Thermoluminescence as a palaeothermometer

R. K. Guimon, K. S. Weeks, B. D. Keck & D. W. G. Sears

Department of Chemistry, University of Arkansas, Fayetteville, Arkansas 72701, USA

Chondritic meteorites are widely regarded as a unique source of information on the chemical and physical processes that occurred in the early Solar System. An important phase in their history is a period of metamorphism[1] and thermoluminescence (TL) has proved a powerful technique for exploring this aspect, especially at the low end of the metamorphic spectrum[2]. The ordinary chondrites as a whole display a 10^5-fold range in TL sensitivity, while the least metamorphosed, or type 3, ordinary chondrites display a 10^3-fold range. Associated with this 10^3-fold range in TL sensitivity are variations in the temperature at which the maximum TL emission occurs, and in the temperature range over which emission occurs[3,4]. We report here the results of annealing experiments on a little-metamorphosed (type 3.5) ordinary chondrite. The TL emission characteristics of the annealed samples show trends very similar to those observed in meteorites which have naturally been metamorphosed to various extents. The trends are also similar to those observed in annealing experiments on terrestrial albite[5–7] where the changes are associated with the low-to high-temperature transformation. These data suggest that the TL phosphor in meteorites is feldspar and that TL can be used to estimate palaeotemperatures for little-metamorphosed and highly-unequilibrated meteorites.

About 1.5 g of the Allan Hills A77011 meteorite was ground (<100 μm) and the magnetic fraction removed. Aliquots weighing 20 mg were then placed in high-purity quartz vials, evacuated to 10^{-5} atmospheres, filled to atmospheric pressure with nitrogen and heat sealed. They were then placed in a wire-wound tube furnace at a variety of temperautres for 100 h. Two vials were annealed at each temperature. After annealing, the vials were allowed to cool in air, reaching room temperature after a few minutes. The TL induced by a 250-mCi ^{90}Sr β source (absorbed dose 25 krad) was then measured 3–5 times. (Our apparatus and techniques are described in ref. 4). Irradiation times were chosen so as to give a signal-to-noise ratio of better than 10, usually it was >50. Data for each vial were averaged and the standard deviation calculated. The data for a given time and temperature were averaged and the total uncertainty calculated by compounding the 1σ uncertainties for the replicate measurements on each vial and for the standard, Dhajala to facilitate comparison between different laboratories.

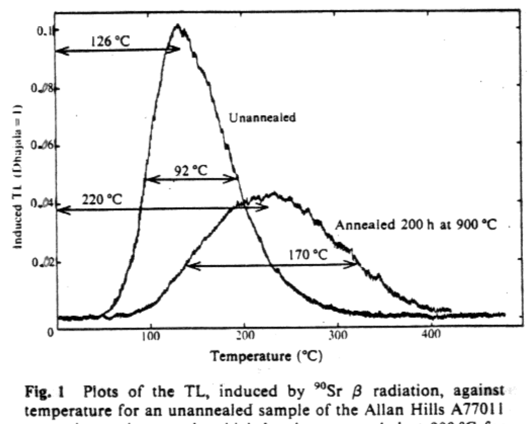

Fig. 1 Plots of the TL, induced by ^{90}Sr β radiation, against temperature for an unannealed sample of the Allan Hills A77011 meteorite, and a sample which has been annealed at 900 °C for 200 h. The annealing treatment has caused the TL peak, in fact, a composite of many smaller individual peaks, to broaden and move to higher temperatures.

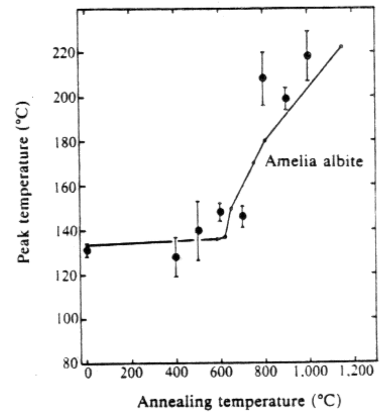

Fig. 2 A plot of the temperature of the TL peak against the temperature at which each sample was annealed for 100 h. The error bars refer to $\pm 1\sigma$ and each data point is the mean of two samples. Also shown are data from ref. 5 for a terrestrial albite from Amelia, Virginia, which was annealed for 24 h in a manner similar to that used here. The change in the TL emission characteristics of the Amelia albite were associated with the low-to-high temperature transformation, suggesting that the TL phosphor in meteorites is feldspar and that the shift in peak temperature on annealing may also be associated with changes in crystallography.

