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THE THERMOLUMINESCENCE CARRIER IN THE DHAJALA CHONDRITE

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Abstract. The thermoluminescence (TL) sensitivities of 58 chondrules separated from the Dhajala (H3.8) chondrite have been measured in order to investigate the cause of the 10^5-fold variation in the TL sensitivity of ordinary chondrites. The TL sensitivities of the individual chondrites range over nearly two orders of magnitude, from 0.002 to 0.12 (where 4 mg of bulk Dhajala powder = 1), with no correlation with chondrule mass, cross-sectional area or diameter. Some chondrules have TL per unit mass ten times that of bulk Dhajala powder and mass-TL balance arguments suggest that these chondrules are a major TL carrier. The composition of 15 chondrules was also determined and it was found that high TL chondrules tend to have Ca contents at the upper end of the range observed. These observations are consistent with the TL sensitivity variation in ordinary chondrites, which is related to metamorphism, being caused by the devitrification of glass to produce feldspar and with feldspar being the dominant TL phosphor in Dhajala. That chondrules are an important TL carrier would also be consistent with primary feldspar as the TL phosphor, but petrologic observations probably make this appear unlikely.

Introduction

The type 3 (unequilibrated) ordinary chondrites provide a major source of information on the early solar system and the physical and chemical processes occurring there. However, disentangling such data is complicated because all but a few, such as Bishunpur and Krymka, display signs of metamorphic alteration. Others, such as Bremervorde and Allan Hills A78084, display many features which resemble the relatively highly metamorphosed type 4 meteorites [Dodd et al., 1967; Wood, 1967; Afiattalab and Wasson, 1980; Huss et al., 1981].

Light metamorphism of this primitive solar system material apparently produces many dramatic effects; it recrystallizes and changes the chemistry of the matrix and it homogenizes the composition of the silicates and metal. Associated with increasing metamorphism are lower abundances of volatile elements and carbon, although it is uncertain whether this is a direct or indirect relationship [e.g. Wasson, 1972; Anders, 1971]. Probably the most spectacular effect, and also the most readily and accurately measured, is that TL sensitivity increases from a Dhajala-normalized value of ~0.003 in Bishunpur to ~2 in Bremervorde and Allan Hills A78084. It displays even higher values (5-50) in type 5 and 6 meteorites [Sears et al., 1980].

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We are attempting to understand the mechanism for this large range in TL sensitivity in type 3 meteorites and its relationship with metamorphism. We believe it will enable new insights into the history of the unequilibrated ordinary chondrites (UOCs) in, perhaps, two ways. First, TL may enable palaeotemperatures to be determined for these meteorites while, for various reasons, other techniques fail [Sears et al., 1982]. Second, it may tell us something about the relationship between various type 3 ordinary chondrites; whether they are related directly by metamorphism or whether each is an unaltered accretionary rock whose components were metamorphosed before accretion [Huss et al., 1981; Anders, 1971; Suess and Wanke, 1967; Fredriksson et al., 1968]. This is because if the mechanism behind the TL range involves intrinsic, perhaps nebula-related differences in the phosphor, then metamorphism alone cannot change a type 3.0 into a type 4. Alternatively, if the mechanism involves progressive creation or destruction of phosphors, then the relationship between types 3.0 and 4 could be simple and involve only metamorphism.

Sears et al. [1980] suggested that a plausible mechanism for the variation of TL sensitivity with mass is that feldspar, which is known to be the TL phosphor in equilibrated chondrites, is formed by the devitrification of glass in response to metamorphism; in the higher petrologic types, this phenomenon can be petrologically observed [e.g., Van Schmus and Wood, 1967]. Except for tiny amounts of shock-produced material, the glass observed in chondrites exists in the chondrules. If TL intensity increases due to the formation of the phosphor through devitrification, one might expect chondrules to be a major TL carrier. In the present paper, we report TL sensitivity measurements on 58 chondrules separated from the Dhajala meteorite. The data were discussed briefly by Sparks and Sears [1982] who made two observations: 1) a large fraction of the TL is concentrated in a small proportion of the chondrules; 2) chondrules show a very large spread in their TL sensitivities. They also pointed out that while the difference in TL sensitivity between various UOCs is related to metamorphism, the range of TL observed within the Dhajala chondrules is not due to individual chondrules experiencing differing degrees of metamorphism, but rather is due to the chondrules showing variations in their ability to respond to metamorphism. In this paper we wish to concentrate on the constraints placed by these data on the mechanism by which metamorphism is related to TL sensitivity, and save for a later time a discussion of how our data relate to the origin and history of chondrules and chondrites.

