

THE NATURAL THERMOLUMINESCENCE OF METEORITES
I. TWENTY-THREE ANTARCTIC METEORITES OF KNOWN ^{26}Al CONTENT

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Abstract. The level of thermoluminescence (TL) naturally present in 23 Antarctic chondrites of known ^{26}Al content has been measured as a means of exploring their thermal and radiation history. Antarctic meteorites tend to have lower natural TL than non-Antarctic meteorites, presumably because of their generally longer terrestrial ages. Seventeen of the meteorites lie on a band of increasing natural TL and ^{26}Al activity; meteorites with ^{26}Al of 45-60 dpm/kg have natural TL values (normalized to the high temperature TL) of 2-4, whereas those with ^{26}Al values of 30-45 dpm/kg have normalized natural TL values of 1-2, and ALHA 76008, with an ^{26}Al content of 11 dpm/kg, has a TL value of 0.87 ± 0.01 . This suggests that the "half-life" for TL decay is comparable to that of ^{26}Al . The six meteorites not lying on the TL- ^{26}Al trend have much lower TL than others of comparable ^{26}Al . For two of them (RKPA 79001 and RKPA 80202, which are tentatively paired) the low natural TL probably reflects a recent severe shock, while another (ALHA 77294) has previously been measured in this lab and had TL consistent with the TL- ^{26}Al trend. The remaining three, ALHA 78006, ALHA 77296, and ALHA 77297 (of which the last two are probably paired), have suffered a recent reheating such as would be expected from orbits of unusually small perihelia.

Introduction

The discovery of large numbers of meteorites in Antarctica has resurrected an interest in terrestrial age determination, since the data are relevant to (1) studies of the secular variation in the source of meteorites to earth [Dennison et al., 1986]; (2) the mechanisms by which ice movements cause the concentration of large numbers of meteorites [Bull and Lipschutz, 1982; Annestad, 1986]; and (3) identifying fragments of a single fall (i.e., "pairing" [Scott, 1984]). The Antarctic meteorite collection is also an important source of new and rare meteorite types [Palme, 1986] and meteorites with unusual histories. The natural thermoluminescence (TL) levels in meteorites provides information concerning their terrestrial age [Sears and Durrani, 1980; Melcher, 1981a; McKeever, 1982] and also other aspects of their thermal and radiation history [McKeever and Sears, 1980; McKeever and Sears, 1979; Melcher, 1981b]. A recent review of natural TL studies of meteorites, and also TL sensitivity studies, is that of Sears and Hasan [1986]. Because of the extremely small sample size required (less than a few milligrams) and the avoidance of the long count times necessary

for cosmogenic isotope measurements, several authors have advocated the measurement of natural TL levels in all returned Antarctic meteorites in a systematic way [Melcher, 1981a; McKeever, 1982; Sutton and Walker, 1986]. The present paper is a study, made at the request of the Meteorite Working Group of NASA/NSF, of the natural TL levels in 23 equilibrated ordinary chondrites of known ^{26}Al contents which were recovered in the Antarctic.

Experimental

Thirty Antarctic meteorite specimens, each of about 200 mg, were removed from 23 meteorites by Jim Gooding, curator of meteorites, Johnson Space Center. Wherever possible, samples were removed at least 2 cm from the fusion crust, and duplicates were taken from widely separated locations. At Fayetteville, all handling was in red light, since exposure to fluorescent lamps or daylight causes a partial draining of natural TL level ("optical bleaching"). The phenomenon has been demonstrated for pottery samples and lunar samples [Aitken et al., 1963; Hoyt et al., 1972] and in Table 1 we show that meteorites are also subject to optical bleaching. Two samples of each meteorite were placed near a window, one covered with an opaque lid to act as a control. We detected a 4-20% decrease in the natural TL levels of several meteorite samples after a 4-day exposure to sunlight and fluorescent lighting. Although these measurements are inadequate to set limits on the amount of draining and the effects of diverse lighting conditions, they clearly indicate the existence of the phenomenon in the meteorite case. The 200 mg chips supplied by the curator were therefore broken and a 10-20 mg chip that had not been exposed to laboratory lighting was removed from the interior. It was ground, the magnetic portion removed with a hand magnet, and the TL measured in three aliquants from each chip. After natural TL measurement, the samples were given a 25 krad dose from Sr-90 beta source and the induced TL measured three times. The apparatus and other techniques are as described by Sears and Weeks [1983].

