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Chemical and physical studies of type 3 chondrites, VII. Annealing studies of the Dhajala H3.8 chondrite and the thermal history of chondrules and chondrites

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Samples of the Dhajala H3.8 chondrite have been annealed for 10 hours at 600, 700, 800, 900 and 1000°C and at 1000°C for 1, 2, 20 and 100 h and their thermoluminescence (TL) properties measured. The TL sensitivity decreased by a factor of 2 after annealing at < 900°, but at higher temperatures fell by an order of magnitude. An abrupt increase in the temperature of the TL peak from 172 ± 9°C to 231 ± 8°C and a steady increase in the width of the peak from 169 ± 7°C to 212 ± 5°C were caused by the annealing treatment. The TL phosphor in Dhajala is thought to be feldspar predominantly in the high-temperature (disordered) form, but the present data indicate that a contribution from the low-temperature form is also present and that this low-temperature component is converted to the high form by the annealing treatment. The low-temperature feldspar is located in a few of the chondrules (~ 20% of those separated from the meteorite) which are also noteworthy for having high TL sensitivities. These chondrules must have suffered greater crystallization of their mesostasis than the other chondrules, and equilibrated to lower temperatures. It is argued that, for compositional reasons, their mesostasis constituted less of a barrier to diffusion and therefore equilibration. Presumably the post-metamorphic cooling rate of the meteorite through the stability field of the low form was slow enough to permit some crystallization, and the width and temperature of the TL peaks for petrologic types 3.5–3.9 are somehow related to cooling rate. Based on TL, there is no indication of a correlation between petrologic type and cooling rate for types 3.5–3.9; this is not consistent with a simple, single internally heated meteorite parent body.

1. Introduction

Previous studies have shown that thermoluminescence (TL) measurements provide new insights into the shock-reheating history and the metamorphism experienced by the ordinary chondrites [1,2]. Annealing experiments on the Allan Hills A77011 (referred to below as A77011) L3.4 chondrite and other type 3 chondrites indicate that the primary TL phosphor, feldspar, exists in two distinct forms, which have different TL properties. These forms are an ordered form which is the stable phase at low temperatures (< 700°C) and a disordered form which is stable at high temperatures. Pasternak [3] observed similar TL changes to those we have observed in meteorites in a study of Amelia albite and found that the changes were associated with the order-disorder transformation.

Thermoluminescence may be emitted over a temperature range of 100–350°C, and may vary in the temperature at which TL emission is a maximum (the "peak temperature") and the temperature range over which it is emitted, i.e. the full-width of the peak at half its maximum intensity (FWHM, or simply "peak width"). A plot of peak temperature against peak width for type 3 ordinary chondrites produces two clusters (Fig. 1) and a sample originally in the lower cluster can be transferred to the other by annealing above the transformation temperature [4,5]. The data therefore have palaeothermometry applications. Meteorites in the upper cluster are, with few exceptions, types 3.6–3.9, whereas those in the lower cluster are types 3.2–3.5. The transition between the low-temperature form and the high-temperature form is rather abrupt, which accounts for existence of a hiatus between the clusters. (Types 3.5 are under-represented in the figure, but this is not the reason for the hiatus between the clusters as there is no tendency for the three type 3.5 chondrites to fill the gap, they clearly belong to

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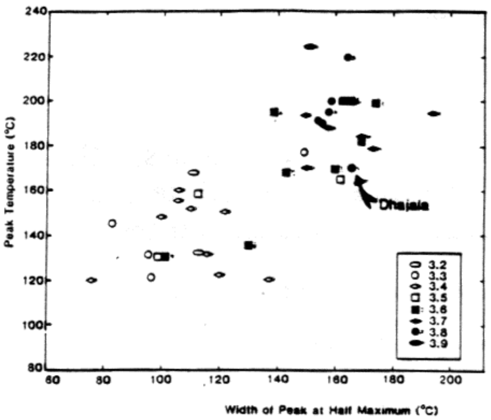

Fig. 1. Plot of the TL peak temperature against the width of the peak at half its maximum intensity for 39 type 3 ordinary chondrites with the petrologic type indicated. The data produce two clusters with the more heavily metamorphosed chondrites belonging to the upper cluster. The Dhajala meteorite plots on the lower edge of the upper cluster [4].

one cluster or the other, or for the petrologic types have any particular distribution within the clusters.)

