## TEMPERATURE GRADIENTS IN METEORITES PRODUCED BY HEATING DURING ATMOSPHERIC PASSAGE

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Abstract—The extent to which high temperatures experienced by the surface of the meteorite during its atmospheric passage have penetrated into the meteorite may be determined by fusion crust and thermoluminescence (TL) methods. The TL results suggest luminous flight times in the order of 10 seconds, but the gradients tend to be 3-5 times less than those predicted theoretically. They appear to have the same dependence on the orientation of the meteorite as the temperature gradients determined from the fusion crust; the steepest gradients being experienced at the front of the meteorite. The fusion crust, besides enabling much of the atmospheric behaviour to be determined, provides a useful source of information complementary to TL work. For example, TL gradients produced by atmospheric heating will only be found when the fusion crust contains an innermost zone.

#### 1. INTRODUCTION

Temperature gradients are produced in a meteorite by heating during its atmospheric passage. The methods so far published for their measurement are broadly of two kinds. They are either petrographic, in which changes in structure and mineralogy resulting from elevated temperatures are used; or they involve the use of thermoluminescence, TL. (TL is the light emitted when electrons drop to the valence band from the conduction band via a luminescent centre after being thermally stimulated from "traps" in the crystal lattice.) It is necessary with the petrographic methods to use a semi-theoretical approach since the gradient is not varying linearly, but approximately exponentially. The observations that have been used tend to be metallurgical, because of the greater sensitivity of iron meteorite structures to the range of temperatures involved (Buchwald, 1961; 1967; Reed, 1972; Lovering et al., 1960; and Marringer and Manning, 1960). Some observations for stony meteorites were made by Clarke et al. (1971).

Temperature gradients are a useful complement to morphological studies since they reflect the orientation of the specimen; steeper gradients being experienced at the front of the stone. They also give a clue to the luminous flight time. Prachyabrued et al. (1971) employed their results in this way, while Vaz (1972) used the similarity in the temperature profiles of Lost City and Ucera to infer similar luminous flight times. In the work described in this

paper the reduction in thermoluminescence near the fusion crust has been examined in several meteorites. These results are compared with those obtained from an examination of the fusion crust and also with predictions from theoretical studies.

### 2. METHODS

Ramdohr (1967) has pointed out a number of mineralogical changes which occur in the fusion crust. These were discussed by Sears and Mills (1973) and led to the temperature gradient determinations shown in Table I. The gradients could be measured roughly by eye directly from the curves of temperature vs. depth, but since these curves vary nonlinearly this is impossible to do with any degree of consistency. Instead one may rearrange the ablation rate equation (see below) and differentiate it to show that

$$\frac{\partial T}{\partial y} = \frac{v_w T_i}{k} \exp\left(\frac{-y v_w}{k}\right)$$

where  $v_w$  is the ablation rate, k the thermal diffusivity  $(K/DC_x)$ , where K is thermal conductivity, D is density and  $C_x$  is specific heat), T is the temperature at depth y and  $T_i$ , the surface temperature. The temperature gradient can therefore be readily calculated at any point for which the temperature and distance are known once the ablation rate has been calculated from the ablation rate equation. The fusion crust provides the data required at each step.

The alternative method of temperature gradient determination involves the measurement of thermoluminescence at various depths below the fusion

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D. W. SEARS

Specimen	Face Type	Temperature gradient (°C/µm) <sup>a</sup>	Ablation rate (cm/sec)
BM 1966,59	Front, Close	12	0.35
BM 1966,65	Front Side, Close	7.6	0.27
BM 1966,57	Lateral, Striated	6.1	0.22
BM 1966,57	Rear, Warty	3.5	0.18

TABLE I
Temperature Gradients and Ablation Rates for the Barwell Meteorite

crust (Vaz, 1971). This results in a curve of TL vs. depth. A calibration curve may then be prepared describing the way in which TL drops when the sample is heated to various temperatures for a short period, and the TL vs. depth curve expressed as temperature vs. depth. The apparatus and the experimental techniques have been described elsewhere (Sears and Mills, 1974), but essentially consists of molybdenum filament electrically heated at  $5.0\pm$ 0.05 °C/sec by an electronic control unit. The emitted light was measured with an EMI 9635B photomultiplier tube. The powder was ground, its magnetic component removed, sieved (50  $\mu$ m) and 10 mg placed on a defined area of the filament. The light emitted was automatically plotted against temperature to produce the glow curve.