Our natural TL data are reported in two ways (Table 2), both normalized to remove the effects of differences in petrologic type, shock history, sample heterogeneity, and powder albedo (including discoloration due to weathering). LT/HT refers to the ratio of the intensity of the low temperature TL peak ($\sim 250^\circ\text{C}$) to that of the high temperature peak ($\sim 350^\circ\text{C}$), while the "equivalent dose" refers to the natural TL at 250°C divided by the induced TL at 250°C multiplied by the test-dose administered. The uncertainties quoted are standard deviations based on triplicate measurements. Precision is generally much higher for the peak height ratio, but the measurement is only applicable to ordi-

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TABLE 1. Optical Bleaching of the Natural Thermoluminescence of 5 Antarctic Meteorites

Meteorite	(LT/HT) _{Dark}	(LT/HT) _{SL}	$\frac{(LT/HT)_{Dark}}{(LT/HT)_{SL}}$
ALHA 77002	1.14 ± 0.03	0.98 ± 0.07	1.16 ± 0.08
ALHA 77012	1.22 ± 0.03	1.18 ± 0.07	1.04 ± 0.06
ALHA 77177	0.89 ± 0.01	0.72 ± 0.07	1.24 ± 0.1
ALHA 77259	0.80 ± 0.04	0.68 ± 0.05	1.19 ± 0.1
ALHA 77261*	1.27 ± 0.06	1.23 ± 0.02	1.04 ± 0.05

(LT/HT)_{Dark} and (LT/HT)_{SL} refer to the peak height ratios for the samples kept in the dark and sunlight/fluorescent lights, respectively. See Table 2 for explanation of uncertainties and other symbols.

* In this case, samples were taken from the interior of the fragment (equivalent to "dark") and surface (equivalent to "SL").

nary chondrites. The equivalence of the two methods in the present case is demonstrated in Figure 1, where LT/HT is plotted against equivalent dose. While most samples fall within two sigma of a correlation line, a few do not. The two methods of normalization are discussed further below. The equivalent dose is critically dependent on the temperature scale for the natural and glow-curves of the induced TL being perfectly aligned, as misalignment of few degrees, which could be caused by differences in thermal lag across the specimen, can cause large errors. A full discussion of the advantages and disadvantages of each method of normalization was published by McKeever and Sears [1980].

Results

The natural TL data are listed in Table 2. The agreement between duplicate chips from the same meteorite is extremely good, generally within 10% for LT/HT. In Table 3 we compare the present data with unpublished data for six of the meteorites that have previously been run in this laboratory. In all but one case (ALHA 77294) agreement is also satisfactory (i.e., within a factor of 2, the maximum difference observed along core samples, Lalou et al. [1970]).

Histograms of the present data are shown in Figure 2, with data for 40 non-Antarctic meteorites from McKeever and Sears [1980]. The Antarctic falls cover two orders of magnitude in natural TL, the peak height ratio going from 4 to about 0.08 with a peak in the histogram at about 2.5. The non-Antarctic meteorites also show considerable spread but are skewed to higher values (range from 6 to 0.05 with a peak at about 4), presumably reflecting smaller terrestrial ages. The present data set is a little small for such comparisons, but a larger data base, which will be published as a sequel to this paper, gives

exactly the same distribution [Hasan et al., 1986].