The scatter in the clusters in the TL peak temperature–peak width plot is much larger than experimental uncertainty can explain, and it seems possible that some "fine structure" may be present. The Dhajala meteorite is a H3.8 chondrite which plots near the lower fringes of the high-temperature cluster and might be expected to undergo changes on annealing if such fine structure is present. Dhajala is also the meteorite on which a detailed study was made of the composition, petrology and TL of separated individual chondrules [6]. The separated chondrules also produce two clusters on plots of TL sensitivity against peak temperature and against peak width, with samples having narrow, low-temperature TL peaks being 10–100 times higher in TL sensitivity than those with broad peaks at high glow curve temperatures (Fig. 2). Significant correlations were found between TL sensitivity and composition, and between TL sensitivity and petrology, which were interpreted in terms of the formation of the feldspar by crystallization of the chondrule mesostasis, the crystallization being most facile in calcic glasses.

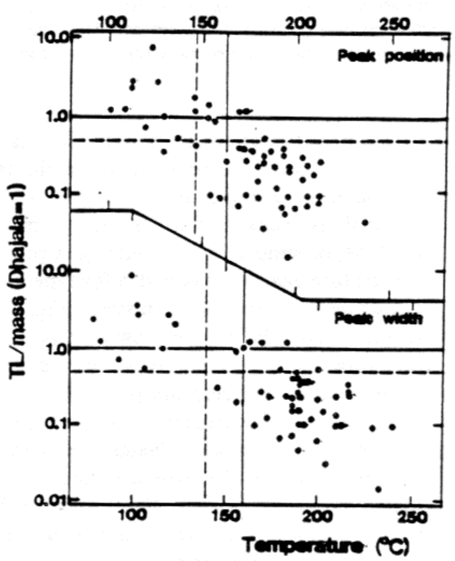

Fig. 2. Plot of the mass-normalized TL sensitivity of 58 separated Dhajala chondrules as a function of TL peak temperature and peak width. The solid cross-hairs locate the position of bulk Dhajala and the broken cross-hairs represent the summation of the data for the individual chondrules [6].

In the present work we annealed samples of the Dhajala meteorite at 600–1000°C for 10 h and at 1000°C for 1, 2, 10, 20 and 100 h to see if any TL changes could be induced which might explain the scatter in the cluster in Fig. 1 and provide further insights into the thermal history of chondrules and chondrites.

2. Experimental

Two chips from the Dhajala meteorite were ground to approximately 100 mesh (150 μm), and the magnetic portions removed with a hand-magnet. The non-magnetic portions were placed in quartz vials which had been acid washed and fired in an oxygen-methane flame to remove impurities. The vials were then evacuated to 100 millitorr and filled with high-purity nitrogen three times. They were then heat sealed, placed in a wire-wound tube furnace and annealed in the way described by Guimon et al. [4] and Sears et al. [7] for the times and temperatures shown in Table 1.

The thermoluminescence of the samples was then measured with the apparatus and techniques, and the data read from the glow curves, in the manner described by Sears and Weeks [8]. The quoted uncertainties are standard deviations displayed by 3–5 replicate measurements, compounded with the uncertainty in the Dhajala standard in the case of TL sensitivity.

3. Results

Our data are presented in Table 1 and are plotted in Figs. 3–8. The annealing treatments caused changes in the TL sensitivity of the samples similar to those we have previously seen in A77011 and Kernouvé [4,7]. The experiments also caused changes in the temperature and width of the TL peak, indicating that some of the scatter in the upper cluster in Fig. 1 is due to factors other than experimental error. Fig. 3 presents two TL glow curves which illustrate some of the effects we have observed.