Experimental Procedures

A 1.6 g portion of the Dhajala chondrite gently crushed and 58 chondrules were removed by hand-picking. An almost log-normal spread of diameters between 0.15 and 1.0 mm was observed, with a mode appearing between 0.4 and 0.5 mm. Identical results were obtained on a second fresh 200 mg sample from which 30 chondrules were extracted (histograms showing our size distributions are available on request). The distribution we observe is in perfect agreement with those obtained by Martin and Mills [1978] and Hughes [1978a,b] who used both chondrule separation and thin section techniques. We are therefore confident that our chondrule sample is representative. We are unable to explain why Martin and Mills [1976] and Gooding et al. [1978] obtained very different chondrule size distributions, especially since the former authors used the same technique and meteorite as Hughes [1978a].

The chondrules were examined under the binocular microscope and cleaned of adhering matrix by picking with a needle. The diameter of each chondrule was recorded. Each condrule was then crushed and a suitable piece retained for thin section production. The remainder was used for TL measurements. This sampling may introduce some error if the sample removed was atypical. Typically we removed one-third to one-fourth of the chondrule for section preparation and since chondrules are not normally heterogeneous on this scale [e.g., see Gooding and Keil, 1981], we doubt that this introduced any serious error. The mass of each chondrule was calculated from its diameter, assuming a density of 3.3 g cm^{-3} [Hughes, 1978b]. Allowing for uncertainty in the radius of $\pm$0.1 mm and in the density of $\pm$0.3 g cm^{-3}, the uncertainty in the calculated mass is $\pm$30% at the upper end of the size range and $\pm$ a factor of 2 at the lower end of the size range. Although large, these uncertainties are not large enough to appreciably affect either the trends observed in our data or our discussions.

The TL was measured in the TL apparatus at Washington University, St. Louis [Melcher, 1981]. Four-milligram aliquots from a gram-sized sample of the meteorite were drained of their natural TL by heating to 500°C, and their TL measured after administering a 25 krad dose from a strontium-90 radiation source. A heating rate of 7°C/s was used. The TL sensitivities quoted are the height of the major TL peak which appears between ~80°C and ~250°C. Measuring the area under the peak yields almost identical results and we have arbitrarily chosen to use the former.

After TL measurement, an attempt was made to transfer the powders to a heating strip in an apparatus similar to that described by Brown [1977] for the preparation of fused glass beads. These were mounted in resin, ground, polished, carbon coated and placed in a Cambridge Stereoscan 600 electron microscope with an automated Tracor Northern NS 880 energy dispersive X ray spectrometer attachment for bulk analysis. Although for our present purposes we are interested primarily in Ca, analyses for Mg, Si, Al, and Fe were also performed. The standards used were Johnstown hypersthene (for Mg, Si, and Fe), anorthite (for Ca), and pure Al. The data were reduced using the Tracor version of the Bence-Albee program. We will present a detailed discussion of the data obtained and our precision and accuracy elsewhere, but for Ca data we think our precision is currently about $\pm$20% of the value quoted. This error estimate is the standard deviation of five analyses of U.S. Geological Survey standard BCR-1 and it includes the effect of errors in analysis and sample preparation.

Results

Our TL sensitivity data are presented as a function of mass in Figure 1. Although the chondrule masses also show a range of about two orders of magnitude, there is clearly no relationship between the two parameters. The high TL chondrules seem to cover almost the entire mass range we observed. An important conclusion, therefore, is that the variation in TL from chondrule to chondrule must be extremely large, so that it overwhelms the very large range in chondrule mass or size. The TL sensitivity data are presented in histogram form in the lower part of Figure 2. Data are quoted relative to a 4 mg aliquot from a gram-sized sample of the Dhajala meteorite. The values spread over nearly two orders of magnitude (0.002 to 0.12) with a mode of ~0.01. There is some hint of a bimodal distribution with 'high TL' chondrules having values ~0.1, and the remainder concentrated between ~0.002 and ~0.02. Mass-normalized TL sensitivities for the individual chondrules are plotted in the upper histogram in the figure. The calculated masses range from 5.3 μg to 1.6 mg and the TL/mass values range over nearly four orders of magnitude. Crushing such small objects produces three or four fragments, rather than fine powder, so that the effect of chondrule size on TL may be better represented by cross-sectional area rather than mass. However the plot of TL against the cross-sectional area of the chondrules is very similar to Figure 1, with no hint of a correlation. Similarly, a plot of TL sensitivity against chondrule diameter shows no indication of a correlation.

