

## THERMOLUMINESCENCE AND THE TERRESTRIAL AGE OF METEORITES

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*The use of thermoluminescence (TL) to determine the terrestrial age of meteorites is investigated. It is found that meteorites can be divided into two groups. One group, in which members lose their low temperature TL rather rapidly (the "low retentivity" group), may be dated up to about 100 years after fall, although with little accuracy. The other (the "high" group) is more retentive, and may still be dated several hundred years after fall. A meteorite of unknown date of fall may be assigned to the high or low group by laboratory determination of the rate of decay of the low temperature TL.*

*Weathering coats the grains with limonite and lowers the intensity of the TL. The percentage reduction is constant for various intensities, but the peak height ratio is changed. Therefore, for weathered specimens, a method which examines the decrease in the intensity of a single peak is preferred to one which depends upon peak height ratios: this is made possible by artificially irradiating the meteorites. The following terrestrial ages for finds were obtained: Plainview 225-300 years; Dimmitt 280-330 years; Calliham 350-400 years. Bluff, Etter, Potter, Shields and Wellman (c) proved to be too old to be dated by our methods ( $\gtrsim 500$  years). None of the low group finds available to us proved to be young enough to be dated precisely. Terrestrial ages indicate an extremely low efficiency of recovery ( $\lesssim 1\%$ ) for meteorites that are not seen to fall.*

*Artificially irradiating the meteorites also revealed the fact that 9 of our 19 meteorites were saturated with respect to thermoluminescence when they entered the atmosphere, and therefore that a technique based on this phenomenon would not be applicable to such specimens to obtain their cosmic ray exposure age.*

### INTRODUCTION

The 'terrestrial age' of a meteorite is that period which has elapsed since it fell to Earth. The terrestrial ages of most meteorites are not known directly since the majority in collections are 'finds' rather than observed 'falls.' The methods so far used to determine this age (Anders, 1963; Boeckl, 1972;

Begemann and Vilcsek, 1969; Chang and Wänke, 1969) employ the decay of spallation-produced radioactive isotopes, and depend on knowing an original value for the decaying isotope. This is determined from falls of known age and assumed constant for all meteorites.

Thermoluminescence (TL) is the luminescence produced on heating a specimen. The thermal energy dislodges electrons which have been excited to "traps" in the crystal lattice by ionising radiation. The electrons drop to the ground state via a luminescent centre and in doing so emit light. TL is therefore a function of the ionising radiation received and the thermal history of the specimen (Randall and Wilkins, 1945). Groups in Berne, Paris and Birmingham (*e.g.* Houtermans and Liener, 1966; Lalou *et al.*, 1970; Christodoulides *et al.*, 1970) have studied the thermoluminescence of meteorites. The latter suggested its use for determining terrestrial age, but no rigorous examination of the possibility appears to have been made. This is the aim of the present paper.

## APPARATUS AND TECHNIQUE

Our equipment is shown diagrammatically in Fig. 1. The specimen (10 mg of 50 micron sieved powder from which any magnetic fraction has been removed) is placed on a defined area of a molybdenum strip, and heated by a high-amperage electric current at a linear heating rate of  $5 \pm 0.05^\circ/\text{sec}$ . It is important that the heating rate is strictly linear because any slight deviation can produce perfectly reproducible but nevertheless spurious "peaks" (see Fig. 2 in Houtermans and Liener, 1966). Lower heating rates improved the resolution at the expense of intensity, and  $5^\circ/\text{sec}$  was a good compromise. The emitted light was recorded with a 9635B EMI photomultiplier tube having a peak sensitivity between 3000 and 5000 Å. It was checked to give a

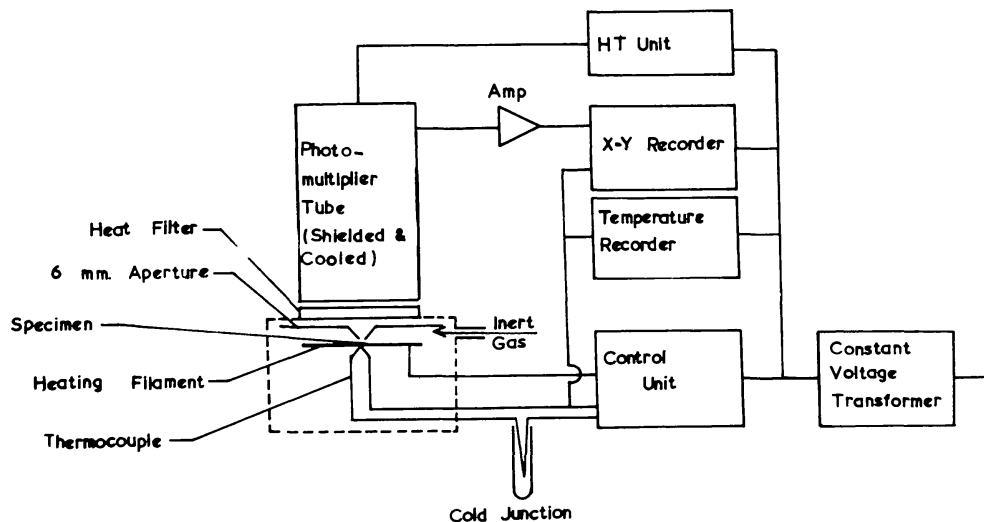


Fig. 1 Schematic diagram of the thermoluminescence apparatus.

