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CHEMICAL AND PHYSICAL STUDIES OF TYPE 3 CHONDRITES: 2. THERMOLUMINESCENCE OF SIXTEEN TYPE 3 ORDINARY CHONDRITES AND RELATIONSHIPS WITH OXYGEN ISOTOPES

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Abstract. Thermoluminescence (TL) sensitivity values for sixteen type 3 ordinary chondrites, fourteen of them from Antarctica, have been measured. The values obtained (normalized to the TL sensitivity of the Dhajala meteorite) range from 1.6 (Allan Hills A77216) to 0.010 (Allan Hills A77176), and include two (Reckling Peak A80207 and Allan Hills A77176) that are particularly low. They fill a hiatus in the TL distribution that previously existed between St. Mary's County and Bishunpur, the latter being a meteorite with one of the lowest TL sensitivities known. The histogram of TL sensitivity values now shows a single distribution with higher values preferred; it resembles the histogram for L chondrites occupying the petrologic types 3, 4, 5, and 6. There is a tendency for the TL sensitivity of meteorites to decrease as $\delta^{18}O$ increases; equilibrated meteorites have TL sensitivities of 5-50 and $\delta^{18}O$ = 4.5 to 5.0‰, while the most primitive meteorites with TL < 0.01 have $\delta^{18}O$ = 5.7 to 6.2‰. Meteorites with intermediate TL have intermediate $\delta^{18}O$. Theoretically it is possible that the range of $\delta^{18}O$ values observed may reflect progressive loss of O in the form of CO at very low temperatures, but very restrictive physical conditions and a complex history seem to be required.

Introduction

It is commonly believed that the type 3 ordinary chondrites can provide many insights into the chemical and physical processes occurring in the early solar system. Important in their study has been the realization that the amount of metamorphic alteration suffered by individual meteorites varies widely. While certain type 3 meteorites, such as Bishunpur, Krymka, and Semarkona, are only slightly, if at all, removed from unaltered condensates [Wood, 1967; Dodd et al., 1967; Huss et al., 1981; Rambaldi and Wasson, 1981; Scott et al., 1982], others such as Dhajala and Bremervörde are almost petrologic type 4 in their chemical and physical properties. A major question of concern to many authors is the extent to which this range of metamorphism reflects progressive metamorphism of a common starting material and to what extent it is the result of the accretion of material with differing metamorphic histories. It is a question not restricted to type 3 chondrites, and it has been the theme of many papers involving mineralogy and petrology [Dodd et al., 1967; Wood, 1967; Huss et al., 1981; Rambaldi and Wasson, 1981; Suess and Wänke, 1967; Fredriksson et al., 1968; Van Schmus and Wood, 1967], volatile trace elements [Larimer and Anders, 1967; Case et al., 1973; Tandon and Wasson, 1968; Lipschutz and Ikramuddin, 1977, and references therein], major and nonvolatile trace elements [Dodd, 1976; Sears, 1982b], noble gases [Zähringer, 1968], and oxygen isotopes [Clayton et al., 1981].

We have been examining the thermoluminescence (TL) systematics of type 3 ordinary chondrites in the belief that they will eventually shed new light on the general topic of metamorphism and the origin of type 3 chondrites. Sears et al. [1980] discovered that the TL sensitivity of type 3 ordinary chondrites is 10^{-2} to 10^{-5} times that of petrologic types 5 and 6. Furthermore, those meteorites that have long been known to be the least metamorphosed (Krymka, Bishunpur, and Semarkona) have the lowest TL in this range, while those with the highest TL observed in type 3 chondrites (Bremervörde, Dhajala, and Mezö-Madaras) are known to be only slightly removed from type 4. Intermediate in TL and extent of metamorphic alteration is another group including Chainpur, Tieschitz, and Manych. In the present paper we report TL sensitivity data for 16 additional type 3 ordinary chondrites. Many have been examined mineralogically and petrographically, in some cases using the same sample as used here. In these cases, there is a strong relationship between TL sensitivity and metamorphism, consistent with our earlier findings. In addition, the new data fill a previous hiatus in the TL distribution and suggest that two of the present samples, Allan Hills A77176 and Reckling Peak A80207 may approach Bishunpur, Semarkona, and Krymka in their low levels of metamorphic alteration. We also discuss other trends in the TL data and the relationship between TL and oxygen isotopes.

Experimental Methods

The TL data were obtained with apparatus purchased from Daybreak Nuclear and Medical Systems. Since these are the first data we have published that were produced entirely with this apparatus, we will describe our procedures in some detail and show our data for the control samples. The apparatus utilizes an EMI 9635Q photomultiplier tube, with the usual magnetic and electric shielding, and photon counting electronics. Photon counting is preferred over current measurement for the low light levels of type 3 ordinary chondrites, but large dead-times make their use ill-advised at high light levels. The heating strip consists of a nichrome plate to which is attached a chromel-alumel thermocouple. We have detected no deviations from linearity in the heating rate. The system is

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Table 1. Thermoluminescence data on control samples

