Earth and Planetary Science Letters, 26 (1975) 97–104

THERMOLUMINESCENCE STUDIES AND THE PREATMOSPHERIC SHAPE AND MASS OF THE ESTACADO METEORITE

D.W. SEARS*

Departments of Astronomy and Geology, University of Leicester, Leicester (Great Britain)

Received November 21, 1974
Revised version received February 26, 1975

The thermoluminescence (TL) intensity of samples taken systematically across a large section of the Estacado meteorite has been measured. A significant proportion of the TL in meteorites is probably caused by cosmic-ray bombardment and variations in the natural TL throughout the slice reflect the preatmospheric shape of the meteorite in that plane. Before it entered the atmosphere Estacado was an elongated shape, approximating to an ellipse of eccentricity 0.8, with a preatmospheric mass of at least 8 tons. The results are consistent with laboratory experiments which indicate that secondary radiation plays an extremely important part in the production of TL.

1. Introduction

Attempts have previously been made to determine the preatmospheric shape of four iron meteorites from their spallogenic gas contents, the most successful results being obtained with the Grant and Carbo irons [1–6]. These studies not only enabled preatmospheric shapes and masses to be estimated but also gave an indication of the various nuclear parameters (reaction cross-sections, etc.) involved in the spallation reactions, and proved the general validity of the spallogenic production model of Ebert and Wänke [7]. Recently systematic measurements of the spallogenic nuclides across stony meteorites have also been made. Interpretation is considerably complicated by the existence of a wide variety of target elements in stony meteorites but from these measurements the Keyes chondrite appears to have been a 70 by 30 cm ellipsoid when it entered the atmosphere [8,9] and Saint-Séverin was probably also an elipsoid [10]. Another valuable tool for the examination of stony meteorites is provided by charged-particle tracks, which have enabled estimates to be made of the preatmospheric shape of the Saint-Séverin meteorite [11] and the preatmospheric mass of the Patwar pallasite [12].

A third cosmogenic phenomenon available for the preatmospheric shape and mass determinations is thermoluminescence (TL). This is the light emitted when electrons which have been thermally excited to the conduction band from traps in the crystal lattice fall to the valence band via a luminescent centre. Any ionising radiation is capable of producing TL and in meteorites an important source is likely to be cosmic-ray bombardment. In addition to this "cosmogenic" component, evidence for which we will discuss later, we expect an important contribution to the TL by the radioactive elements within the meteorite (a "radiogenic" component). This work concerns an attempt to use TL to evaluate the preatmospheric shape and mass of the Estacado meteorite from the TL.

2. Method

A variety of meteorites with large cut faces were sampled and their TL measured. Only the very smallest, which were recent falls, had appreciable low temperature TL, but all displayed some at higher temperatures (300–400°C). Estacado, besides presenting the largest cut face, had by far the most in-

---

* Present address: Department of Metallurgy, The University, Manchester M13 9PL, England.

tense high-temperature TL and was therefore chosen for this study. A large (17-kg) slice of this meteorite is deposited in the British Museum (BM 1906, 259). It had previously been removed, approximately centrally, from a 290-kg stone measuring 58.5 × 45.7 × 44.4 cm which was found in Hale County, Texas, in 1883. The meteorite and slice have been described by Howard and Davison [13], and recently the stone was analysed by Mason and Wiik [14]. It is an H6 chondrite [15]. A second stone weighing 122 kg and measuring 30 × 30 × 50 cm was acquired in 1912 by the American Museum of Natural History. The total known post-atmospheric mass is therefore 412 kg.

A polythene sheet on which had been drawn a 5-cm grid was placed over the slice. 2–3 mm long cores, 5 mm in diameter, were then taken over the grid using a "Diagrit" diamond drill operated in a hand brace and lubricated with distilled water. The technique had previously been checked to ensure that natural TL was not drained and that triboluminescence was not created. 72 samples were taken in this way. Additional samples were taken at the three fusion crust edges present on the slice (Fig. 1) to enable measurement of "edge-effects", but these results will not be dealt with here. The 72 samples were crushed and their magnetic component removed. They were then sieved (50 μm) and the TL measured in the normal way, duplicate measurements being made for all samples. The apparatus is described elsewhere [16,17,26]. Many samples had their TL measured repeatedly to ensure constancy in the sensitivity of the equipment. The drained material was then irradiated to about 50 krad by ^{60}Co γ-rays and its "artificial TL" recorded. The irradiated samples were allowed to decay at room temperature for 50 hours before TL measurement so that differential decay during a run was insignificant. The "temperature sensitization", caused by irradiating previously drained material, was thus assumed constant for all specimens. This point will be discussed later.

