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Measuring metamorphic history of unequilibrated ordinary chondrites

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A thermoluminescence sensitivity technique is used to give a new measurement of the degree of metamorphism of unequilibrated ordinary chondrites. Consequently the petrological assignment of these meteorites is modified.

THE wide range of textures in chondritic meteorites indicates a variety of metamorphic histories[1–4]. Most meteorites are metamorphosed and it is estimated that they have experienced maximum temperatures of around 1,200 °C (ref. 5). A few (described as type 3 in the scheme of Van Schmus and Wood[6]) have apparently experienced little or no metamorphic heating and their relatively unaltered state makes them interesting subjects for study[7]. We report here measurements which seem to enable the degree of metamorphism experienced by these unequilibrated ordinary chondrites to be quantified. Based on our findings, we propose a modification to the petrological type assignment of these meteorites.

We have used a thermoluminescence (TL) sensitivity technique, sensitivity being defined as the amount of TL induced in a sample by a standard radiation dose. Any previously accumulated TL is first drained by annealing the sample to ⩾500 °C. The TL sensitivity, or TL per rad of absorbed radiation, is an intrinsic property of the luminescent mineral in the meteorite. It should not be confused with the level of natural TL, which is additionally governed by a complex interplay between the thermal and radiation environment of the meteorite.

This work was prompted by a study of H chondrites organized by Hutchison[29]. H. chondrites are distinguished from the other ordinary chondrites (L and LL) on the basis of bulk chemistry, but the TL properties of the three ordinary chondrite classes are remarkably similar. This is presumably because TL is produced almost entirely by feldspar[12], which is present in the same proportions and has similar composition in each group[8]. When the results of our H-group study were available, it was decided to examine some type 3 meteorites. Most of the observed falls studied by Dodd *et al.*[9] were included (Table 1). Our measurements were made at the University of Leicester, UK[10] and at Washington University, St Louis[11]. Samples were lightly crushed and the metal and adhering sulphide removed with a hand-magnet. Three samples produced brown powders due to small quantities of terrestrial corrosion (Kernouvé, Butsura and Limerick). These were shaken for a few seconds in 10 M HCl to restore their normal grey colour, thus reducing an albedo effect which reduces TL[12]. The nonmagnetic portions of the chondrites were ground fine enough to pass through a 100-μm sieve and then heated to 500 °C to remove their natural TL. Leicester samples were given a test dose of 50 krad from a ^{60}Co γ source, whereas the St Louis samples were given a test dose of 25 krad with a ^{90}Sr β source. Samples of the Dhajala meteorite were run on both sets of apparatus and used for normalization to enable a comparison of data from the two laboratories. Using different test doses introduces a small error, but this is not thought to be significant in view of the large effects discovered. The TL sensitivity measurements are reproducible to within ±5%.

Results

Three plots of TL sensitivity against sample temperature ('glow curves') are shown in Fig. 1. These three curves demonstrate the range of shapes observed, although most meteorites produce glow curves which closely resemble those from Khohar. Variations in glow-curve shape are caused by the presence of several separate TL peaks whose relative contributions vary slightly. The shape variations seem unrelated to any of the factors considered here. We have also looked at the best method of extracting data from the curves; two possibilities are to measure the peak height or the area under the curve. We found no difference in the two approaches and present here the former. It also makes very little difference if the TL is measured at a given temperature rather than at the peak, as the glow curves are generally so similar and the effects discovered so large.

The thermoluminescence data are shown in Fig. 2 and listed in Table 1. The average TL sensitivity of the highly metamorphosed type 5 and 6 meteorites is appreciably higher than for the type 3 samples. Furthermore, the TL sensitivities range over a factor of ⩽10 in samples of type 4–6 compared with a range of ~1,000 for type 3 chondrites.