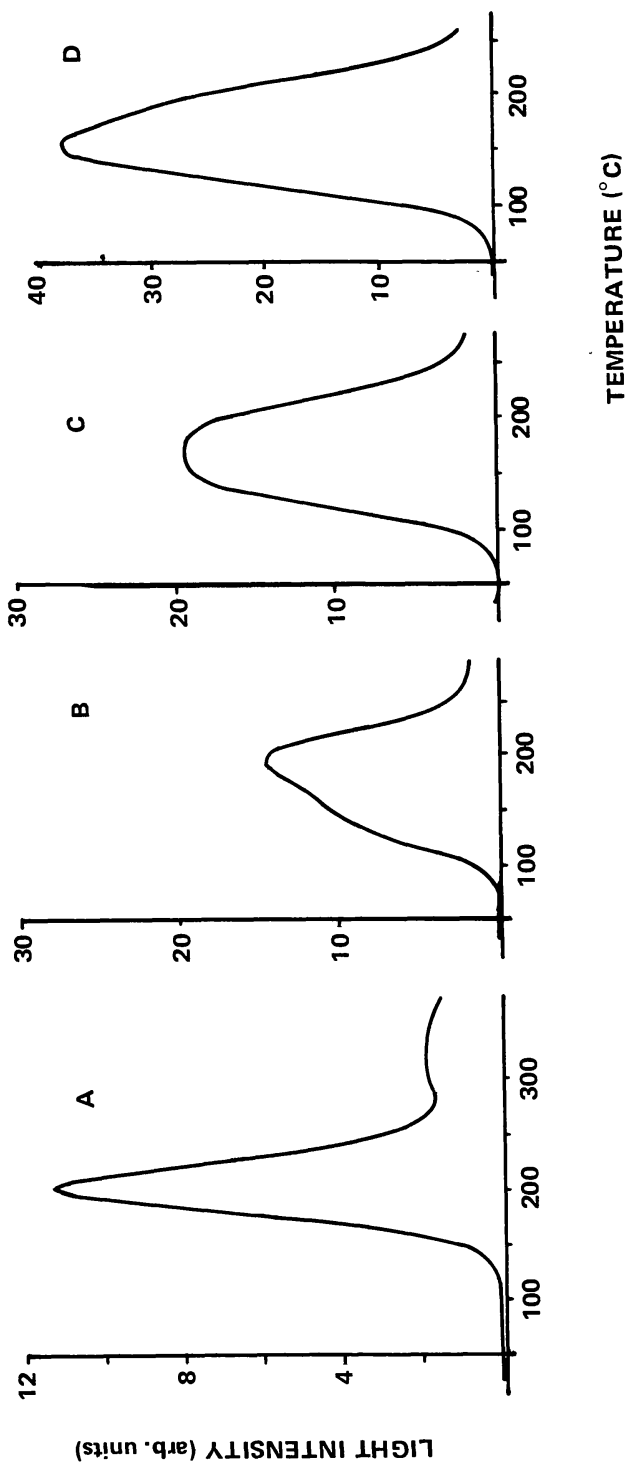


Fig. 2 The low temperature region of the Tennesilm meteorite glow curves. A) natural, B) natural + 23 krad, C) natural + 46 krad, D) natural + 69 krad. Thereafter the TL is saturated and the form of the glow curve changes little. The doses of  $\gamma$ -rays from  $^{60}\text{Co}$  bomb are accurate to  $\pm 2$  krad.

linear response at the light levels concerned. A Chance HA3 heat filter was placed between the photomultiplier tube and the specimen. The results are plotted automatically on an x-y recorder as light intensity *vs.* temperature to produce the “glow curve”. A standard lamp (a Saunders-Roe “Betelight”) is used before each set of runs to check that the sensitivity is constant. Results that are to be compared directly are obtained at one session, duplicating occasionally to ensure constancy. Irradiation is performed with a  $^{60}\text{Co}$  source delivering  $\gamma$ -rays at about 700 krad/hour, and irradiated specimens are allowed to decay at room temperature for about 30 hours. This ensures that decay during a run is minimal, as it is only relative differences in decay which will affect the results used here. Figure 2 illustrates the importance of using low doses when examining the shape of the glow curve. Irradiation to high doses enhances the lowest peak considerably more than the others until it eventually dominates the glow curve. We do not work in red light since we have failed, using both irradiated and natural meteorite powder, to detect any draining by our laboratory fluorescent lights. Where possible, specimens were taken at least two centimetres from the fusion crust to exclude material that might have experienced atmospheric heating and be already partially drained. We have checked for possible tribo-thermoluminescence induced by the mild grinding procedure, but have not found any.

## RESULTS

### *Natural thermoluminescence*

The TL of common chondrites has been shown to be due to feldspar (Lalou *et al.*, 1970). The TL occurs in two temperature regions, one at about 200 °C, *LT*, and one at about 360 °C, *HT*. The peaks vary slightly in position depending on the amount of overlap between them.

Figure 3 shows the natural glow curves of the 19 specimens of known terrestrial age used in this study. They appear to fall into at least two main groups: one in which *LT* falls below *HT* in less than 100 years, and one in which *LT* is still much stronger than *HT* after 200 years. These differences are perhaps more noticeable in Fig. 4, a plot of  $\log_e$  peak height ratio *vs.* terrestrial age. The ‘high retentivity’ group lies close to a straight line, whereas the ‘low’ group shows considerable scatter.

### *Artificial thermoluminescence*

Christodoulides *et al.* (1970) suggested a way of determining the initial glow curve. Assuming *HT* has not faded (its half-life is considerably greater than *LT*) the radiation dose that is equivalent to the present TL of a specimen can be determined. If this is then given to drained material its luminosity will be restored to the initial level. However, using the following technique, these

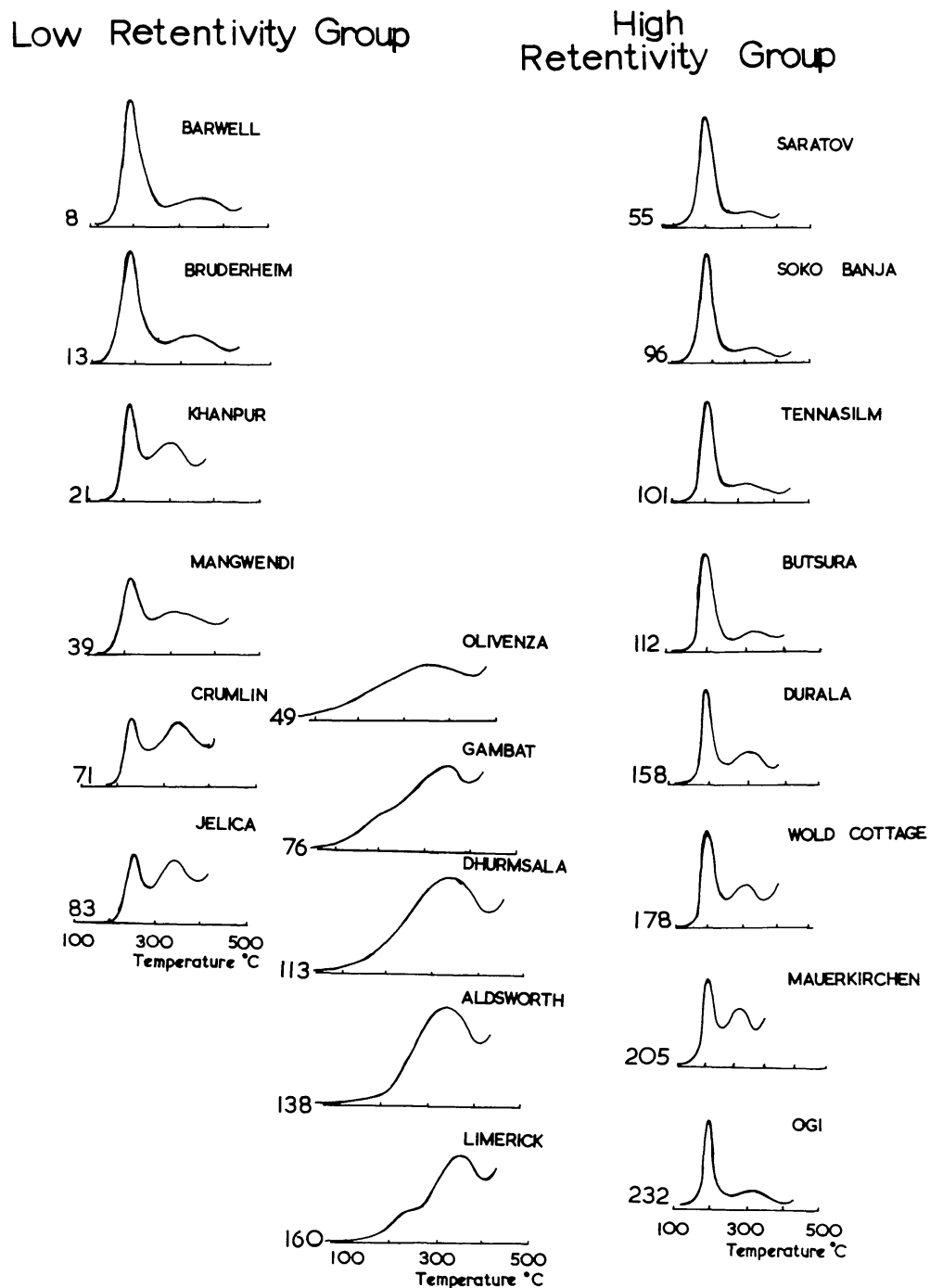


Fig. 3 Sketches of the glow curves of 19 meteorites of known terrestrial age. The numbers denote this age in years. Details of the meteorites, peak positions and intensities are listed in Table 1. The vertical axis changes slightly, the height of  $HT$  is approximately constant. The curves turn up at the right with the onset of black-body radiation.