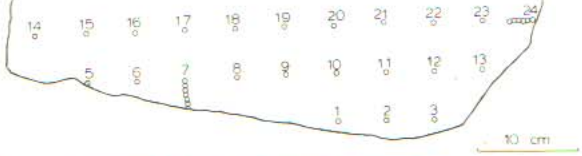

Fig. 1. Location of the specimens over the slice.

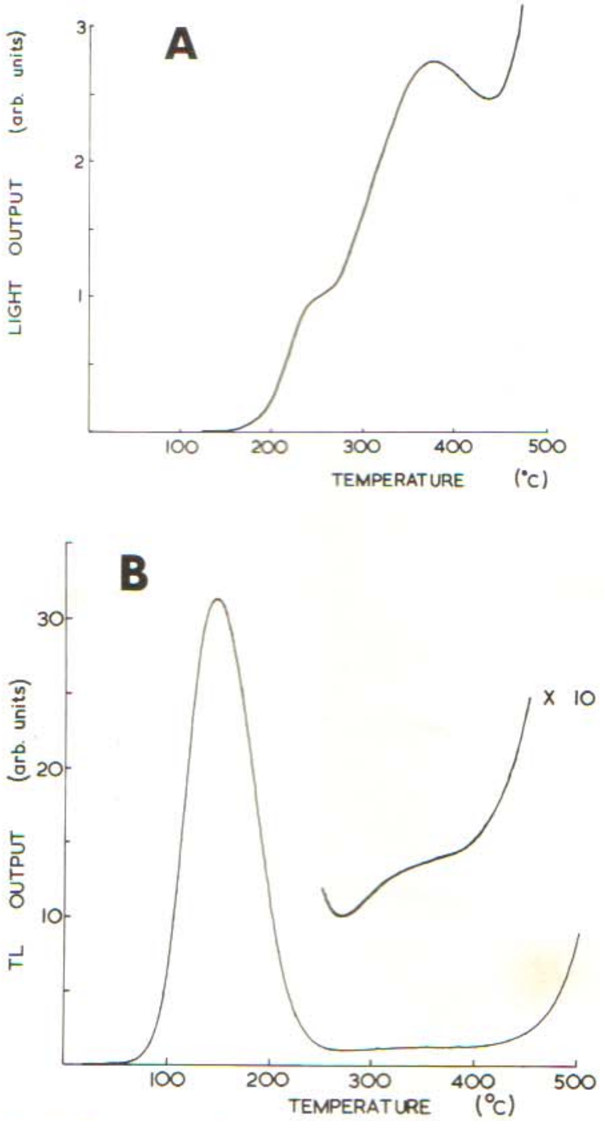

Fig. 2. Glow curves of material from the Estacado meteorite. A. A curve for natural material. B. A curve for material which has been drained of its natural TL by heating to 500°C and irradiated to about 50 krad by ^{60}Co γ-rays.

3. Results

Typical glow curves are presented in Fig. 2. The curves for the natural powder have a broad peak at about 370°C and a very weak 250°C peak, probably associated with weathering [16]. The total luminosity, i.e. the area under the curve, was measured for each curve and the results are presented in Fig. 3. The areas under the curves obtained from specimens that had been drained by heating to 500°C and irradiated to about 50 krad with ^{60}Co γ-rays were also measured. The curves of irradiated specimens have a sharp peak

