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THERMOLUMINESCENCE AND THE TERRESTRIAL AGE OF METEORITES: SOME RECENT RESULTS

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This paper reports the results of an examination of the thermoluminescence (TL) of 23 meteorites which were observed to fall, and 17 meteorite finds which have had their terrestrial age determined by the ^{14}C method. The terrestrial ages of the observed falls range from 1 to 205 years, whilst the ^{14}C terrestrial ages range from 1200 ± 2000 to >20,000 years. A statistically significant correlation has been observed between the natural TL – as expressed as the ratio of the intensity of the low-temperature TL peak to that of the high-temperature peak – and the terrestrial age of the 40 meteorites. Furthermore, peak height ratios in excess of 3.0 are only observed in falls which fell within the last 250 years, suggesting that finds with peak height ratios as large as this, such as Allan Hills A77003 and Plainview (1917), fell within the last few hundred years. The present results are consistent with evidence that meteorite TL decay is a non-first-order process. The implications of the results for estimates of the terrestrial ages of 8 meteorite finds, for which there are no ^{14}C data, are also discussed.

1. Introduction

The principles for using thermoluminescence (TL) to determine terrestrial age are, in some respects, similar to those utilising cosmogenic isotopes [1]. The TL is maintained at a high level in space by cosmic ray bombardment. Once on Earth, where the irradiation level is low and the environmental temperature is high, the TL begins to decay. The terrestrial age of a find can, in principle, be estimated from the TL level observed in it, providing one knows or can determine the level at the time of fall, and the decay rate of the relevant TL "traps" at the Earth's ambient temperature.

Five years ago Sears and Mills [2] reported on a detailed investigation of the application of TL to the determination of the terrestrial ages of meteorites. In the intervening period two concepts have emerged – these are discussed below – which have an important bearing on this work. Moreover, with the recovery of a great many Antarctic finds, we believe the time is ripe to re-examine the topic. This paper reports the results of the re-examination.

Sears and Mills [2] considered two methods for determining terrestrial age from TL. The first, following a suggestion by Christodoulides et al. [3] is based on the assumption that the natural dose in the meteorite, as determined from the high-temperature traps, would, if imparted in the laboratory at room temperature, reproduce the in-space level of TL in the low-temperature parts of the glow curve. In fact this is not true, since at the very low dose rate in nature the equilibrium TL level is much lower than that attained in the laboratory at the very high dose rates employed [4]. The second simply measures apparent changes in the TL glow-curve shape as the terrestrial age increases. This is the method we re-examine here, taking into account two recent developments alluded to above. The first development is the realisation that the decay of TL in meteorites does not follow a simple logarithmic law (first-order kinetics), but that as the decay proceeds, the logarithm of the TL decreases more slowly. This is apparent in the isothermal decay curves of reference 6 and a recent, detailed analysis of meteorite glow curves by McKeever [5] suggests that TL decay in meteorites is a second-

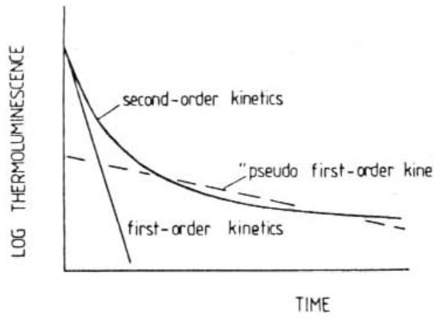

Fig. 1. Schematic diagram illustrating two modes by which the low-temperature TL peak in meteorites (LT) could decay. Sears and Mills [2] assumed first-order kinetics, so that after ~500 years LT would no longer have been measurable. It now seems that decay follows non-first-order kinetics, probably second-order, and can be used to date much greater periods.

order process. Fig. 1 is an attempt to illustrate the situation schematically. As a consequence we can determine fall times much greater than previously believed, although it is not possible to say just how much greater these times might be. It also means that we cannot use laboratory methods to estimate the rate of TL decay, because of experimental difficulties, especially the enormous time periods involved.

