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Metamorphism and aqueous alteration in low petrographic type ordinary chondrites

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Abstract–In order to investigate the relative importance of dry metamorphism and aqueous alteration in the history of chondrules, chondrules were hand-picked from the Semarkona (petrographic type 3.0), Bishunpur (3.1), Chainpur (3.4), Dhajala (3.8) and Allegan (5) chondrites, and matrix samples were extracted from the first three ordinary chondrites. The thermoluminescence (TL) properties of all the samples were measured, and appropriate subsets of the samples were analyzed by electron-microprobe and radiochemical neutron activation and the water and H-isotopic composition determined.

The TL data for chondrules from Semarkona and Bishunpur scatter widely showing no unambiguous trends, although group B1 chondrules tend to have lower sensitivities and lower peak temperatures compared with group A5 chondrules. It is argued that these data reflect the variety of processes accompanying chondrule formation. The chondrules show remarkably uniform contents of the highly labile elements, indicating mineralogical control on abundance and volatile loss from silicates and loss and recondensation of mobile chalcophiles and siderophiles in some cases. Very high D/H values (up to ~8000‰ SMOW) are observed in certain Semarkona chondrules, a confirmation of earlier work.

With increasing petrographic type, mean TL sensitivities of the chondrules increase, the spread of values within an individual meteorite decreases, and peak temperatures and peak widths show trends indicating that the TL is mainly produced by feldspar and that dry, thermal metamorphism is the dominant secondary process experienced by the chondrules. The TL sensitivities of matrix samples also increase with petrographic type. Chainpur matrix samples show the same spread of peak temperatures and peak widths as Chainpur chondrules, indicating metamorphism-related changes in the feldspar are responsible for the TL of the matrix. The TL data for the Semarkona and Bishunpur matrix samples provide, at best, only weak evidence for aqueous alteration, but the matrix contains H with approximately terrestrial D/H values, even though it contains much water. Secondary processes (probably aqueous alteration) presumably lowered the D/H of the matrix and certain chondrules. While chondrule properties appear to be governed primarily by formation processes and subsequent metamorphism, the matrix of Semarkona has a more complex history involving aqueous alteration as a meteorite-wide process.

INTRODUCTION

The chondrites provide much information on conditions and processes in the early Solar System and even on the interstellar medium (Kerridge and Matthews, 1988). However, there is uncertainty as to which properties reflect nebular processes (*i.e.*, preaccretionary processes responsible for the "precursors") and which result from processes occurring on the parent body. Preaccretionary processes generally include mixing of different precursors, gas-solid reactions occurring during condensation in the nebula and during chondrule formation. Parent-body processes usually involve reactions between condensed phases, including liquids. The many and varied effects of dry metamorphism in ordinary chondrites have been well documented (*e.g.*, McSween *et al.*, 1988), but aqueous alteration effects have been observed in type 3 ordinary chondrites only recently (Hutchison *et al.*, 1987). To complicate matters further, most chondrites are breccias, and since it may involve both devolatilization and recondensation, brecciation may sometimes result in effects normally associated with the nebula.

Figure 1 summarizes much of the present discussion concerning the metamorphic and aqueous alteration history of type 3 ordinary chondrites. Conventionally, it has been assumed that the type 3 ordinary chondrites represent a simple single metamorphic series extending from type 3.0 to type 6. However, Hutchison *et al.* (1987) mentioned the possibility of type 3.0 being produced from higher types by aqueous alteration. Also, Guimon *et al*. (1988) found that while fairly severe laboratory heating treatments caused an increase in TL sensitivity and other changes associated with increasing petrographic type, consistent with the conversion of type 3.3 to type 6 by metamorphism, low-temperature hydrothermal treatments cause a decrease in TL sensitivity consistent with the conversion of type 3.3 to type 3.0 by aqueous alteration (Fig. 1b, after Sears and Dodd, 1988). Thus, the possibility exists that type 3 ordinary chondrites show some similarity to carbonaceous chondrites that McSween (1979) described in terms of the cartoon in Fig. 1a.

For the present study, we removed chondrules from four type 3 and one type 5 chondrite, and matrix samples from three type 3 ordinary chondrites and determined their induced TL properties.