A detailed examination of the accuracy of the TL sensitivity measurements for type 4–6 meteorites has been given else-

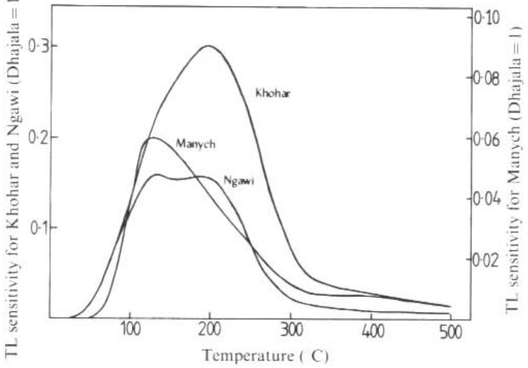

Fig. 1 Plots of thermoluminescence (TL) sensitivity—the TL induced in a sample by a standard laboratory test dose—against temperature to which the sample has been heated ('glow curves'). The TL is emitted mainly in a broad peak between 100 and 300 °C. Most chondrites studied here have glow-curve shapes resembling Khohar, but a few, like Manych and Ngawi, have shapes which differ in detail.

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where[13]. The standard deviation in 10 different meteorites which were expected to have similar TL sensitivity was 21%. Type 3 meteorites are highly inhomogeneous and only small quantities were available for our measurements, so a second chip taken from a different location on the available sample was run. The root-mean-square deviation on the duplicate values is 48%. The error bars on Figs 3 and 4 are ±1 σ of the means.

Discussion

Several parameters for assessing the extent of equilibration in type 3 meteorites have been discussed, but none of them covers the range observed for TL sensitivity or is as free from ambiguity as TL (Fig. 3). The best indication of unequilibration available to Van Schmus and Wood[6] was the inhomogeneity of the silicates. This is expressed as the per cent mean deviation (PMD) shown by the fayalite content of the olivine or pyroxene. The olivine data of Dodd *et al.*[9] are compared with the TL data in Fig. 3*a*; the pyroxene data give a similar plot, but the range in pyroxene PMD is smaller. The inverse correlation indicates that as the meteorites equilibrate, TL sensitivity increases and the silicates become more homogeneous.

In an analogous way, the range of Co contents of the matrix kamacite becomes smaller as the meteorite becomes more equilibrated[7]; we have calculated the PMD of Co in the kamacite and added our own unpublished Tieschitz datum (Fig. 3*b*). Again, there is a clear negative relationship. Figure 3*c* compares our data with the percentage matrix recrystallization[14]. There is some uncertainty in this measurement because it is necessary to discriminate between chondrule fragments and other non-matrix components, but there is a trend in which TL sensitivity increases with the percentage of recrystallized matrix. Huss *et al.*[14] have shown that the FeO content of olivine in the matrix (normalized to the same parameter for the whole rock) decreases with equilibration. One possibility is that FeO is reduced out of the matrix and into the metal, consistent with the views of Wood[15] and Afiattalab and Wasson[7]. This parameter also shows an inverse correlation with TL sensitivity (Fig. 3*d*).

Changes in bulk chemistry can be related to metamorphism in ordinary chondrites, although the causes of these relationships

Table 1 Samples used in the present study, their TL sensitivity, and the laboratory at which the measurements were made