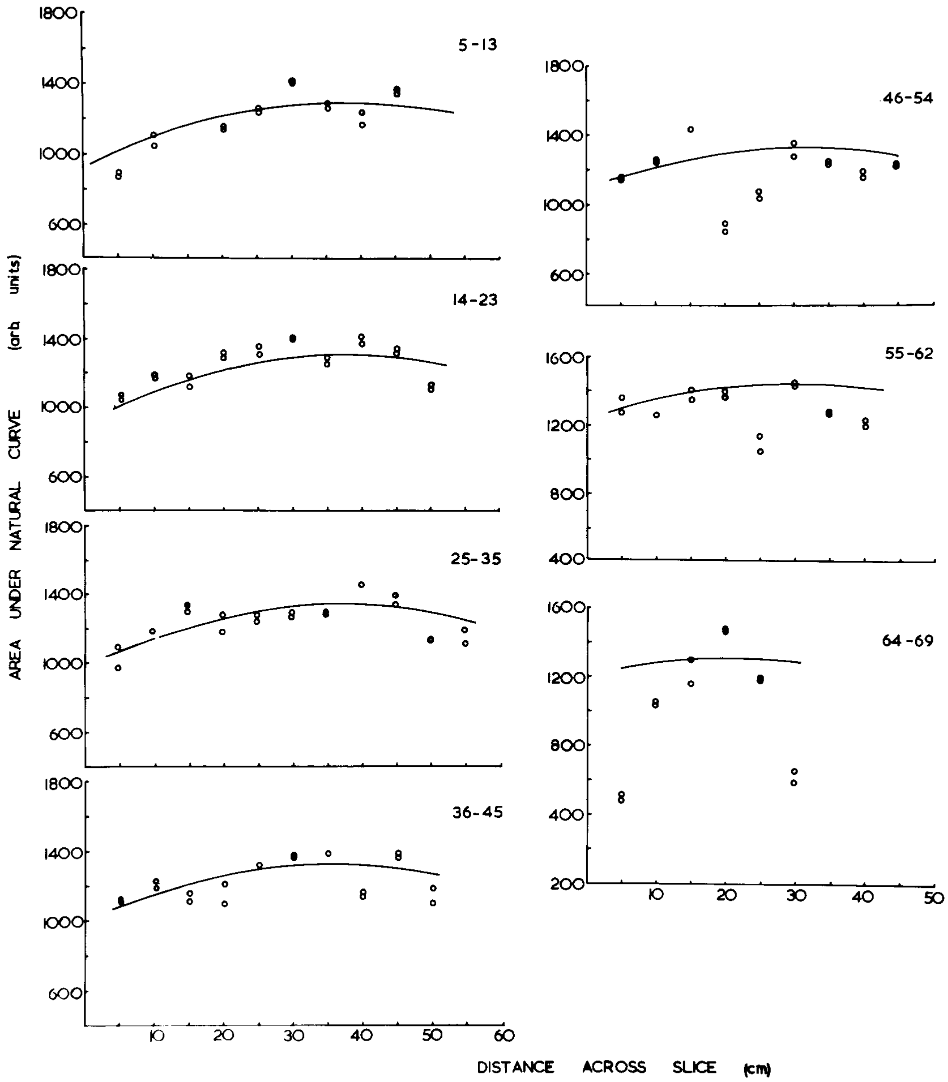

Fig. 3. Areas under the natural glow curves as a function of distance across the slice for each row of specimens. The lines are least-squares fits of second-order curve to all the data. Duplicate measurements are shown for each specimen.

[illegible] C' level?

at 140°C (LT) as well as some high-temperature TL (HT). LT is known to consist of two or possibly three peaks between 140 and 200°C [16].

The data were fed into the KWIK R8 computer program which fits a trend surface to a two-dimensional layout of data by the method of least squares [18,19]. Curves described by a polynomial equation of any given order can be fitted to the data and the position of the second-order curve along each of the rows is also shown in Fig. 3. The test coefficients for attempts to fit first, second, third and fourth-order curves to the areas under the natural curves, the areas under LT and HT in artificial curves, and the ratio of HT in the natural to HT in the artificial curves were calculated. Although the variation is not great and in places there is much scatter the correlation coefficients for curves fitted to the *natural* TL are encouraging. First, second, third and fourth-order curves have correlation coefficients of 0.70, 0.84, 0.92 and 0.94 respectively. However, for the artificial results the coefficients are low, often less than 0.5. Davis [19] points out that correlation coefficients of 0.3 have been achieved with fourth-order curves on randomly generated data. The F-test similarly indicates a significant fit has been achieved for the area under the natural curves but that the area under the artificial curves and the artificial to natural TL ratio do not give significant trends.