Two curves for the TL induced by the β irradiation as a function of glow curve temperature are presented in Fig. 1. Ordinary chondrites display a broad band of TL at 100–300 °C which is composite of many smaller, individual peaks. The glow curves for an unannealed sample and for one of our most severely annealed samples are shown in Fig. 1. The TL peak is relatively sharp in the unannealed sample and appears at a lower temperature in the glow curve than the TL peak of the annealed materials. In the examples in Fig. 1, the annealed sample has its peak 94 °C higher in the glow curve, and its peak is 78 °C wider at half its maximum intensity, than that of the unannealed sample. Our annealed samples in general have glow curves which resemble one or other of the curves in Fig. 1, the less severely annealed samples resembling the unannealed curve—with some variability in the width—and the more severely annealed samples resembling the annealed curve.

Figure 2 shows a plot of the peak temperature of the TL emission as a function of the temperature to which the samples have been annealed. (Peak temperature is that at which the TL emission is at a maximum.) There is a slight indication of a <20 °C increase in the position of the peak as the sample was annealed at temperatures up to 700 °C. The samples annealed at higher temperatures uniformly have TL peaks 80–100 °C higher than in the unannealed sample. We have also indicated on the plot Pasternak's data for samples of low albite from Amelia, Virginia[5–7]. Pasternak's samples were wrapped in Pt foil and annealed in He for 24 h. The agreement is striking and suggests that some change is occurring to the TL phosphor in both the feldspar and the meteorite between 600 and 800 °C. Pasternak performed X-ray diffraction studies on the Amelia albite and found that these changes in TL properties were associated with the transformation from the low-temperature (ordered) form to the high-temperature (disordered) form. However, the association was not a simple one as the TL changes occurred at faster rates than the order–disorder transformation: Pasternak suggests that the TL changes reflect crystallographic changes which precede the transformation, such as the formation

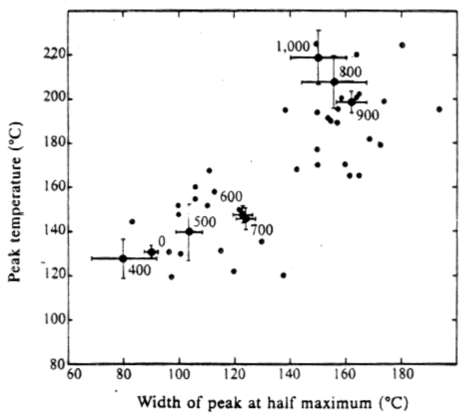

Fig. 3 Peak temperature plotted against the width of the TL peak at half its maximum intensity for the present data (large symbols) and for type 3 ordinary chondrites (small symbols, data from refs 3 and 4). Alongside the present data points are numbers indicating the temperature (°C) at which the samples were annealed for 100 h; 0 indicates the unannealed samples. Errors are $\pm 1\sigma$; two samples were run at each temperature. The meteorites show the same temperature–width relationship as the annealed samples, and the same tendency to bimodality, perhaps associated with the low- and high-temperature forms of the TL phosphor.

of defects. The low to high transformation temperature in albite or meteoritic feldspar (oligoclase in equilibrated meteorites, but unknown in others) is poorly known, but appears to be between 500 and 700 °C in albite[8]. Kinetic factors complicate the comparison of the meteorite and the Amelia anealing data with the feldspar phase diagram, but if the TL changes are associated with known crystallographic changes in the phosphor, the possibility exists that TL can be used for geothermometry.