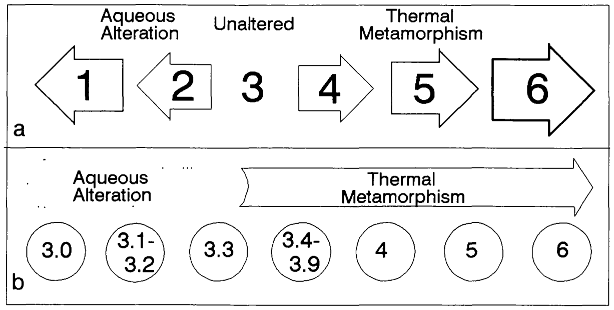

FIG. 1. A cartoon showing ideas concerning the relative importance of aqueous alteration and metamorphism in (a) carbonaceous chondrites (McSween, 1979) and (b) the type 3 ordinary chondrites (Sears and Dodd, 1988). The type 3 ordinary chondrite scenario is based on laboratory annealing experiments. These experiments demonstrate that although the series type 3.3 to type 6 appears to be the result of dry thermal metamorphism, it is possible that the type ≤ 3.3 represent either metamorphism along the series 3.0 to 3.3 or aqueous alteration from 3.3 to 3.0 (Guimon *et al.*, 1988). Petrographic observations suggest that Fig. 1b might be an oversimplification in that some type 3.1 and 3.2 chondrites are unaffected by aqueous processes, while Tiechitz (type 3.4) may have experienced aqueous alteration.

Pieces of many of these samples were examined petrographically, and many of the chondrules from three of the type 3 chondrites were analyzed by radiochemical neutron activation analysis. Twenty-two chondrules and four matrix samples from Semarkona were also analyzed for water content and H isotopes. Preliminary discussions of various aspects of this work have been presented (Sears *et al.*, 1988a,b; Morse *et al.*, 1987, 1988; Morse, 1991). We conclude that although type 3.0 chondrules closely resemble their preaccretionary form and, thus, may be considered the most "primitive" of the chondrules in ordinary chondrites, the matrix suffered extensive aqueous alteration following accretion on the meteorite parent body. We also conclude that it is doubtful that any type 3 ordinary chondrite contains matrix that is entirely unaltered nebular dust.

EXPERIMENTAL

Chondrule Sampling

Chondrules and matrix fragments were hand-picked from Semarkona, Bishunpur, Chainpur, Dhajala and Allegan. A fragment of meteorite, ~200 mg, was placed in a V-shaped, stainless steel plate and then gently crushed in a vise. The samples were selected under a low-powered binocular microscope using cleaned stainless steel dental tools. Chondrules were selected by their sphericity and coherence and matrix by its uniform very fine-grained dark appearance after crushing. Each sample was briefly described, and matrix adhering to chondrules was removed to the extent possible. Chondrules could not be extracted from Bishunpur in the way described above because they would fragment rather than separate cleanly from the unique glassy, and especially tough, matrix (Alexander *et al.*, 1989b). Instead, we located chondrules on fracture faces and removed them by chiselling. Semarkona, Bishunpur and Chainpur chondrules were weighed on a Cahn balance, while the masses of the Dhajala and Allegan chondrules were calculated from their diameter assuming sphericity and a density of 3.3 g cm^{-1}. Each sample was wrapped in thick aluminium foil and split into three parts by crushing with a dental mallet. The fragment that best sampled the chondrule core-to-rim was used to make a polished section. The largest remaining fragment was used for H-isotope analysis, and the remainder was used for TL measurements. The TL samples were then used for radiochemical neutron activation.

Petrography and Mineral/Phase Analysis

Each chondrule fragment was placed on the side of ~0.5-mm oblique resin-filled hole in a plastic microscope slide. After tumbling to ensure good adhesion, the fragment was oriented so as produce the most representative section. With about six samples per slide, the chondrules were ground and polished in the normal way. Petrographic descriptions were recorded, back-scattered electron images were prepared, and the phenocrysts and mesostasis were analyzed by electron-microprobe analysis. A summary of our data appears in Table 1; more details may be found in Morse (1991). Some chondrules were not classified because of the lack of analytical data. About 5% of the chondrules (usually small fragments) were lost in polishing, and some were not mounted for analysis for other reasons (too small, lost in handling, *etc.*).