Evans et al. [1982] have measured the ²⁶Al activity of the present samples from which the present splits were taken (Table 2), and their results are compared with our natural TL data in Figure 3. The ²⁶Al measurements are made on the entire recovered mass; the mass used for the TL measurement is listed in the Table. Of the 23 samples, 17 show evidence for a relationship between natural TL and ²⁶Al. Samples with ²⁶Al activities of 45-60 dpm/kg have TL ratios of 2-4, those with ²⁶Al of 30-45 dpm/kg have TL ratios of 1-2 and ALHA 76008 with an ²⁶Al activity of 11 dpm/kg has a natural TL value of 0.87 ± 0.01. A regression line, fitted by the method of least squares, is significant at the 99% level. There is an indication that H chondrites tend to lie above this line and L chondrites below, which suggest that were the ²⁶Al activity to be corrected for differences in target composition the correlation would be even better. The remaining samples plot below this band, presumably due to unusual thermal/radiation histories. Four of these may be fragments of two meteorites (RKPA 80202 and RKPA 79001 may be paired, as are ALHA 77296 and ALHA 77297), while our previous unpublished determination for ALHA 77294, 2.12 ± 0.06, plots it along with meteorites of comparable ²⁶Al.

Discussion

Terrestrial Age

With the exception of meteorites with natural TL ratios < 0.8 (or equivalent doses < 1.0 krad), which are thought to have been recently reheated, the distribution of natural TL values in meteorites resembles that of ²⁶Al. This is apparent in the similarity of the histograms, especially in the differences between Antarctic and non-Antarctic meteorites (Figure 2, this work, and Figure 40 in Evans et al. [1982]) and the tendency for natural TL to correlate with ²⁶Al in Figure 4. This would seem to imply a "half-life" for natural TL decay of ~10⁶ year. (An anonymous reviewer calculates 3 x 10⁵ year.) The existence of a significant correlation between natural TL and ²⁶Al suggest that natural TL levels at the time of fall, and terrestrial storage conditions, were similar for these 17 meteorites.

Previous attempts to determine the decay kinetics of natural TL with reference to Antarctic meteorites have been made by Melcher [1981a] and McKeever [1982], both using a combination of experimental and theoretical methods. The decay rate is heavily dependent on the assumed storage temperature. Assuming an effective storage temperature of -20°C, Melcher [1981a] suggested that the natural TL of Antarctic meteorites would decay by a factor of 5 in 10⁶ years and that a storage temperature of -15°C would produce the observed range in natural TL values, while McKeever's [1982] more sophisticated theoretical treatment shows that, at -5°C, LT/HT will fall from a level of 3.0 to 1.0 in about 1.1 x 10⁶ years. The present TL data (with those not lying on the TL-²⁶Al trend excluded) are compared with the terrestrial ages calculated by Nishizumi [1984] from ⁵³Mn, ¹⁰Be, ²⁶Al, ³⁶Cl, and ¹⁴C data