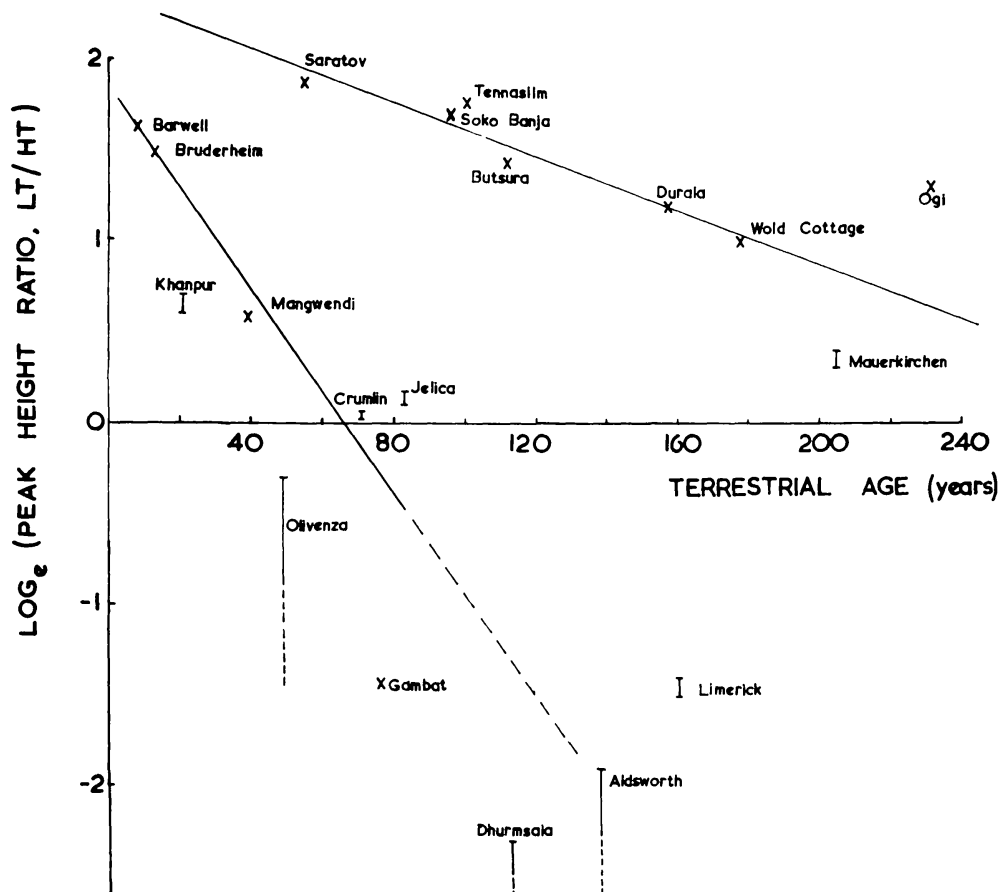


Fig. 4 Natural logarithm of the peak height ratio ( $LT/HT$ ) as a function of terrestrial age for the meteorites of known terrestrial age.

two operations can be performed at the same time and the use of previously drained material avoided, since this introduces poorly understood complications. Various doses of radiation are given to the meteorite and curves of luminosity against superimposed dose produced for each of the two peaks (Fig. 5). By extrapolating the  $HT$  line back to zero TL the initial dose may be found. The intercept of the  $HT$  line and the dose axis is assumed to equal the natural equivalent dose  $D$ . The height of  $LT$  equivalent to this dose,  $\ell_0$ , may then be determined from the  $LT$  line.

The main problem of this technique for determining  $D$  is that irradiation appears to broaden the peak and move it to lower temperatures, inferring that more than simple enhancement is occurring. This effect was explained by Liener and Geiss (1968) as being due to the irradiation times in the laboratory being so much shorter than those in space, resulting in an extra peak below 200 °C. The contribution from this extra peak was minimised by measuring height rather than the theoretically more meaningful area, since the extra peak appears to broaden  $LT$  to a greater extent than it adds height.

Figure 6 shows a plot of  $\log_e [\ell_0/\ell_{LT}]$  vs. terrestrial age. Certain meteorites (Olivenza, Dhurmsala, Aldsworth) show a very large  $\ell_0/\ell_{LT}$  ratio,

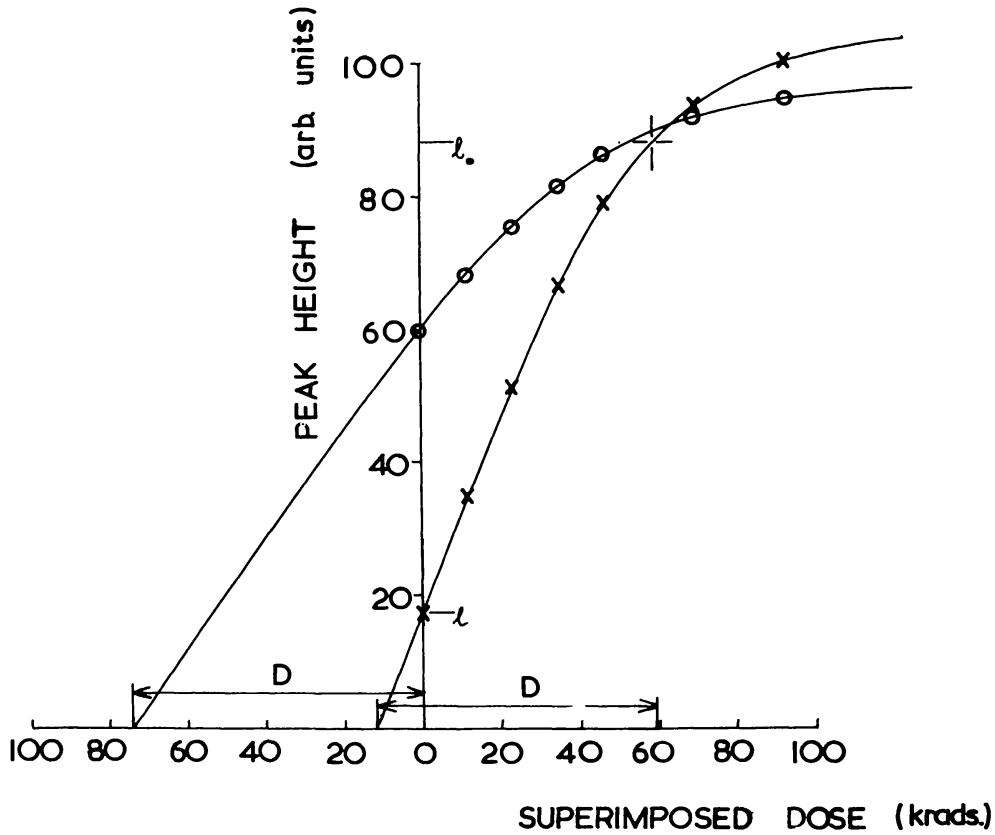


Fig. 5 Dose curves, peak height vs. superimposed dose, for the Soko Banja meteorite. Crosses denote heights of  $LT$ , circles represent heights of  $HT$  times twenty.