Thermoluminescence Measurements

The TL apparatus and procedures and the cathodoluminescence (CL) imaging were described by Sears and Weeks (1983) and Sears *et al.* (1989). The natural TL was removed by heating the sample to 500 °C in the TL apparatus and TL induced by a 250 mCi ^{90}Sr β source was measured. Bulk samples of the Dhajala chondrite were used for normalization and for daily and long-term stability check of the apparatus. Three to five measurements were made for each chondrule TL chip. Averages and standard deviations were determined. On two occasions, the TL chip was large enough for further subdivision, and each subchip was measured three times. As with earlier work, we report the maximum intensity of the TL normalized to Dhajala (the TL sensitivity), the full-width at half-maximum of the major TL peak (or peak width) and the temperature of the maximum of the TL peak (peak temperature). Because chondrule masses ranged over three orders of magnitude, TL sensitivities were mass-normalized (Sears *et al.*, 1984). Our TL data for Dhajala and Allegan chondrules were the subject of previous reports where details of the Dhajala data may be found (Sears *et al.*, 1984; Keck *et al.*, 1986; Guimon and Sears, 1987). Data for the remaining samples appear in Tables 2 and 3.

Isotopic Analysis

Hydrogen-isotope analysis was performed on 26 Semarkoma samples using a modified 602E SIRA mass spectrometer (Morse, 1991). The sample preparation and inlet system were maintained at 100 °C. Reference gas from a commercial cylinder ($\delta D \sim -650$‰) was loaded into the variable volume bellows at the beginning of each day. The reference gas also facilitated the H_3^+ correction (Fisher and Brown, 1971).

Samples were placed in a quartz bucket, loaded into the furnace, and pyrolyzed in 45-min steps. The gases released during each step (H_2O, a little CO_2 and SO_2, and traces of CH_4 and H_2) were collected in a liquid-nitrogen cryotrap for 5 min. All but the water were pumped away by heating to –110 °C, and the water was then released by heating to 100 °C. After being collected in a stainless steel U-tube (20 cm x 1/16" O.D. x 0.030" I.D.) at liquid nitrogen temperature, the water was reduced to H_2 by passing through a Zn furnace consisting of 10 cm of 4 mm I.D. quartz tube with the central 2 cm packed with HNO_3-cleaned Zn shot. From the Zn furnace, the H passed directly into the changeover valve of the mass spectrometer.

An estimate of the amount of water produced was made from the intensity of the mass 2 peak, 60 μg of water giving a source pressure of about 4×10^{-7} mbar and a major beam current of about 2×10^{-9} μA for ~30 min. A Cajon fitting with a septum allowed 60-μg standard water samples to be injected with a 1-μL calibrated syringe.

The main problems encountered with this technique were a "memory effect" (*i.e.*, incomplete removal of the water and H generated during previous measurements), a relatively large blank for the Zn furnace and fractionation of the sample gas 10‰/min. Careful control of the timing of injection, frequent use of SMOW samples, an empirical calibration and a blank correction were necessary. A variety of operating conditions and a U furnace at 620 °C did not remove the memory and blank problems. A summary of our data appears in Table 4. Details can be found in Morse (1991) and Morse *et al.* (1993).

Chemical Analysis

After TL measurement, volatile or otherwise mobile elements were determined in the chondrules separated from Semarkona, Bishunpur and Chainpur using the radiochemical neutron activation analysis methods described in Zolensky *et al.* (1992). The results appear in Table 5. Neutron activation analysis of samples previously used for TL measurements shows that the latter do not affect compositional data (our own unpublished work). The heating conditions necessary for devolatilization were experimentally determined by Ikramuddin *et al.* (1977) and are considerably more severe than the momentary heating at 500 °C required by the TL measurement.

RESULTS

Petrographic Data and Chondrule Classification

Table 1 lists textural descriptions, mesostasis CaO and Na_2O, olivine FeO and CaO and low-Ca pyroxene FeO and CaO where olivine was totally absent from the chondrule. The textural types described by Gooding and Keil (1981) are present in approximately the proportions they observed, except that because of the method

TABLE 1. Petrographic and electron-microprobe data for some of the chondrules from type 3 ordinary chondrites in the present study.* +