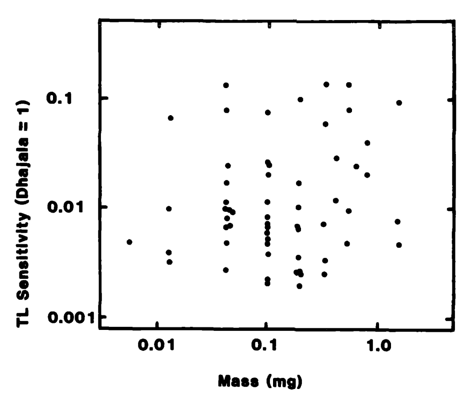

Fig. 1. Dhajala-normalized TL sensitivity of the separated chondrules plotted against chondrule mass.

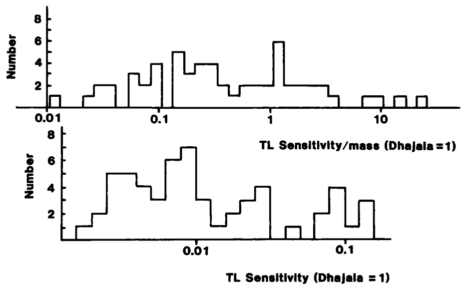

Fig. 2. Histograms showing the thermoluminescence sensitivity of separated chrondules from the Dhajala meteorite. Below, the TL is expressed relative to that displayed by 4 mg of bulk Dhajala non-magnetic powder. Above, this normalized value has been divided by the calculated mass of the chondrule.

About one-fourth of our samples, generally the larger ones, were successfully prepared for chemical analysis and yielded reasonable data. The major difficulty was handling the material, but fortunately the chondrules analyzed had TL values which virtually completely covered the full range observed in the study. The only element to show behavior seemingly related to TL was Ca (Figure 3). All the analyzed chondrules with TL > 0.05 have a CaO content in excess of 3 mg/g, while eight out of eleven of the low TL chondrules (TL < 0.05) have a CaO content below 3 mg/g.

Discussion

Although there is some uncertainty as to the cause, we believe there is little doubt that the range of TL sensitivity displayed by UOCs is related to metamorphism. The heterogeneity displayed by olivine, pyroxene, and metal compositions vary systematically with TL sensitivity, as do matrix recrystallization, matrix chemistry, bulk carbon, and primordial inert gas contents [Sears et al., 1980]. Considered alone, some of these trends are only suggestive (e.g., metal heterogeneity), while many are very strong (e.g., silicate heterogeneity, matrix chemistry). Together the evidence that the TL

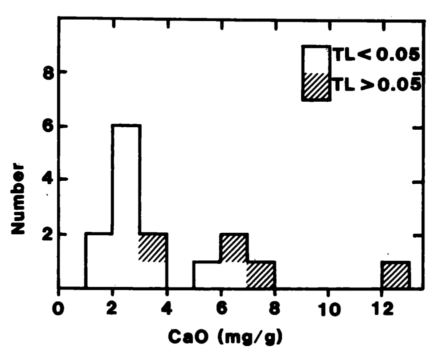

Fig. 3. Histogram of the CaO contents of 15 chondrules separated from the Dhajala H3.8 chondrite. CaO was determined by energy-dispersive X ray spectrometry of fused glass beads.

variation is somehow associated with metamorphism is compelling. This is expressed in another manner in Table 1. Fourteen type 3 ordinary chondrites are listed with the petrologic assignment of Sears et al. [1980] based on TL and other properties, of Afiattalab and Wasson [1981] based on metal heterogeneity, and of Huss et al. [1981] based on matrix composition, and matrix recrystallization, and other properties. Comparison of the three sequences shows significant disagreements for only two chondrites in each case; nonrepresentative sampling can probably be blamed for most of the discrepancies. The only thing these diverse properties have in common is that they are metamorphism-sensitive.