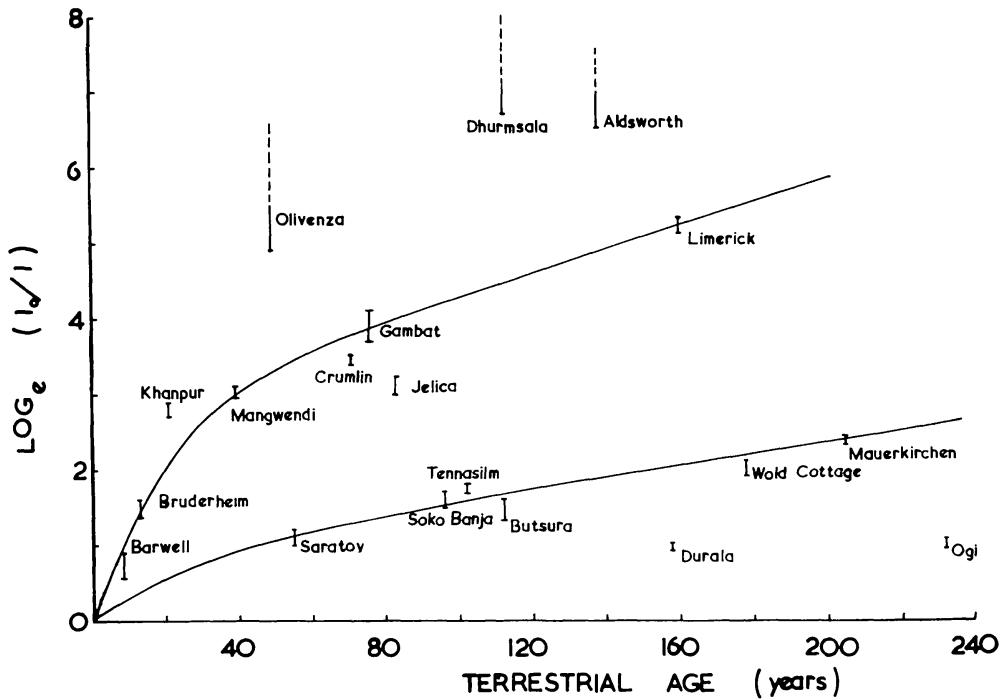


Fig. 6 Natural log of  $q_0/q_{LT}$  as a function of terrestrial age.

which together with the natural curves, Fig. 3, hints at a third group of meteorites.

It is interesting to note that many of the meteorites examined here had saturated TL when they entered the atmosphere; that is, the natural dose is on the plateau of the peak height vs. dose curve, Fig. 5, Table 1. This increases the accuracy with which  $\ell_0$  can be measured by decreasing its dependence on dose. Valladas and Lalou (1973) pointed out that it also means that these meteorites cannot be used to determine cosmic ray exposure age.

#### *Determination of terrestrial age*

The next step is to determine how a find can be assigned to one or other of the main groups, so that its terrestrial age can be found. No difference, within our experimental limits, was apparent in  $LT$  position for high or low group meteorites, or in the energy of the peak as determined by the initial rise method, Table 1 (Garlick and Gibson, 1948). However, the experimentally determined  $\tau$  (the mean-life, time to fall  $1/e$ ) appeared to be different for meteorites in the high and low retentivity groups, Fig. 7. Using three meteorites from each group,  $\tau$  at  $120^\circ\text{C}$  was determined by annealing the powder in the dark for up to 70 hours in an Abderhalden drying pistol surrounded by refluxing tetrachloroethylene. A temperature of  $120^\circ\text{C}$  was chosen because it should readily disclose if the main difference in Fig. 6 was due to different mean-lives. The values of  $\tau$  at  $120^\circ\text{C}$  are given in Table 1 and indicate room temperature mean-lives of 6-10 years for the low group and about 50 years for the high group. These results suggested it was possible to assign a meteorite to one of the two groups on the basis of the mean-life of its low-temperature TL peak.

The cause of the two groups is uncertain. However, the low group contains many meteorites which are breccias or have been shocked, and this suggests that shock may be responsible for the division (Sears and Mills, 1974).

Several finds were then examined, their descriptions and mean-lives being given in Fig. 8 and Table 2. It is apparent that Plainview, Calliham and Dimmitt are very young since they still retain a high peak height ratio  $LT/HT$ . In Table 3 those in the high group are given an age from their peak height ratio using Fig. 4. We also present their  $\log_e[\ell_0/\ell_{LT}]$  terrestrial age values. The disagreement between the ages found by each method is at first sight unexpected. However, the possibility that it was associated with the degree of weathering the finds had received was examined in the following way. We placed a piece of the Barwell meteorite on damp filter paper for two weeks. After this treatment it had acquired localised rusting but not an overall brown coloration. This sample, and a piece that had been kept dry, were prepared in the same manner for TL measurement, irradiated, and the dose curves produced, Fig. 9. As expected, the TL from the rusted specimen had