			Ol/Px		Mesostasis	
#	group	txt	Fa/Fs	CaO	Na_2O	CaO
Semarkona Chondrules						
1	B1	PO	20	0.21	1.6	4.4
2	B1	PP	12	0.11	1.4	2.3
6	B1	PO	9	0.13	1.6	1.3
9	---	RP	19	---	---	---
11	B1	PO	16	0.11	0.65	7.6
14	B1	PO	8	0.12	0.71	6.1
17	A1	---	1.5	---	0.46	14.3
19	A1	POP	1.5	0.25	0.4	14.3
20	B1	PP	---	---	1.3	2.3
22	B1	PO	10	0.15	1.8	1.6
23	A2	POP	8	0.13	1.2	6.2
26	B1	PP	14	---	2.3	2.9
28	B2	---	---	---	4.0	3.5
29	B1	---	16	---	3.1	7.4
33	B1	C	33	---	0.26	1.1
35	---	C/RP	30	---	0.31	0.72
36	B1	POP	13	0.05	1.8	3.4
37	A4	O	17	0.29	---	---
39	A5	PP	11	---	6.8	4.4
40	A5	POP	11	0.28	7.2	4.1
41	B1	POP	10	0.07	1.7	0.67
42	B1	PO	18	0.23	1.7	0.35
44	---	C	27	---	0.37	2.0
45	---	C/RP	27	---	0.33	1.6
46	---	C/RP	27	---	0.28	1.1
47	A1-4	PP	3	---	4.9	7.6
49	A1-4	POP	7	---	---	---
50		I	8.7	---	0.31	13.4
51	A5	I	<.05	---	0.05	18.4
52	B1	PP	15	---	0.79	2.3
53	B1	PP	24	---	0.47	1.9
54	B1	PP	25	---	0.63	3.0
55	A3	POP	7	0.29	1.2	6.2
58	A1	POP	2	0.17	---	---
59	A1	POP	2	0.16	<0.05	14.6
60	B1	PO	22	0.40	0.69	1.2
64	A3	POP	2.2	0.12	1.2	6.9
65	B1	PP	25	---	2.6	0.39
66	B1	PP	8	0.40	1.8	4.0
67	B2	PP	5	---	4.7	1.5
68	A5	PP	23	---	11.4	0.86
69	A4	POP	21	0.23	0.30	10.4
70	B1	POP	7	0.13	5.2	4.5
71	A1-4	PP	2.5	---	3.4	10.8
72	B1	POP	20	0.18	2.5	5.8
Semarkona Matrix Samples						
4	--	IC	---	---	---	---
16	--	IC	---	---	---	---
18	--	IC	---	---	---	---
40	--	R	---	---	---	---
Bishunpur Chondrules						
2	B1	P/O	40	---	1.5	2.0
3	B1	G/IC	28	---	0.50	1.4
4	B1	BP	*10*	---	2.1	2.3
5	A1-4	μC/P	*1.5*	---	0.76	4.3
7		GP	*30*	---	---	---
8	B1	BO/P	30	---	3.7	2.5
10	B1	P/BP	*14*	---	2.0	3.3
12	B1	RP	*13*	---	3.2	0.89
13	B1	PP	*33*	---	1.9	2.1
15		P/O	28	---	---	---
18	B1	RP	*30*	---	2.3	1.5
19	--	RP	*33*	---	---	---
20	A2	RP	*20*	---	1.3	2.0
21	B1	GOP	8	---	2.2	3.4
22	A5	POP	3	<.05	---	---
23		RP	*31*	---	1.0	0.78
25	A3	BO	0.5	0.33	6.8	11.1
26		RP	*28*	---	4.1	1.1
27	B1,2	μCP	*30*	---	---	---
28	A2	POP	2	0.23	7.3	10.1
29	A1	PP	*0.5*	---	2.6	10.5
30	B1,2	POP?	23.5	0.17	---	---
Chainpur Chondrules						
20	B1	PO	19	0.13	3.2	4.1
21	A5	PP	3	<.05	5.2	9.8
23	---	RP	*35*	*0.05*	---	---
24	B1	POP	15	0.09	2.7	3.1
26	B1	POIK	6	0.12	2.8	1.9
41	B1	μC	33	3.6	4.3	2.8
42	B1,2	μC/C	33	1.5	---	---
43	B1	POP	29	0.12	4.9	1.6
44	B1	BP	35	0.19	4.2	0.96
45	A5	Cl	9.0	0.14	6.7	7.3
46	B2	RP	*32*	0.11	2.1	1.3
52	A5	PP	*2*	*0.19*	10.8	1.3
54	A5	RP	*16*	*1.1*	3.2	3.1
57	A5	μCP	27	0.12	7.4	4.3
58	---	μCP	*25*	*2.5*	---	---
59	A	GOP	7	0.08	---	---
60	B1	POP	15	0.17	5.7	0.07
61	B1	PO	8	0.08	3.2	3.2
63	B1	POP	7	0.07	8.0	0.68
65	A3	POP	3	0.35	2.4	5.6
Allegan Chondrules						
16	A5	---	*17*	*0.73*	7.9	5.1
33	A5	---	18	0.05	7.2	3.2
38	A5	---	19	0.03	6.3	6.7
43	A5	---	12	0.05	7.2	16.9
46	A5	---	18	0.08	2.3	3.2