Sample	Source*	TL sensitivity (Dhajala = 1)	Peak Position (°C)	FWHM (°C)
Dhajala	PRL (T67)		163	167
			173	157
			153	155
			158	157
			165	157
			170	159
			173	169
			173	148
Mean			165	160
Standard Deviation			7.6	6.7
Chainpur	ASU 618.8	0.092	173	102
		0.075	165	92
		0.084	123	85
		0.085	93	85
		0.088	129	87
		0.076	117	97
		0.081	122	92
Mean		0.083	132	91
Standard Deviation		0.006	28	6.4
Krymka	AS 1707	0.0031	165	194

* PRL, D. Lal, Physical Research Lab, Ahmedabad, Navrangpura, India. ASU, C. Moore, Arizona State University (via J. T. Wasson). AS, Academy of Sciences, USSR (via J. T. Wasson).

evacuated to 50 μm of Hg, and oxygen-free nitrogen purchased from Airco is bled into the sample chamber. The apparatus was modified in two ways: the gas and vacuum valves were replaced with metering valves and a 4-mm copper aperture was placed over the heating strip to shield the photomultiplier tube (PMT) from black body radiation from the filament. In early phases of the present work, samples were placed in an aluminum dish 5 mm in diameter, 2 mm in depth, and with a base thickness of 0.01 mm. However, it was discovered by running the control samples listed in Table 1 that our lowest TL samples suffered considerable interference from the TL of the dish, so samples whose TL were below 0.05 were run in a copper dish of similar dimensions.

Where possible two samples of each meteorite were obtained from different naked-eye lithologies. The samples used and their weights are listed in Table 2. A short description was recorded and then the samples were gently crushed in an agate pestle and mortar. Metal was removed with a hand-magnet wrapped in weighing paper and the nonmagnetic fraction gently crushed again. This reduced the grain size to ~100 mesh. The sample was thoroughly stirred and 4 ± 0.5 mg placed in the dish for TL measurement. The sample was then heated to 500°C in the TL apparatus, and exposed to a standard radiation dose.

Irradiations were performed with a 250 mCi ^{90}Sr β source placed ~4 mm from the sample. The irradiation cell is uncalibrated, but the administered dose is calculated to be on the order of 20 krad. The dose administered is unimportant, provided it lies on the linear portion of the TL versus dose curve. This was checked for five samples of highly differing TL. As in previous work we express our TL intensities relative to a 4-mg sample of Dhajala; the homogenized powder used in earlier work was also used here. This practice eliminates the need for absolute calibration of the TL apparatus every time it is used, while making interlaboratory comparisons feasible.

Results

Data of three kinds are read from the glow curve (Figure 1): (1) the intensity of the TL at maximum emission normalized to that of the Dhajala meteorite; (2) the temperature at which the TL emission is maximum; (3) the width of the TL 'peak' (in fact, it is the sum of several peaks) at half its maximum intensity (FWHM). The accuracy with which it is possible to determine the TL sensitivity for a given meteorite has been expressed as the quantity $(100/n)[\Sigma(\Delta x/x)^2]^{1/2}$, where Δx refers to the deviation from the mean, x, for the n meteorites for which duplicates were run. In cases where 200-mg samples were used this value is ~25%; for cases where 50-mg samples were used it is ~50%. The reproducibility of the (Dhajala-normalized) TL sensitivity for an individual sample is taken to be 7%, which is the standard deviation for seven determinations of the TL sensitivity of a Chainpur sample (Table 1). Two of the control samples were aliquots of the samples used in earlier work on other apparatus and the agree-

Table 2. Thermoluminescence and other data on 16 type 3 ordinary chondrites in order of decreasing TL

Sample	Source*	Mass† (mg)	TL Sensitivity‡ (Dhajala = 1)		Peak Position (°C)		FWHM§ (°C)	
			Duplicates	Mean	Duplicates	Mean	Duplicates	Mean
Allan Hills A77216	MWG (38)	46	2.4	1.6	187	191	152	154
	MWG (39)	398	0.74		194		155	
Allan Hills A79022	MWG (16)	211		0.96		189		157
Reckling Peak A79008	MWG (9)	226	1.3	0.89	199	194	145	150
	MWG (10)	199	0.48		189		155	
Bremervörde	BM (33910) via UNM(B1)	108		0.60				
Allan Hills A77304	MWG (40)	524	0.62	0.60	192	195	187	192
	MWG (44)	271	0.58		197		196	
Reckling Peak A80205	MWG (7)	54	0.51	0.55	170	185	172	171
	MWG (8)	70	0.58		199		169	
Outpost Nunatak A80301	MWG (9)	65	0.47	0.52	182	179	172	173
	MWG (8)	43	0.56		175		174	
Allan Hills A77197	UNM	95		0.38		187		
Reckling Peak A80256	MWG (8)	137		0.38		199		174
Parnallee	BM(34792)	50		0.37		136		184
Suwahib (Buwah)	BM (1931,428) via UNM (SB2)	195		0.25		182		164
Allan Hills A77013	UNM	145		0.14		165		162
Allan Hills A76004	MWG (12)	63	0.012	0.034	139	131		
	MWG (14)	126	0.055		122			97
Inman	UNM (C71-12,D)			0.031		177		150
Reckling Peak A80207	MWG (6)	70	0.012	0.013		168		111
	MWG (7)	165	0.014					
Allan Hills A77176	UNM	120		0.010		131		114

* MWG, Antarctic Meteorite Working Group of NASA/NSF. UNM, E. Scott, Univ. of New Mexico. BM, R. Hutchison, British Museum, Natural History. (Catalog or fragment numbers in parenthesis.)