As with the other samples we have studied, the TL sensitivity of Dhajala dropped to about 50% of its unannealed value after relatively mild annealing (~ 600°C) and remained near this value after annealing at 700 and 800°C, but after annealing at higher temperatures it fell abruptly (Fig. 4). The

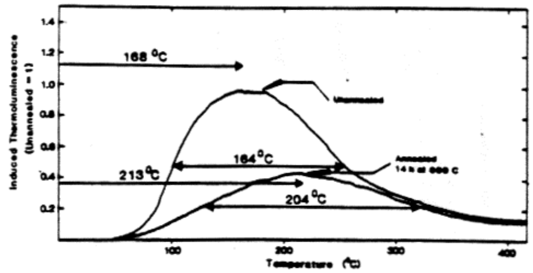

Fig. 3. Glow curves, plots of the TL emitted as a function of temperature of heating in the TL apparatus, for a natural sample of the Dhajala meteorite and for a sample which has been annealed at 800°C for 14 h.

TABLE 1

Thermoluminescence data for samples of the Dhajala meteorite after annealing [a]

Annealing temperature (°C)	Annealing time (h)	TL sensitivity (unannealed = 1)	Peak temperature (°C)	FWHM [b] (°C)
Unannealed	-	0.88 ± 0.06	172 ± 3	171 ± 6
Unannealed	-	1.05 ± 0.05	177 ± 13	186 ± 3
Unannealed	-	1.16 ± 0.05	160 ± 5	154 ± 12
Unannealed	-	0.94 ± 0.05	178 ± 11	165 ± 4
600	10	0.60 ± 0.04	165 ± 9	197 ± 10
600	10	0.45 ± 0.05	170 ± 11	191 ± 11
700	10	0.52 ± 0.05	183 ± 9	211 ± 16
700	10	0.52 ± 0.05	193 ± 3	199 ± 7
800	14.5	0.46 ± 0.08	213 ± 4	204 ± 13
800	14.5	0.42 ± 0.05	203 ± 3	182 ± 5
900	10	0.18 ± 0.05	217 ± 3	199 ± 1
900	10	0.14 ± 0.06	213 ± 3	190 ± 4
1000	1	0.17 ± 0.02	217 ± 5	204 ± 8
1000	1	0.23 ± 0.03	217 ± 3	216 ± 7
1000	2	0.16 ± 0.02	218 ± 1	205 ± 6
1000	2	0.12 ± 0.02	217 ± 1	207 ± 8
1000	10	0.12 ± 0.08	216 ± 4	199 ± 8
1000	10	0.18 ± 0.07	226 ± 12	225 ± 1
1000	20	0.14 ± 0.03	229 ± 5	182 ± 6
1000	20	0.11 ± 0.01	234 ± 11	197 ± 14
1000	100	0.14 ± 0.06	221 ± 2	202 ± 9
1000	100	0.12 ± 0.01	229 ± 2	169 ± 2

[a] Uncertainties quoted are $\pm 1\sigma$ based on 3–5 replicate measurements.

[b] Full width of the TL peak at half its maximum intensity.

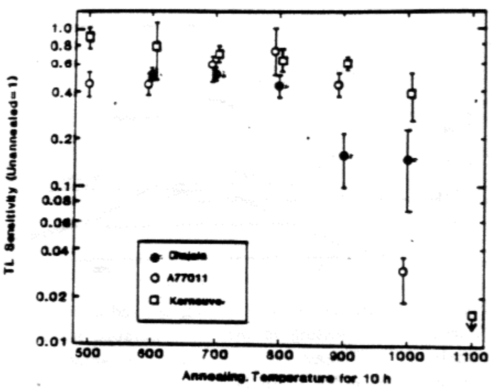

Fig. 4. Plot of the TL sensitivity of the Dhajala meteorite as a function of annealing temperature. Also shown are the data for another type 3 chondrite, Allan Hills A77011 [4], and an equilibrated ordinary chondrite, Kernouvé [7].

