Geochimica et Cosmochimica Acta Vol. 46, pp. 2471–2481
0016-7037/82/122471-11$03.00/0

Chemical and physical studies of type 3 chondrites-I: Metamorphism related studies of Antarctic and other type 3 ordinary chondrites

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(*Received August* 10, 1981; *accepted in revised from August* 30, 1982)

Abstract—Thermoluminescence sensitivity measurements have been made on 18 unequilibrated ordinary chondrites; 12 finds from Antarctica, 5 non-Antarctic finds and 1 fall. The TL sensitivities of these meteorites, normalized to Dhajala, range from 0.034 (St. Mary's County) to 2.3 (Allan Hills A78084), and, based primarily on these data, petrologic type assignments range from 3.3 (St. Mary's County) to 3.9 (Allan Hills A78084). Although the very low levels of metamorphism experienced by types 3.0 to ~3.4 evidently cause large changes in TL sensitivity, the new data demonstrate that they are unable to cause any appreciable homogenization of silicate compositions. We have therefore slightly revised the silicate heterogeneity ranges corresponding to the lower petrologic types.

We have discovered that the temperature of maximum TL emission and the broadness of the major TL peak, vary systematically with TL sensitivity; as TL increases these parameters first decrease and then increase. Several mechanisms which could account, partially or completely, for the relationship between TL sensitivity and metamorphism are discussed. Those which involve the formation of feldspar—the TL phosphor in equilibrated meteorites—seem to be consistent with the trends in peak temperature and peak width since experiments on terrestrial albite show that the TL peak broadens and moves to higher temperatures as the stable form changes from the low (ordered) state to the high (disordered) state. (The post-metamorphism equilibration temperature of type ~3.5 meteorites would then correspond to the transformation temperature for the high to low form of meteorite feldspar.) Other factors which may be involved are obscuration of the TL by carbonaceous material, changes in the composition of the phosphor and changes in the identity of the phosphor.

INTRODUCTION

IT IS FREQUENTLY assumed that each class of chondrite meteorites owes its unique combination of bulk chemistry and oxidation state to the distance from the Sun at which it formed. If this is so, then subtle variations in their chemical and physical properties may be interpreted as reflecting conditions at diverse locations in the early solar nebula, provided, of course, it is possible to "see through" the effects of any subsequent metamorphism or other forms of alteration they have experienced. The least metamorphosed, and generally least altered meteorites, are the CI chondrites, this is not withstanding the aqueous alteration they have apparently suffered, (aqueous alteration appears to have redistributed elements without destroying the nebula fractionations, *e.g.*, Kallemeyn and Wasson, 1981; McSween, 1979). In view of their volatile-rich nature and highly oxidized state, they probably formed at fairly low pressures and temperatures, somewhat further from the Sun than the ordinary chondrites (Larimer and Anders, 1967; Wasson and Wetherill, 1979).

The most primitive ordinary chondrites, those termed unequilibrated ordinary chondrites (UOC) by Dodd *et al.* (1967) and assigned to petrologic type 3 by Van Schmus and Wood (1967), show a wide range in the extent to which they have suffered from metamorphism. The range is much larger than displayed by higher petrologic types, and it is necessary to subdivide this petrologic type in order to maximize the value of data obtained from them. At least three schemes, including our own, have been proposed (Afiattalab and Wasson, 1980; Sears *et al.*, 1980; Huss *et al.*, 1981). We proposed a decimal subdivision of type 3 in which the least metamorphosed UOC is type 3.0, and the most metamorphosed is type 3.9. Thus when seeking primordial material from the ordinary chondrite nebula location, it is the low petrologic types 3.0–3.1 that are the most valuable; the identification of such meteorites is one of our principal concerns. We are also interested in the effects of mild metamorphism on the primitive material of the solar system, since this too may lead to an understanding of the evolution of the meteorite parent bodies.

Petrologic studies indicate that when primitive, highly heterogeneous material—such as that found in petrologic type 3.0 chondrites—undergoes metamorphism, various mineralogical and petrologic changes occur. One such effect is that the silicates appear to become more homogeneous in composition. Probably the most dramatic change is displayed by thermoluminescence sensitivity. The 13 type 3

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Table 1. The effect of weathering on TL sensitivity measurements and its removal by acid washing.

Sample	Class	Weathering Grade	TL* Sens.
78084,91	H	C	
Acid-washed			2.3
Unwashed			0.90
77260,22	L	C	
Acid-washed			0.12
Unwashed			0.055
77214,14	L	C	
Acid-washed			0.099
Unwashed			0.034

* Normalized to two measurements of Dhajala which differed by 4%.

ordinary chondrites examined by Sears *et al.* (1980) showed a $>10^3$-fold variation in TL sensitivity, whereas each of the higher petrologic types showed a range of ~10. This behavior is probably associated with changes in the nature and amount of the main TL phosphor(s). In this paper we first report new TL sensitivity measurements on 17 finds and 1 fall. This brings the total number of the type 3 meteorites studied by this technique to 31; less than a dozen remain to be examined. Second, we have made petrologic type assignments on the basis of TL sensitivity, and on silicate heterogeneity. Third, we discuss the correlation that exists between TL sensitivity and glow curve shape. The difference in the glow curve shapes of types ≤3.4 and types ≥3.5 meteorites is tentatively attributed to an order-disorder transition in plagioclase, the TL phosphor. The temperature at which this transition occurs in terrestrial albite is known from laboratory experiments (500–700°C), but controlled studies of the meteoritic feldspar order-disorder transition would be required before palaeotemperatures can be estimated for UOC from their TL properties.

EXPERIMENTAL

The TL sensitivity measurements were carried out in the TL laboratory at Washington University, St. Louis. The apparatus has been described in detail elsewhere (Melcher, 1981a). Powdered samples of ~50 μm grains were prepared by crushing ~200 mg chips and removing the magnetic material with a hand magnet. Aliquots of ~4 mg were placed in 4 mm diameter aluminum pans. These were heated on a nichrome plate at 7.3°C/sec in a chamber which was evacuated and then filled with oxygen-free argon. The samples were heated to 550°C to drain their naturally accumulated TL, and then given a dose of 2.5×10^4 rads with a ^{90}Sr β source. The TL induced by this dose was then measured using an EMI 9635QB photomultiplier tube. The induced TL, normalized to that induced in Dhajala, is defined as the TL sensitivity of the sample.