† Refers to mass homogenized for sampling. Measurements were made on 4 mg.

‡ 1σ uncertainties on the mean value assigned to the meteorite are 50% (50 mg samples) to 25% (200 mg samples).

§ Full-width-half-maximum.

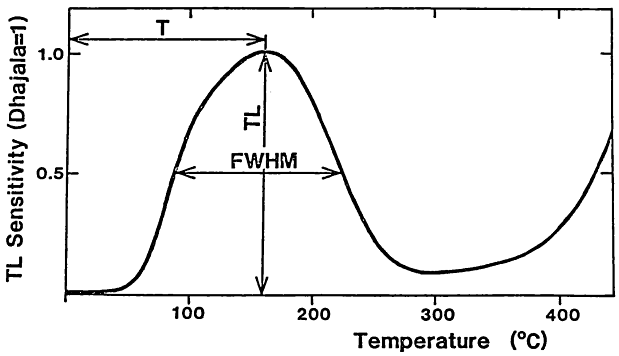

Fig. 1. Glow curve for the Dhajala meteorite showing the way in which TL sensitivity (TL), peak position (T), and full-width-half-maximum (FWHM) are measured.

ment is extremely good (Table 1; previous Chainpur value 0.078, previous Krymka value 0.0027). A third, Parnallee, is a different fragment from a sample previously run and it also agrees well with our earlier determination (0.42).

The uncertainty in the peak position and FWHM has been estimated from eight measurements of the Dhajala meteorite measured on four separate occasions (Table 1). The 1σ uncertainty on the peak position is 5% and for FWHM it is 4%. Seven measurements of the Chainpur meteorite, which has a TL that is much lower than that of Dhajala, yielded 1σ uncertainties for the peak position of 21% and FWHM of 7%.

The TL sensitivities are listed in Table 2 and presented in histogram form in Figure 2 along with data from our previous two studies [Sears et al., 1980, 1982]. The new data fill the hiatus that previously existed between St. Mary's County and Bishunpur, so that we now see a continuous spectrum in TL sensitivity over three orders of magnitude. The distribution is not log-normal, however, but skewed to higher values. It is possible that a few of the higher TL meteorites are different fragments of the same fall, but even if all the Antarctic meteorites were plotted as a single averaged value, the distribution remains skewed. In this respect it resembles the distribution of L chondrites over the petrologic types 3, 4, 5, and 6.

In Figures 3 and 4 we make a comparison of TL sensitivity, peak position, and peak width. For the sake of completeness we also show data we have published previously. Trends are not so easily delineated as in our previous study because of the several low TL samples we have discovered here, but for samples with TL > 0.05 the general tendency for peaks to broaden and move to higher temperatures as TL increases is still present. Samples with TL < 0.05 scatter randomly on the plot.

Discussion

TL Sensitivity and Petrology

Since our previous discussion of the preliminary version of these data [Sears and Weeks, 1983], petrologic data for 11 of the present samples have been gathered by Scott [1983] and

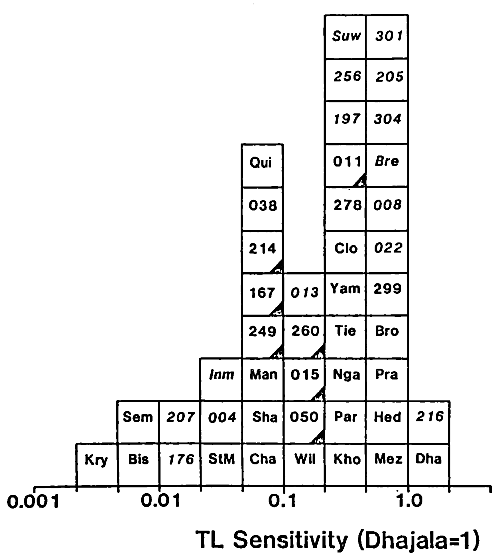

Fig. 2. Histogram of TL sensitivity data for type 3 ordinary chondrites. The meteorite names are abbreviated by the first three letters or last three digits of their names. Identifications in italics refer to samples in the present study and may be identified by reference to Table 1. Others refer to samples from the earlier studies [Sears et al., 1980, 1982]. The small triangles identify samples which are probably paired.