The second development is related to the first. Sears and Mills [6] found that the rate of TL decay in a group of three meteorites was perhaps a factor of ten lower than in another group of three. It seemed that, before determining the terrestrial age of any find, an estimate would have to be made of its TL decay rate. Melcher and Sears [7] repeated these measurements using different techniques and found that ten meteorites, including the six previously examined by Sears and Mills [2] had uniform decay rates. It is now apparent that the three meteorites with high decay rates also had higher surviving TL when the measurements were made and the apparent differences were due to the fact that decay followed non-first-order kinetics. Sears and Mills [2] were effectively measuring different parts of the second-order decay curve in Fig. 1 when comparing different meteorites.

In this paper we compare the TL of 40 meteorites which were either seen to fall, or which have had their terrestrial age determined from their ^{14}C contents. We shall demonstrate that there is a statistically significant correlation between the level of TL in meteorites and their terrestrial age. It seems that the actual decay curve – which probably resembles the second-order curve in Fig. 1 – can be approximated by a straight line, similar to that labelled "pseudo first-order kinetics" and with a half-life of ~5000 years. In contrast, the first-order decay curve has a half-life around 50 years. It follows that recently fallen meteorites may lie well above the imaginary "pseudo first-order" curve. In this paper we also discuss the implications of the results for terrestrial age estimates for eight meteorite finds.

2. Methods

The TL apparatus and procedures are essentially the same as described elsewhere [8], and only a brief description is necessary here. Samples are heated at a rate of $5^\circ C\ s^{-1}$ in an oxygen-free-nitrogen atmosphere and the TL measured by an EMI 9084 QB photomultiplier tube. A chance HA3 heat filter and Ilford 621 instrument filter are used to prevent thermal damage of the PMT and to suppress black body radiation. A chromel-alumel thermocouple is used to measure the temperature of the nichrome heating strip and the TL is plotted directly as a function of temperature of an X-Y plotter. Samples are washed in 10 M hydrochloric acid and then prepared as fine-grain discs in the way described elsewhere [9].

3. Results

Fig. 2 shows the glow curves of three meteorites of known terrestrial age; all the samples used here are ordinary chondrites. The natural glow curves consist effectively of two apparent, broad peaks; one at about 230°C and another, much broader hump at around 400°C. For the sake of brevity, they will be referred to in this paper as LT and HT. In fact, each probably consists of several independent peaks.

The three meteorites plotted in Fig. 2 have very different terrestrial ages: (1) Kernouvé, whose fall was observed in 1869; (2) Clovis, a find with a ^{14}C terrestrial age of 2000 ± 2000 years; and (3) Potter, a find with a ^{14}C terrestrial age greater than 20,000

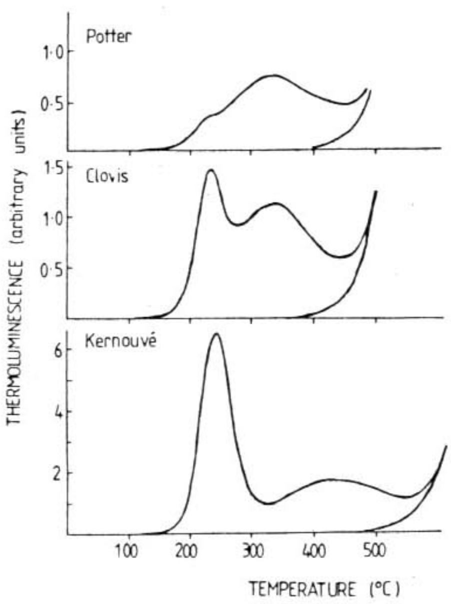

Fig. 2. Glow curves (graphs of thermoluminescence emitted against the temperature to which the sample has been heated) for three meteorites of very different terrestrial age. Kernouvé was observed to fall in May 1869, whilst Clovis and Potter are finds for which the terrestrial ages, determined from ^{14}C, are 2000 ± 2000 years and greater than 20,000 years respectively. The background, due to black body radiation, is also shown. Notice differences in the three ordinate scales.