Lalou et al. [1970] have shown that in equilibrated ordinary chondrites the major TL phosphor is feldspar. Some observations by Sears et al. [1982] suggest that feldspar is also the TL phosphor in type 3 chrondrites. Metamorphism not only causes an increase in TL but it also causes the TL peak to broaden and move to higher temperatures. Such behavior was also observed in a terrestrial low (ordered) albite as a result of conversion to high (disordered) albite by annealing for 60 days at 1000°C [Pasternak et al., 1976; Pasternak, 1978]. Sears et al. [1982] therefore suggested that throughout the type 3 chondrites there was a change in the crystallography of the feldspar from the ordered form (types 3.3-3.5) to the disordered form (types > 3.6). The range of TL sensitivities observed in type 3 chondrites suggests that either the feldspar is being formed by processes which accompany metamorphism (particularly the lowest levels of metamorphism), or there are chemical changes in the feldspar which are only indirectly associated with metamorphism. Since feldspar is a strong candidate for the TL phosphor in most UOCs, we will first discuss the occurrence of this mineral and how our data may be accordingly interpreted. We will then discuss other possible phosphors.

Broadly speaking, it is possible to recognize three kinds of feldspar in chondrites.

(1) That which forms in chondrules by the devitrification of feldspathic glass during metamorphism. This is the most frequently discussed type of feldspar and the existence of glass compared with feldspar was an important parameter in Van Schmus and Wood's [1967] definition of petrologic types. Its mode of origin and its relationship with glass and metamorphism has been discussed by Miyashiro [1962], Mason [1965,1967], Dodd et al. [1967], Wood [1967], Dodd [1969], Van Schmus and Ribbe [1968], and Van Schmus [1969, and references therein]. Direct observations of feldspar in type 3 chondrules are few because of its microcrystalline nature, however Ashworth [1981] reports high voltage transmission electron microscopy (TEM) observations of feldspar in the chondrules of Tieschitz (H3.6) and Bremervorde (H3.9).

(2) That which is observed in the matrix. Observations of such feldspar have been reported by Huss et al. [1981], who also saw evidence that the feldspar was forming as a response to metamorphism-related reactions, a possibility

Table 1. Thermoluminescence Sensitivity Values of 14 UOCs and Their Subdivision on the Basis of Metamorphic Equilibration

	TL (Dhajala=1)	Type*	Category†	Metal Uniformity††
Bremervorde	2.6	3.9	III	
Dhajala	1.0	3.8	IV	--
Mezo-Madaras	0.9	3.7	II	high
Hedjaz	0.82	3.7	I	--
Khohar	0.44	3.6	II	high
Parnallee	0.42	3.6	III	moderate
Ngawi	0.25	3.6	II	low
Tieschitz	0.23	3.6	II	--
Chainpur	0.078	3.4	I	moderate
Sharps	0.071	3.4	I	high
Manych	0.070	3.4	I	--
Bishunpur	0.0054	3.1	I	low
Semarkona	0.0045	3.0	I	low
Krymka	0.0027	3.0	I	--

* Sears et al. [1980].
† Huss et al. [1981].
†† Afiattalab and Wasson [1981].

originally discussed by Dodd et al. [1967]. Using TEM data, Ashworth [1978] has also observed feldspar in the matrix of Parnallee (LL3.6) and Hedjaz (L3.7). It is significant that even with TEM techniques feldspar is only observed in the chondrules or matrices of the more heavily metamorphosed chondrites (type > 3.5). If there is a linear relationship between TL sensitivity and feldspar content, a type 3.7 chondrite has ~1% of the feldspar of an equilibrated chondrite (10 wt %) and ~100 times as much as the least metamorphosed chondrites.

(3) Feldspar which was present before metamorphism and with an abundance that is unrelated to metamorphism. Dodd et al. [1967] describe such 'primary' feldspar in a chondrule in Hallingeberg and it has also been observed in Tieschitz by Hutchison et al. [1979 and personal communication].

It is difficult to estimate the relative amounts of these three types of feldspars, although both matrix and primary feldspar seems to have been seldom observed. Compositionally, chondrules and matrices should have similar feldspar content when fully crystallized, although there are systematic differences in certain relevant elements and the feldspar may be compositionally different. Thus calcium is similar in the fine-grained dark matrix and whole rock for Dhajala, although K, Na, and Al are ~50% lower in the matrix [Huss et al., 1981].