Since the area under the natural curves produces fits with good correlations with second, third or fourth-order curves, it is necessary to decide which best reflects the overall trend in the data and is least perturbed by spurious points. In this instance it is

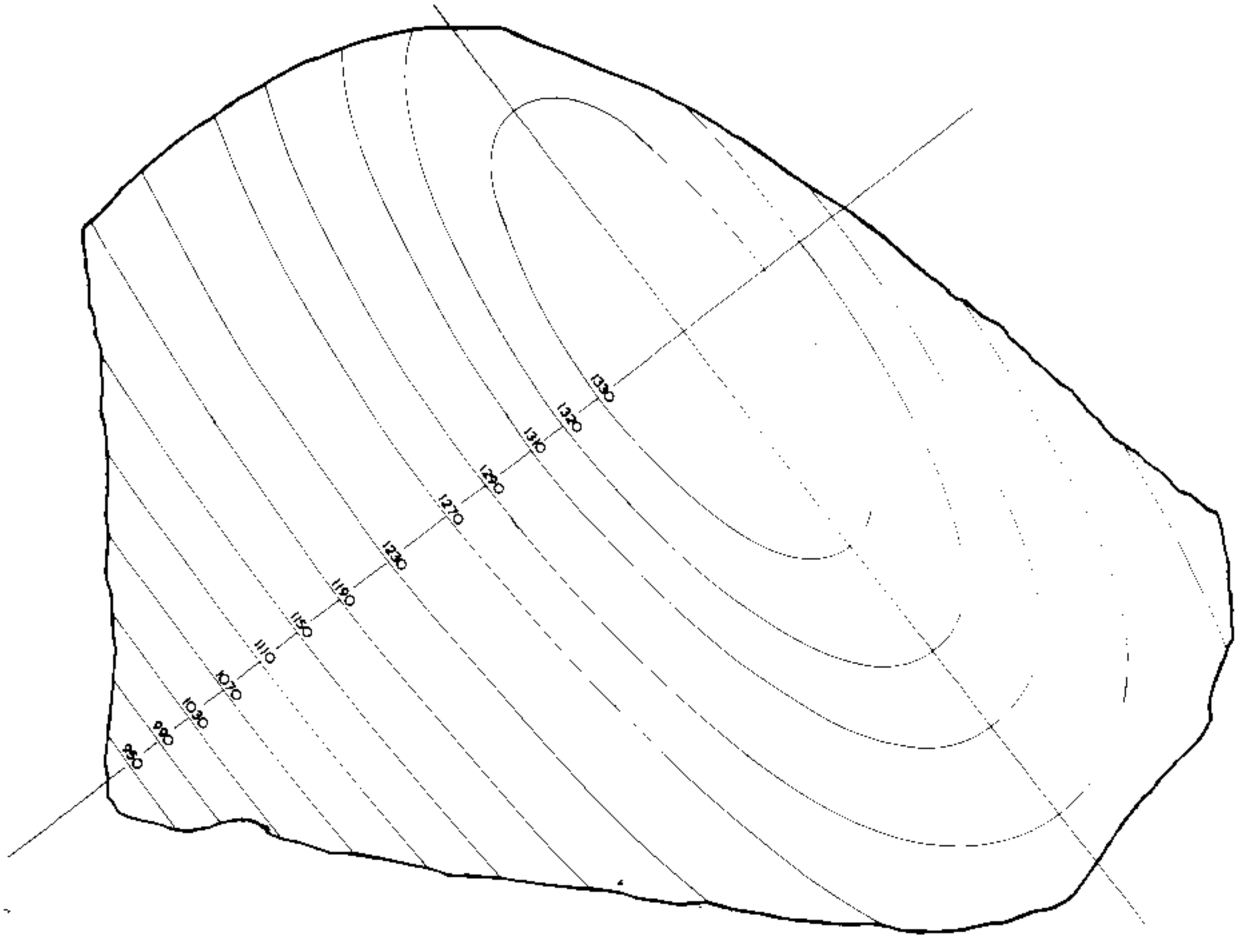

Fig. 4. The second-order fit to the area under the natural glow curves. The contours probably reflect the preatmospheric shape of the meteorite.

clear that the third-order fit is already picking up individual isolated high points, caused, for example, by local enrichment in the feldspar content. The best overall trend is therefore that given by the second-order fit (Fig. 4).