TABLE 2. Thermoluminescence Data of 23 Antarctic Meteorites

Meteorite*	Recovered Mass (g)	Mass Studied (mg)	Class	LT/HT	E.D. (krad) at 250°C	²⁶ Al dpm/kg ⁺	Terr. Age (10 ⁵ y) [§]
ALHA 76008,21	281	279	H6	0.87 ± 0.01	150 ± 6	11 ± 1	100 ± 70
ALHA 77002,47	285	210	L5	1.3 ± 0.03	372 ± 26	30 ± 3	700 ± 160
ALHA 77004,19	2230	250	H4	2.32 ± 0.02	447 ± 26	52 ± 5	170 ± 70
ALHA 77004,18		230	H4	2.23 ± 0.02	384 ± 60	52 ± 5	
ALHA 77191,16	642	210	H4	2.23 ± 0.02	377 ± 13	56 ± 4	-
ALHA 77258,28	597	250	H6	2.87 ± 0.07	633 ± 16	29 ± 2	270 ± 70
ALHA 77261,26	411	280	L6	1.11 ± 0.02	387 ± 27	36 ± 4	310 ± 80
ALHA 77262,43	861	240	H4	3.66 ± 0.14	560 ± 7	47 ± 5	-
ALHA 77294,39	1351	250	H5	0.48 ± 0.01	106 ± 11	61 ± 4	30 ± 2
ALHA 77296,21	963	240	L6	0.20 ± 0.01	12 ± 0.2	67 ± 4	80 ± 40
ALHA 77297,33	951	250	L6	0.29 ± 0.02	27 ± 6	70 ± 7	-
ALHA 78043,28	680	250	L6	0.92 ± 0.006	198 ± 94	38 ± 3	490 ± 80
ALHA 78076,24	275	210	H6	3.34 ± 0.11	793 ± 28	52 ± 4	130 ± 70
ALHA 78102,17	336	250	H5	1.65 ± 0.03	340 ± 17	35 ± 3	210 ± 80
ALHA 78105,26	941	260	L6	2.75 ± 0.03	825 ± 132	61 ± 7	260 ± 70
ALHA 78112,26	2485	200	L6	2.05 ± 0.05	818 ± 33	42 ± 3	230 ± 70
ALHA 78112,25		240	L6	1.73 ± 0.05	956 ± 27	42 ± 3	
ALHA 78114,21	808	220	L6	1.16 ± 0.06	220 ± 19	38 ± 2	460 ± 80
ALHA 78115,25	847	260	H6	2.87 ± 0.1	892 ± 21	43 ± 3	< 90
ALHA 78251,26	1312	220	L6	3.01 ± 0.03	752 ± 95	56 ± 6	--
ALHA 78251,25		210	L6	2.88 ± 0.02	936 ± 49	56 ± 6	-
META 78003,23	1726	250	L6	2.35 ± 0.04	546 ± 36	50 ± 3	-
META 78003,24		250	L6	2.51 ± 0.01	674 ± 70	50 ± 3	-
META 78006,21	409	210	H6	0.206 ± 0.02	15 ± 4	60 ± 4	<100
META 78028,82	2065	227	L6	1.62 ± 0.03	294 ± 40	56 ± 3	32 ± 1
META 78028,81		227	L6	1.60 ± 0.06	350 ± 2	56 ± 3	
RKPA 79001,23	3006	210	L6	0.57 ± 0.01	76 ± 41	58 ± 4	-
RKPA 79001,24		200	L6	0.62 ± 0.02	130 ± 11	58 ± 4	-
RKPA 80202,12	544	220	L6	0.062 ± 0.001	8 ± 1	53 ± 3	-
RKPA 80202,13		250	L6	0.094 ± 0.001	19 ± 2	53 ± 3	-

Errors quoted are one sigma based on triplicate measurements.

* ALH - Allan Hills, MET - Meteorite Hills, RKP - Reckling Peak. The curatorial fragment numbered is indicated after the comma.

⁺ Data from Evans et al. [1982], except for ALHA 77294, ALHA 77296, RKPA 79001, and RKPA 80202, which are based on personal communications from L. A. Rancitelli.

[§] From Nishiizumi [1984], based on ⁵³Mn, ¹⁰Be, ²⁶Al, ³⁶Cl, and ¹⁴C activities.

in Figure 4. ALHA 76008 is one of several Antarctic meteorites that has experienced a two-stage irradiation history and its low ²⁶Al is attributable to a low cosmic-ray exposure [Nishiizumi et al., 1979]; its ²¹Ne exposure age is 1.5×10^5 y. This being so, it seems fairly clear that its low TL may also reflect under-saturation in space. ALHA 76008 has therefore been omitted from Figure 4. Most of Nishiizumi's ages are $\sim 1-2 \times 10^5$ years, but four are $> 3 \times 10^5$ y (ALHA 77002, $7.0 \pm 1.6 \times 10^5$; ALHA 77261, $3.1 \pm 0.8 \times 10^5$; ALHA 78043, $4.9 \pm 0.8 \times 10^5$; ALHA 78114, $4.6 \pm 0.8 \times 10^5$ y). All four have values of LT/HT around 1.0. Those with terrestrial ages of $1-2 \times 10^5$ y have LT/HT values of 2-4.

Indicated on the plot are McKeever's [1982] theoretical curves for TL decay at a variety of storage temperatures. These suggest that the effective terrestrial storage temperature for Antarctic meteorites is $\sim 5^\circ\text{C}$. This probably reflects the temperatures during the relatively short period on the surface of the ice, rather than the longer period at lower temperatures and great depths.