	Class†	Source and catalogue no.‡	Sample weight (g)	TL sensitivity (Dhajala = 1)		Lab. §
				Replicates	Mean	
Kernouvé	H6	BM 43400	~0.100		25	L
Barwell	L5	BM 1965, 57	~0.100		16	L
Ambapur Nagla	H5	BM 81117	1.57		15	L
Appley Bridge	LL6	BM 1920, 40	3.86		14	L
Allegan	H5	BM 1920, 281	~0.100		8.6	L
Ogi	H6	BM 55256	~0.100		8.3	L
Jilin (Kirin)	H5	CAS	~0.100		7.7	L
Butsura	H6	BM 34795	0.69		7.5	L
Limerick	H5		~0.100		6.0	L
Quenggouk	H4	BM 33764	~0.100		6.0	L
Mangwendi	LL6	BM 1934, 839	0.450		6.0	L
Olivenza	LL5	BM 1925, 430	0.870		6.0	L
Beaver Creek	H4	BM 73646	0.995		2.6	L
Bremervörde	H3	BM 33910	0.880		2.6	L
Saratov	L4	BM 1956	~0.100		2.5	L
Monroe	H4	BM 25462	0.796		1.8	L
Dhajala	H3	PRL (T67)	0.30	1.0	1.0	L/SL
Mezö-Madaras	(L)3	HNM	0.030	1.3	0.9	SL
			0.037	0.96		
			0.027	0.82		
			0.031	0.51		
Hedjaz	(L)3	MHNP 2132	0.038	0.85	0.82	SL
			0.042	0.78		
Khohar	L3	ASU 623.1	0.032	0.30	0.44	SL
			0.039	0.58		
Parnallee	(LL)3	BM 34792	0.034	0.57	0.42	SL
			0.034	0.26		
Ngawi	(LL)3	USNM 2483(4)‖	0.029	0.16	0.25	SL
			0.025	0.34		
Tieschitz	H3/L*	BM 1975, M11		0.092		L
		NHMc 2140	0.038	0.20	0.23	SL
			0.039	0.23		
			0.034	0.27		
			0.035	0.22		
Chainpur	(LL)3	ASU 424.1(4)‖	0.032	0.061	0.078	SL
			0.058	0.078		
Sharps	H3	NHNM 640	0.039	0.044	0.071	SL
			0.032	0.098		
Manych	(L)3	AS 2331	0.032	0.061	0.070	SL
			0.058	0.078		
Bishunpur	(L)3/H*	ASU 618.8‖	0.030	0.0073	0.0054	SL
				0.0034		
Semarkona	(LL)3/H*	USNM 1805	0.032	0.0061	0.0045	SL
			0.036	0.0029		
Krymka	(L)3	AS 1707	0.032	0.0035	0.0027	SL
			0.053	0.0019		

† Ref. 6. The chemical classifications of type 3 chondrites are often uncertain. Those marked with an asterisk are based on Co content of kamacite (ref. 7, except our own unpublished value for Teischitz).

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§ L, Leicester; SL, St Louis.

‖ Samples were powders taken from a larger reservoir of homogenized, nonmagnetic powder of unknown sample weight.

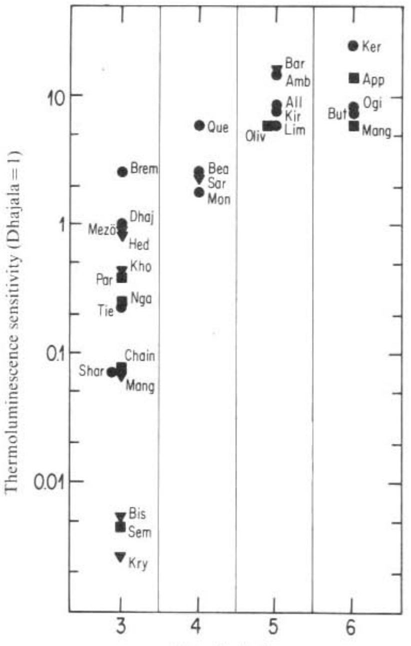

Fig. 2 Plot of TL sensitivity against petrological type for 29 meteorites, normalized to Dhajala. Note the much greater range displayed by type 3 compared with the other types. In this and Fig. 3, meteorites are identified by the first three letters of their names (see Table 1) or some simple variant. ■, LL chondrites; ▼, L chondrites; ●, H chondrites.

are debatable[4,16–18]. The carbon[6] and the primordial ^{36}Ar (ref. 19) contents of chondrites decrease with petrological type. These are compared with TL sensitivity in Fig. 4 (the sources of the ^{36}Ar and C data are given in the legend). These, too, display clear inverse relationships with TL sensitivity.