4. Discussion

4.1. Preatmospheric shape

Durrani and Christodoulides [20] have estimated that for the Allende meteorite the dose received from internal radioactivity is about 30 mrad/yr and that received from cosmic-ray bombardment to be in the order of a few rad/yr. Although these values were obtained for a different meteorite it is unlikely that the figures for Estacado differ by more than an order of magnitude. The time period involved in the former process is about 100 times greater than that involved in the latter and the total dose received in each case is probably comparable. One may therefore expect TL to have been produced by both mechanisms. Relationships between TL and exposure age [21] and TL and K–Ar age [22] have previously been investigated and, although correlations are not good, the results confirm that both cosmogenic and radiogenic TL are present. Other investigations also provide evidence for a cosmogenic contribution to the TL in meteorites. These are the systematic variations in the intensity of the TL found in the Ucera, Saint-Séverin, Allende and Plainview meteorites [23–26] which are most readily explained in terms of the production of some of the TL by cosmic-ray bombardment. One may expect the contours of equal TL intensity measured in Estacado to reflect the preatmospheric shape of the meteorite.

Some attenuation of the TL caused by *internal* radioactivity near the surface may also be expected. Although the presence of such an effect would not affect our conclusions about preatmospheric shape and minimal preatmospheric mass, it is doubtful that any signs of it survive atmospheric ablation. The depth to which this effect will appear is equal to the range of the ionising radiation concerned, and for α and β particles is negligible. The range for γ-rays, however, is of the order of a decameter, but even this will probably have been removed by atmospheric ablation since *peak* ablation rates are in the order of 2.5 cm/sec [26]. The large-scale variation observed in the Estacado slice is therefore due to cosmic-ray effects.

The only other factor which could be argued to be responsible for the contours in Fig. 4 is sample inhomogeneity. Unfortunately, the overlap of peaks caused by the combination of minerals in common chondrites made it impossible to measure the feldspar distribution by X-ray diffraction. Similarly it was impossible to assess the feldspar content chemically with the quantities of material available. However, it seems very unlikely that a feldspar enrichment over such a large area could be the explanation. Certainly there was no visible sign of a 10–20 cm xenolith in the slice. Finally, the artificially induced TL was not higher in this region, as would be expected from a feldspar enrichment.

The results obtained here show a build-up in TL intensity with depth in a similar manner to the low-energy spallogenic nuclides [8,10]. This is generally believed to be because as measurements are made further into the meteorite secondary radiations become increasingly important. Spallation reactions usually require the bombarding particles to have an energy of at least a few hundred MeV. The production of TL by the promotion of electrons to traps, on the other hand, involves an energy transition of a few eV, the size of the forbidden gap. We may argue therefore that since secondary radiations become more important for lower-energy products they will be of paramount importance for the production of TL. The only published thick target measurements for TL production are those of Lalour et al. [28] whose results also suggest that secondary radiation plays an important part. Unfortunately their results have only been presented in summary form and concern only a unidirectional beam of incident particles. Our results with the Estacado meteorite are consistent with the belief that secondary radiations are important for the production of TL.

The artificial results are a little disappointing in that neither the areas under the artificial curves nor the ratio of natural to artificial TL show a trend. The absence of a trend in the artificial TL suggests that the trap numbers do not vary monotonously across the slice, and that a significant number are not cosmogenic. Alternatively, any trend in the trap numbers may be obscured by sample inhomogeneity. If this is the case then one may expect the ratio to show a good trend, which it does not. The fact that the natural TL is that of HT is probably making the use of artificially irradiated materials unreliable in this case. HT is very difficult to measure in the glow curves of artificially irradiated specimens because of the intense LT thermoluminescence. However, there are a number of additional difficulties in using artificially irradiated materials. Some meteorite material is sensitised by being heated and re-irradiated The luminosity of Barwell, for example, was increased by a factor of eight this way [29]. This "temperature sensitisation" is little understood and may, for example, vary considerably between different specimens taken from the same meteorite. Secondly, traps may be created by the irradiation process, and not simply populated as we are assuming here. Finally, the absence of a trend may be caused by the irradiation saturating the traps with electrons. In summary, it is very difficult to isolate and remove the effects of the large number of variables to which artificial TL is subject. Consequently the presence of trends will be very difficult to detect. Fortunately, however, this is not true of natural TL.