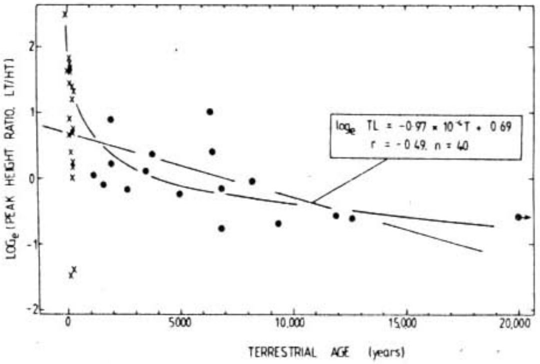

Fig. 3. Natural logarithm of the peak height ratio of 40 meteorites as a function of the terrestrial age, T. The crosses refer to meteorites whose fall was observed (Table 1), the dots refer to finds whose terrestrial ages have been determined by the ^{14}C method (Table 2). The regression line, which is given in the insert, is highly significant (with $r = -0.49$ at 38 degrees of freedom). The curved line is a more reasonable guess at the trend, assuming that the TL is decaying in a non-first-order fashion.

years. LT appears to have substantially faded in the older specimens. However, it is also apparent from Fig. 2 that some method of normalizing the TL is required. For example, in Clovis the intensity of HT – which should not have faded appreciably if LT is still apparent – is only about half the value of that in Kernouvé (note the difference in the ordinate scale). This may be due to a number of causes, the main one being the intrinsically lower TL efficiency of meteorites which have been shock-reheated or which are of petrologic type 3 (lightly metamorphosed) [10,11].

One method of normalization, which is suggested by Fig. 2, is to take the peak height ratio, LT/HT, where both are measured in the natural glow curve. This method effectively looks at changes in the shape of the glow curve as the terrestrial age increases and the LT/HT ratio decreases. This approach assumes that LT and HT in a given meteorite are produced by the same phosphor; and assumption which, from mineral separation experiments, seems to be generally true [12]. Because it compares structural changes in the glow curve, the method can only be applied to meteorites which yield glow curves of similar shape when they are given a standard test dose after having been drained of their natural TL. Fortunately, this is true of all the three ordinary (H, L, LL) chondrite classes [13], which constitute some 85% of all stony meteorites.

The peak-height ratio of our 17 "found" meteorites is plotted against their ^{14}C terrestrial ages, as measured by Suess and Wänke [14] and by Boeckl [15], in Fig. 3. Also plotted, are the data for 23 meteorites whose fall was observed within the last 200 years [2,6]. A regression line has been fitted to the data and its correlation coefficient calculted. Both with, and without, the 23 recent falls there is a highly significant correlation between ln(LT/HT) and terrestrial age, T; $r = -0.49$, $n = 40$, which, for 38 degrees of freedom, denotes a high degree of significance for the value of the correlation coefficient. Without the falls, $r = -0.38$ ($n = 17$), which again

TABLE 1

Details of 23 observed falls used in this study

Meteorite	Source * and Catalogue No.	Class	Terrestrial age (years)	Thermoluminescence peak height ratio (LT/HT)
Kirin	CAS	H5	1	13.20
Saratov	BM 1956, 169	L4	55	6.17
Tennasilm	BM 1913, 218	L4	101	5.89
Soko Banja	BM 51857	LL4	96	5.37
Dhajala	PRL	H3	1	5.18
Barwell	BM 1965, 57	L5	8	5.13
Quenggouk	BM 33764	H	119	5.13
Bruderheim		L6	13	4.47
Butsura	BM 34795	H6	112	4.17
Alta'ameen	AR	LL5	1	4.15
Kernouvé	BM 43400	H6	107	3.98
Ogi	BM 55256	H6	232	3.72
Durala	BM 32097	L6	158	3.31
Wold Cottage	BM 1073	L6	178	2.29
Khanpur	BM 1933, 158	LL5	21	1.95
Beaver Creek	BM 73646	H4	83	1.95
Mangwendi	BM 1934, 839	LL6	39	1.78
Mauerkirchen	BM 19967	L6	205	1.48
Ambapur Nagla	BM 81117	H5	81	1.29
Jelica	BM 65605	LL6	80	1.19
Crumlin	BM 86115	L5	71	1.00
Gambat	BM 83864	L6	76	0.23
Limerick		H5	160	0.25