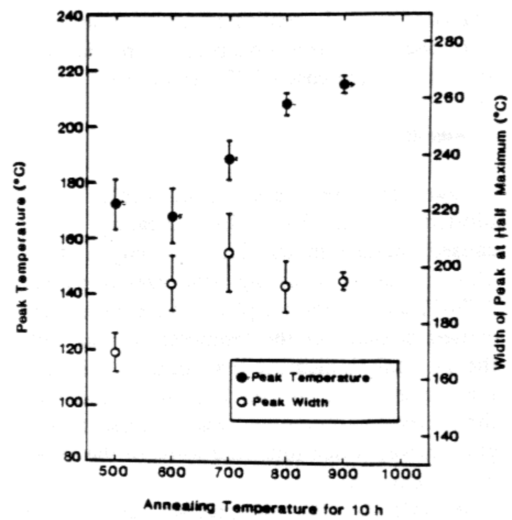

Fig. 6. Plot of the TL peak temperature (solid symbols, left-hand scale) and peak width (open symbols, right-hand scale) as a function of the temperature to which the samples had been annealed for 10 h (14 h in the case of the 800°C data). The peak temperature increased steadily from 172 to 221°C as the annealing temperature increased, while the width of the peak increased abruptly from 169 to 195°C after annealing at 600°C and thereafter remained fairly constant.

abrupt decrease occurred around 1100°C in the case of Kernouvé and at about 1000°C for A77011 with Dhajala being intermediate, dropping more abruptly at 900°C than the others, but falling less than A77011 at 1000°C.

We were able to detect a slight time-dependency in the decrease of the TL sensitivity of the samples after annealing at 1000°C for various times (Fig. 5). After annealing for 100 h the TL sensitivity was almost an order of magnitude lower than its unannealed value, and a factor of 2 lower than for the sample annealed for 1 h. In contrast, TL sensitivity of the A77011 meteorite fell almost

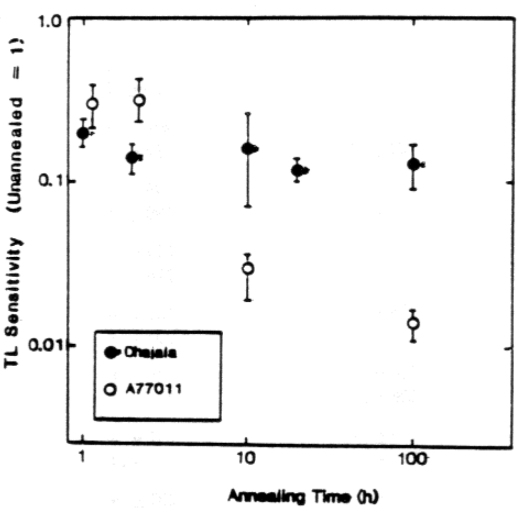

Fig. 5. Plot of the TL sensitivity of the Dhajala meteorite after annealing at 1000°C for various times. Also shown are data for the Allan Hills A77011 meteorite also annealed at 1000°C [4].

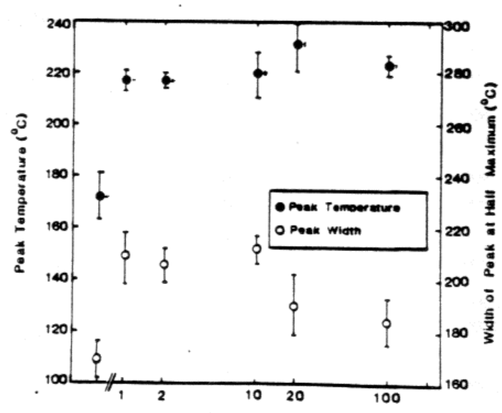

Fig. 7. Plot of the TL peak temperature (solid symbols, right-hand scale) and peak width (open symbols, left-hand scale) as a function of time annealed at 1000°C. The peak temperature increases from its unannealed value of 172°C to 220°C and remains constant with longer annealings. The width of the peak also shows an abrupt increase after 1 h (from 172 to 210) and then gradually falls to 185°C with larger annealings.