Thermoluminescence data are most commonly presented as plots of the light intensity emitted by the sample as a function of the sample temperature as it is heated from room temperature to ~550°C. The resulting "glow curve" will exhibit peaks at temperatures related to the energies of the trapped electrons in the sample.

The major experimental problem anticipated in this study was removing the effect of rust in the samples. Sears (1978) has shown that rust causes a $^1/_2$–$^1/_{10}$ reduction in the TL of meteorite samples. The effect may be removed either by preparing the samples as very fine (~10 μm) monolayers of powder, or by washing the powdered samples in 10 M HCl for two minutes. Normally, the first procedure is successful, but the reproducibility of TL measurements on such samples is poor (±15%). Samples prepared in the second way give very reproducible results, but frequently still contain rust. The ease of sample cleaning was a major surprise in the present study. A single acid wash completely removed the rust from all of our samples except two non-Antarctic meteorites (Willaroy and Brownfield) which were still brown colored after three acid washes. Despite the weathered appearance of most Antarctic meteorites (most were assigned to the C weathering category by Score *et al.*, 1981), these meteorites have apparently suffered a much more superficial weathering than the non-Antarctic finds we studied. In order to evaluate by how much our TL sensitivity values for Brownfield and Willaroy are underestimates, samples of three highly weathered meteorites were run without previous acid washing (Table 1). These samples showed 2–3 fold lower TL than their acid-washed counterparts. Hence, we suspect that the weathered state of Willaroy and Brownfield has lowered their TL values by a factor of ~3, resulting in the assigned petrologic type being low by 0.1. This has been taken into account in the final type assignment.

RESULTS

Thermoluminescence sensitivity

Our TL data are listed in Table 2 along with silicate heterogeneity data. Duplicate samples were run for all except Allan Hills A77050,3, and an attempt was made to take samples from widely separated locations on the meteorite. Where possible, two samples which appeared different under the binocular microscope were selected. In general, duplicates show better agreement than in our previous study. An estimate of the uncertainty of the mean of two measurements was calculated from $1/n[\Sigma(\Delta\chi/\bar{\chi})^2]^{1/2}$ where $\Delta\chi$ is the deviation, $\bar{\chi}$ the mean of two determinations and n the number meteorite samples (18 in the present case). The value we obtained, 23%, is appreciably better than the value of 48% obtained by Sears *et al.* (1980), despite the additional handling procedures necessary here. This is probably due to the use of larger samples in the present study (~200 mg compared with 30 mg).

In Fig. 1 are plotted the raw TL data with no corrections for weathering. The samples range in TL sensitivity from 2.3, normalized to Dhajala, to 0.034. Based only on TL, this corresponds to a range in petrologic type of 3.9 to 3.3. The present suite of samples does not include any meteorites as unequilibrated as Semarkona, Bishunpur or Krymka (3.0–3.1) (Sears *et al.*, 1980). The smaller range of TL sensitivities is unrelated to the larger sample size used. Not only are the range of values observed in the Dhajala samples similar in both studies, but our duplicates are chosen to fully represent the ~3 g mass from which the duplicates are taken. The TL sensitivity of St. Mary's County suggests that it is more metamorphosed than is perhaps expected on the basis of the work of Noonan *et al.* (1977) who observed that this meteorite has greater inhomogeneity in its silicate composition than almost any other meteorite. There exists a hiatus in TL sensitivity between Semarkona, Bishunpur and Krymka and the other type 3 meteorites, which has been only partially eroded by these new data.

At the upper end of the TL sensitivity range, we find that Allan Hills A78084 resembles Bremervörde in being a type 3.9 meteorite. On the basis of Dodd *et al.*'s (1967) silicate heterogeneity data we expected Prairie Dog Creek and Clovis to also be type 3.9, but their TL sensitivities are clearly too low for this. Even assuming the TL sensitivity of Prairie Dog Creek is a factor of three low because of incomplete removal of rust, its type can be no higher than

Table 2. Samples run in the present study, in order of decreasing TL sensitivity, and other relevant data.

						Silicate inhomogeneity‡						
						Olivine			Pyroxene			
Sample#	Class*	W.G.**	Sample Wt.	TL sens.+	Mean	No.	C(Fa)	An.	No.	C(Fs)	An.	Pet. Type
78084,35	H	C	0.200	2.3	2.3	10	1.3	1	14	28	1	3.9
78084,91			0.229	2.3								
Prairie Dog Creek		-	0.191	0.55	0.55		12	3		46	3	3.8
			0.066	0.54								
Brownfield@		-	0.500	0.49	0.48							3.6
				0.47								
77299,44	H	A	0.165	0.25	0.47	20	14	1	8	21		3.7
77299,45			0.185	0.69								
Yamato 74191,80	L	-	0.105	0.42	0.41							3.6
			0.078	0.40								
Clovis		-	0.500	0.29	0.29		10	3		40	3	3.6
				0.28								
77278,31	LL	A	0.166	0.35	0.25	84	24	2	14	32	1	3.6
77278,40			0.103	0.15								
77011,7	LL	C	0.154	0.18	0.22	15	48	1	18	64	1	3.5
77011,19			0.159	0.26								
Willaroy@	H	-	0.088	0.15	0.20	31	27	1	6	57	1	3.5
			0.106	0.26								
77050,3		-	0.093	0.18	0.18							3.5
77015,23	L	C	0.256	0.15	0.15	14	63	1	19	59	1	3.5
77015,25			0.226	0.15								
77260,23	L	C	0.256	0.094	0.11	21	45	1	13	84	1	3.4
77260,22			0.184	0.12								
77249,11	L	C	0.231	0.11	0.095	14	42	1	18	60	1	3.4
77249,21			0.301	0.083								
78038,12	LL	C	0.311	0.047	0.078	18	50	1	15	64	1	3.4
78038,14			0.282	0.11								
77214,38	L	C	0.384	0.058	0.078	14	65	1	10	54	1	3.4
77214,14			0.175	0.099								
77167,11	L	C	0.413	0.071	0.075	24	86	1	20	84	1	3.4
			0.211	0.080		100	57	2				
Quinyambie		-	0.110	0.060	0.047	9	37	1	22	43	1	3.4
			0.099	0.035								
St. Mary's Co.	LL	-	0.029	0.040	0.034	269	66	4	274	82	4	3.3
			0.031	0.028								