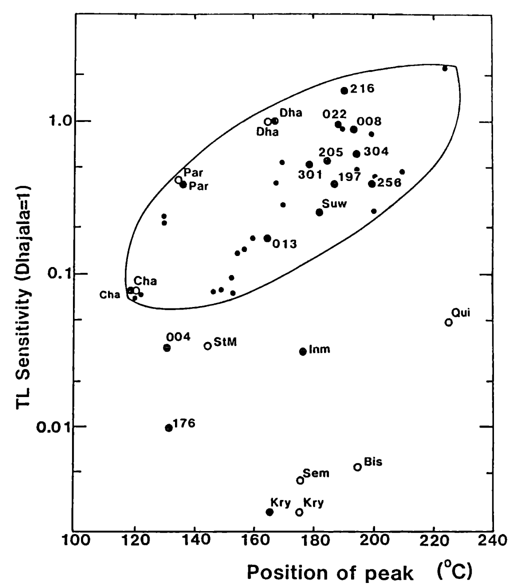

Fig. 3. Thermoluminescence sensitivity plotted against the temperature at which TL emission is at a maximum. Large filled symbols refer to samples in the present study, small symbols refer to samples from the studies of Sears et al. [1980, 1982]. Large open symbols refer to samples lying outside the balloon or to samples whose duplicates were run here.

McKinley and Keil [1983]. In Figure 5 we have superimposed the new data on the TL sensitivity versus olivine heterogeneity plot of Sears et al. [1982]. To avoid confusion and increase clarity, we have omitted error bars from the earlier data which are plotted with small symbols. The large symbols represent the present samples. The reproducibility of each TL value is comparable to the size of the symbols. The accuracy of the C.V.(Fa) values (standard deviation/mean, see Appendix) is 14% at the 1σ level. This has been determined from the data for Allan Hills A77011, A77015, A77260, A77249, and A77167 [see Sears et al., 1982] on the assumption that they are fragments of a single fall. Tie-lines in Figure 5 connect samples from a single meteorite.

The tendency for TL sensitivity to decrease as silicate heterogeneity increases, which was observed in earlier studies, is also apparent in the present data. Where available, detailed petrographic descriptions are consistent with the suggestion that a decrease in silicate heterogeneity, and increase in TL sensitivity, are associated with increasing levels of metamorphism experienced.

Two well-studied chondrites that lie at the low end of the TL sensitivities observed in the present study, are Inman and Allan Hills A76004. According to Keil et al. [1978], Inman contains abundant carbon (0.35 wt %) and much fine-grained matrix as well as highly inhomogeneous silicates (we calculate C.V.(Fa) = 40% from their Figure 5). The carbon content and C.V.(Fa) suggest types 3.3 and 3.5 respectively, while the TL indicates an assignment of type 3.3. Allan Hills A76004 was described by Olsen

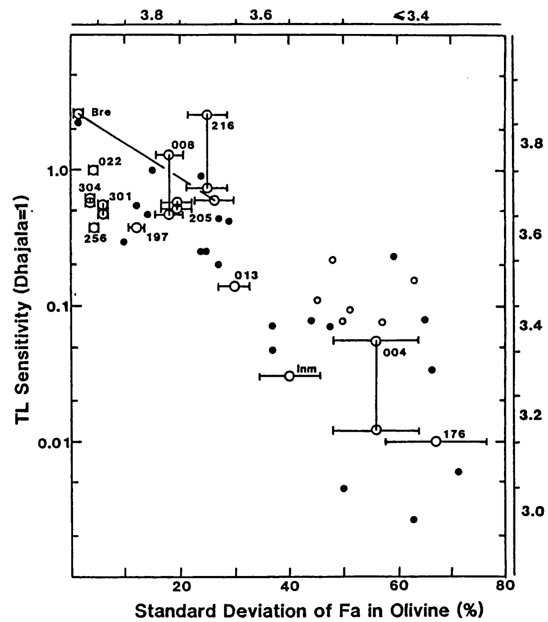

Fig. 5. Thermoluminescence sensitivity versus olivine heterogeneity. The large symbols refer to the present samples, duplicates are connected with tie-lines, and the small symbols refer to data from Sears et al. [1980] and Sears et al. [1982]. Silicate heterogeneity data for the present samples are from Scott [1983] and McKinley and Keil [1983]. Also indicated are the ranges corresponding to each petrologic type.

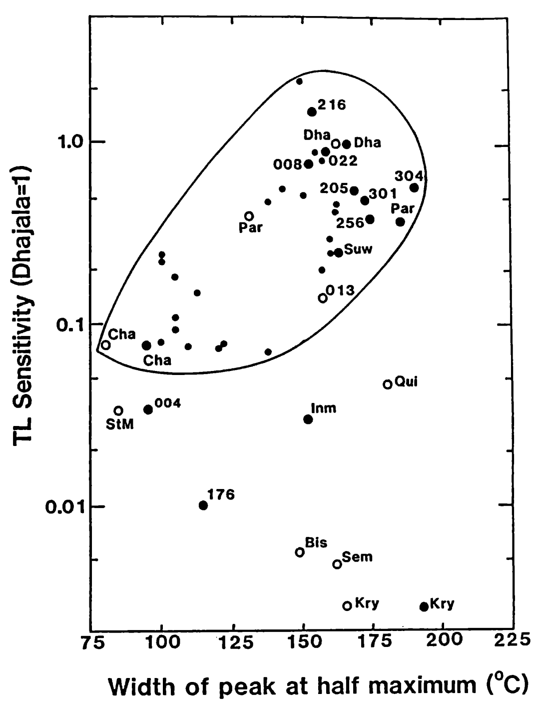

Fig. 4. Thermoluminescence sensitivity plotted against the width of the TL peak at half its maximum intensity. Symbols as in Figure 3.

et al. [1978] as containing much glass and having highly inhomogeneous silicates with C.V.(Fa) = 65% (converted from 50 PMD wt FeO, Olsen personal communication, 1980). Scott [1983] finds C.V.(Fa) = 56% for this meteorite. The heterogeneity figures are both equivalent to type <3.4, while the TL is that of a type 3.3.