Shock

The low TL values of RKPA 79001 and RKPA 80202 are most probably due to shock reheating. The petrographic evidence for shock in these possibly

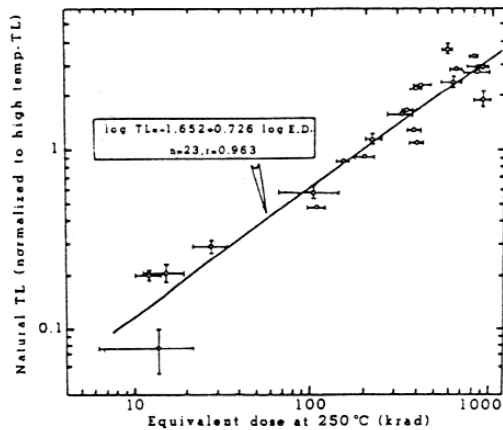


Fig. 1. A plot of natural TL (normalized to the high temperature TL, LT/HT) against equivalent dose (ED, in krad), measured at glow-curve temperature of 250°C, for the 23 Antarctic meteorites. Peak height ratio can be determined with greater accuracy than equivalent dose, but in principle, it can be influenced by solid state changes in the phosphor. The essential equivalence of the two methods of reducing the data is demonstrated by the strength of the correlation.

paired samples [Score et al., 1981, 1982; Scott, 1984] is considerable; the feldspar has been converted to maskelynite and there are extensive glassy veins. For comparison, none of the other meteorites in the present suite of samples show petrographic evidence for shock, according to Mason's description in the Antarctic Meteorite Newsletter [Score et al., 1981, 1982]. A tenfold decrease in LT/HT was observed experimentally by Sears et al. [1984] in a sample of the Kernouve meteorite shocked to 27 GPa, while pressures of 16 GPa did not significantly change the ratio. Maskelynitization is known to require shock pressures of ~30 GPa [Ostertag, 1983].

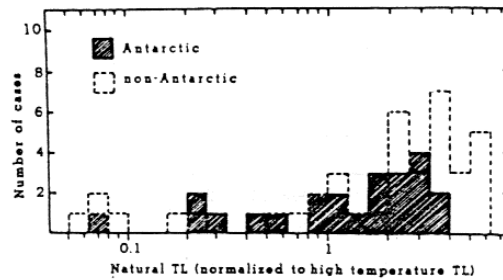


Fig. 2. Histogram of the natural TL data (LT/HT) for 23 Antarctic ordinary chondrites (solid line) and for 40 non-Antarctic ordinary chondrites (broken line, data from McKeever and Sears [1980]). Antarctic meteorites are displaced to lower values than non-Antarctic, suggesting a greater proportion of large terrestrial ages.

Shock also decreases the ability of the meteorite to display TL (i.e., TL sensitivity), but this effect has been removed by normalizing the TL data to the high-temperature TL. Most heavily shocked meteorites (> 30 GPa) have LT/HT ratios comparable to unshocked meteorites [Sears et al., 1984], suggesting that the equilibrium value is not affected by shock. The low values observed in the present RKPA samples must therefore reflect a shock event that occurred recently, so that the TL has not had sufficient time to return to equilibrium. The time taken for meteorites to reach TL equilibrium is currently only poorly known; most theoretical models suggest 10^5 - 10^6 y. Apparently the reheating event that drained the natural TL in these meteorites occurred $< 1.5 \times 10^6$ y. Most heavily shocked L chondrites yield ^{40}Ar - ^{39}Ar ages for the time of the shock event of 5×10^8 y [Bogard et al., 1976], so apparently the shock event suffered by RKPA 79001 and 80202 was not that which shock-blackened many L chondrites. The factor of ~10 difference in the natural TL of RKPA 79001 and RKPA 80202 may indicate that these are not paired meteorites, but it is also