All Allan Hills meteorites are written simply as the specimen and sample number. They were obtained from the Meteorite Working Group of the NSF. The sources of the remaining samples are as follows: Dhajala (sample T67), Professor D. Lal, Physical Research Laboratory, Ahmedabad, Narrangpura, India. Prairie Dog Creek, Dr. M. Prinz, American Museum of National History, New York. Yamato 74191,80, Dr. K. Yanai, National Inst. of Polar Res., Tokyo, Japan. Brownfield and Clovis, Dr. H. Suess, Univ. of California, San Diego, U.S. Willaroy and Quinyambie (USNM 6076), Dr. B. Mason, Smithsonian Inst., Washington, D.C. Allan Hills A77050, 3, Dr. E.R.D. Scott, Inst. of Meteoritics, Univ. of New Mexico, Albuquerque, N.M. St. Mary's County (USNM) 5930), Dr. R. Clarke, Smithsonian Inst., Washington, D.C.

+ Normalized to two measurements of Dhajala which differed by 4%.

* Noonan et al. (1977), Score et al. (1981), Yanai (1979).

** Weathering Grade from Score et al. (1981); A is unweathered, C is highly weathered.

@ Acid washing was unable to remove the brown coloration due to weathering of these meteorites. Their TL sensitivity data is therefore probably low; judging from the three meteorites for which samples were run in both the acid washed and unwashed state (Table 1), they are probably low by a factor of 2-3. The preferred petrologic types, assigned on the strength of a corrected TL sensitivity, are indicated.

‡ "No." refers to a number of points analyzed, "An." to analyst; 1. B. Mason (unpublished). 2. J.N. Grossman (this work). 3. Dodd et al. (1967). 4. Noonan et al. (1977). C(Fa) and C(Fs) refer to the coefficients of variation (percent standard deviation) of the fayalite and ferrosilite contents of the olivine and pyroxene, respectively.

3.8. Similarly it is doubted that Clovis is as equilibrated as type 3.8.

Other properties are known which can be related, directly or indirectly, to metamorphism and we suspect more remain to be discovered. For example, the strength of the correlation between Na and Al content of separated chondrules increases with metamorphism, being weakest in Semarkona (3.1), stronger but still poor in Chainpur (3.4), intermediate in Tieschitz (3.6) and strongest in Allegan (5) (Osborne *et al.*, 1973; J. N. Grossman, unpublished).

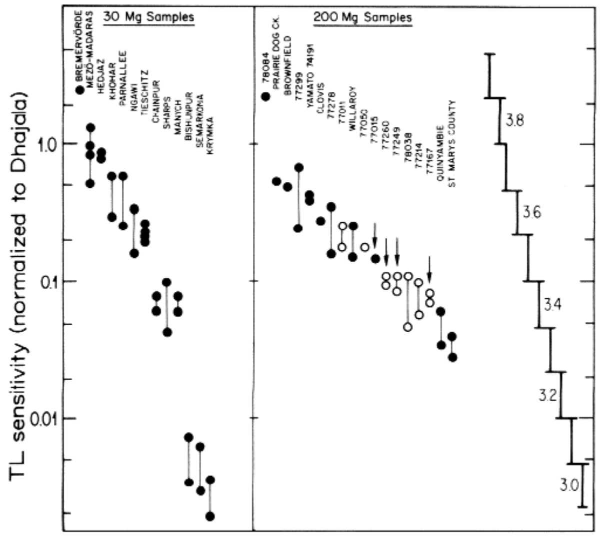

FIG. 1. The TL sensitivity of the present samples compared with the 13 falls studied by Sears *et al.* (1980). No corrections for weathering have been made. For all samples shown, except Allan Hills A77050, duplicates were run; although frequently these were indistinguishable in TL. The open symbols refer to meteorites which McKinley *et al.* (1981) believe to be fragments of a single fall; those indicated with an arrow mark refer to meteorites which B. Mason (pers. comm.) believes are a single fall. There is reasonable agreement between the two authors. In subsequent figures open symbols refer to these possibly associated fragments. The bars and numbers on the right refer to the petrologic type intervals defined by Sears *et al.* (1980). Each Allan Hills meteorite is identified by its number only; this should be prefixed by "Allan Hills A".

Glow curve shape

When given the same test dose of radiation, most ordinary chondrites yield glow curves which are very similar in shape. This is not true of the unequilibrated ordinary chondrites which produce three easily distinguishable glow curve shapes (Fig. 2). Since metamorphism causes such large changes in TL sensitivity, it is plausible that these variations are also related to mineralogical changes occurring in the meteorites.

We will characterize the glow curve shapes by two parameters: (1) the width of the peak and (2) the temperature at which the peak is at a maximum. Both these features are a consequence of the broad band of TL between 80 and 250°C in the glow curve being a composite of several narrow, overlapping peaks (Sears and Mills, 1974; McKeever, 1980). We prepared plots of TL sensitivity against width of the TL peak at half maximum (FWHM) (Fig. 3) and against the temperature of the peak maximum (Fig. 4), including the data from Sears *et al.* (1980). Trends of TL sensitivity against FWHM and peak temperature are clearly present. For types ≥3.4, as the TL sensitivity of the meteorites increases, the position of the apparent peak moves to higher temperatures and it becomes broader. Meteorites of lower petrologic type (≤3.3) such as Bishunpur, Krymka and Semarkona do not follow these trends possibly because the dominant TL phosphor is not crystalline plagioclase. These meteorites comprise a separate and distinct group near the bottom of Fig. 3 and Fig. 4. Quinyambie, on the other hand, is truly anomalous and may be the result of severe annealing (see below).