Figure 2 demonstrates that certain chondrules contain a major TL phosphor. Nineteen chondrules contain higher TL/mass than bulk Dhajala powder; four chondrules have TL/unit mass a full order of magnitude larger than bulk powder. Such chondrules may contribute as much as 50% of the total TL of the meteorite, even though their abundance is so low (4 out of 58). If chondrules with a TL/mass of 10 were the only source of TL in the bulk sample, and the other material acted simply as a dilutant, then such chondrules need to be only ~10% of the bulk sample to account for its TL. Estimates of the abundance of chondrules in meteorites are conspicuously absent from the literature, even for type 3 chondrites with relatively distinct chondrules. However, Lux et al. [1981] mentioned 50-60 vol.% as a typical figure for the bulk meteorite, so we may say ~75% of the nonmagnetic portion used for TL measurements was originally composed of chondrules. The chondrules with a TL/mass of 10 are ~7% of our present sample (4 out of 58) or ~5% (75% x 0.07) of the bulk sample. When one considers the contribution of the remaining 15 chondrules, whose TL/mass is greater than bulk Dhajala, it is possible that all the TL resides in the chondrules. The average TL/mass of these 15 chondrules is almost exactly twice that of bulk Dhajala powder, slightly more than required to account for the TL of a sample which is 75% chondrules. We conclude that chondrules are a major, if not the major, carrier of TL in the Dhajala meteorite.

Almost by definition, one distinctive characteristic of chondrules is that they contain glass. A priori, the conclusion that chondrules are an important TL carrier therefore carries the implications that feldspar of the first type described above may be the important TL phosphor in Dhajala, and that the mechanism behind the relationship between TL sensitivity and metamorphism is the devitrification of this glass. One necessary caveat is that primary feldspar has also been observed in chondrules. However, the observations of primary feldspar are so few and the observations of glass so numerous that this seems to be an unlikely option, at least in Dhajala, where we predict that the TL phosphor should be relatively abundant.

Our Ca data are of interest in this connection. The simplest interpretation of these data is that the TL phosphor is either a Ca-bearing mineral (or associated with a Ca-bearing mineral) and that high TL chondrules simply contain more of the phosphor. In its simplest form, this explanation fails because the variation in TL is more than 100 times greater than the variation in Ca. However it is known that high-Ca glasses devitrify more readily than low-Ca glasses, so that the Ca data may be interpreted as further evidence for the devitrification mechanism. In the type 4 meteorite Kramer Creek, where devitrification is sufficiently advanced to be petrologically visible, Gibson et al. [1977] observed that chondrules with a mesostasis which was 16% CaO contained small plagioclase crystals in that mesostasis, while in other chondrules where the mesostasis was glassy, the CaO content was only 1-6%.

Other possible mechanisms for interpreting the relationship between TL sensitivity and metamorphism were described by Sears et al. [1982]. One is that although feldspar is the phosphor, factors other than devitrification account for the intensity change. Possible factors are obscuration of the TL in the lower types by carbon, or differences in trace element contents. Carbon is ~0.4 wt.% of the least-metamorphosed meteorites, but ~0.1 wt.% in type 4 chondrites. It is present in the form of graphite and perhaps carbonaceous compounds. A second possibility is that the feldspar differs in composition throughout the type 3 meteorites. Although nothing is known about the composition of feldspar in type 3 chondrites, there are systematic whole-rock variations in the contents of the trace elements In, Tl, Bi, Pb, and Cd. Typically, type 4 chondrites may contain these elements in $\sim 10^{-4}$ times their CI values, while the least-metamorphosed ordinary chondrites may contain them in CI abundance [e.g., Wasson, 1972]. However, these particular elements are siderophile or chalcophile, and one would not normally associate them with feldspar. Although they cannot be entirely ruled out on the basis of present data, both these explanations seem rather contrived.

Other Phosphors

In addition to feldspar in its three occurances, there are probably other TL phosphors which may be important in type 3 meteorites. Whitlockite is the only other phosphor observed in equilibrated chondrites [McKeever and Sears, 1980]. It has red cathodoluminescence and appears as irregular grains in the matrix. In equilibrated meteorites its contribution to the total TL is negligible, since its abundance is low and the TL equipment is highly insensitive to red radiation. Most Ca-bearing minerals are also potential TL phosphors, and calcic pyroxene is such an example. In the Allende meteorite, forsterite was also observed to have red cathodoluminescence by Sears and Mills [1974], and the heterogeneity of type 3 chondrites is such that a few forstertic olivine grains may occur. It is unclear whether the trends in TL against peak width and peak position could be produced by any of these potential phosphors. However, the type 3.0-3.2 meteorites do not follow these TL trends and there is no compelling evidence against any of them. It may be assumed that even if feldspar is the phosphor in type $\geq$3.2 chondrites, the abundance of this mineral in type $\leq$3.2 is so low that other phosphors become important in their case.

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