We conclude, therefore, that the contours in the natural TL are related to the meteorite's preatmospheric shape. They suggest a non-spherical, elongated object approximating in the plane of the slice to an ellipse with an eccentricity of about 0.8.

4.2 Minimal preatmospheric mass

By extrapolating the contours so as to completely enclose the slice we may estimate the minimal preatmospheric mass of the Estacado meteorite. In the plane of the slice the minimal preatmospheric dimensions would be 130 × 70 cm. The derived preatmospheric masses are 8–13 tons, depending on whether the ellipse is rotated about its major or minor axis. A possible guide to this is the post-atmospheric shape [13] which suggests the latter value. Assuming complete recovery of the material which fell these figures correspond to 95 and 97% mass-loss. Complete recovery was almost certainly not the case, which makes this mass-loss an overestimate. Conversely, the fact that minimum dimensions are obtained means the values are underestimated.

4.3 Actual preatmospheric mass

In order to follow the Hoffman-Signer-Nier approach [1–6] for determining the actual preatmospheric mass, one needs a depth variation equation for TL in meteorites. Unfortunately no such work has been reported. However, as an initial attempt we may adapt the Ebert and Wänke [7] equation, stressing that its use here is as an empirical relationship. Then:

$$\frac{\mathrm{TL}}{\mathrm{TL}_{\mathrm{R}}} = (\mathrm{e}^{-k_{\mathrm{a}}} + B\mathrm{e}^{-k_{\mathrm{s}}})^{x-1}$$

where TL is the TL intensity at a fractional depth x; B, k_{a} and k_{s} are constants and TL_{R} is the TL intensity at the centre of the preatmospheric meteoroid ($x = 1$). The first exponential term represents the attenuation of primaries with depth and the second the build-up of secondaries with depth. Fortunately, many of the conditions assumed by Ebert and Wänke for the production of spallogenic nuclides are also applicable to TL. For example the exponential attenuation and build-up with no energy distribution change, the existence of secondary radiations of lower energy than the primaries ($B < 1$) but which are still capable of interacting, and the constancy of k_{a} and k_{s}. The main distinction between TL and spallogenic nuclide production is the contribution to the TL by internal radioactivity, and we have estimated that probably the cosmogenic and radiogenic contributions are of equal order of magnitude. No determinations have been made of the natural radioactivity of Estacado, or of the efficiency of natural radioactivity in producing TL. We will therefore derive a variety of preatmospheric masses, governed by the proportion of TL ascribed to internal radioactivity assuming this to be uniform across the slice. Although this technique will not give us a single answer it will allow some grasp of the relative importance of radiogenic and cosmogenic TL and enable limits to be fixed.

An exact fit between the theoretical and observational curves is not expected because the former is the result of a combination of exponential terms while the latter is a second-order expression. Nevertheless, a very good approximation to the TL variation along the minor axis of the ellipse (Fig. 5) is obtained by putting $k_{\mathrm{a}} = 0.35$ and $k_{\mathrm{s}} = 1.25$ and the B values in Table 1 (Fig. 6). The same values of k_{a} and k_{s} were found to fit the observational data regardless

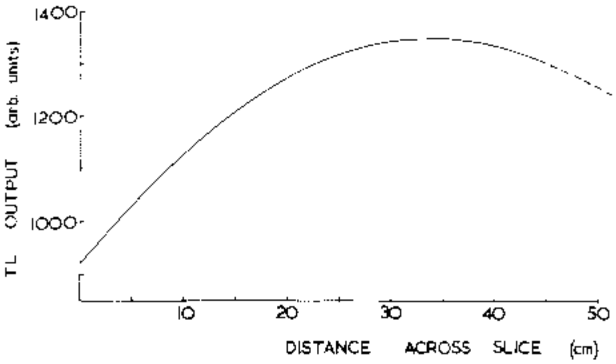

Fig. 5. The TL profile along the minor axis of the ellipse obtained by second-order fit to the data.