* BM – Dr. R. Hutchison, British Museum, Natural History; AR – Dr. Y. Al-Rawi, College of Science, Baghdad, Iraq; PRL –Dr. Bhandari, Physical Research Laboratory, Ahmedabad, India; CAS – Dr. Ouyang Ziyuan and Dr. Lin Shunsheng, Chinese Academy of Sciences, Kweiyang, China.

has statistical significance. However, as discussed in section 1, the decay of meteorite TL is not a first-order process, so we have also indicated in Fig. 3 a second curve, drawn by eye, which probably represents the TL decay more reasonably. The terrestrial age of the Estacado meteorite has previously been discussed with reference to its TL properties [24]. While there is no doubt that Estacado has particularly intense TL, its peak height ratio is not inconsistent with its ^{14}C terrestrial age of 6800 years.

An alternative means of normalizing the data is to divide the natural TL by the TL exhibited by the same specimen after it has been drained of its natural TL and then given a test dose of radiation [16]. Such a procedure is preferable in that it can be applied to any region of the glow curve. However, this method failed to produce any correlation in the present case. There are several reasons why this should be so. Firstly, this procedure is critically dependent on the temperature scale for the natural and the artificial glow curves being perfectly aligned. A glance at the Kernouvé glow curve (Fig. 2) will demonstrate that a misalignment of only a few degrees can cause a large error in the TL level measured near a peak. Errors in the temperature scale of this order could be caused by thermal lag across the specimen or heating strip. Secondly, unusual artificial glow curves have sometimes resulted from particularly weathered samples;

TABLE 2

Details of 17 finds with measured ^{14}C content used in this study *

Meteorite	Class	Year found	^{14}C terrestrial age (years)	Thermoluminescence peak height ratio (LT/HT)
Texline	H	1937	6400 ± 3000	2.75
Faucett	H		2000 ± 2000	2.47
Tulia	H5	1917	6400 ± 3000	1.50
Dimmitt	H3, 4	1947	3800 ± 2600	1.45
Clovis	H3	1961	2000 ± 2000	1.23
Gilgoin	H5	1889	3500 ± 2500	1.12
Elm Creek	H4	1906	1200 ± 2000	0.96
De Nova	L	1940	8300 ± 3000	0.90
Kingfisher	L	1950	1600 ± 2000	0.90
Weldona	H	1934	6800 ± 3000	0.86
Alamogordo	H5	1938	5000 ± 2600	0.79
Keyes	L6	1939	2700 ± 2000	0.66
Brownfield	H3	1937	12,700 ± 4500	0.56
McKinney	L4	1870	10,200 ± 3500	0.56
Potter	L6	1941	⩾20,000	0.55
Leon	H		9200 ± 3100	0.50
Estacado	H6	1883	6800 ± 3000	0.47

* All samples were supplied by Professor H.E. Suess and Dr. M. Herndon. The same specimens were used for the ^{14}C determinations [14,15].

presumably the rust contains no natural TL, but is sensitive to radiation. A similar difficulty in normalizing the TL in this way was encountered by Hoyt et al. [17] although in their case the interfering phosphor was an indiginous mineral. Another source of error could be a shift in the position of natural and artificial peaks during fading because of overlapping peaks or non-first-order kinetics.

4. Discussion

Studies of the natural TL levels of meteorites share with many cosmogenic studies the problems of differential shielding from cosmic rays and, occasionally, from unusual orbits. Several studies on meteorites [12,22] and experiments involving thick target measurements using accelerators [23] demonstrate how TL levels vary as a function of depth. Variations in the order of 30% (Estcado) and 50% (St. Séverin) have been observed in the largest specimens. This variation is comparable to the spread about the regression line displayed by the finds whose terrestrial age was determined by the ^{14}C method. Variable shielding would therefore seem to be the major cause in the scatter in Fig. 3, but small differences in orbit must also be a contributing factor. This is especially true of meteorites with particularly low TL where heating to temperatures around 300–350°C seems a more reasonable cause; this may well have been the result of close passage to the Sun [18]. Two of our 23 falls (Gambat and Limerick) appear to be of this kind. When assigning a large terrestrial age by TL, the assumption is implicit that the meteorite in question has not been reheated. Our present data suggests that this assumption will be good nine times out of ten, but the problem may be more serious for LL chondrites [6] and we have deliberately excluded them from the present study.