A relationship between TL sensitivity and petrological type was first suggested by Liener and Geiss[20], but their data fail to display such a relationship because only two of their 70 specimens are unequilibrated[12]. The relationship between TL sensitivity and petrological type is believed to be associated with changes in the crystalline structure of the phosphor, which in most ordinary chondrites is feldspar[21]. However, in unequilibrated ordinary chondrites, the feldspathic material is in the form of a glass[6]. Glasses are intrinsically poorer phosphors than crystalline substances of the same composition because the TL mechanism involves lattice defects or impurity ions in a crystal lattice[16]. The small TL sensitivity that glasses possess is probably due to localized areas of short-ranged order. The transition from glass to crystalline feldspar takes place in meteorites of type 3. In type 4, glass is generally absent, having been replaced by microcrystalline feldspar, although there may be occasional instances of turbid glass. In meteorites of type 5, glass is entirely absent and in type 6, the feldspar has grown to clear, coarse grains[6]. It seems reasonable, therefore, that TL sensitivity should be rather sensitive to the extent of equilibration in type 3 meteorites.

It has been proposed[22,23] that radiation damage will also cause changes in TL sensitivity. Evidence for this has long been sought[20,22,23], but the largest effects so far observed[24] are at most of the order of 25%. In fact, there is some evidence[12] that they may be zero for the TL emitted between 100 and 300 °C (Fig. 1).

The reheating and shock believed to have been suffered by the black chondrites causes a decrease in TL sensitivity comparable with the range observed here. The cause is similar to that responsible for the relationship between TL and petrological type; shock causes a vitrification of the feldspar[25]. The shock events required to affect TL are very large (≥250 kbar) and also cause loss of ^{40}Ar—reflected in K–Ar ages very much less than 4,600 Myr—and characteristic changes in the metallography. None of these meteorites are among those examined for shock effects by Heymann[24], although Wood[15] describes Manych as reheated and Huss *et al.*[14] mention shock effects in their descriptions of type 3 chondrites. Of the three meteorites with the lowest TL sensitivity in the present suite of specimens, ^{40}Ar data are available for two of them. Assuming a K content of 800 p.p.m., taking ^{40}Ar contents from ref. 26 and using the usual decay constants and other data, these meteorites have K–Ar ages of 4.060 Myr (Bishunpur) and 4,100 Myr (Krymka), where in each case the uncertainty is probably ±15%. It seems unlikely that these meteorites have had their TL appreciably lowered by a recent shock event of the magnitude responsible for the low TL sensitivities which have been observed in L chondrites[12]. However, we cannot rule out some complex relationship between very early shock and petrological type which also implicated TL.