We will attempt to rationalize these trends below, but it is clear that whatever the explanation, this behavior emphasizes the unique character of the 3.0–3.1 meteorites. Their glow curve peaks are markedly broader and more hummocky than those meteorites just above them in petrologic type (Fig. 2).

The location of St. Mary's County on these plots is also worthy of comment. In both its FWHM and peak position it plots on the main trend and well removed from Bishunpur, Semarkona and Krymka. Thus in both its TL intensity and glow curve shape it should be considered a type 3.3 meteorite rather than type 3.0–3.1.

DISCUSSION

The basis of petrologic type assignment

The petrologic type assignments made by Sears *et al.* (1980) were based on several parameters, in addition to TL sensitivity. With a few exceptions, only TL sensitivities and silicate heterogeneity data are available for the present group of meteorites. It is therefore pertinent to consider in detail the consistency of the two techniques when used to measure the extent to which a type 3 chondrite has been metamorphosed.

Table 3 shows the petrologic type assignments based on various types of data for the type 3 meteorites considered by Sears *et al.* (1980). For each parameter, a TL sensitivity against parameter trend was identified and intervals were chosen to define the petrologic types. These definitions are listed in Table 2 of Sears *et al.* (1980) with modifications to the silicate heterogeneity parameter listed below. From these, a petrologic type can be quoted for each parameter independently of the others (Table 3). This exercise serves to demonstrate two points: first, assignment of a petrologic type between 3.0 and 3.9

can be made on the basis of most of the parameters listed in Table 3 independent of the others. It is also clear that some parameters are better than others for this purpose. This is because the precision and accuracy of the techniques vary, and some mineralogical and petrological systems are more sensitive to the relevant levels of metamorphism than others. It also has to be borne in mind that some systems respond over a wider range of metamorphic intensities than others. Matrix recrystallization, for example, separates less metamorphosed from the most metamorphosed but lacks resolution below type ~3.3 and above type ~3.6. Apparently, most matrix recrystallization occurs during a relatively limited range of metamorphic temperatures. On the strength of present data, metal heterogeneity and carbon data are probably the weakest parameters. Metal composition shows only a weak correlation with the other parameters, probably because data are few (Afiattalab and Wasson, 1980). Carbon content does not show as much spread, and consequently, although showing gross agreement, its petrologic type assignments tend to be discrepant in detail. As will become evident below, the existence of volatile-rich clasts can also make this parameter unreliable.

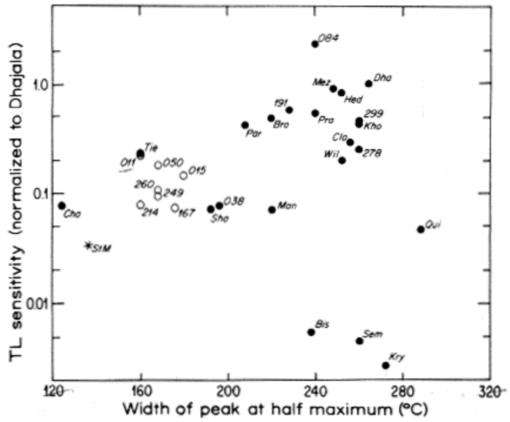

FIG. 3. TL sensitivity compared with the width of the 80–250°C peak. Meteorites are identified by the first three letters or the last three numbers of their names (see Fig. 1). Most meteorites plot in a band in which TL sensitivity increases as the width of the peak increases. The particularly unequilbrated meteorites, Bishunpur, Semarkona and Krymka form a separate cluster. Quinyambie, an anomalous case, is discussed in the text. Error bars for TL are the same as in Figs. 5a,b. Error bars for peak width have not been evaluated but they are probably ±20°C.

The FeO/(FeO + MgO) ratio of the matrix, normalized to the same parameter for the whole rock, varies steadily with the other parameters and is fairly reliable. Primordial ^{36}Ar is similar in this respect. However, in their "resolution", *i.e.*, the total range displayed by all meteorites compared with the repro-

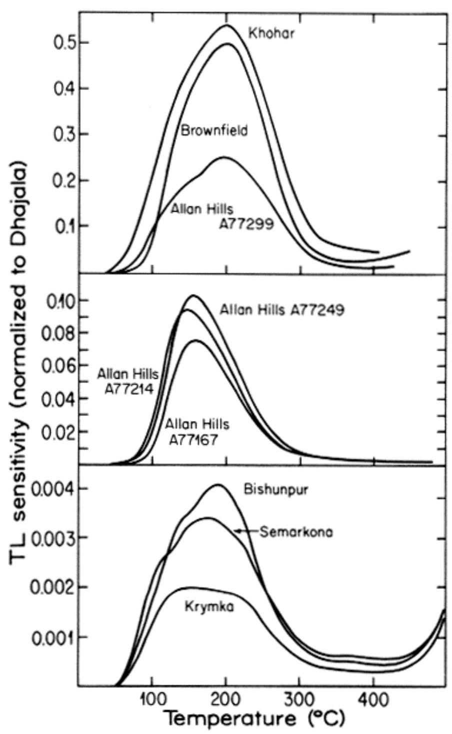

FIG. 2. Glow curves for six type 3 meteorites with very different TL sensitivities. Type ~3.6 meteorites (top) have a broad peak which has a maximum at ~200°C. Type 3.4 meteorites (middle) have a much narrower peak at ~160°C. The most unequilibrated meteorites (type 3.0–3.1, bottom) have very broad, hummocky peaks which have a maxima at ~200°C.