Our main objective in the present study was to see if, despite these complications, there existed a simple relationship between TL and terrestrial age. We believe that the present results demonstrate that it does. The data in Fig. 3 can be summarized in terms of two observations: (1) for the 40 meteorites in our present study there is a significant correlation between peak height ratio and terrestrial age – the regression line in Fig. 3 corresponds to a half-life of ~5000 years; (2) peak height ratios greater than 3.0 are only observed in meteorites with a terrestrial age of less than 250 years (more than half the observed falls in Table 1 have ratios >3.0). Laboratory determinations of TL decay rates allow an estimate for the half-life at room temperature of 10-100 years [6], whilst the TL of observed falls with terrestrial ages between 0 and 250 years led Melcher to assign a half-life of ~40 years [20].

These data are consistent with evidence, cited earlier, that meteorite TL decay is not a first-order process. It seems that we can effectively approximate the actual decay curve, which will resemble the second-order curve in Fig. 1, by a combination of the "first-order curve" – which gives rise to observation 2 – and the "pseudo first-order curve" (observation 1).

We are now in a position to consider the implications of these results for the determination of terrestrial age. According to the numbers quoted in Hutchison et al. [19], nearly one-half of the 1400 stony meteorites in our museums were not seen to fall and are therefore of unknown terrestrial age. The vast majority of these were found in the Prairie States of the U.S.A. [1, section 2.2.4]. Since the Hutchison et al. [19] catalogue was compiled, it has become apparent that large numbers of meteorites are concentrated in certain regions of the Antarctic continent; over 1300 meteoritic pieces have so far been recovered. (However, it is uncertain how many discrete falls these represent, because meteorites normally fragment into hundreds, or perhaps thousands, of pieces during fall.) There are, therefore, two major regions of the world where stony meteorites have been found in great quantities.

The meteorites used to derive Fig. 3 were either largely found in the prairies of central U.S.A. or were observed falls in inhabited parts of the Earth. The mean annual temperature of the Prairie regions of the U.S.A. – in which most of the finds were made – is 20.4°C [26], which is not appreciably different from the probable room temperature at which the falls were stored. Most of our falls were transferred to the same room of the British Museum within weeks or months of fall. They will all, therefore, have been stored in their natural state at much the same temperature since fall. The curve may be meaningfully compared only with data from other meteorites found in similar regions of the Earth. Table 3 lists the data for five meteorites found in the U.S.A.: Plainview, Calliham, Shields, Wellman (c) and Etter. Glow curves for these meteorites were published by Sears and Mills [2]. Their peak height ratios lead us to assign terrestrial ages of 19,500, 21,400 amd 23,700 years to Wellman (c), Etter and Shields, respectively, using the regression line in Fig. 3. The 95% confidence levels for the slope and intercept correspond to errors of ±8000 years on these values, and suggest upper limits for the terrestrial ages of Plainview and Calliham of 250 and 2500 years, respectively. Values smaller than these were previously assigned to these meteorites by Sears and Mills [2], based on the assumption of first-order kinetics and laboratory estimates of decay rates.

Fig. 4 gives the glow curves for three meteorites which were found in the Allan Hills region of Antarctica by the Japanese-American expedition of 1977-78. A difficulty in comparing these data with Fig. 3 is that the storage temperatures for Antarctic specimens will have been much lower than those used to derive Fig. 3.

Melcher quotes a value of –20°C for the mean annual temperature at the find site, but it is the maximum temperatures experienced for any length of time which are of importance and Annexstad and Yanai [25] report the frequent occurrance of pools of water there. Our estimates will clearly, however, be lower limits. Following the same procedure as before, and calling our values lower limits, we arrive at the terrestrial ages in Table 3.