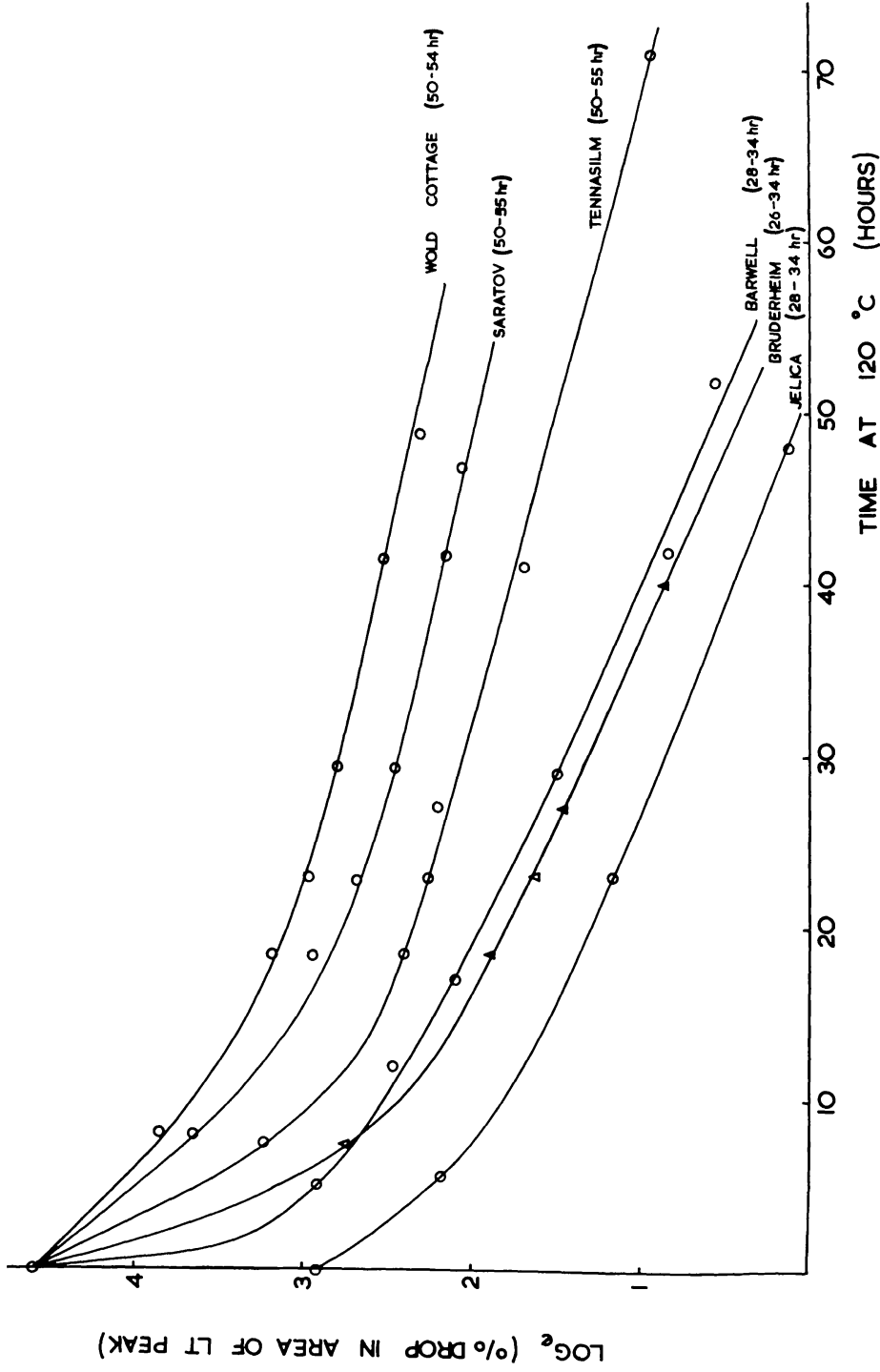


Fig. 7 Isothermal annealing curves for three high and three low group meteorites. The abscissa is the natural log of the percentage drop in the area under half *LT*. Overlap with *HT* has been allowed for where necessary. The curve for Jelica has been displaced vertically for clarity.

Table 1  
 Details of the thermoluminescence of meteorites of known terrestrial age

Group <sup>(1)</sup>	Terrestrial Age in years	Natural Peak Height Ratio	Natural Curve Peak Temperatures, °C	$E_{LT}^{(2)}$ (eV)	Mean-Life at 120 °C in hours	Natural equivalent dose in krad	$\rho_0^{(3)}$ in arbitrary units
Saratov	55	6.14	200	320	50-55	46-49	25-28
Soko Banja	96	5.5-5.3	190	320	50-55	70-81	80-88
Tennasilm	101	5.85-5.7	205	330	50-55	134-145	65-70 sat. <sup>(5)</sup>
Butsura	112	4.17	190	235		52-120	45-58 sat.
Durala	158	3.3-3.26	180	320		81-105	84-86 sat.
Wold Cottage	178	2.78-2.72	205	345	50-54	58-250	87-97 sat.
Mauerkirchen	205	1.37-1.54	220	365		75-230	96-98 sat.
Ogi	232	3.67-3.63	200	345		81-105	57-61 sat.

Barwell	L6	8	5.12	200	360	1.24	28-34	15-27	40-60
Bruderheim	L6	13	4.44	190	335	1.18	26-34	70-75	50-60
Khanpur	LL5	21	2.0-1.8	205	330			105-116	108-112
Mangwendi	LL6	39	1.78	210	335			116-140	110-130
Olivenza	LL5	49	≤0.74	(225) <sup>(6)</sup>	375			96-116	102-104 sat.
Crumlin	L5	71	1.06-1.02	225	350	0.86		145-155	92-95 sat.
Gambat	L6	76	0.237	220	320			35-46	9-12
Jelica	LL6	83	1.19-1.12	230	395	1.42	28-34	38-42	75-85
Dhumsala	LL6	113	0.1-0.0	(225)	345			52-150	80-130
Aldsworth	LL5	138	0.15-0.0	(225)	340			170-180	105-115
Limerick	H5	160	0.26-0.24	235	345			43-49	43-48 sat.

## Footnotes:

- (1) As defined by Van Schmus and Wood (1967).
- (2) As determined by Initial Rise Method (Garlick and Gibson, 1948).
- (3) See text for definition.
- (4) Two determinations using different curves.
- (5) These meteorites are saturated at the natural equivalent dose, *i.e.*, additional dose makes no difference to  $\lambda$ .
- (6) Where no peak is obvious, intensity of  $LT$  is measured at 225 °C.

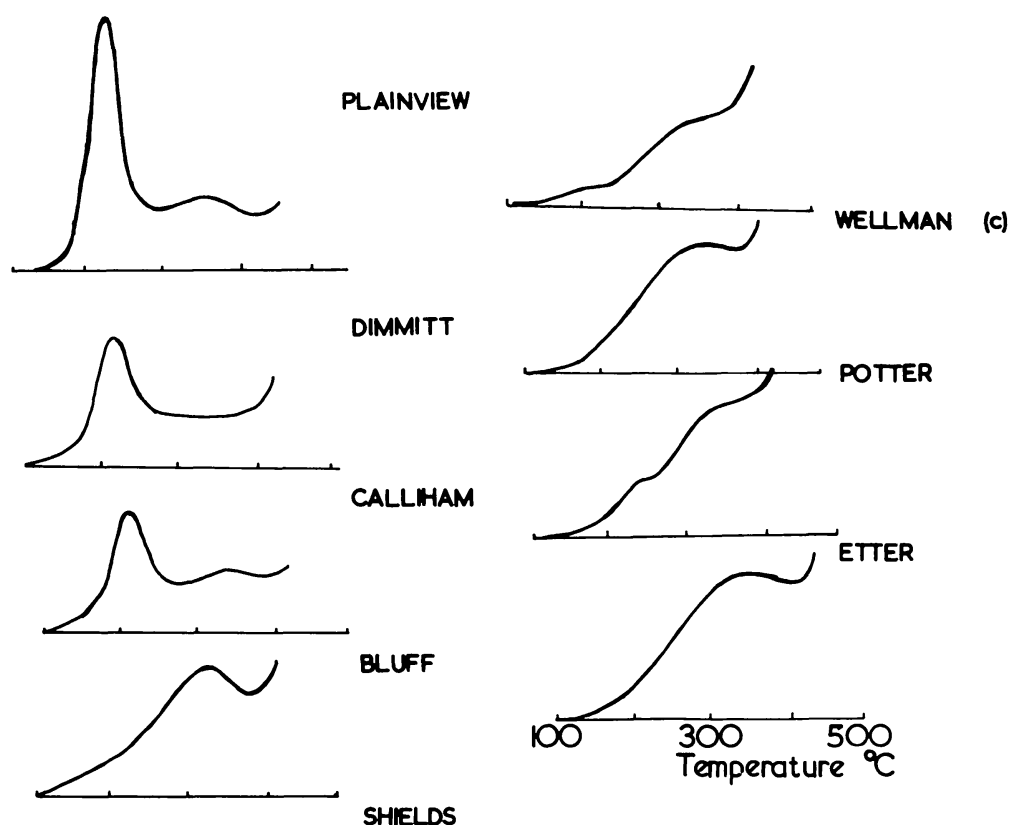


Fig. 8 Sketches of the glow curves of several finds. Details of the meteorites, peak positions and intensities are given in Table 2. The curves turn up at the right with the onset of black body radiation.