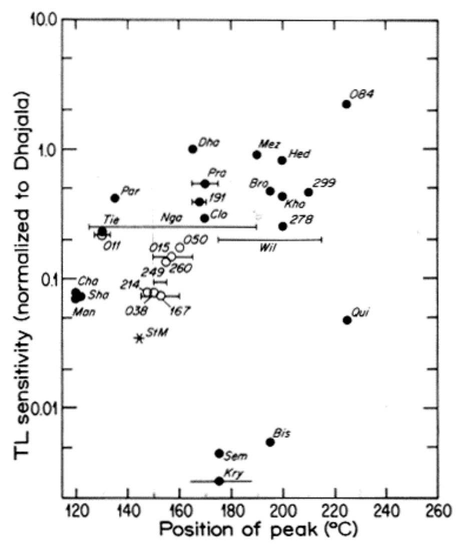

FIG. 4. TL sensitivity compared with the position of the temperature at which TL is a maximum. Most meteorites lie in a band in which the peak moves to higher temperatures as its TL intensity increases, but the type 3.0–3.1 meteorites lie in a group away from this trend. Quinyambie is again discordant. Error bars for TL are the same as in Figs. 5a,b. Error bars for peak position are comparable with the symbol size except where indicated.

Table 3. Assignment of petrologic type on the basis of each parameter separately*.

	TL Sens.	C(Fa)	C(Co)	% Mat. recryst.	F/FM	C	$^{36}Ar_p$
Bremervorde	3.9	3.9	---	3.6	3.8	3.8	3.8
Dhajala	3.8	---	---	3.6	---	---	3.7
Mezo-Madaras	3.7	3.6	3.7	3.4	3.6	3.2	3.4
Hedjaz	3.7	---	---	---	---	---	3.8
Khohar	3.6	3.8	3.5	3.6	3.5	3.5	3.5
Parnallee	3.6	3.8	3.7	3.6	3.6	3.9	3.6
Ngawi	3.6	3.3	3.4	3.6	3.3	3.7	---
Tieschitz	3.6	3.0-3.4	3.8	3.1-3.3	3.3	3.7	3.6
Chainpur	3.4	3.5	3.4	3.1-3.3	3.1	3.2	3.1
Sharps	3.4	3.3-3.4	3.5	3.1-3.3	3.3	3.0	---
Manych	3.4	3.6	---	---	---	---	3.6
Bishunpur	3.1	3.0-3.4	3.0	3.4	3.1	3.1	3.2
Semarkona	3.0	3.0-3.4	3.1	3.0	3.3	3.1	---
Krymka	3.0	3.0	---	3.1-3.3	3.0	3.7	3.2

* References to data used to make the assignments: TL sensitivity, Sears et al. (1980); C(Fa) in olivine, Dodd et al. (1967); C(Co) in kamacite, Afiattalab and Wasson (1980), % matrix recrystallization and FeO/(FeO + MgO) in fine grained matrix normalized to whole rock (F/FM) from Huss et al. (1981); weight percent carbon from Moore and Lewis (1967) except Sharps which is from Fredriksson et al. (1968); primordial argon content calculated by Sears et al. (1980) from a compilation of inert gas contents by Schultz and Kruse (1978).

ducibility of measurements on a single meteorite, where available, TL sensitivity is the superior parameter and the one which can best be used in the absence of others. It is also one of the most accurate because large masses may be homogenized for testing—in this respect it resembles the ^{36}Ar measurement—whereas other techniques necessarily use more restricted sampling. We are confident that silicate heterogeneity and TL sensitivity data are adequate to establish a petrologic type.

The second point which is made by the data in Table 3 concerns the accuracy with which one can ascribe a precise petrologic type to a whole meteorite, as opposed to an individual sample from a meteorite. With the exception of the matrix data, each datum in Table 3 was probably obtained on a separate sample of each meteorite. Different techniques yield essentially the same petrologic type assignments for a given meteorite even though different samples of the meteorite were used. Thus, it is meaningful to subdivide petrologic type 3 meteorites in the way we have suggested.

Table 3 is misleading, however, in one respect; it perhaps implies greater uncertainty in the petrologic type assignment than is true. This is due to the weaknesses in the various techniques as discussed above. In assigning a petrologic type, attention should be paid to the precision of the techniques. TL sensitivity is by far the most precise technique since it has a 10^3-fold range and better than a factor of two agreement between replicates. We believe type assignments on the basis of TL data are good to ±0.1. This is the uncertainty at the 1σ level reported by Sears *et al.* (1980).

[1] This procedure yields standard deviations which are probably good to 5% absolute, or better. The uncertainty is a consequence of the Fa or Fs distributions not necessarily being normal.

Silicate heterogeneity data and petrologic type assignments

In Table 2 we list silicate heterogeneity parameters and TL sensitivity. These are also compared in Fig. 5 where we plot TL sensitivity against the standard deviation of the fayalite component of the olivine and of the ferrosilite component of the pyroxene; both are expressed in mole % and as a percentage of the mean. For comparison, we also show the data for falls obtained by Sears *et al.* (1980). The silicate heterogeneity data for these meteorites are largely from Dodd *et al.* (1967), whose percent mean deviation figures have been converted to percent standard deviations of the mole % Fa or Fs using an empirical conversion plot.[1] We have chosen to use percent stan-

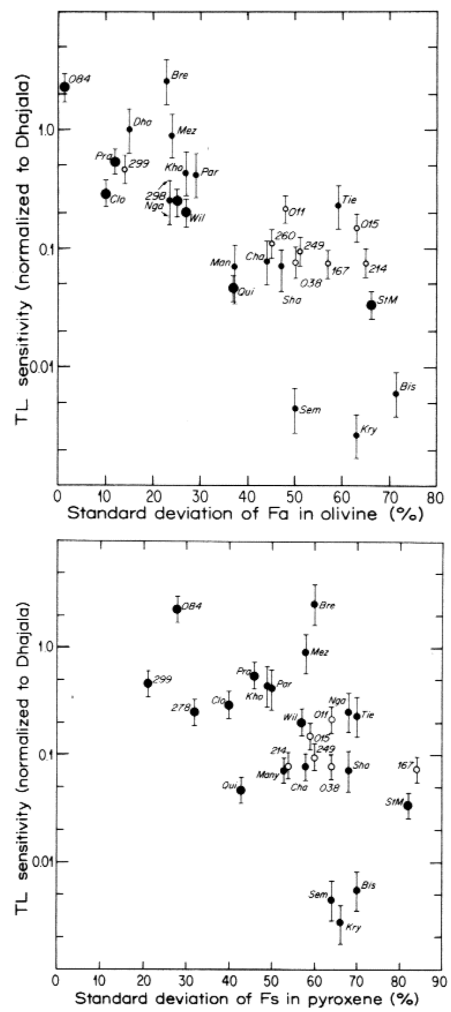

FIG. 5. TL sensitivity compared with a parameter which measures the heterogeneity of the silicates; (a) olivine, (b) pyroxene. Large symbols refer to TL data from the present study and silicate data from the sources listed in Table 2; while small symbols represent meteorites whose TL data is from Sears *et al.* (1980) and whose silicate data is primarily from Dodd *et al.* (1967).

dard deviation (otherwise known as the coefficient of variation), rather than standard deviation, since it is no less statistically valid and it enables comparison of distributions about very different means.