## 3. RESULTS

#### 3.1 The Barwell Meteorite

A sample of the Barwell meteorite with frontal fusion crust was used in this investigation (Leicester University Geology Department acquisition number 43801). The bar taken was at an angle of about 45° to the surface of the crust and provided thirty 1 mm thick sections of about ½ cm² area. Cutting was performed with a Metals Research Ltd. Microslice II diamond saw giving a 0.10 mm wide cut. The specimen was mounted for cutting with adhesive tape and elastic bands.

The natural TL (Figure 1) occurs in two temperature regions 200–250 °C (LT) and 300–370 °C (HT). The exact position of the peak varies, depending on the amount of overlap with adjacent peaks (Sears and Mills, 1974). The thermoluminescence of LT and probably also HT, is the result of a number of peaks and there is evidence that this is also true of lunar samples in which the TL is similarly caused by

feldspar (Hwang, 1974; Durrani et al., 1974). It is for this reason that I prefer the less precise terms LT and HT. The existence of many peaks in LT makes its behaviour difficult to interpret in terms of the commonly used theory of Randall and Wilkins (1945) which assumes single isolated peaks. A better approximation to this situation is obtained by measuring peak height, rather than area, since adjacent peaks add more to the width than to the height of the peak (see Figure 2 of Sears and Mills, 1974).

The results for Barwell are presented in Figure 2. Figure 2a shows the variation in the height of LT along the bar. The scatter is removed by expressing the height of LT relative to HT (Figure 2b) or relative to the height of LT after irradiation of the drained powder to a standard dose of about 50 krad (Figure 2c). The scatter is due to sample inhomogeneity, presumably variations in the feldspar content. The peak height ratio produces a curve with less scatter, but since HT may also vary across the specimen, could result in the gradient being slightly underestimated. This is not a serious problem here, however, since compared to LT, HT is highly insensitive to the temperatures LT is being used to measure, although it is just as sensitive as LT to sample inhomogeneity.

Having measured the drop in TL as the specimens approach the fusion crust, it is necessary to convert the reduction in TL to the temperature that is responsible for that drop. Powder from a central portion of the stone was therefore heated for five seconds at various temperatures and its glow curve was measured. The heating was performed by placing the powder on the filament of the TL apparatus and holding the temperature steady for the required time. The period of time chosen is clearly very important and is discussed below. A calibration curve of TL vs. temperature of "pre-heating" is thereby produced (Figure 3) and by converting the y-axis of the

<sup>&</sup>lt;sup>a</sup> At the depth at which nickel-iron and troilite/nickel-iron eutectics begin to flow into the meteorite matrix (Sears and Mills, 1973).

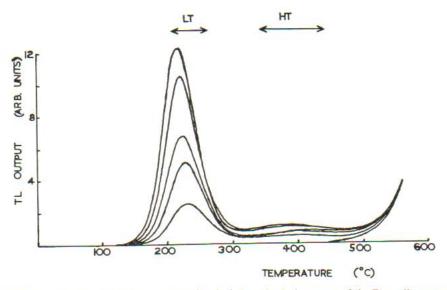


FIG. 1. Glow curves for material taken from various depths below the fusion crust of the Barwell meteorite, indicating the two regions (LT and HT) in which luminescence occurs. Note the slight movement of LT to higher temperatures as it drops in intensity. Variations in the height of HT reflect sample inhomogeneity (see Fig. 2).

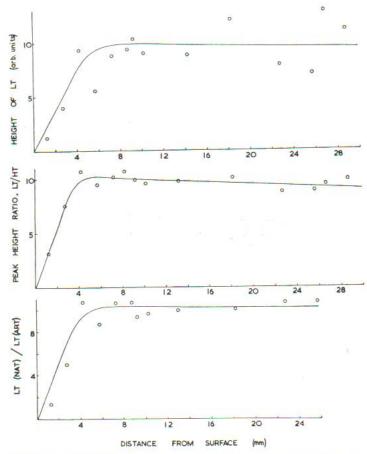


FIG. 2. Draining of the thermoluminescence by atmospheric heating near the fusion crust in a frontal specimen of the Barwell meteorite.