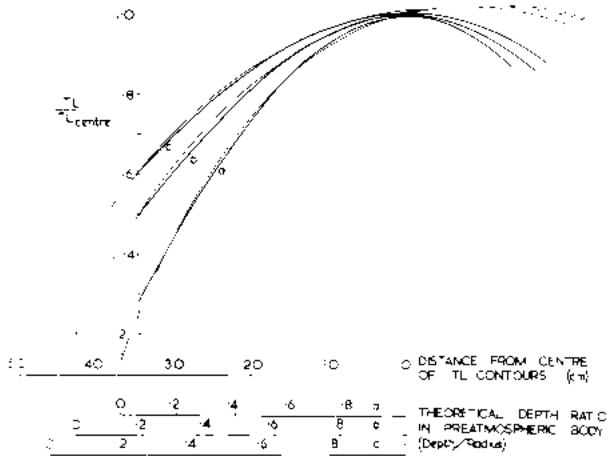

Fig. 6. The results of fitting a theoretical expression (see text) to various TL profiles. These have been obtained from Fig. 5 by subtracting 250, 500 and 750 arbitrary units of area under the curve (curves *a*, *b* and *c*) to represent various extents of contribution to the TL by internal radioactivity. The axes *a*, *b* and *c* apply to the theoretical curves.

TABLE 1

Results of fitting the Ebert-Wänke equation

Assumed radiogenic contribution*	*B*	Preatmospheric radius (cm)	Preatmospheric mass (tons)	Mass-loss (%)
250	0.89	47	20–34	98–99
500	0.92	42	14–24	97–98
750	0.95	37	10–15	96–97

* In arbitrary units of TL. The total (radiogenic + cosmogenic) TL is 959–1450.

of the amount of TL ascribed to internal radioactivity. This is presumably because, as in the spallogenic nuclide case, these values are governed by the nature of the target material. The large values of B and the relative values of k_λ and k_s are consistent with our belief in the importance of secondary radiations in the production of TL. The values for the preatmospheric radius obtained this way, and the equivalent mass ranges calculated assuming an ellipsoid of eccentricity of 0.8 are also given in Table 1. It is interesting to observe that an assumed radiogenic contribution to the TL in excess of about 800 arbitrary units of TL (i.e. just over half the maximum TL observed in the slice) would lead us to a preatmospheric radius less than the post-atmospheric radius. 800 therefore represents an upper limit to the amount of TL in this meteorite caused by its internal radioactivity. Because of the approximations in this technique, and the empirical nature of the expression (as used here) only a unidirectional beam of incident cosmic radiation has been considered and the results must be treated with some caution.

5. Conclusions

From the studies of the TL in the Estacado meteorite it appears that in the plane of the slice Estacado was an elongated non-spherical shape, probably an ellipse of about 0.8 eccentricity, and that the preatmospheric mass was at least 8 tons and may have been up to 34 tons. The major cause of uncertainty in the latter value is the unknown extent of the contribution of radiogenic particles to the TL. The results are also consistent with laboratory experiments which indicate that secondary radiation plays an important part in TL production in meteorites.

Acknowledgements

I am greatly indebted to Dr. R. Hutchison (British Museum, Natural History) for allowing me to sample the slice of Estacado, to Dr. A.A. Mills for his guidance with techniques and to Dr. S.A. Durrani (University of Birmingham) and an anonymous referee for their comments on the manuscript which resulted in a considerable improvement. I also thank Mr. C.

English and Mr. R. Wilson for their help and Professor Symons and the staff of the Chemistry Department, University of Leicester, for use of the ^{60}Co bomb.