The terrestrial ages of several Antarctic meteorites, including the three studied here, have also been determined from their TL by Melcher [20]. Lacking a calibration curve analogous to Fig. 3, Melcher [20] assumed an Antarctic storage temperature of –20°C, a first-order mean-life (time to decay 1/e) at room temperature of 40 years and a trap depth of 1.1 eV. Assuming zero age for the meteorite with highest TL, the results ranged from 35,000 to 230,000 years. This procedure suffers mainly from its heavy depen-

TABLE 3

Terrestrial age estimates based on the present study

Meteorite	Source *	Class	Year found	Thermoluminescence peak height ratio (LT/HT)	Estimated terrestrial age (years)
Allan Hills A77003	MWG(22)	?L3	1977-78	12.0	<250
Plainview (1917)	BM1959, 805	H5	1917	3.70	<250
Calliham	AML	L	1958	2.60	<2500
Allan Hills A77272	MWG(14)	L6	1977-78	1.30	>4400
Allan Hills A77214	MWG(21)	L or LL	1977-78	1.15	>5700
Wellman (c)	AML	H	1964	0.30	19,500 ± 8000
Etter	AML	H	1965	0.25	21,400 ± 8000
Shields	AML		1962	0.20	23,700 ± 8000

* MWG – Meteorite Working Group of the U.S. National Science Foundation (the number allocated by the curator to the piece used here is indicated in parentheses); BM – Dr. R. Hutchison, British Museum, Natural History; AML – Mr. G. Huss, American Meteorite Laboratory.

dence on the assumed storage temperature and trap depth, and our lack of knowledge concerning the decay of meteorite TL. For example, an assumed storage temperature of 0°C, instead of −20°C, results in calculated terrestrial ages which range from 1000 to 10,000 years. A 10% change in the assumed trap depth results in calculated terrestrial ages also in the order of 10^3 to 10^4 years. The use of first-order decay equations also means that, even allowing for the appropriate storage temperature and trap depths, the estimates are lower limits. As Melcher [20] intimates, a preferable procedure would seem to be the derivation of a curve analogous to Fig. 3 based only on Antarctic meteorites. Both sets of data are consistent with the terrestrial age estimates for these meteorites, based on ^{26}Al contents. They were Allan Hills A77003, <50,000 years; Allan Hills A77214, <30,000 years and Allan Hills A77272, <70,000 years [21].

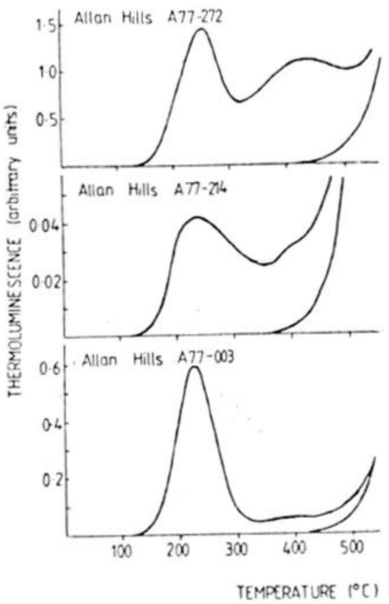

Fig. 4. Glow curves for the three Antarctic meteorites examined here. The same arbitrary units are used in all cases for the TL intensity, but notice the differences in the scale.

5. Conclusions

There is a statistically significant correlation between the thermoluminescence peak height ratio and the terrestrial age of 40 meteorites which were either observed to fall or which have had their terrestrial age determined by the ^{14}C method. We conclude therefore that TL provides a method for determining the terrestrial ages of meteorites. Present data suggest that falls which occurred within the last ~10^4 years may be dated, perhaps even earlier. Based on the observed correlation, we have made estimates for the terrestrial ages of 8 finds and these are listed in Table 3.

Thermoluminescence peak height ratios (LT/HT) greater than about 3.0 are only observed in meteorites which fell within the last 250 years, so it would seem that meteorite finds with peak height ratios in excess of this, for example, Allan Hills A77003 and Plainview (1917), are particularly recent falls.

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