dropped. The percentage reduction was the same for specimens which had received various doses, the intensity of the rusted specimen being always about one-third that of the fresh specimen. However, the peak height ratios were clearly different, being 24, 31 and 39 for rusted specimens after 23, 46, 69 krad of  $\gamma$ -radiation compared with 16, 27 and 31 for fresh specimens. That is, the meteorite that had been allowed to rust appeared younger on the basis of its peak height ratio. Rusting has little effect on the  $\ell_0/\ell_{LT}$  values since the drop due to obscuration appears to be approximately constant at all intensities. The obscuration seems to be a selective filtering effect, varying with wavelength of the TL peak. Valladas and Lalou (1973) have recently shown that *LT* and *HT* have different coloured luminescence. Therefore, the first column in Table 3, where the peak height ratio is employed to date the meteorites, is not considered reliable. We prefer the results determined by  $\ell_0/\ell_{LT}$  and use them in Table 4.

#### ACCURACY AND SOURCES OF ERROR

The measurement of peak heights is complicated by the fact that many peaks are present and these usually overlap. The presence of peaks below

Table 2  
 Details of the thermoluminescence of meteorites of unknown terrestrial age

Group	Year found	Natural Peak Height Ratio	Natural Curve Peak Temperatures, °C		Mean-Life at 120 °C in hours	Natural equivalent dose in krad	$\lambda_0^{(3)}$ in arbitrary units
			LT	HT			
Plainview	1917	3.7	220	365	39-52	55-65	31-35
Calliham	1958	2.95-2.35	210	350	40-60	145-165	7.5-8.5
Etter		0.25	(225)	330	45-55	20-24	3.2-3.8
Potter	1941	0.48-0.40	250	365	20-28	26-30	6.6-7.7
Bluff	1878	0.05-0.15	(225)	335	22-30	22-26	4.0-5.0
Dimmitt	1947	3.5-3.0	220	315-340	45-55	86-100	5.0-6.0
Shields		0.13-0.28	225	345	50-54	36-40	8.0-8.5
Wellman(c)		0.43-0.17	(225)	310	40-50	9-20	4.0-9.0

See Table 1 for footnotes.

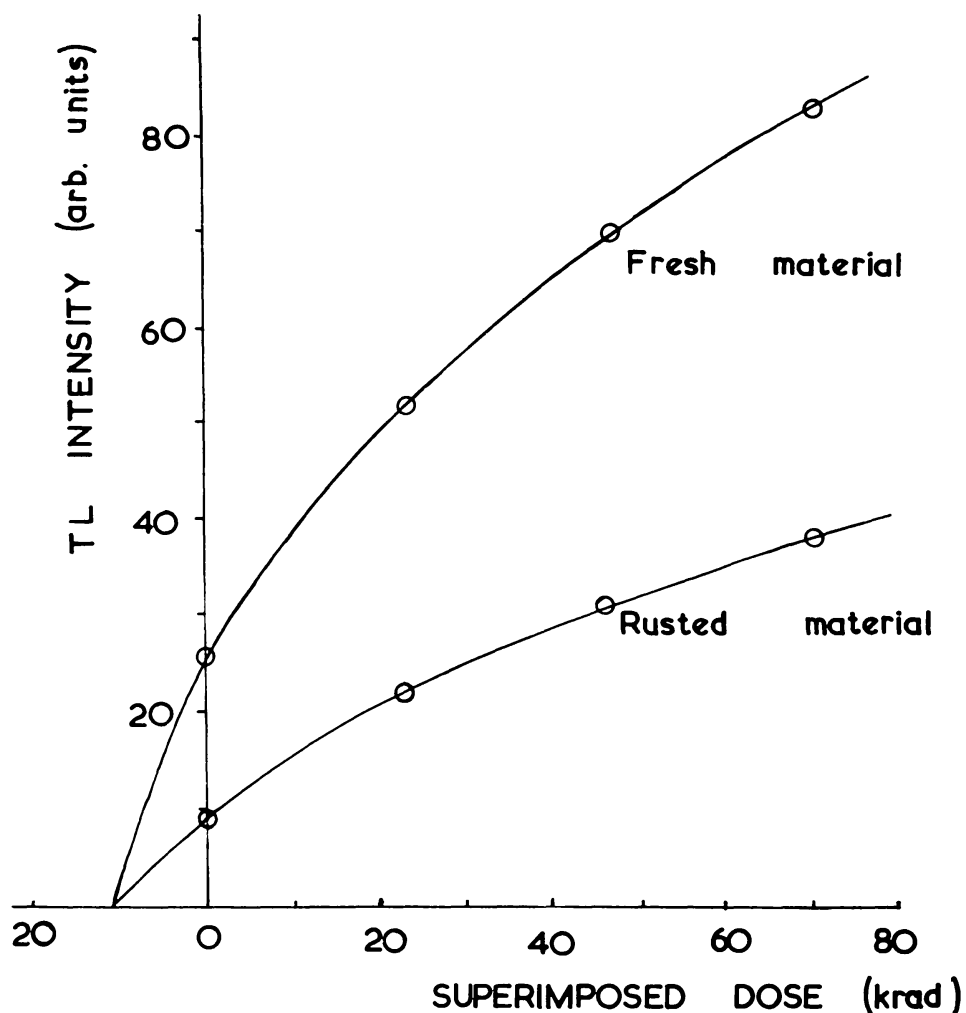


Fig. 9 The effect of rusting on the dose curve. Both curves are for the Barwell meteorite. The rusted specimen was produced by being left on wet filter paper for two weeks.

Table 3  
The terrestrial ages of stony meteorites in  
Table 2 as determined by two thermoluminescence methods

Meteorite	From peak height ratio and Fig. 3	From $\log_e(\ell_0/\ell_{LT})$ and Fig. 8
Plainview	135-145	225-300
Calliham	170-210	350-400
Etter	$\geq 300$	$\geq 350$
Potter	—	$\geq 300$
Bluff	—	$\geq 200$
Dimmitt	140-170	280-330
Shields	$\geq 300$	$\geq 500$
Wellman (c)	$\geq 300$	$\geq 350$

**Table 4**  
**Thermoluminescence ages of stony meteorites compared**  
**with radiocarbon ages**

Meteorite	TL age in years	Radiocarbon age in years	Ref.
Plainview	225-300	$\leq 2000$	a
		$\leq 3800$	b
Calliham	350-400	—	
Etter	$\geq 350$	—	
Potter	$\geq 300$	$\geq 20,000$	a
		$\geq 21,000$	
Bluff	$\geq 200$	1300-6500	c
Dimmitt	280-300	0-4000	a
		1200-6400	c
Shields	$\geq 500$	—	
Wellman (c)	$\geq 350$	—	

References: a. Suess and Wänke (1962)  
 b. Goel and Kohman (1962)  
 c. Boeckl (1972)

200 °C is evident in three ways: i) the dose curves are occasionally supralinear, Fig. 10; ii) the shape of some glow curves clearly show its presence, Fig. 2; iii) there is a shift in the low temperature peak height *vs.* peak temperature, Fig. 10. Hwang (1973) has argued for the existence of two peaks below *LT* in lunar samples using completely different evidence. However, these low-temperature peaks should not affect the results because they are caused by the same mineral as the 200 °C peak and are present in both falls and finds. The curves derived from falls therefore include any effects from these extra peaks. However, this may not be true of extra peaks at higher temperatures than *LT*. In achondrites, Fig. 12, and lunar samples (see, for example, Hoyt *et al.*, 1970) there is a peak at 260 °C which may be confused with *LT* when it occurs in chondrites. Only one of our 28 samples has a peak at 260 °C; namely Potter, which is also the most highly weathered meteorite. Overlap may move *LT* from 200 °C, *e.g.* Barwell, to 235 °C, *e.g.* Limerick, as illustrated in Fig. 10. However, confusion with a possible peak at 260 °C may be avoided by careful observation of the temperature of *LT*.