Our data show the same broad tendency for TL sensitivity to decrease as the silicate heterogeneity increases as observed by Sears *et al.* (1980). The explanation we offered before was that as metamorphism homogenized the mafic silicate composition, it also devitrified feldspathic glass to produce the TL phosphor, crystalline feldspar. The new data however, seem to indicate that the correlation breaks down in the most highly unequilibrated meteorites (those with standard deviations ~ 50% or TL ~ 0.05) because the silicate distribution becomes essentially random.

Before accepting this as a real effect we have looked into the possibility that it is due to experimental error. The trends could be restored for example if the heteogeneity data for Allan Hills A77167 and St. Mary's County were erroneously high by ~30%, if the same data for Bishunpur, Krymka and Semarkona were 30% too low, or if the TL data for Allan Hills A77167 and St. Mary's County were an order of magitude too high. We are certain that the TL data are reliable within the stated 48% uncertainty quoted by Sears *et al.* (1980). Not only is it experimentally unlikely that such an error could occur, but we also observed that the glow curve shape parameters (Figs. 3 and 4) indicate that St. Mary's County and Allan Hills A77167 are more like Chainpur, Sharps and Manych (type 3.4) than Bishunpur, Semarkona and Krymka (type 3.0–3.1). At Mason's and Dodd's suggestion, we have made further electron probe measurements on Allan Hills A77167, Bishunpur and Krymka. The St. Mary's County data are thought to be very reliable, since they are based on over 150 points obtained with an automated ARL probe (Noonan *et al.*, 1977). Our data for 77167 are given in Table 2; for Bishunpur and Krymka our σ(Fa) was 71% (72 points) and 63% (73 points), respectively. While these are both ~15% higher than Dodd *et al.*'s (1967) data yield, they are not enough to change significantly the patterns in Fig. 5.

Given that the effect is real—that at the least metamorphosed end of the range, TL sensitivity seems to be detecting changes no longer measureable with the silicate heterogeneity parameters,—what is the explanation? As discussed below, the most plausible explanation for the range of TL sensitivities observed in type 3 meteorites, and its correlation with other

Table 4. Standard deviations of the fayalite of each petrologic type.

	C(Fa)
3.9	5-10
3.8	10-20 (11-20)
3.7	20-30 (21-30)
3.6	30-40 (31-40)
3.5	40-50 (41-50)
3.4	>50
3.3	>50
3.2	>50
3.1	>50
3.0	>50

Table 5. Assignments based on TL sensitivity and olivine heterogeneity.

	TL Sens.	Olivine C(Fa)
78084	3.9	4
Prairie Dog Creek	3.8	3.9
77299	3.7	3.9
Clovis	3.6	3.9
77278	3.6	3.8
Willaroy	3.5-3.6	3.7
77011	3.5	3.0-3.4
77015	3.5	3.0-3.4
77260	3.5	3.8
77249	3.4	3.5
78038	3.4	3.0-3.4
77214	3.4	3.5
77167	3.4	3.0-3.4
Quinyambie	3.4	3.0-3.4
St. Mary's Co.	3.3	3.0-3.4

metamorphic parameters (Sears *et al.*, 1980), is that it is caused by a metamorphism-induced devitrification of glass. It is not unreasonable that homogenization of silicate compositions, which requires atomic diffusion over distances of several hundred μm, is a less facile process than devitrification of feldspathic glass which requires atom displacements over a few A distance. We have performed simple numerical experiments and found that even randomly distributed data produced a σ of only 50%; higher values can only be obtained by producing a distribution with several outliers, suggesting inadequate sampling.

Table 4 shows our currently preferred σ(Fa) values for each petrologic type and Table 5 compares the petrologic type assignments for the present samples made on the basis of TL alone and silicate composition alone. Most of the assignments agree within 0.2, which we think is reasonable in view of the small number of silicate data points sometimes available. Four meteorites agree within 0.1, the stated uncertainty in the TL data.

McKinley *et al.* (1981) recently presented new data bearing on the petrologic type of Allan Hills A77011 and concluded that it is type 3.2 ± 0.2. Matrix crystallization, FeO/(FeO + MgO) of the fine-grained matrix and the heterogeneity of the kamacite suggest that it is 3.5, 3.7 and 3.5, respectively, consistent with the present TL assignment (3.5). Their σ(Fa) value for the olivine is based on a greater number of analyses than ours and is consistent with a petrologic type of 3.0–3.4. We believe that the meteorite is best described as type 3.5 ± 0.1. The remaining data discussed by McKinley *et al.*, which led them to suggest that the meteorite was as unequilibrated as 3.2, was the abundance of carbon and primordial argon. Huss has emphasized that the possibility of primordial compositional differences must also be borne in mind (pers. comm.), and we entirely agree. Allan Hills A77011 is known to contain volatile-rich clasts and in this instance the compositional data are reflecting the presence of these clasts rather than metamorphic history. Sharps has a carbon value (0.6 wt %) which also leads to a petrologic type assignment which is anomalously low compared with other petrologic type parameters (Table 3), and this meteorite is also known to contain volatile-rich clasts (Fredriksson *et al.*, 1968; Rubin *et al.*, 1981).