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* Group, compositional groups according to Sears *et al.* (1992); txt = textural descriptions according to Gooding and Keil (1981) with the following additions; P = pyroxene; μC = microcrystalline; P/O = pyroxene-olivine intergrowths; POIK = poikylitic; Cl, noritic clast; BP = barred pyroxene; I = other igneous textures; O = other; GP = granular pyroxene. The query by Bishunpur chondrule 30 indicates texture uncertain due to poor polish. The matrix samples are described as "inter-chondrule"(IC) and "rim" (R). Fa refers to olivine phenocryst composition unless in italics in which case the value refers to Fs; Na_2O and CaO refer to mesostasis compositions or whole chondrule analyses in the case of cryptocrystalline textures. Typically 4–5 olivine grains and 2–3 mesostasis regions were analyzed for each chondrule.

+ Petrographic data for the Dhajala chondrules discussed here were presented in Sears *et al.* (1984). Most Dhajala chondrules are groups A5, A4 and B3. Very limited data were also obtained for the Allegan meteorite whose chondrules are equilibrated and members of group A5.

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used to extract chondrules from Bishunpur, pyroxene-rich chondrules were over represented. To a good approximation, the trends observed in our earlier studies (DeHart *et al.*, 1992; Sears *et al.*, 1992) are also observed here. Olivine and mesostasis data are plotted in Fig. 2. Semarkona mesostasis compositions are spread over the normative quartz-albite-anorthite triangle and chondrule olivines contain >0.1 wt% CaO. With increasing petrographic type, olivines lose CaO and their FeO approaches the value observed in equilibrated chondrites, while the chondrule mesostasis compositions migrate towards oligoclase compositions.

Figure 2 enables the present chondrules to be assigned to the compositional groups of Sears *et al.* (1992), which are also given in Table 1. Semarkona and Bishunpur mainly contain chondrules of groups A1, A2, A5 and B1, while our Chainpur chondrules are predominantly groups B1 and A5. All the chondrules separated from the Allegan chondrite are compositional group A5. The

TABLE 2. Induced thermoluminescence data for chondrules separated from type 3 and the Allegan ordinary chondrites.*

