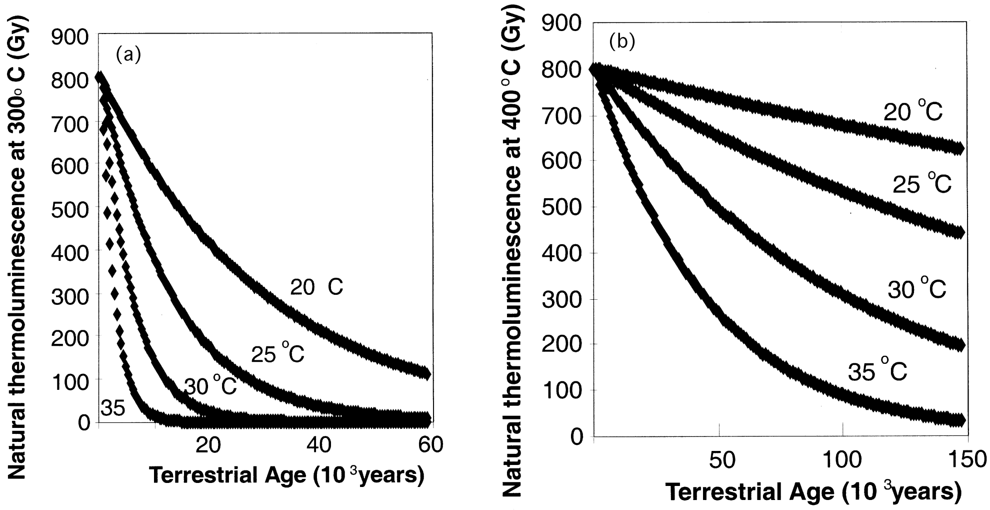

Fig. 7. Natural TL decay curves for various temperatures (K) using (A) Peak 1 trap parameters and (B) Peak 2 trap parameters (Table 1). An apparent radiation dose rate of 5×10^{-2} mGy/year is assumed. Temperatures and dose rates are based on conditions in hot deserts. The decay of Peak 1 and Peak 2 can be used to measure terrestrial ages of meteorites, if storage temperature can be estimated.

North Africa, and the Southwestern US (e.g., Sipiera et al., 1987; Bevan and Binns, 1989; Jull et al., 1993) and the collection of these meteorites continues to grow. These meteorites typically have terrestrial ages, *estimated from ^{14}C activities in mineral phases in the meteorites*, of up to 20,000 years (Jull et al., 1993, 1998) and experience temperatures in excess of 30°C (Benoit et al., 1993). Under these conditions, natural TL at glow curve temperatures $<300°C$ is drained too quickly for dating most meteorites, but the present data allow modeling of TL decay of the more stable high temperature portion of the glow curve (Fig. 7). For meteorites from hot deserts, natural TL dating could either be used in place of ^{14}C dating, or could be used in conjunction with ^{14}C dating, allowing more detailed study of terrestrial thermal history, including burial and surface exposure history similar to that possible using the low temperature portion of the glow curve for Antarctic meteorites (Benoit, 1995).

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