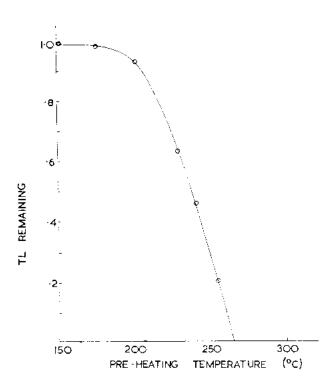


FIG. 3. The height of LT after the powder has been heated at various pre-heating temperatures for 5 seconds prior to measuring its TL. The powder was taken well from the fusion crust of the Barwell specimen.

TL vs. depth curve, and correcting for a bar normal to the surface, the temperature profile can be determined (Figure 4).

Temperatures as high as 250 °C have been experienced no further than 0.5 mm from the present surface, but temperatures exceeding 150 °C have penetrated to about 5 mm. The mean gradient at a depth exceeding 1 mm is 15-20 °C/mm.

The semi-theoretical results calculated from the fusion crust measurements are presented in Table I. Since the temperature at the troilite/troilite-nickel iron melting boundary is probably the one most reliably known it is to this boundary that the method has been applied. The results illustrate the strong dependence, at the shallow depth represented by the fusion crust, of the temperature gradient on the ablation rate. The gradients from the fusion crust and TL may be expected to differ because the fusion crust results apply to a depth of a few hundred micrometers, while the TL figures apply to depths of many millimeters and, as we shall see, the temperature drops off approximately exponentially with depth.

#### 3.2 The Allende Meteorite

In a study described by Sears and Mills (1974b) several 4 × 4 mm bars were taken from a slice of an Allende stone (NMNH 3636). The bars intercepted the fusion crust at 8 locations. I mm thick slices were prepared from each of these bars and atmosphere-induced TL gradients sought. In four cases the exceptional inhomogeneity shown by Allende obscured the gradients. However, the remainder had detectable gradients and these are presented in The temperatures are approximate Figure 5. because of the uncertainty in the average value for the meteorite (possibly  $\pm 20\%$ ) but relatively the accuracy is better. The shape of the stone suggested an orientation, although not very convincingly, and the fusion crust supported this; face d has a temperature gradient of  $5^{\circ}$ C/ $\mu$ m (for an ablation rate of 0.39 cm/sec) and face b a value of 4.1 °C/  $\mu$ m (for an ablation rate of 0.31 cm/sec). The TLmeasured temperature gradients are in agreement with these trends and support the suggestion that dis the front and b the rear of the stone.

## 3.3 The Plainview Meteorite

Two bars were removed from an approximately rectangular 4 mm thick section of the meteorite (BM 1959,805). They intercepted the crust at four locations, at approximately 90° to each other. However, a temperature gradient could be detected by TL for only one of them. In section the fusion crust always shows a well developed outermost zone, but contains an innermost (FeS/NiFe-FeS eutectic flow) zone only for the surface at which TL could detect a temperature gradient. It appears that with the exception of this one face the stone was not exposed to heat long enough to produce temperature gradients or an innermost zone, although it was able to produce a uniform coating of fusion crust.

## 3.4 The Estacado Meteorite

Temperature gradients were sought in specimens taken from three fusion-crusted edges of a 17 kg slice of the Estacado meteorite (BM 1906,259). The meteorite was badly weathered, and only HT was present in the glow curves. The fusion crust specimens showed none of the characteristic features; only large bands of limonite and altered meteorite matrix were present. No TL gradients due to atmospheric heating could be detected in this meteorite. (The higher temperatures needed to diminish the HT peak would affect only the outermost millimeter or so.)

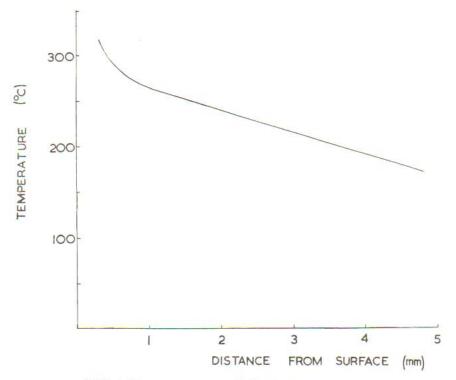


FIG. 4. The temperature profile for the Barwell meteorite.