On the other hand, Allan Hills A77216, Bremervörde, and Allan Hills A79022 have petrographic descriptions that place them fairly high in the metamorphic spectrum. Both the sample of Bremervörde used here and the sample used by Sears et al. [1980] have TL sensitivities near the top of the type 3 range, and they appear on the basis of silicate homogeneity to be rather highly equilibrated (Table 3). Huss et al. [1981] noted that the amount and composition of opaque and recrystallized matrix, and the C, inert gas content, and matrix olivine heterogeneity of Bremervörde were all consistent with this meteorite being one of the most highly metamorphosed type 3 chondrites. Allan Hills A77216, which was paired with Allan Hills A77215, A77217, and A77252 by Score [1980], was described by her as 'nearer to petrologic type 3 than type 4'. Its silicate heterogeneity was given as C.V.(Fa) = 25% (converted from PMD (FeO) = 20%; see Appendix), equivalent to type 3.7. Allan Hills A79022 appears to be a breccia also lying near the type 3-type 4 boundary, for while Score et al. [1981] found turbid partially devitrified glass and silicate heterogeneity equivalent to a type 3 (Fa_{1-28}, Fs_{9-22}), Scott et al.

Table 3. Thermoluminescence and silicate heterogeneity data for Allan Hills A78084 and two Bremervörde samples

	Allan Hills A78074	Bremervörde (present sample)	Bremervörde [Sears et al., 1980, sample]
TL (Dhajala = 1)	2.3	0.6	2.6
C.V.(Fa)*	0.32%	26%	1.5%
Petrologic type	4	3.7	4

* Coefficient of variation (standard deviation/mean) expressed as a percentage (see Appendix). Data provided by Ed Scott.

[1982] found a C.V.(Fa) = 1.0%, equivalent to a type 4. The TL sensitivity for our single sample is certainly appropriate for a high type 3 meteorite, being equivalent to type 3.7.

Of the seven samples for which we were able to run duplicates, three showed agreement between duplicates within experimental error and four showed differences of a factor of 3 or 4 between duplicates. At least three of the four with poor agreement between duplicates are known to be breccias. The Sears et al. [1980] Bremervörde sample had a TL sensitivity comparable to Allan Hills A78084, which Scott et al. [1982] found to be type 4. Ed Scott therefore measured the silicate heterogeneity of our Bremervörde sample and sent us another fragment of the meteorite with silicate heterogeneity equivalent to type 3.7 material. The present sample had a TL sensitivity much lower than our earlier sample (Table 3); apparently, Bremervörde contains both type 3.7 and type 4 material. Allan Hills A77216 is a gas-rich breccia with solar flare tracks [Goswami, 1981], and Allan Hills A76004, while not known to be gas-rich, is known to be a particularly heterogeneous breccia [Olsen et al., 1978; Scott et al., 1982]. It is probable that the differences between fragments from the same meteorite are due to many meteorites being breccias whose components differ in metamorphic history. In these cases, it is unclear to which of our duplicates, if any, the silicate data for a given meteorite most reasonably apply. We suspect, on the basis of our TL data, that Reckling Peak A79008 is also brecciated, while Allan Hills A77304, Reckling Peak A80205, and Outpost Nunatak A80301 are not. The fresh sample of Parnallee included in the present sample has identical TL to the previous sample we have measured, which suggests that on the scale of our ~5 g fragment, this meteorite is not brecciated.

A detailed study of the TL systematics of one gas-rich breccia (Plainview) was made by Sears [1974, 1982a]. The TL sensitivities of 79 samples were measured, about half being light equilibrated clast and half dark matrix material. The light clasts were a factor of between 2 and 3 higher in TL sensitivity than the dark matrix; this is similar to the difference observed here between duplicate samples

Table 4. Petrologic type assignments based on TL sensitivity and silicate heterogeneity

Meteorite	Petrologic type: TL	Petrologic type: C.V.(Fa)	Petrologic type: Preferred Value	Comments
Allan Hills A77216*	3.8	3.7	3.7(3.7-3.9)	Range based on TL duplicates
Allan Hills A79022*	3.7	4.0	3.7(3.7-4.0)	Range based on TL - C.V. difference
Reckling Peak A79008*	3.7	3.8	3.7(3.7-3.8)	Range based on TL duplicates
Bremervörde*	3.7	3.7	3.7(3.7-4.0)	Range based on TL duplicates
Allan Hills A77304	3.7	4.0	3.8	
Reckling Peak A80205	3.7	3.8	3.7	
Outpost Nunatak A80301	3.7	3.9	3.8	
Allan Hills A77197	3.6	-	3.6	
Reckling Peak A80256	3.6	4.0	3.8	
Suwahib (Buwah)	3.6	-	3.6	
Allan Hills A77013	3.5	-	3.5	
Allan Hills A76004*	3.3	3.5	3.2(3.2-3.4)	Range based on TL duplicates
Inman	3.3	3.5	3.2	
Reckling Peak A80207	3.2	3.7	3.2	
Allan Hills A77176	3.2	-	3.2	

* Breccias; where possible an estimate of the range present has been made.

from breccias. It is not clear from TL data whether the matrix has suffered marginally less metamorphism than the clasts (equivalent to a difference in petrologic type of 0.1-0.2), or whether it is appreciably less metamorphosed but contaminated with equilibrated material. The TL measurement is made on a bulk sample, and tiny additions of equilibrated material will cause large increases in TL (a 10^{-3}% addition of type 6 material to type 3.0 material would double its TL).