2 orders of magnitude below the unannealed value after 100 h of annealing at 1000°C, and more than an order of magnitude difference was produced by annealing for 100 h compared with 1 h.

The temperature of the TL peak increased gradually from 172 ± 9 to 221 ± 10°C with increasing annealing temperature in the 10 h annealing series, whereas the width of the peak increased abruptly from 169 ± 7 to 195 ± 3°C after annealing at 600°C and thereafter remained essentially unchanged (Fig. 6). After annealing for 1 h at 1000°C the temperature of the TL peak increased from the unannealed value of 172 ± 9 to 217 ± 4°C and longer periods of annealing at this temperature caused no further changes (Fig. 7). The width of the peak also showed an abrupt increase after 1 h at 1000°C, from the unannealed value of 169 ± 7 to 210 ± 9°C, but after longer annealing periods showed a very slight, gradual decrease from 210 ± 8 after 1 h to 185 ± 9°C after 100 h.

4. Discussion

4.1. TL sensitivity changes with annealing

The cause of the 40–50% decrease in TL sensitivity of A77011, Kernouve and Dhajala after annealing at temperatures of < 900°C is unclear, although the activation energy provides some clue as to the cause. Assuming first-order kinetics, and using the data in the present Fig. 4 and methods of Sears et al. [7], we calculate a value of 6.4 ± 3.4 kcal/mol for this decrease in the case of the Dhajala meteorite. We were unable to obtain an activation energy for the TL sensitivity decrease over this temperature range for A77011, but for Kernouvé we obtained values of 7.7 ± 1.8, 11.0 ± 6.5 and 6.6 ± 3.4 kcal/mol using various treatments of the data. (All uncertainties $\pm$ 1 sigma.) The similarity of the values suggests the same mechanism may be involved in each meteorite, despite very different original TL sensitivities, peak temperatures and peak widths. We previously pointed out that such low activation energies were inconsistent with fusion or the breakage of Si–O bonds and implied processes such as the diffusion of small ions through the lattice or subtle crystallographic rearrangements.

The abrupt decrease ⩾ 900°C is clearly due to a different process. For Kernouvé, the drop in TL sensitivity occurred near the melting point of sodic feldspar and can be attributed to the fusion of the phosphor. For A77011, we believe that the feldspar exists at submicron grains widely distributed throughout the glassy mesostasis and it would probably be resorbed before it reached the melting point. We estimate, assuming that the TL sensitivity is directly proportional to its crystalline feldspar content, that about 5% of the feldspathic material in the Dhajaia meteorite is crystalline feldspar and the remainder is feldspathic glass. The grains would presumably be present in a variety of sizes, some large enough to resist resorbtion until the melting point was reached, and others not. The intermediate behavior of Dhajala, as far as the abrupt high-temperature decrease in TL sensitivity is concerned, is therefore not surprising.

4.2. TL peak temperature and peak width changes on annealing

The most straightforward interpretation of the changes in peak temperature is that while the Dhajala meteorite contains predominantly the high form, there is also a component of low form present which the annealing treatment destroyed. (Most probably, by conversion to the high form.) The peak temperature of the unannealed Dhajala sample was intermediate to those associated with the low and high forms (160–170°C) but after annealing > 800°C for > 10 h it moved to values associated with the high form (210–220°C).