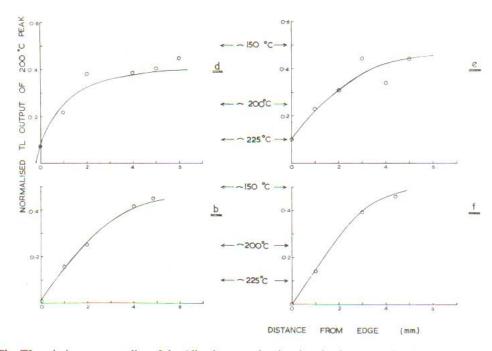


FIG. 5. The TL variation across a slice of the Allende meteorite showing the drop near the edge caused by atmospheric heating. Also shown are approximate temperatures thought to be required to produce that drop. The temperature gradients indicated differ in such a way as to infer that d is the front and b the rear of the stone.

160 D. W. SEARS

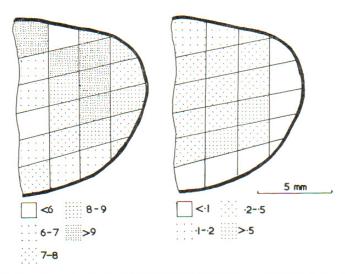


FIG. 6. The distribution of thermoluminescence across a small Holbrook stone. a (left) the intensity of the TL produced by material that has been drained and irradiated to about 300 krad. This reflects the feldspar distribution. b (right) the ratio of natural to artificial TL plotted across the slice. This describes the heat penetration. The TL has been considerably reduced near the primary fusion crust (heavy line) but not near the secondary crust. The TL intensity is in arbitrary units.

Weathering has a complicated effect on the TL of meteorites. Sears and Mills (1974) stated that it changes the peak height ratio, presumably by filtering the light from one peak more than the other. We also believe that the peak at about 250 °C, usually apparent as an inflexion on the low temperature side of HT, is due to weathering. It is only present in weathered meteorites and is more intense near the weathered crust of Estacado.

### 3.4 The Holbrook Meteorite

A 2 mm thick slice was taken from a small (5 g) completely encrusted stone (Leicester University Geology Department acquisition number 24239). The slice was too small to be cut into bars, but instead was cut into 23  $2 \times 2$  mm squares. The distribution of the natural TL, and the TL produced by irradiating the drained powder to about 50 krad, is given in Figure 6. The natural TL distribution reflects the penetration of heat into the meteorite, plus the distribution of feldspar, while the artificial TL reflects mainly the distribution of feldspar over the slice. The flat surface of the approximately hemi-spherical slice appears not to have been exposed to heat and in the hand specimen is clearly seen to be a secondary face, whereas the rest was a

primary crust. In section the secondary crust was found not to possess an innermost zone (Figure 7).

#### 4. DISCUSSION

## 4.1 Theory of Temperature Gradients and the Ablation Rates of Meteorites

The conductivity equation, describing the temperature T at depth y, can be written to include a term for ablation in the following form:

$$\frac{\partial^2 T}{\partial y^2} + \frac{v_w}{k} \frac{\partial T}{\partial y} - \frac{1}{k} \frac{\partial T}{\partial t} = 0$$

where the symbols are as defined earlier and t is time (Carslaw and Jaeger, 1959). The solution to this is

$$\begin{split} T &= \frac{1}{2} T_i \!\! \left\{ \! \text{erfc} \left( \! \frac{y + v_w t}{2(kt)^{1/2}} \! \right) + \exp \left( \! \frac{-v_w y}{k} \! \right) \text{erfc} \right. \\ & \times \left( \! \frac{y - v_w t}{2(kt)^{1/2}} \! \right) \end{split}$$

where

erfc 
$$x = \frac{2}{\pi^{1/2}} \int_{x}^{\infty} \exp(-x^2) dx$$
.