Feldspar has not been petrographically observed in meteorites of petrologic type 3.5 or lower, except as occasional grains of anorthite usually referred to a 'primary'. However, transmission electron microscope observations of feldspar have been made in meteorites of types >3.7 and feldspar is readily observable in types 5 and 6 where it seems to have formed by the devitrification of glass. Mineral separation experiments have shown that the TL phosphor is located in the low density, feldspar-rich component in equilibrated meteorites[9]. Therefore, we have previously suggested that the TL range in type 3 ordinary chondrites reflects progressive formation of feldspar by the devitrification of glass, and that the peak shape reflects the crystallography of the feldspar[3]. The present data are experimental evidence of that interpretation.

In Fig. 3 we compare the TL peak temperature with the width of the TL peak at half its maximum intensity. The data plot in two fields, the 800, 900 and 1,000 °C annealed samples have peaks at 200–220 °C and widths of 150–170 °C, while the unannealed and <700 °C annealed samples have peaks at 130–150 °C and widths of 80–120 °C. There seems to be a correlation between peak temperature and width for unannealed and <700 °C annealed samples. A ~50% increase in width corresponds to a ~15% increase in peak temperature. The change in peak width caused by the annealing treatment is, therefore, almost as marked as the change in peak position plotted in Fig. 2. We also plot in Fig. 3 data for 39 type 3 ordinary chondrites taken from refs 3 and 4. We have omitted meteorites with TL sensitivities <0.01 because the TL-peak position and TL-peak width relationships break down for these very low TL meteorites where we suspect that a different TL phosphor becomes important. The meteorite samples display the same general trend as our annealed samples, and show a tendency for the peak width and peak position to produce two clumps which may be related to the low- and high-temperature form of the TL phosphor. The data suggest that variations in thermal history are responsible for the peak position–peak width trends observed in type 3 ordinary chondrites[3] and that while some type 3 ordinary chondrites have experienced temperatures above 700 °C others have not. The trends in the chondrites are consistent with the idea that the least metamorphosed meteorites have a low-tempe[illegible] form of feldspar as their TL phosphor. The implications of these data for our understanding of the metamorphic history of chondrites will be discussed elsewhere. However, one caveat is that our annealing times are very much less than the duration of metamorphism, but if, as we suspect, we are dealing with an equilibrium process, then laboratory measurements will be applicable. A study of the kinetics of the process is required.

Palaeothermometry of type 3 ordinary chondrites has always been a problem. The conventional approach of applying thermodynamic calculations to mineral pairs fails because the meteorites are non-equilibrium assemblages[10]. The oxygen-isotope thermometer works very well for petrologic types 4–6, but the heterogeneity problem is particularly acute for the type 3 chondrites because of the variable amounts of an ^{16}O-rich component[11,12]. We think that TL sensitivity may afford a new means of palaeothermometry in at least one rock type, the type 3 ordinary chondrites, where conventional methods fail. It might be usefully applied to other mildly metamorphosed rocks which contain feldspar as their dominant phosphor.

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1. Dodd, R. T. *Geochim. cosmochim. Acta* **33**, 161–203 (1969).
2. Sears, D. W., Grossman, J. N., Melcher, C. L., Ross, L. M. & Mills, A. A. *Nature* 287, 791–795 (1980).
3. Sears, D. W., Grossman, J. N. & Melcher, C. L. *Geochim. cosmochim. Acta* **46**, 2[illegible]–[illegible]81 (1982).
4. Sears, D. W. G. & Weeks, K. S. *J. geophys. Res. Suppl.* **88**, B301–B311 (1983).
5. Pasternak, E. S. thesis, Univ. Pennsylvania (1978).
6. Pasternak, E. S., Gains, A. M. & Levy, P. W. *Geol. Soc. Am. Abstr. with Program* 8, [illegible] (1976).
7. Levy, P. W. *PACT J.* **3**, 466–480 (1978).
8. Smith, J. V. *J. Geol.* **80**, 505–525 (1972).
9. Lalou, C., Nordmann, D. & Labyrie, J. *C. r. hebd. Séanc. Acad. Sci., Paris* D **270**, 2401–2404 (1970).
10. Dodd, R. T. *Meteorites* (Cambridge University Press, 1981).
11. Onuma, N., Clayton, R. N. & Mayeda, T. K. *Geochim. cosmochim. Acta* **36**, 157–168 (1972).
12. Mayeda, T. K., Clayton, R. N. & Olsen, E. J. *Meteoritics* **15**, 330–331 (1980).

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