The activation energies for the increase in peak temperature displayed by the Dhajala meteorite are the same as those responsible for the changes in the peak temperature of A77011, which also indicates that a similar mechanism is responsible. In the case of A77011, the peak increase is associated with the first stages in the conversion of the low form to the high form [4]. The activation energy for the peak temperature increase from 172 ± 9 to 215 ± 3 in Dhajala was found to be 15.7 ± 4.4 kcal/mol, using the data in Fig. 6 and assuming first-order kinetics, while for A77011, two sets of data yielded activation energies of 7.1 ± 2.7 and 10.9 ± 4.5 kcal/mol for an increase in peak temperature from 130 ± 6 to 214 ± 15°C [4].

The width of the TL peak increases after annealing at 600°C and then levels off at a value of 200°C (Fig. 6), as if low form were being converted to the high form and that 200°C is the width of the high form. However, after annealing for 1 h at 1000°C, which broadened the peak to 210°C, longer annealing periods caused the peak to narrow a little (Fig. 7). This may be an indication that we are not dealing with a simple, single high form of the mineral, but rather that some low-temperature form remains, which is only slowly transforming to the high form.

Fig. 8 is a plot of peak temperature against peak width for the annealed samples of Dhajala and A77011; this enables better comparison of the present data and the type 3 ordinary chondrite data shown in Fig. 1. The annealing at 0–700°C for 10 h moved the Dhajala data from bottom

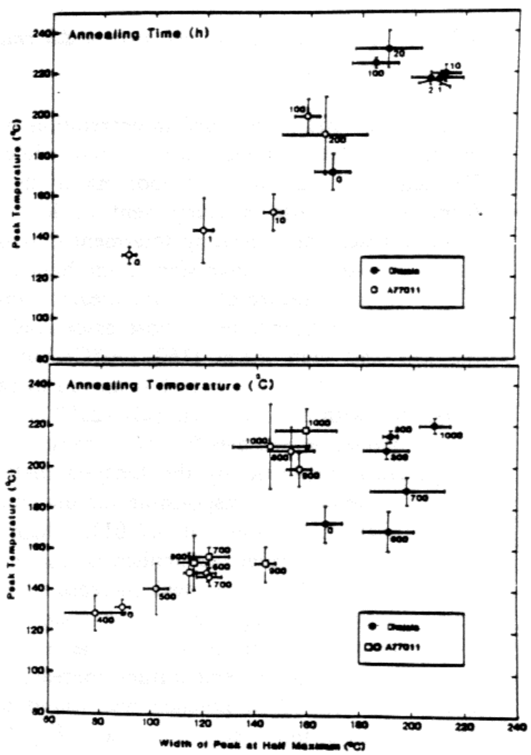

Fig. 8. Plot of TL peak temperature against peak width for Dhajala (filled symbols) and Allan Hills A77011 (open symbols, data from [4]) after annealing for the times indicated (upper figure), at 1000°C in the case of Dhajala and 900°C for 77011, and for the temperature indicated (°C, lower curve, circles 10 h, squares 100 h).

center of the upper cluster, round the high-temperature edge of the cluster to the top right-hand corner. Annealing at 1000°C for 1 h caused it to broaden, but annealing for longer times caused the peak width to narrow and the peak, to move to ~ 230°. The TL peak of A77011 also shows signs of some fine-structure within the two clusters. For the 0–600°C for 100 h series and the 0–900°C for 10 h series there is an indication that higher temperatures favor greater width and higher peak temperatures. For the 900–1000°C for 100 h series, on the other hand, there is an indication that higher temperatures favor slightly smaller widths and higher temperatures.

The TL peaks for the high-temperature cluster of A77011 are displaced about 40°C to lower values compared to those of Dhajala, again an indication that annealing above the transformation temperature has not produced the same pure high form in both meteorites. Indeed, since Dhajala started out containing much higher abundances of both forms, compared with A77011, this 40°C difference could represent difficulty in quantitative destruction of the low form.

In summary, the trends displayed by the TL peak temperature and width in response to the annealing treatment can be understood in terms of the TL being produced by mixtures of the high- and low-temperature forms of the TL phosphor, feldspar. The two forms have peak temperatures of 120 and 220°C and may be relatively narrow (for example 80–100°C, the width of the peak in unannealed A77011) but appear broader whenever both forms are present because of overlap between the peaks. Although the two clusters consist predominantly of one form or the other, there is always a small amount of the other component which causes the fine structure to the two clusters and the movements within the clusters caused by the annealing treatments.