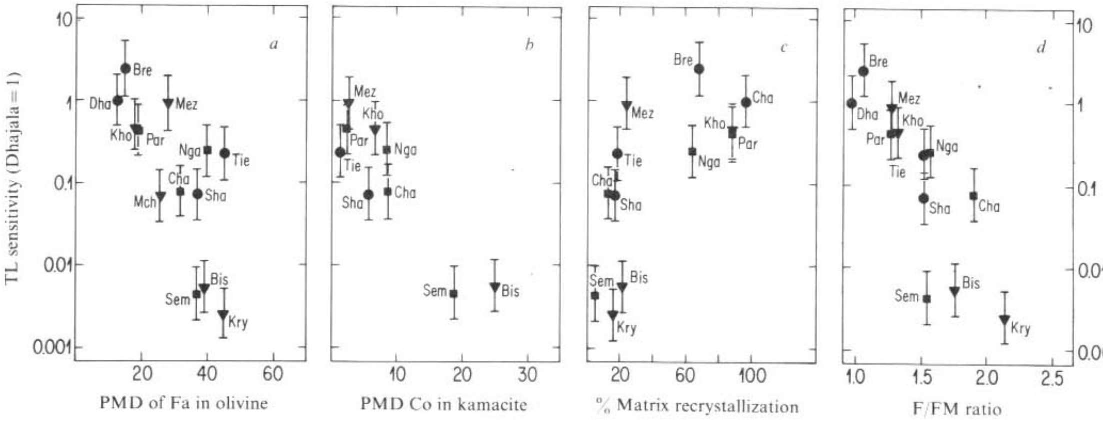

Fig. 3 Plots comparing the TL sensitivity of the type 3 meteorites studied here with literature data which attempt to measure the extent of metamorphic equilibration. *a*, Per cent mean deviation of the FeO content of the olivine[9]. *b*, Per cent mean deviation of the Co content of the kamacite (calculated from data in ref. 7 with our own unpublished data for Tieschitz). These two parameters decrease during metamorphism as the silicate and metal compositions become more homogeneous. *c*, Per cent matrix recrystallization[14]. *d*, FeO/(FeO + MgO) in the matrix normalized to the same parameter for whole-rock[14]. This last parameter decreases with equilibration as FeO is apparently reduced out of the matrix. ■, LL chondrites; ▼, L chondrites; ●, H chondrites.

Table 2 Subdivision of petrological type 3

Type	Member*	TL sensitivity (Dhajala = 1)	PMD olivine†	PMD metal‡	Matrix recrystallization (%)§	F/FM ratio‖	C w/w%¶	^{36}Ar (10^{-8}cm^{3}STP g^{-1})#
3.9	*Clovis no. 1* *Prairie Dog Creek* *Carraweena* *Bremervörde*	2.2–4.6 ✓	5–15	<1.5	>60	<1.0	≤0.21	<4
3.8	Dhajala	1.0–2.2 ✓	15–20	1.5–2.5	>60	1.0–1.1	0.21–0.24	4–8
3.7	Mezö–Madaras Hedjaz	0.46–1.0 ✓	20–25	2.5–4.0	>60	1.1–1.2	0.24–0.27	8–13
3.6	Khohar Parnallee Ngawi Tieschitz	0.22–0.46 ✓	25–29	4.0–6.0	>60	1.2–1.3	0.27–0.30	13–18
3.5		0.10–0.22 ✓	29–33	6.0–8.0	~50	1.3–1.4	0.30–0.33	18–27
3.4	Chainpur Sharps Maynch	0.046–0.10 ✓	33–36	8.0–10	~20	1.4–1.5	0.33–0.38	27–35
3.3		0.22–0.046	36–38	10–13	10–20	1.5–1.6	0.38–0.43	35–45
3.2		0.010–0.022	38–40	13–17	10–20	1.6–1.7	0.43–0.50	45–55
3.1	Bishunpur	0.0046–0.~~0~~010	40–42	17–21	10–20	1.7–1.9	0.50–0.60	55–65
3.0	Semarkona Krymka	<0.0046	>42	>21	<10	>1.9	≥0.60	≥65

* Meteorites in italics were not examined by TL and their present assignment is tentative being based entirely on literature data.
† % mean deviation of fayalite in olivine[9].
‡ % mean deviation of Co in kamacite[7].
§ Ref. 14.
‖ FeO/(FeO + MgO) in the matrix normalized to the same quantity for the whole-rock[14].
¶ Ref. 27.
\# Ref. 26.

Subdivision of petrological type 3

In many respects, and especially in their TL properties, the unequilibrated ordinary chondrites display a more excessive behaviour than shown by any other petrological type. For instance, Bishunpur bears as little similarity to Dhajala, another type 3, as Dhajala does to Kernouvé, a type 6 chondrite. To emphasize this point, we believe that some subdivision of petrological type 3 is desirable. There is no doubt that the metamorphism experienced by type 3 chondrites is gradational and shows no hiatuses. This is reflected in most of the data in Figs 2–4. (The only exception seems to be a possible discontinuity in matrix recrystallization.) This is also true of the more equilibrated meteorites. The sharpness of chondrule outlines and an olivine PMD of 5 are examples of parameters used to define higher petrological type boundaries (5–6 and 3–4 respectively), but neither is a sharp, single-temperature effect and both reflect gradational proceses. The lack of hiatuses in the equilibration parameters for type 3 chondrites, such as TL sensitivity, is not an argument against subdivision.