TABLE 3. Comparison of the Data for Six Samples From the Present Work With Data Obtained Previously by This Lab (Unpublished Results)

Meteorite	LT/HT		E.D.	
	present	previous	present	previous
RKPA 79001	0.59 ± 0.01	0.41 ± 0.03	103 ± 30	126 ± 29
META 78028	1.61 ± 0.05	1.80 ± 0.02	322 ± 28	474 ± 103
ALHA 77296	0.20 ± 0.01	0.096 ± 0.01	12 ± 0.2	20 ± 3
ALHA 77297	0.29 ± 0.02	0.15 ± 0.01	27 ± 6	38 ± 5
ALHA 77294	0.48 ± 0.01	2.12 ± 0.06	106 ± 11	765 ± 15
ALHA 77002	1.30 ± 0.03	1.16 ± 0.03	372 ± 26	455 ± 156

Uncertainties and symbols as in Table 2.

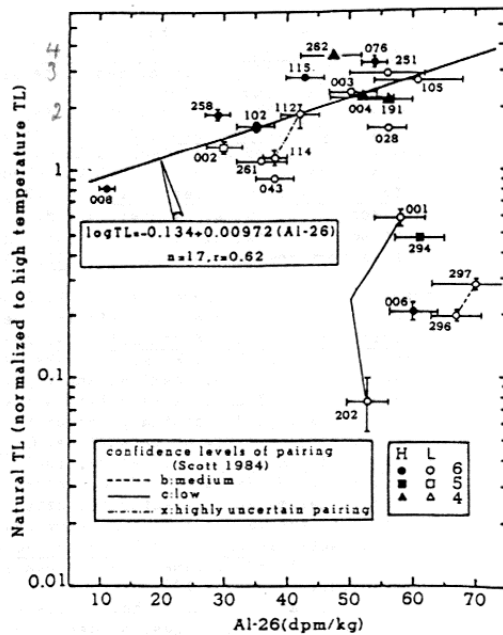


Fig. 3. A plot of natural TL (LT/HT) against ^{26}Al activity [Evans et al., 1982] for 23 Antarctic meteorites. Meteorites are identified by the last three digits of the meteorite's name (see Table 2). Seventeen meteorites lie on a correlation line between the two parameters, while six have lower values due to unusual radiation and thermal histories. Scott [1984] suggests that ALHA 77296 and ALHA 77297 are probably paired (confidence level a), while RKPA 79001 and RKPA 80202 are tentatively paired (confidence level b). However, ALHA 77112 and ALHA 77114 are highly uncertain paired (confidence level x).

possible that the difference reflects the heterogeneity of the shock and reheating suffered by the meteorite.

Meteorite Orbit

The remaining four meteorites with anomalously low TL in Figure 3 show no signs of shock reheating. Two of them (ALHA 77296 and ALHA 77297) are probably paired according to Scott [1984] and one of them (ALHA 77294) may have low TL for other reasons (see experimental section and below). There are therefore two objects, ALHA 77296/7 and META 78006, that appear to have experienced reheating but not shock within the last 10^5 - 10^6 years. Simplified TL theory indicates that passage to within 0.7 a.u. at the sun would be sufficient to cause the observed levels of TL draining [McKeever and Sears, 1980; Melcher, 1981b], and an unusual orbital history might also explain the slightly high levels of ^{26}Al (ALHA 77296/7 has 67/70 dpm/kg of ^{26}Al , while other Antarctic meteorites of comparable terrestrial age, $0.8 \pm 0.4 \times 10^5$ years according to Nishiizumi [1984] have 45-60 dpm/kg). Malakal has similarly low TL (LT/HT = 0.05 [McKeever and

Sears, 1980; Melcher 1981b]) and high ^{26}Al (79 = 2 dpm/kg [Cressy and Rancitelli, 1974]). On the basis of ^{22}Na , ^{26}Al , and ^{53}Mn data, Cressy and Rancitelli [1974] proposed that Malakal had experienced an orbit change about 2 m.y. ago, before which it was in a high cosmic-ray flux environment. However, Nishiizumi et al. [1979] argued that the ^{53}Mn data are not consistent with a high-inclination, high-dose rate orbit.