4.3. *Implications for chondrule history*

The TL sensitivity of individual chondrules separated from the Dhajala meteorite by Sears et al. [6] also produce two clusters on a peak temperature against peak width plot, one with a peak temperature and width centering on 120 and 100 respectively, and another with the temperature centering on about 180°C and the width centering

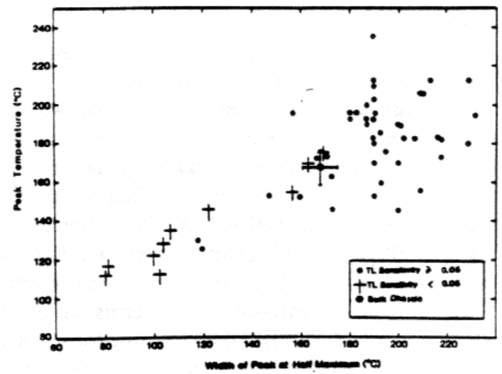

Fig. 9. Plot of TL peak temperature against peak width for 55 chondrules separated from the Dhajala meteorite [6]. The data form two clusters, with the chondrules with higher TL sensitivity tending to plot in the lower cluster. Three chondrules are missing from the 58 reported by Sears et al. because of incomplete data, however, on the basis of Fig. 1 it is clear that one has a TL sensitivity of < 0.05 and would plot on the upper cluster and two have TL sensitivities > 0.05 and would plot in the lower cluster.

on about 200°C (Fig. 9). Importantly, the chondrules plotting in the lower cluster have TL sensitivities an order of magnitude or so higher than those plotting in the upper cluster (see also Fig. 2). This is in contrast to the situation for the type 3 ordinary chondrites (Fig. 1), where meteorites in the upper cluster have TL sensitivities and order of magnitude *higher* than those in the lower cluster.

The differences in TL sensitivity from one chondrule to another, as with the meteorite to meteorite differences, very probably reflect differences in the abundance of crystalline feldspar [6]. Apparently, therefore, the data in Fig. 9 indicate that while most chondrules contain feldspar which is predominantly in the high form and relatively low in TL sensitivity, the meteorite contains a few (about 20% of the chondrules separated) which contain low-temperature feldspar and have high TL sensitivities. In principle, annealing a chondrule in the lower cluster above 700°C should move it to the upper cluster, but without significant change (more than a factor two) in its TL sensitivity. We conclude that the chondrules in the lower cluster of Fig. 9 underwent greater crystallization than those in the upper cluster, and that this occurred predominantly below the transformation temperature, while the chondrules with lower TL sensitivity essentially ceased crystallization above the transformation temperature.

One might conclude that the chondrules with high TL sensitivity (> 0.05) cooled more slowly than the chondrules with TL sensitivity (< 0.05). The problem with this idea is the proportion of equilibrated (equivalent to high TL sensitivity) chondrules in the type 3 chondrites increases as the proportion of recrystallized matrix and homogeneity of the metal increases, which suggests that the process of chondrule "equilibration" occurred in situ. An alternative to differential cooling rates is suggested by the observation that the mesostasis of the chondrules with high TL sensitivity is significantly more calcic than that of the chondrules with low TL sensitivity and would devitrify more readily [6]. There would, therefore, be a much larger number of pathways for grain boundary diffusion in the brighter chondrules to enable equilibration within the chondrule and between the chondrule and surrounding matrix. The reason why the high TL sensitivity chondrules equilibrated to low temperatures is therefore that grain boundary diffusion was effective, whereas for the low TL sensitivity chondrules the pathways were few or absent, and equilibration, including crystallization, may have had to depend on volume diffusion.