References

1 E.L. Fireman, Distribution of ^{3}He in the Carbo meteorite, Nature 181 (1958) 1725.
2 E.L. Fireman, The distribution of helium-3 in the Grant meteorite, Plan. Space Sci. 1 (1959) 66.
3 J.H. Hoffman and A.O. Nier, Production of helium in iron meteorites by the action of cosmic rays, Phys. Rev. 112 (1958) 2112.
4 J.H. Hoffman and O.A. Nier, The cosmogenic ^{3}He and ^{4}He distribution in the meteorite Carbo, Geochim. Cosmochim. Acta 17 (1959) 32.
5 J.H. Hoffman and A.O. Nier, Cosmic-ray-produced helium in the Keen Mountain and Casas Gandes meteorites, J. Geophys. Res. 65 (1960) 1063.
6 P. Signer and A.O. Nier, The distribution of cosmic-ray produced rare gases in iron meteorites, J. Geophys. Res. 65 (1960) 2947.
7 K.H. Ebert and H. Wänke, Über die Einwirkung der höhenstralung auf Eisenmeteoriten, Z. Naturforsch, 21a (1957) 766.
8 W.R.J. Wright, L.A. Simma, M.A. Reynolds and D.D. Bogard, Depth variation of cosmogenic noble gases in the 120-kg Keyes chondrite, J. Geophys. Res. 78 (1973) 1308.
9 B.M. Trivedi and P.S. Goel, Nuclide production rates in stone meteorites and lunar samples by galactic cosmic radiation, J. Geophys. Res. 78 (1973) 4885.
10 L. Schultz, D. Phinney and P. Signer, Depth dependence of spallogenic noble gases in the St. Séverin chondrite, (abstract only) Meteoritics 8 (1973) 435.
11 Y. Cantelaube, P. Pellas, D. Nordemann and J. Tobailem, Reconstitution de la météorite Saint-Séverin dans l'espace, in: P. Millman, ed., Meteorite Research (1969) 705.
12 P.B. Price, R.S. Rajan and A.S. Tamhane, On the pre-atmospheric size and maximum space erosion rate of the Patwar stony-iron meteorite, J. Geophys. Res. 72 (1967) 1377.
13 K.S. Howard and J.M. Davison, The Estacado Aerolite, Am. J. Sci. 22 (1906) 55.
14 B. Mason and H.B. Wiik, The composition of the Richardton, Estacado and Knyahinya meteorites, Am. Mus. Novitates No. 2154 (1963).
15 W.R. van Schmus and J.A. Wood, A chemical-petrologic classification for the chondritic meteorites, Geochim. Cosmochim. Acta 31 (1967) 747.
16 D.W. Sears and A.A. Mills, Thermoluminescence and the terrestrial age of meteorites, Meteoritics 9 (1974) 47.
17 A.A. Mills, D.W. Sears and R. Hearsey, Apparatus for the measurement of thermoluminescence, in preparation.
18 J.E. Esher, P.J. Smith and J.C. Davis, KWIK R8, A FORTRAN IV program for multiple regression and geologic trend analysis, Kansas Geol. Surv. Computer Contrib. 28 (1968).
19 J.C. Davis, Statistics and Data Analysis in Geology (J. Wiley and Sons, 1973) Chapter 6.
20 S.A. Durrani and C. Christodoulides, Allende meteorite: age determination by thermoluminescence, Nature 223 (1969) 1219.
21 F.G. Houtermans and H. Liener, Thermoluminescence of meteorites, J. Geophys. Res. 71 (1968) 3378.
22 H. Liener and J. Geiss, Thermoluminescence measurements on chondritic meteorites in: D.J. McDougall, ed., Thermoluminescence of Geological Materials (1968) 559.
23 J.E. Vaz, Asymmetric distribution of thermoluminescence in the Ucera meteorite, Nature Phys. Sci. 230 (1971) 23.
24 C. Lalou, D. Nordemann and J. Labyrie, Etude préliminaire de la thermoluminescence de la météorite Saint-Séverin, C. R. Acad. Sci. (Paris) Ser. D. 270 (1970) 2401.
25 D.W. Sears and A.A. Mills, Thermoluminescence studies of the Allende meteorite, Earth Planet. Sci. Lett. 22 (1974) 391.
26 D.W. Sears, Ph.D. Thesis, Leicester University (1974).
27 D.W. Sears and A.A. Mills, Temperature gradients and atmospheric ablation rates for the Barwell meteorite, Nature, Phys. Sci. 242 (1973) 25–26.
28 C. Lalou, U. Brito, D. Nordemann and M. Mary, TL induite dans les cibles epaises par des protons de 3 GeV, C. R. Acad. Sci. (Paris) Ser. D. 270 (1970) 1706.
29 C. Christodoulides, S.A. Durrani and K.V. Ettinger, Study of the thermoluminescence in some stony meteorites, Modern Geol. 1 (1970) 247–259.