Afiattalab and Wasson have already proposed subdivision on the basis of metal homogeneity; they divided their nine unequilibrated ordinary chondrites into: four with 'high'; two with 'moderate'; and three with 'low' metal uniformity[7]. Huss *et al.* have proposed subdivision into four categories which they described with Roman numerals[14]. The utility of any subdivision scheme is governed not only by its necessity but also by its simplicity and consistency with existing schemes. Ideally, any subdivision should merge into existing schemes so naturally that it is largely self-explanatory. Terms like 'highly', 'moderate' and 'somewhat' unequilibrated are cumbersome; they cannot always be neatly incorporated into graphs or tables. Letter abbreviations, such as L3h, L3m and L3l, are confusing because current practice is to use letters for chemical class and numbers for petrological types. The simplest approach is to modify the present system by using decimal numbers. We propose that type 3 be considered to consist of 10 subdivisions going from type 3.0 to type 3.9. It might be logical to go from 2.5 to 3.4, so that the numbers round off to 3, but we believe our proposed range will be more popular.

Table 2 summarizes our proposed subdivision scheme for type 3 ordinary chondrites, based on the trends observed in Figs 3 and 4. Three of the subdivisions are unpopulated at present, but we see no obvious reason why these will not eventually be filled. One subdivision contains members which we have not examined, but literature data (mainly PMD olivine) suggest that they should be assigned to petrological type 3.9. They were included for the sake of completeness because they are more equilibrated type 3 members than those examined here. It is possible to assign a petrological subtype on the basis of each of the parameters in Table 2, and such assingments are generally in very good agreement with the TL sensitivity assignment. (We have arbitrarily chosen the upper value where data fall on the boundary between subtypes.) In general, most petrological subtype assignments are within ±0.1 of those listed in Table 2, and this is the uncertainty we would assign to the petrological

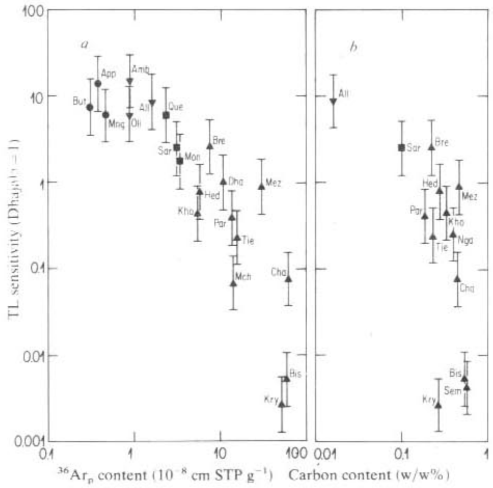

Fig. 4 Two bulk chemical parameters which are known to decrease with increasing equilibration during metamorphism, compared with TL sensitivity; *a*, primordial ^{36}Ar content, calculated from data in ref. 26 and unpublished supplements, assuming primordial ^{36}Ar/^{38}Ar = 5.6 and spallogenic ^{36}Ar/^{38}Ar = 0.65 (ref. 28); and *b*, carbon content. As with TL, the range of primordial ^{36}Ar and C contents is much greater in meteorites of type 3 than any other type. Types: ▲, 3; ■, 4; ▼, 5; ●, 6.

subtype of a given meteorite. There are only three meteorites for which the literature data assignments differ systematically from the TL assignment; the Mezö–Madaras subtype could be decreased by 0.1, the Parnallee value increased by 0.1 and the Semarkona value increased by 0.2. Clearly there would be no problem in assigning a petrological subtype in the absence of TL data, provided there were other good quality data available.

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