Petrologic Types

Despite these difficulties associated with sample heterogeneity it remains desirable and feasible to assign a petrologic type to the overall meteorite [see Table 3 in Sears et al., 1982]. This is because although differences between duplicates of a factor of 3 or 4 sometimes occur, the total range of TL sensitivity in type 3 chondrites covers a factor of 10^3. In Table 4 we list our currently preferred petrologic type assignments for the present samples based on TL sensitivity and C.V.(Fa). As a result of new silicate heterogeneity data and, in one instance, improved TL data, these assignments differ occasionally from those we tentatively proposed earlier [Sears and Weeks, 1983]. Our approach to assigning petrologic types has been described in previous papers [Sears et al., 1980, 1982]. In the present paper we have adopted the additional procedure that in the case of breccias the lowest reliable petrologic type observed is assigned to the meteorite as a whole and the apparent range indicated in parenthesis. Five of the non-brecciated samples considered here have assignments based on TL that are lower than the assignments suggested by silicate heterogeneity; the differences are 0.3, 0.1, 0.2, 0.4, and 0.2 for Allan Hills A77304, Reckling Peak A80205, Outpost Nunatak A80301, Reckling Peak A80256, and Inman, respectively. Based on the accuracy of the TL data for nonbreccias, the uncertainty in the petrologic type assignment is ±0.1; for the silicate heterogeneity assignment the uncertainty is probably somewhat greater, say ±0.2 (see Scott, 1983). The differences are therefore significant and suggest that the petrologic type boundary definitions for the more equilibrated meteorites may need some small modification when more data in this metamorphic interval are available. Our suggested petrologic type for these meteorites is the TL value plus 0.1.

TL Trends with Peak Height and Width

The present data enable us to add nothing new to our interpretation of the tendency for peaks to broaden and move to higher temperatures as TL increases [Sears et al., 1982]. The TL peak is a composite of several separate peaks that are capable of varying independently [Sears and Mills, 1974; McKeever, 1980]. The TL phosphor in type 3 ordinary chondrites is unknown, although a major TL carrier in the Dhajala 3.8 chondrite is located in the chondrules [Sparks et al., 1983]. The TL phosphor in the LL5 Saint-Séverin chondrite was shown to be in the low density, feldspar-rich fraction by Lalou et al. [1970]. This provides a plausible mechanism for the relationship between TL and metamorphism, and its association with chondrules, since throughout the type 3 and type 4 meteorites feldspathic glass devitrifies and feldspar becomes more abundant [Van Schmus and Wood, 1967]. It also provides an interpretation for the tendency of the peak to broaden and move to higher temperatures, since in laboratory experiments terrestrial feldspar showed the same behavior. This was associated with phase transformations in the feldspar [Pasternak et al., 1976; Pasternak, 1978].

The scatter shown by the low TL samples would imply that the TL phosphor is different in identity or physical form from that present in the other samples. Whitlockite has been shown to be luminescent in equilibrated ordinary chondrites, and forsterite, enstatite, and silica, which are present in type 3 ordinary chondrites, luminesce under an electron beam in the microprobe [Sears, 1974; Ed Scott, personal communication, 1983], so that alternative phosphors seem to be a plausible, albeit at the moment speculative, explanation for the behavior of very low TL samples.

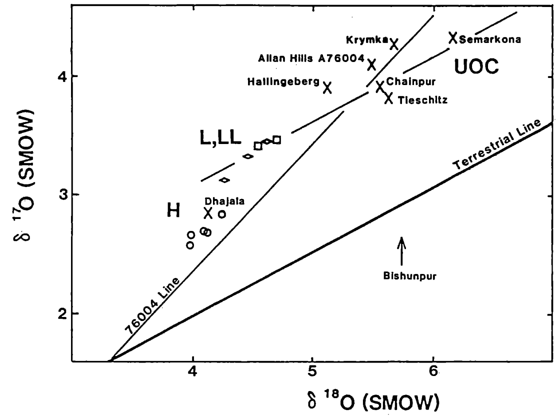

Fig. 6. Three-isotope plot for ordinary chondrites and the terrestrial mass fractionation line. Data for equilibrated meteorites (circles, H chondrites; squares, L chondrites; diamonds, LL chondrites) calculated from Clayton et al. [1976] assuming the following percent modal abundances: olivine-H 35, L 48, LL 58; pyroxene-H 25, L 23, LL 16; plagioclase-all classes 10. Data for type 3 chondrites (crosses) from Clayton et al. [1981] and for Allan Hills A76004 from Mayeda et al. [1980].