Shielding

Just as production rates of cosmic-ray produced nuclides in meteorite samples vary with the size and shape of the meteoroid, one expects changes in the TL level within a meteorite due to cosmic dose rate variations. A 10-15% build-up in the natural TL from the edges towards the center of the Estacado meteorite was observed by Sears [1975]; however, Estacado has only high-temperature natural TL and this is expected to be less depth-dependent than the low temperature TL. A factor of two increase was observed in two 50-cm core samples taken from the St. Severin meteorite [Lalou et al., 1970]. These and several studies on smaller samples were recently reviewed by Sears and Hasan [1986]. Bar and Herr [1974] observed a 39% decrease in the natural TL between 17 and 60 cm in simulated lunar soil exposed to 600 MeV protons. The factor of five difference in LT/HT between the present sample of ALHA 77294 and that which we have previously measured (0.48 ± 0.01 , compared with 2.12 ± 0.06) may be due to differences in shielding. It is also possible, however, that the present sample came from within a centimeter or so of the fusion crust, and had suffered heating during atmospheric descent. Further studies of this meteorite may prove worthwhile.

Aluminum-26 activity is fairly insensitive to depth because, over considerable distances, the attenuation of the primary radiation is balanced by the build-up of secondary radiations. Cressy [1975] found that the ^{26}Al activity went from

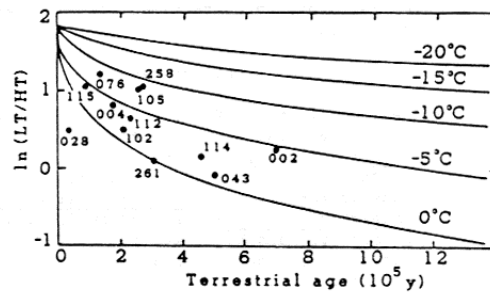


Fig. 4. Natural TL levels in meteorites as a function of terrestrial age, for a variety of storage temperatures, according to the theoretical model of McKeever [1982]. Superimposed on the theoretical curves are data for 11 of the present samples that plot in the $\ln(LT/HT)$ versus ^{26}Al trend (terrestrial ages based on ^{26}Al , ^{10}Be , ^{26}Al , ^{37}Cl , and ^{14}C from Nishiizumi [1984]); ALHA 76003 omitted for the reasons given in the text. The data are consistent with the theoretical calculations, assuming effective storage temperatures of 0° to -5°C .

68.9 ± 5.4 dpm/kg at the center of the Keyes chondrite to 50.9 ± 3.7 dpm/kg near its surface, while Barton et al. [1982] found a 20% decrease in the ^{26}Al activity from the center to the edges of a 45 x 55 cm slab of the Estacado meteorite. Similarly, Heymann and Anders [1967] found that ^{26}Al activity was independent of recovered mass. Theoretical calculations seem essentially consistent with these results [Reedy, 1985].

Shielding may therefore account for much of the scatter in Figure 3, but is probably more important for TL than ^{26}Al . More important in causing scatter in the ^{26}Al data are the experimental uncertainties, due mainly to counting statistics.

Compositional and Petrological Effects

In principle, one might expect both chemical class and petrologic type to affect the scatter observed in Figure 3. The ^{26}Al activity is governed by target chemistry and H chondrites have abundances of Si (a major target for ^{26}Al production) 20% below those of the L chondrites [Mason, 1979]. This could account for the prevalence of H chondrites above the natural TL versus ^{26}Al regression line in Figure 3.

The possible influence of petrologic type on Figure 3 is more subtle and results from glow-curve shapes being metamorphism-controlled to some extent [Guimon et al., 1985]. However, such an effect is very weak or absent in the present data as there is little or no correlation between petrologic type and location on the plot. In any event, such effects do not influence the equivalent dose measurement and the quality of the correlation between equivalent dose and peak height ratio (Figure 1) also indicates that variations due to this effect are unimportant.

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