Mechanism for the relationship between TL sensitivity and metamorphism

Mineral separation experiments demonstrate that in equilibrated meteorites the major TL phosphor is feldspar (Lalou *et al.*, 1970). Mason (1965), Dodd *et al.*, (1967) and Van Schmus and Wood (1967) have noted that feldspar is abundant in equilibrated meteorites but relatively scarce in UOC, and that in some intermediate meteorites, say type 4, appears to have grown by devitrification of glass. The same authors note that glass is abundant in the chondrules of some UOC; Score *et al.* (1981) mention glass in their descriptions of all but two of the Allan Hills specimens discussed here. Observations of feldspar in type 3 meteorites are few and restricted to electron microscope studies of the higher petrologic types, for example, Tieschitz (3.6), Bremervörde (3.9), Hedjaz (3.7) and Parnallee (3.6) (Ashworth, 1977; 1981). Sears *et al.* (1980) therefore suggested the devitrification of glass (low TL sensitivity) to feldspar (high TL sensitivity) as a plausible mechanism for the large changes in TL sensitivity.

A fundamental, temperature-dependent structural change which occurs to feldspar concerns the degree of order in the Al-Si distribution through the feldspar lattice; high temperatures favor a disordered structure whereas low temperaures favor an ordered structure. Smith (1972) has summarized the numerous data concerning the synthesis of albite from albitic glass as indicating that either the transformation from low to high albite occurs at some temperature between 500–700°C, or that the transformation is particularly rapid during this temperature interval. Pasternak and coworkers (Levy, 1978; Pasternak, 1978) performed annealing experiments on a terrestrial low-albite from Amelia, VA, and examined the effect on TL properties. They found that annealing at 1070°C for 25 days caused the TL peak to move to higher glow curve temperatures and broaden; the same trends that are apparent in Figs. 3 and 4 of the present work. Pasternak *et al.* (1976) also measured the X-ray diffraction patterns of Amelia albite after annealing and observed a change from low to high albite. There is some reason for thinking, therefore, that the peak shift to higher temperatures and the increase in peak width with increasing petrologic type may be due to a low-oligoclase/high-oligoclase transformation. Certainly, such an idea is not inconsistent with current views and data on the kinetics of meteorite TL and luminescence activators in other systems in which feldspar is the phosphor (Sippel and Spencer, 1970; Geake *et al.*, 1971; Valladas and Lalou, 1973; Sears and Durrani, 1980; McKeever, 1980; Melcher, 1980, 1981b). If this is the correct interpretation of our data, then TL has potential use as a palaeothermometer for type 3 meteorites.[2] Meteorites of type 3.3 to 3.5 appear to contain feldspar which equilibrated below the low/high transformation temperature (this temperature would be 500–700°C if the laboratory data reviewed by Smith could be applied to ordinary chondrites), whereas meteorites of types $\gtrsim$3.5 have feldspar which equilibrated at temperatures above this so that their feldspar is in the high (disordered) structural state. The glow curve shapes of meteorites of type $\leq$3.2 suggest that they have not been annealed sufficiently to produce crystalline feldspar. Their TL emission is probably produced by feldspathic glass and other minerals with very low TL sensitivity.

It is worth emphasizing that meteorites of petrologic types 5 and 6 produce glow curves which are similar to those of type $\gtrsim$3.5 in their width and peak position (see, for example, Fig. 1 of Sears, 1978) and the feldspar in these types 5 and 6 always has a high-oligoclase structure (Van Schmus and Ribbe, 1968).

This explanation for the trends in peak position and width says nothing about the means by which the feldspar formed. Huss *et al.* (1981) observed that plagioclase occurs in the recrystallized matrix and not in the opaque (non-recrystallized) matrix. Apparently some feldspar was formed by chemical reactions during metamorphism. This process would cause an increase in TL sensitivity and it would produce the trends observed in Figs. 4 and 5. Observations of devitrifying glass in the type 3 meteorites are more frequent than observations of feldspar in the matrix, but this may be a selection effect since while chondrules have been much studied, the matrix has not. Similarly, if the feldspar were being formed in various amounts at various temperatures by nebula processes, its crystallography would depend on temperature and the observed trends would still occur.

It is possible that several other factors may be involved in determining the TL sensitivity of a type 3 meteorite, although it is not necessarily clear at the moment how any of these could cause the TL peak to broaden and move to higher temperatures. The inverse correlation observed between carbon abundance and metamorphism suggests that carbonaceous material may coat the grains and produce an effect by obscuration (R. M. Walker, pers. comm.) or by changing the sample's albedo (see Sears, 1980). This possibility could be readily tested in the laboratory. Less amenable to experimentation is the possibility of systematic, metamorphism-related changes in the minor or trace element content of the TL phosphor which could also change its TL properties in a systematic way. This is difficult to measure directly because the phosphor may constitute a very minor phase. More readily explored is the idea that several

[2] It is interesting to note that if the TL sensitivity is a measure of the amount of feldspar present, then we are detecting and, perhaps identifying the crystallography of a phase which is $\lesssim 10^{-3}$ wt% of the meteorite. We understand that measurements with a TEM, or similar instrument, are unable to detect and measure such a minor constituent.

different TL phosphors are present in type 3 meteorites, and that each dominates over a different range of metamorphism. This is especially true of type 3.0–3.1 meteorites where the intensities are very low and the glow curve shapes do not follow any particular trend. This can be checked by cathodoluminescence measurements, provided sufficiently sensitive equipment is available.