TL Sensitivity and Oxygen Isotopes

Type 3 ordinary chondrites are up to 1‰ higher in $\delta^{18}O$ and 0.5‰ higher in $\delta^{17}O$ than equilibrated chondrites (Figure 6). The type 3 ordinary chondrites appear to scatter about a mass fractionation line passing through equilibrated chondrites; in view of the extreme ^{18}O heterogeneity observed in Allan Hills A76004, the scatter is probably due to sample heterogeneity. In Figure 7 we compare our TL sensitivity data with $\delta^{18}O$. Only published L

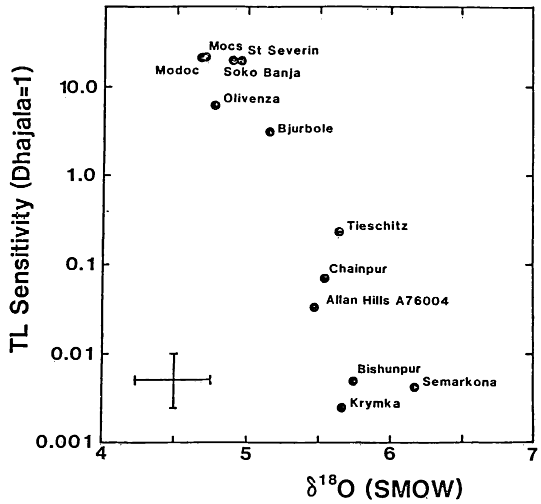

Fig. 7. Plot of TL sensitivity against $\delta^{18}O$.

chondrite data are shown on this plot because otherwise the ^{16}O difference between the classes confuses the picture. Tieschitz is included because in terms of its oxygen isotopes and its metal composition it is apparently an L chondrite, although in other respects it appears to have H group characteristics. For several of the equilibrated meteorites for which oxygen isotope data are available (Mocs, Modoc, Saint-Sévérin, and Bjurbole), TL has not been measured and group means have been used. (This is an acceptable procedure since, while the the total TL range covers a factor of 10^5, within types 5 and 6 it is <10.) The least equilibrated meteorites, which also have the lowest TL sensitivity values, have the highest $\delta^{18}O$ observed, while meteorites with intermediate TL have intermediate $\delta^{18}O$ values. The equilibrated meteorites have highest TL and lowest $\delta^{18}O$ values.

As discussed in the introduction, an important question concerns whether the range of metamorphism observed throughout the type 3 chondrites, and the TL range, reflects progressive metamorphism of a single starting material, and to what extent it reflects the assemblage of material of varying metamorphic histories [a recent discussion is that of Huss et al., 1981]. TL measurement on separated chondrules from Dhajala seems to indicate the possibility that at least some of the TL variability with a given chondrite is associated with primary, chemical differences [Sparks et al., 1983]. It is therefore pertinent to the present paper to consider how the oxygen isotope differences between type 3 and equilibrated chondrites could have arisen. There is no known solid solar system material lying at the opposite end of the mass fractionation line to the type 3 ordinary chondrites that could, by chemical exchange, bring the type 3 chondrites down to the region occupied by the equilibrated meteorites. Clayton et al. [1981] therefore suggested that the type 3 chondrites may be related to the equilibrated chondrites by the loss of ^{17}O and ^{18}O from the type 3 chondrites in the form of CO(g). A plausible reaction is:

$$C(s) + FeO(s) = CO(g) + Fe(m) \quad (1)$$

where carbon is in the form of graphite and FeO as olivine or pyroxene. The amount of oxygen removable depends on the available carbon, assuming the reaction goes to completion (Table 5). For meteorites with intermediate TL and $\delta^{18}O$ (Tieschitz, Chainpur, and Allan Hills A76004) about 1% of the total oxygen can be removed, while for the low TL-high $\delta^{18}O$ meteorites (Bishunpur and Semarkona) about 2% of the total oxygen can be removed. The change in $\delta^{18}O$ as a result of the loss of CO(g) depends on the partition of oxygen isotopes between olivine or pyroxene and CO(g) and is strongly temperature dependent. Use of Onuma et al.'s [1972] compilation of oxygen partition data yields the results in Table 6. At 800 K, 4.2-5.0% of the total oxygen present in the meteorite must be removed to cause a change in $\delta^{18}O$ of 0.5‰; a change of 1‰ requires that twice as much oxygen be removed. It is impossible to remove this much oxygen by the above reaction. However, at 400 K only 1.9-2.8% of the total oxygen has to be removed to cause a change of 0.5 ‰ in $\delta^{18}O$. This does seem plausible in view of the available carbon, especially since the partition data has to be extrapolated from much higher temperatures and factor of 2 uncertainty is not unreasonable.