The new data are therefore consistent with our earlier suggestion that the co-existence of equilibrated and unequilibrated chondrules in a single meteorite is not necessarily evidence that chondrules experienced diverse metamorphic histories prior to their incorporation in the meteorite.

4.4. *Implications for meteorite history*

We conclude from the TL data that the Dhajala meteorite, and the other chondrites in the upper cluster of Fig. 1, experienced a period of time in the stability field of the high-temperature form of feldspar; i.e. > 600°C if the feldspar phase diagram is used as a guide [9], and > 700°C if our annealing experiments on A77011 are applicable. This was necessary to produce a broad TL peak at relatively high peak temperatures. However, since the present annealing experiments removed some of the low-temperature TL and produced the trends described above, we conclude that these meteorites contain a component which crystallized below the transformation temperature.

The low-temperature component is present in a small subset of the chondrules, which also have high TL sensitivities. If, as we argued in the preceding section, the chondrules are cogenetic, then the meteorite must have cooled slowly enough to permit significant crystallization below the transformation temperature. The width of the peak, i.e. the relative abundance of the high- and low-temperature forms, provides some clue to the cooling rate of the meteorite. There is no tendency for any preferred distribution of petrologic types within the upper cluster in Fig. 1, which would imply no systematic variation in cooling rate and petrologic type within these type 3 chondrites. In the higher petrologic types, there seems to be a systematic decrease in cooling rate and metamorphism, implying greater burial depth for the higher types [10].

If the chondrules were not cogenetic, then it would seem that all the chondrites in the upper cluster of Fig. 1 picked up a roughly comparable proportion of equilibrated and unequilibrated chondrules. This is contrary to petrographic observations, which indicate that the higher petrologic type 3 chondrites contain a higher proportion of equilibrated chondrules. The ratio of equilibrated chondrules to chondrules studied is 0/10 for a type 3.3, 5/100 for a type 3.4, 10/20 for four type 3.6–3.7 and 100% equilibrated for type 4 and 5 [11–13], while according to Huss et al. [14] the percentage recrystallization is 10–20, 20–~50, ⩾ 60 and 0 for petrologic types 3.3, 3.4, 3.6–3.7, 4–5, respectively.

5. Conclusions

The TL sensitivity of the Dhajala H3.8 chondrite decreased after annealing < 900°C by about 50%, in which respect it was similar to A77011 (L3.4) and Kernouve (H6). After annealing > 900°C the stability of the TL sensitivity was intermediate to that of the other two meteorites, which is thought to be related to the petrographic setting of the feldspar, the TL phosphor. In the equilibrated chondrite (Kernouvé), the feldspar exists as relatively large crystals, and loses its TL when it melts, while for A77011 it exists as submicron grains widely distributed in glass and probably loses its TL when it is resorbed.

The annealing treatment caused the width of the TL peak to increase abruptly and the peak temperature to increase gradually by factors of 1.4 and 1.2, respectively, as the annealing temperature was increased from 600 to 1000°C. After annealing at 1000°C the TL peak moved to 220°C and narrowed slightly in a time-dependent manner. These trends are interpreted in terms of the existence of low- and high-temperature forms of the feldspar the relative proportions of which are altered by annealing.

Separated chondrules from the Dhajala meteorite contain mainly the high-temperature form, but a few of the chondrules contain predominantly the low form; the latter also have higher TL than the others by a factor of about 10. Apparently, the low TL chondrules crystallized predominantly above the transformation temperature for a relatively short period of time, while the others equilibrated for a relatively long period of time mainly below the transformation temperature. The chondrule to chondrule difference in response to metamorphism probably reflects the composition of the mesostasis and the ease with which it crystallizes. This being so, the width of the TL glow curve is related to the cooling rate of the bulk meteorite. If the heat source which caused metamorphism was internal radioactivity then one might expect a correlation between petrologic type and cooling rate, since both would depend on burial depth. However, based on these TL data there appears to be no relationship between cooling rate and petrologic type for the 3.5–3.9 chondrites.

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