Meteorites which are fragments of the same fall

Mason has frequently observed (Marvin and Mason, 1980; Score *et al.*, 1981; and B. Mason, pers. comm.) that many Allan Hills meteorites may be fragments of a single fall on the basis of his petrographic and electron microprobe studies. McKinley *et al.* (1981) have similarly argued that their observations on the abundance of inclusions of graphite and magnetite are evidence that many Allan Hills UOC are members of a single fall. We will not discuss this point at length, other than to summarize the relevance of the present data to the question. Suggested associations are summarized in Fig. 1 by the symbols described in the figure caption. The majority of the meteorites thought by these authors to be members of a single fall, Allan Hills A77260, 77249, 78038, 77214 and 77167, have similar TL sensitivities. Allan Hills A77011, 77050 and 77015 have identical TL sensitivity to each other but higher by a factor of about 2 from those above. It is possible for replicates of the same meteorite to differ in TL sensitivity by a factor of 2, but in the present data, it is rare (only three cases out of 17). We think that our data probably indicate that these meteorites are from two falls rather than one, but clearly there is nothing in these data to suggest that each of these meteorites is a separate fall. The normalized level of natural TL (*i.e.*, the "equivalent dose") can also be used to look for evidence of associations. Equivalent dose data exists for three of the Antarctic meteorites studied here: Allan Hills A77214, A77278, and A77299 (Melcher, 1981b). Although the TL sensitivities of Allan Hills A77299 and A77278 are similar, their equivalent doses at 200°C differ by more than a factor of three which possibly suggests that they are not members of the same fall. However, variations of this order within a single meteorite are not unknown (Lalou *et al.*, 1970). Allan Hills A77214 appears to represent yet another fall on the basis of both equivalent dose and TL sensitivity. At this point no single parameter appears capable of conclusively identifying members of a fall and consequently all available evidence must be evaluated. It should be noted that all those meteorites which are probably fragments of a single fall cluster much more tightly in TL sensitivity than in their σ(Fa) and σ(Fs) values.

Quinyambie

Quinyambie does not conform to the trends in Figs. 3 and 4 and is anomalous in several respects. Its glow curve contains a broad peak with a maximum which is at a relatively high temperature.

A possible explanation is that Quinyambie has suffered annealing of a kind different from that responsible for metamorphism. Laboratory experiments have shown that when highly equilibrated meteorite samples are annealed or artificially shock loaded in the laboratory their TL sensitivity decreases. According to Sears (1980), a temperature of $\sim$1000°C for 1 hour, or a shock pressure of 335 kbar for $\sim$5 μsec (which is presumably accompanied by some annealing) causes a ten-fold decrease in the TL sensitivity of equilibrated meteorites. A detailed study is still in progress, but it seems likely that the lower temperature part of the TL emission (say $\lesssim$150°C) would be destroyed at lower pressures and temperatures than the higher temperature part (say $\gtrsim$150°C). This would explain the TL data. However, we have examined a Smithsonian thin section of Quinyambie (USNM 6076) but were unable to find evidence for a shock event capable of affecting TL sensitivity.

A second possible explanation for Quinyambie would be the presence of an additional TL phosphor (either natural or a contaminant) with peak emission at 200°C. If this emission were combined with a typical type 3.4 glow curve, the broad Quinyambie glow curve could be produced. A third possible explanation would be the existence of anomalously large amounts of minor TL phosphors and a marked deficiency of feldspar.

The importance of shock events in the history of several meteorite classes has often been discussed. Since feldspar is a compressible mineral it readily absorbs shock energy and may invert to maskelynite. Probably as a consequence, heavily shocked meteorites have a TL sensitivity 10 times, or more, lower than unshocked meteorites (Sears, 1980; Sears *et al.*, 1982). In the case of type 3 meteorites shock events may play an even more important role since shock heating may be the main source of heat for the metamorphism (Huss, 1980; Sears and Marshall, 1980; Huss, 1980; Huss *et al.*, 1981). However, although caused by essentially the same phenomena, the two types of shock have affected the TL very differently. In the case of type 3 meteorites, it has provided heat which devitrified isotropic glass, while in the case of black chondrites it has produced an anisotropic glass from crystalline feldspar. Apparently, the difference is in the intensity of the shock, the nature of the material being shocked, and the cooling rate following it. Type 3 meteorites cooled rather slowly ($\sim$1°C/10^6 y, Wood, 1967), while heavily shocked chondrites cooled very rapidly ($\sim$1°C/10^3 y, Smith and Goldstein, 1977).

CONCLUSIONS

1) The 17 UOC meteorites studied here display a correlation between TL sensitivity and the heterogeneity of the silicate compositions similar so that observed by Sears *et al.* (1980) in 13 UOC falls. The correlation appears to break down in the most heterogeneous meteorites. Metamorphism does not appear to begin to homogenize silicate compositions until metamorphic temperatures equivalent to type $\sim$3.4 are reached, and therefore this parameter cannot resolve petrologic types in the range 3.0–3.4. Based on variations in TL sensitivity and silicate homogeneity we assign the petrologic types given in Table 2.

2) The shapes of the glow curves of type 3 meteorites show systematic changes associated with metamorphism. Comparison with laboratory annealing experiments suggests that the change in shape could be associated with the order/disorder transition in feldspar which would imply that meteorites of type 3.3 to 3.5 have feldspar in a form analogous to low-albite, meteorites of type $\sim$3.5 have feldspar in the high form and meteorites of type $\lesssim$3.3 have feldspathic glass and/or other minerals as TL phosphors. We discuss various other mechanisms for producing the observed TL variations, but, for types $\geq$3.2 do not find them as plausible as the devitrification of feldspathic glass.

3) Several Antarctic meteorites which were paired by various authors have been examined. The TL data show a much more restricted range in TL sensitivity

than in silicate heterogeneity, and by and large, are consistent with the proposed pairings.

4) Quinyambie is the only meteorite which is not consistent with the observed trends. Although several possible explanations are put forth, none is totally satisfactory and the behavior of this meteorite remains unclear.

Acknowledgements—We are grateful to the donors of the meteorites listed in Table 2 and Dr. B. Mason for his interest, and for providing much unpublished data. We are also grateful to K. Keil, B. Mason, E. Scott, R. M. Walker and J. T. Wasson, for hospitality, financial support and discussion, R. S. Clarke for rapidly responding to our request for thin sections and A. Gaines for supplying a copy of the Pasternak thesis. The manuscript has also profited much from thoughtful reviews by J. V. Smith, G. Huss, D. S. Burnett, B. Mason, J. A. Wood, E. Scott and W. R. Van Schmus. The research was also partially supported by NASA grant NGR 05-007-329.

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