Several other considerations are necessary before anything more definitive can be concluded from these data, however. There are questions such as whether gas-dust equilibrium could be maintained at such low temperatures or whether diffusion or kinetics become the dominant factor

Table 5. Weight percent of total oxygen which can be 'lost' as CO(g)

	Petrologic type	$\delta^{18}O$	C(wt %)*	O loss (wt %)
Bishunpur	3.1	5.74	0.53	1.9
Semarkona	3.0	6.16	0.57	2.0
Tieschitz	3.6	5.63	0.25	0.88
Chainpur	3.4	5.54	0.44	1.5
Allan Hills A76004	3.3	5.49	0.35	1.2

* Moore and Lewis [1967], Olsen et al. [1978].

Table 6. Weight percent of total oxygen which has to be lost as CO(g) to cause a change in $\delta^{18}O$ of 0.5% *

Temperature(K)	Solid Phase	
	Olivine	Pyroxene
800	4.2	5.0
700	3.5	4.2
600	2.9	3.5
500	2.4	3.1
(400)†	(1.9)	(2.8)

* Based on laboratory partition coefficients for olivine-CO and pyroxene-CO exchange [Onuma et al., 1972].

† Requires considerable extrapolation from laboratory data.

in determining isotopic distributions. It also has to be explained why the highly metamorphosed meteorites, after achieving low temperature equilibration with the gas, did not maintain equilibrium at higher temperatures. There is also uncertainty as to whether the reaction represents open system metamorphism or a nebula reaction; if the latter is the case, then rather restrictive pressure-temperature conditions must be met for a CO-C buffered nebula in a system of cosmic composition (see Sears [1978], Figure 5). Overall these considerations seem to render the conversion of a type 3 meteorite into an equilibrated meteorite more difficult than the calculations in Tables 4 and 5 suggest. Perhaps some process other than the C-CO reaction was involved, or perhaps the type 3 chondrites bear no direct genetic relationship with the equilibrated chondrites. Gas-solid reactions involving H_2O do not lead to large isotope fractionations. Reactions involving less abundant oxygen phases, such as SiO(g), or as yet unidentified heavy-isotope-poor solids, need to be investigated. Our preliminary data on siderophile element abundances [Sears, 1982] and published data on bulk iron contents [Dodd, 1976], do not seem consistent with a simple metamorphic relationship between type 3 and type 6 meteorites.

Conclusions

The present study provides further support for our earlier observations of a relationship between TL sensitivity and metamorphism and permits two observations that may eventually enable identification of the responsible mechanism. First, the hiatus which previously existed in the TL spread has been filled. Second, trends in peak position and peak width with TL sensitivity are still present for samples with normalized TL > 0.05 but absent for samples with TL below this. Additionally, as the TL sensitivity displayed by the chondrites decreases the $\delta^{18}O$ (and $\delta^{17}O$) increases, so that those meteorites distinguished as having extremely low TL, being the most highly unequilibrated in their mineralogy and being volatile rich, have $\delta^{18}O$ ~1‰ higher than equilibrated meteorites. Theoretically, the high $\delta^{18}O$ of the most unequilibrated meteorites could be reduced to the equilibrated values by exchange with CO and its subsequent loss, but very low temperatures and very specific physical conditions are required.

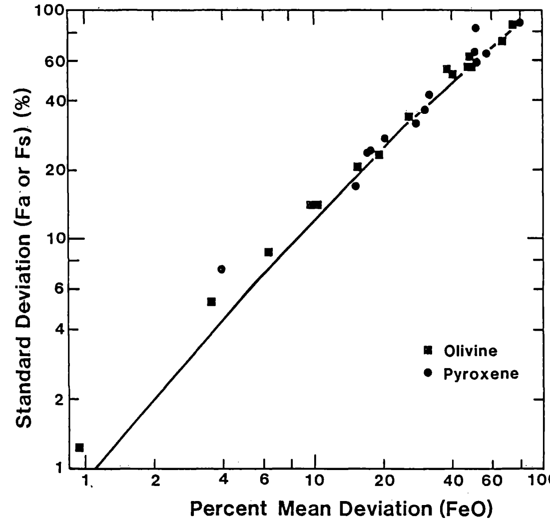

Fig. A1. An empirical plot for converting percent mean deviation of the FeO content of olivine or pyroxene into the standard deviation expressed as a percentage of the mean (C.V.) of the fayalite content of the olivine or ferrosilite content of the pyroxene. See text for sources of data.

Appendix

We agree with McSween and Grimm [1983] that standard deviation is to be preferred over percent mean deviation of the FeO content of the silicate for reporting silicate heterogeneity, although we prefer to use the standard deviation of the fayalite or ferrosilite rather than Fe or FeO. Scott [1983] has also considered this point and come to the same conclusion. Since it is always necessary to quote the mean, in certain applications we use the coefficient of variation of the fayalite content of the olivine or ferrosilite content of the pyroxene [C.V.(Fa) or C.V.(Fs)] which is the standard deviation divided by the mean. Existing PMD(FeO) data have been converted into C.V.(Fa) or C.V.(Fs) using the empirical conversion plot shown in Figure A1. The line is based on invented distributions and the points (crosses for olivine, dots for pyroxene) are calculated from measured data for Antarctic meteorites of widely differing heterogeneity provided by B. Mason. The line agrees quite well with conversions based on Mason's data. It also comes close to the predicted slope of 0.80 calculated assuming normal distributions [Scott, 1983].

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