This requires considerable computation but may be simplified by considering the two cases;





FIG. 7. Fusion crust specimens of the slice of the Holbrook meteorite shown in Figure 6, as seen in reflected light. The outer 2 mm is visible. (a) A well developed primary crust showing all three zones, (b) a secondary crust with no innermost zone.

i) Steady state conditions  $(\partial T/\partial t = 0)$ ; then the temperature distribution is

$$T = T_i \exp\left(-v_w y/k\right)$$

which is the equation applicable to the fusion crust (Bethe and Adams, 1959; Adams, 1959).

ii) Zero ablation; when a steady state is not established and the temperature distribution is governed by the time during which heat flows in. Then

$$T = T_i \operatorname{erfc}\left(\frac{y}{2(kt)^{1/2}}\right)$$

The results obtained from these two equations are shown in Figure 8, which also includes the results for iron meteorites taken from Lovering et al. (1960).

4.2 The Innermost Zone of the Fusion Crust and the Existence of TL Gradients

It is now possible to understand quantitatively why the absence of the innermost zone in the fusion crust is associated with the absence of a temperature-induced TL gradient. This zone takes a period in the order of one second to form (Sears and Mills, 1973). The temperature gradients calculated above show that periods considerably in excess of this, say 10 to 100 seconds, are required to produce temperature gradients in the meteorite. A useful check to make before looking for atmosphere-induced temperature gradients in meteorites with TL would appear to be the existence of an innermost zone in the fusion crust.

# 4.3 Comparison of Theoretical and TL-observed Temperature Gradients

The temperature profiles predicted by theory and those measured by TL are compared in Figure 9. Two main observations may be made. Firstly that there is good agreement between theory and experiment in the temperatures experienced at depths of 5-10 mm (about 200 °C). From this we may conclude that steady-state conditions do not apply to the regions in which the TL gradient is caused by atmospheric heating. Instead, the TL measurements infer a luminous flight time in the order of 10 seconds, since 1 second would allow a temperature of 200 °C to penetrate less than a millimeter or so, and 100 seconds would permit temperatures this high to penetrate 20 mm. However, the Allende results show that orientation, and so presumably ablation, is still a major factor in controlling the temperature gradient.

Secondly, the gradients at depths of 5-10 mm measured by TL are always much lower than those predicted by theory. The TL results are 15-20 °C/ mm (Barwell), 20 °C/mm (Plainview), 15-20 °C/mm (Allende), and published values for Lost City are 25 °C/mm (Prachyabrued et al., 1971), and 15 °C/ mm (Vaz, 1971), The theory predicts gradients of about 70 °C/mm at these depths. This is obviously an important discrepancy to explain if the atmospheric TL gradients are to be completely understood. The major factors in the theoretical treatment are the thermal properties, for example a higher thermal conductivity would give a lower temperature gradient. However, the thermal conductivity would need to be greater than for pure iron in order to resolve the discrepency this way. Instead, it is necessary to examine more closely the TL-derived temperature gradients.

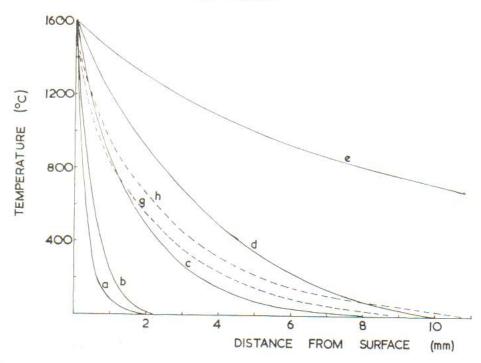


FIG. 8. Theoretical temperature gradients in meteorites. a, b, and c are steady-state solutions assuming ablation rates of 0.25, 0.15, and 0.05 cm/sec. d and e are non-steady-state results assuming 10 and 100 seconds luminous flight times respectively. The dotted lines are from Lovering et al. (1960) and are appropriate to iron meteorites; g is for a steady-state with an ablation rate of 0.18 cm/sec and h is the non-steady-state result for a 3 second luminous flight time.