The error bars on the  $\ell_0/\ell_{LT}$  values in Fig. 6 are determined from the measurement of  $\ell_0$  and  $\ell_{LT}$ . The error in  $\ell_{LT}$  is determined from duplicate curves, where the repeatability was about 5%.  $\ell_0$  usually contains greater error and is determined from the dose curves, Fig. 5. It is governed by the accuracy by which the lines may be extrapolated to zero height to find the natural

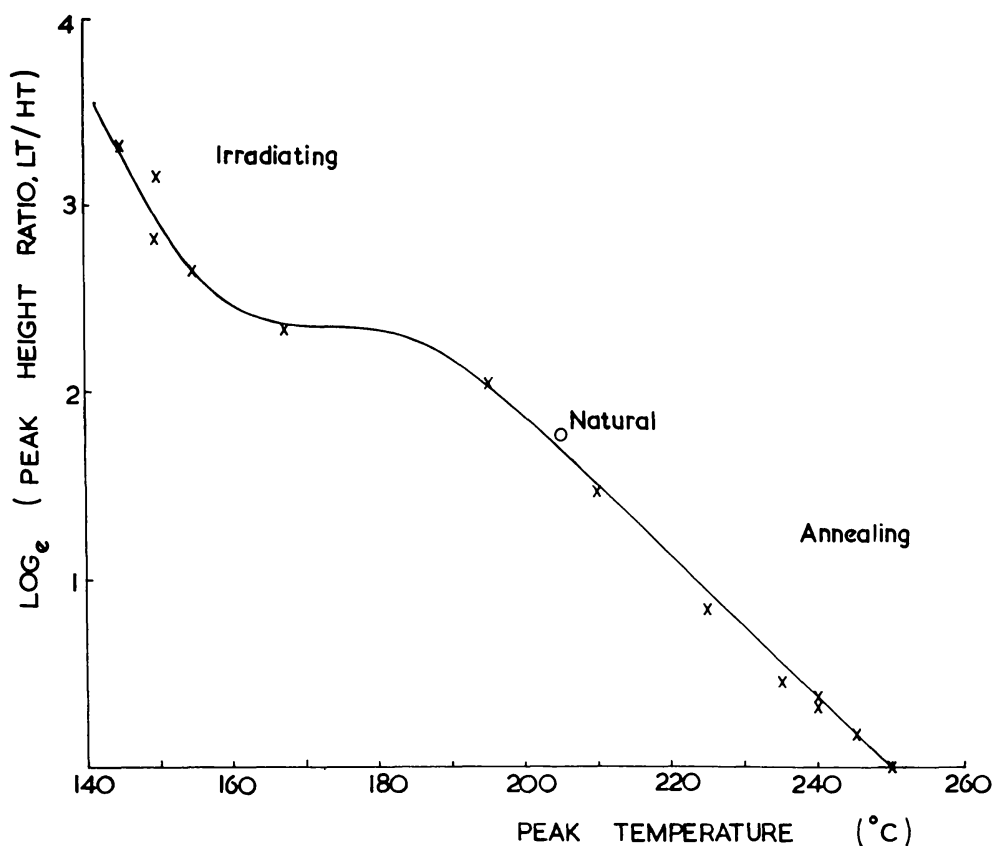


Fig. 10 Natural log of the peak height ratio vs. the position of the peak of *LT*. The movement of the peak due to overlap with *HT* is monotonous. The discontinuity at 170°C is caused by the appearance after irradiation of a discrete second peak at lower temperatures.

dose. For saturated meteorites, where the natural dose was on the plateau of the peak height vs. dose curve, Fig. 5,  $\ell_0$  may be determined very accurately.

The error bars have been slightly underestimated since there is another source of error which cannot be determined, namely the different terrestrial histories of the meteorites. This is mainly the temperature at which the meteorites have been kept, since the decay rate is temperature sensitive. The effect is probably less for meteorites in the high group because of their longer mean-lives. Six of the eight high group meteorites fall on or very near the line and we think, therefore, that this line is reasonable. Ogi displays anomalously high present TL in both Fig. 6 and Fig. 8. Abnormal histories, *e.g.* heating on a hot-plate after cutting, probably account for the remaining discrepancies. The scatter of the low group may be caused by additional factors, but clearly the use of TL to determine accurately the ages in this group will be difficult. To allow for finds that have suffered appreciably different temperatures we have made a generous allowance for the scatter on the curves in determining the age ranges in our results (Table 4).



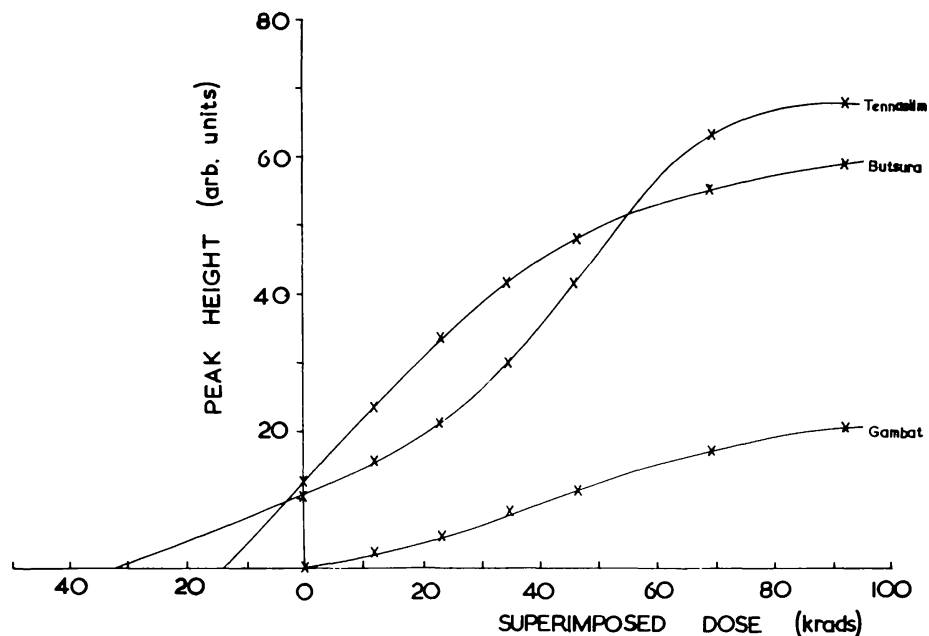


Fig. 11 Dose curves, peak height vs. superimposed dose, for *LT* for meteorites. Tensasilm shows abnormally bad supralinearity, a few have slight supralinearity like Gambat, but most have curves in which the slope is initially linear like Butsura.