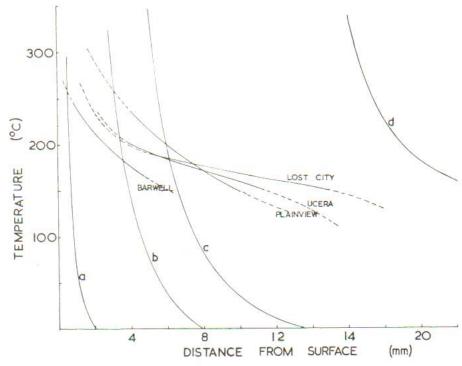


FIG. 9. A comparison of the theoretical temperature profiles with those obtained by TL measurement. a and b are for ablation rates of 0.25 and 0.05 cm/sec, c and d are for zero ablation for 10 and 100 second luminous flight times respectively. The TL-derived gradients for Lost City and Ucera are from Vaz (1971 and 1972).

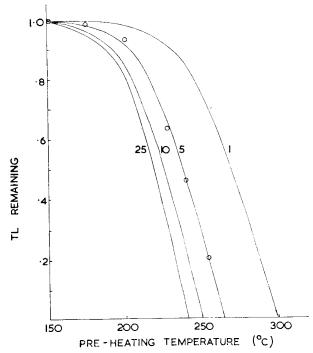


FIG. 10. The calculated height of LT after preheating to various temperatures for 1, 5, 10, and 25 seconds. The circles represent the experimental values shown in Figure 3.

## 4.4 Pre-heating Curve and Pre-heating Times

The major unknown in the TL procedure for measuring temperature gradients is the duration of preheating. This will be examined in this section. From the theory for the production of TL derived by Randall and Wilkins (1945) it can be readily shown that the number of trapped electrons remaining after heating for  $t_p$  seconds at a temperature of  $T_p$  is

$$n_o = n_i \exp(-st_p \exp(-E/kT_p))$$

where  $n_i$  and  $n_o$  are the number of trapped electrons before and after pre-heating, s and E are the trap parameters (the "s factor" and energy) and k is the Boltzmann constant (Garlick, 1949). Since the intensity of the TL is proportional to the number of excited electrons (i.e. first order decay)

$$\frac{I_p}{I} = \exp\left(-st_p \exp\left(-E/kT_p\right)\right)$$

describes the proportional drop in TL intensity after pre-heating. s is about  $10^{11} \sec^{-1}$  (Prachyabrued et al., 1971) and E is about 1.2 eV (Sears and Mills, 1974a) so that the drop in TL vs. pre-heating temperature may be calculated (Figure 10). The agreement

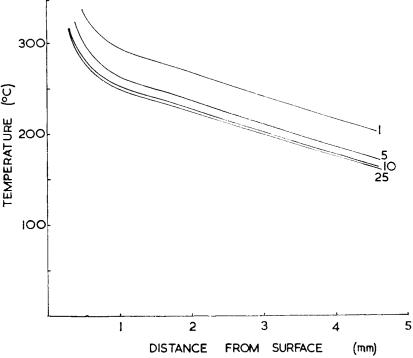


FIG. 11. Temperature profiles for the Barwell meteorite assuming various luminous flight times (and the theoretical calibration curves in Figure 10). Using different times changes the range of the temperatures concerned but does not change the gradient.

164 D. W. SEARS

with the experimental results is excellent, and lends support to the theory and the E and s values used. The need to know the pre-heating time is also emphasised by this figure. The pre-heating time for the meteorite is probably very similar to the luminous flight time.

A short luminous flight time would therefore increase the temperatures that the TL would appear to be measuring. However it would not change the value of the TL-derived temperature gradients but merely displace them vertically (Figure 11), because the slopes of the preheating curves are similar for all values of  $t_p$ .

The explanation for the difference between the theoretical and observed temperatures gradient would appear to lie clsewhere in the experimental procedure, possibly with the finite thickness of the specimens cut from the bar.

## 5. CONCLUSIONS

Although in detail there are worrying discrepancies between the theoretical temperature gradients and those measured by TL, in broad terms there is a measure of agreement. Temperatures in the order of  $200\,^{\circ}\text{C}$  have usually penetrated no further than  $5\text{--}10\,\text{mm}$ , which is consistent with a luminous flight time of the order of  $10\,\text{seconds}$ . Orientation is an important factor in determining these gradients, and they may therefore be used to complement morphological and fusion crust studies. Since the production of an innermost zone in the fusion crust takes a shorter time than a measurable temperature gradient, the existence of the former is a prerequisite to the existence of the latter. This may be used as a check prior to TL measurement.

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