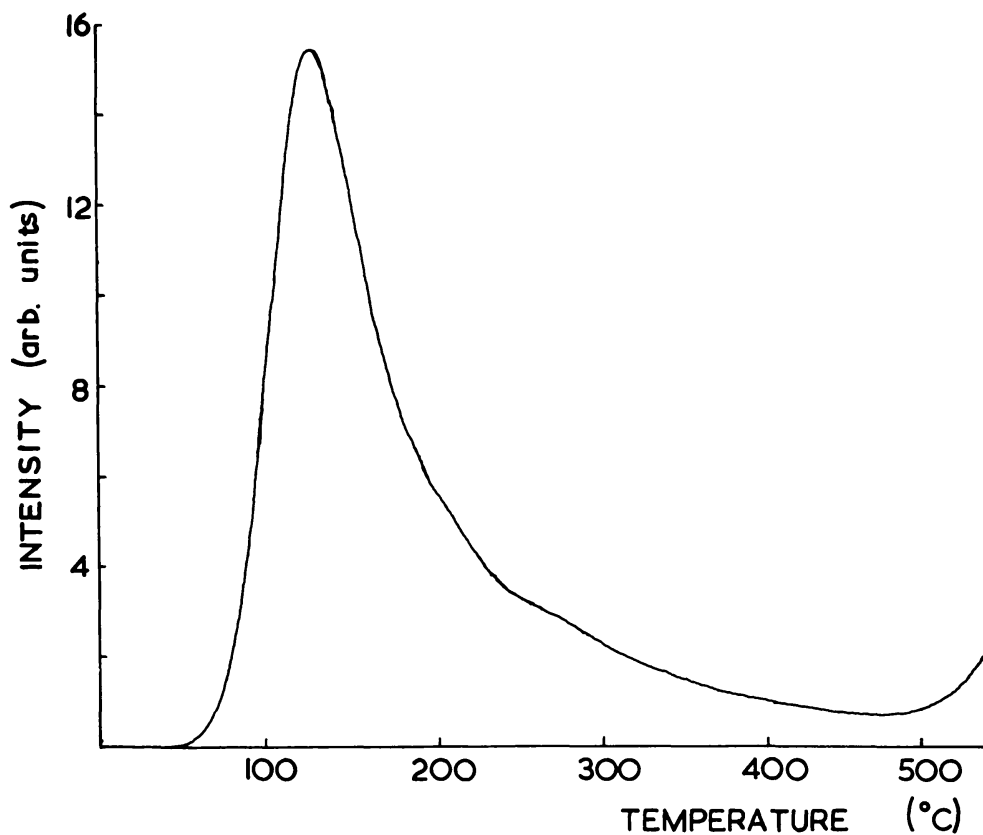


Fig. 12 Glow curve for the Kapoeta pyroxene-plagioclase anchondrite.

## DISCUSSION AND CONCLUSIONS

The only method for determining the terrestrial age of stony meteorites, prior to that investigated here, utilises the decay of carbon-14. It is applicable only to ages exceeding a few thousand years, because the half-life of carbon-14 is 5760 years and the initial value for the abundance of the isotope is not known directly. TL makes available a method for the few-hundred-year range for certain meteorites and enables a lower limit to be established for others.

Dimmitt, Plainview, Potter and Bluff have been dated by the carbon-14 method, and the values so obtained are compared with those from TL in Table 4. Plainview is an interesting case since a fireball was seen and a stone recovered in 1902/1903 (Nininger, 1952). However, at least two more finds have been made in the Plainview area, and it is not certain which belongs to the observed fireball. Our result suggests that the Plainview meteorite we examined (Plainview 1917) was not associated with the 1902/1903 fireball.

The carbon-14 method has produced two surprising results of terrestrial ages in excess of 10,000 years, *viz.*, Potter and Woodward County (Suess and Wänke, 1962; Goel and Kohman, 1962). Boeckl (1973) found the average results for 19 terrestrial age determinations to be about 5000 years. The results presented here show that, although terrestrial ages of the order of a few thousand years may be common, a significant number may fall in the few hundred year range.

From these results we may estimate the efficiency of recovery of finds, assuming the meteorite influx derived from observed falls is correct (Hawkins, 1960; Brown, 1960). The surface density of finds can be calculated for the Prairie states from, for example, the compilation in Mason's (1962) book. Hawkins meteorite flux equation

$$N = N_0 - \log m$$

where  $N$  is the number of meteorites falling per unit area per unit time of mass equal to or greater than  $m$ , and  $N_0$  is a constant, can be written

$$\frac{n}{A\beta T} = N_0 - \log m$$

where  $n$  is the total number of stony meteorites actually found in the Prairie states (76),  $A$  is the land area concerned,  $\beta$  the efficiency of recovery, and  $T$  the typical terrestrial age. Numerous factors control  $\beta$ ; the population, soil colour, land use, climate and history of the region for example. The rate of recovery (and therefore  $\beta$ ) is probably higher in the Prairie states than anywhere else in the world. Consequently, most of the stones for which we

have terrestrial ages were found in the Prairie states. Accepting typical terrestrial ages for stones from this area as a few thousand years we find  $\beta$  to be  $\lesssim 1\%$  (Table 5). A similarly low  $\beta$  is found for irons, but we will here restrict ourselves to a discussion of stones. Brown's results give a slightly lower meteorite flux but make no significant change to the  $\beta$  estimated here. This figure may be underestimated if the influx rate is lower than the flux equation predicts. That this is so is suggested by the meteor camera network operated in the Prairie states. After eight years' operation the Prairie Network had witnessed only five meteorite-sized objects, whereas 1.5 per year were predicted by the influx statistics (McCrosky *et al.*, 1971). The true influx of meteorites would therefore seem to be lower than the presently accepted statistics suggest.

### ACKNOWLEDGEMENTS

Eleven meteorites were supplied by Dr. R. Hutchison (British Museum, Natural History). For these and numerous helpful discussions we are extremely grateful. The other meteorites were obtained from the collections of the Departments of Astronomy and Geology, University of Leicester. Our thanks are also due to the Birmingham group, especially W. Prachyabrued, for discussions, and to Professor Symons and the staff of the Chemistry Department, University of Leicester, for permission to use and help with the  $^{60}\text{Co}$  source.

The catalogue numbers for the British Museum specimens are: Saratov BM 1956, 169; Tennesilm BM 1913, 218; Butsura BM 34795; Durala BM 32097; Wold Cottage BM 1073; Mauerkirchen BM 19967; Ogi BM 55256; Crumlin BM 86115; Gambat BM 83864; Aldsworth BM 61308; and Plainview BM 1959, 805.

Table 5  
Terrestrial ages expected from the observed  
surface density and fall rate of meteorites

Percent Recovered	Stones Estimated age in years	Irons Estimated age in years
.5	2,600	22,900
1	1,320	11,500
2	661	5,740
5	265	2,270
10	105	1,140
15	88.3	760
20	53.8	570

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