#	Mass (mg)	TL sens. (Dhaj = 1)	Temp (°C)	Width (°C)
Semarkona				
1	1.18	0.000443	---	---
2	0.424	0.00123	130	50
6	0.335	0.000008	---	---
9	0.0758	0.03296	163	88
11	0.0689	0.000038	---	---
15	0.42	0.000621	---	---
17	0.0802	0.0586	142	92
20	2.95	0.00023	179	93
22	0.0386	0.0271	186	---
23	0.25	0.00377	---	---
26	0.0784	0.000033	---	---
28	0.109	0.000024	---	---
29	0.46	0.00102	---	---
33	0.0872	0.003	175	160
34	0.0915	0.016	103	84
36	0.226	0.00927	112	45
39	0.673	0.0513	186	71
		0.00485	189	116
40	0.324	0.00367	179	125
42	0.392	0.000007	---	---
44	0.4372	0.000470	---	---
45	0.39	0.00482	155	125
46	0.433	0.232	200	198
47	0.0376	0.00583	185	165
50	0.0342	0.0214	145	113
51	0.251	0.0025	155	145
53	1.67	0.000907	156	90
54	1.98	0.00145	158	100
55	0.472	0.000886	146	103
58	0.779	0.000782	149	120
59	1.66	0.000583	181	150
	0.95	0.132	106	---
	0.95	0.00055	139	---
60	0.0913	0.000429	120	55
65	1.12	0.00117	---	---
66	0.444	0.000006	---	---
67	0.609	0.00193	158	120
68	0.325	0.00602	203	135
69	1.31	0.000399	140	110
70	0.296	0.115	187	142
71	0.0914	0.000028	---	---
72	1.64	0.000159	150	100
Bishunpur				
2	0.793	0.00033	---	---
3	1.01	0.000441	201	127
4	0.183	0.000143	---	---
5	0.0394	0.000663	---	---
7	0.2809	0.00154	158	85
8	0.268	0.000098	---	---
9	0.213	0.00736	188	---
10	0.34	0.000077	---	---
11	0.0334	0.00547	126	40
12	2.41	0.000543	180	146
		0.00391	180	131
15	0.106	0.000246	---	---
18	0.723	0.00253	136	75
19	1.07	0.000491	144	90
		0.000025	---	---
20	0.451	0.000058	---	---
21	0.0546	0.00335	114	30
22	0.0149	0.00175	---	---
23	0.513	0.000051	---	---
24	0.423	0.00166	170	115
25	0.0506	0.000516	---	---
26	0.664	0.000512	124	50
27	0.322	0.000081	---	---
29	0.14	0.00502	142	75
30	0.702	0.00744	175	130
31	0.089	0.000294	---	---
Chainpur				
1	170	2.51	104	63
2	0.324	1.57	119	77
3	1.15	0.0108	215	181
4	3.24	0.00641	123	94
5	1.83	0.0262	133	112
6	0.446	0.904	118	73
11	2.99	0.00996	116	74
12	2.25	0.00227	132	136
15	0.0195	0.121	119	100
16	0.891	0.00171	116	75
18	0.126	0.0228	127	95
20	0.333	0.00283	129	97
21	0.407	0.855	127	74
44	4.42	0.00098	167	139
45	1.86	0.00189	117	75
48	4.98	0.00255	128	100
49	1.07	0.00219	138	110
51	3.82	0.00143	123	79
52	0.769	0.00918	114	73
54	0.662	0.00237	120	100
55	0.767	0.0133	125	118
56	0.049	0.0162	118	63
57	0.19	0.194	124	77
58	0.19	0.194	108	61
59	0.055	0.901	130	70
61	0.525	0.00149	110	70
62	0.244	0.711	104	60
63	0.039	0.0261	155	100
64	3.42	0.214	105	62
65	0.115	0.151	111	33
Allegan				
1	0.75	1.66	184	145
2	0.65	0.00407	---	---
3	0.06	0.301	185	149
4	0.25	0.815	183	143
5	0.005	0.0815	186	124
6	0.15	1.51	186	141
7	2.18	11.9	196	148
8	0.32	1.36	183	144
9	0.46	3.58	178	148
10	0.51	6.27	183	146
11	0.10	1.38	187	146
12	0.47	0.943	183	143
13	0.78	4.40	184	143
15	0.13	0.684	182	149
16	0.1	0.464	184	141
19	0.24	32.1	185	145
20	0.39	2.64	187	146
21	3.61	21.1	189	152
22	0.75	2.68	182	139
23	0.1	1.00	185	145
24	0.18	0.688	184	148
25	2.17	7.89	197	150
26	4.93	25.7	203	153
27	1.06	9.44	190	148
28	0.39	2.32	189	147
29	0.29	1.69	194	149
30	0.21	0.790	181	149
31	0.91	8.90	190	147
32	0.26	2.14	182	144
33	0.26	2.93	173	144
34	0.91	8.90	200	150
36	0.22	2.25	180	143
37	1.13	17.4	191	151
38	0.59	6.44	198	146
39	0.19	0.262	180	141
40	0.26	1.48	183	146
41	0.32	4.45	187	147
42	0.51	2.88	190	148
43	0.31	1.81	183	147
44	0.39	1.81	179	145
45	0.17	2.00	187	148
46	0.68	8.40	187	147
47	0.22	0.593	187	142
48	0.43	2.94	187	147
49	0.11	0.945	183	145
50	0.09	0.942	181	148

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* The 2σ uncertainties are typically ±5 in the last figure. Data for Dhajala can be found in Sears *et al.* (1984).

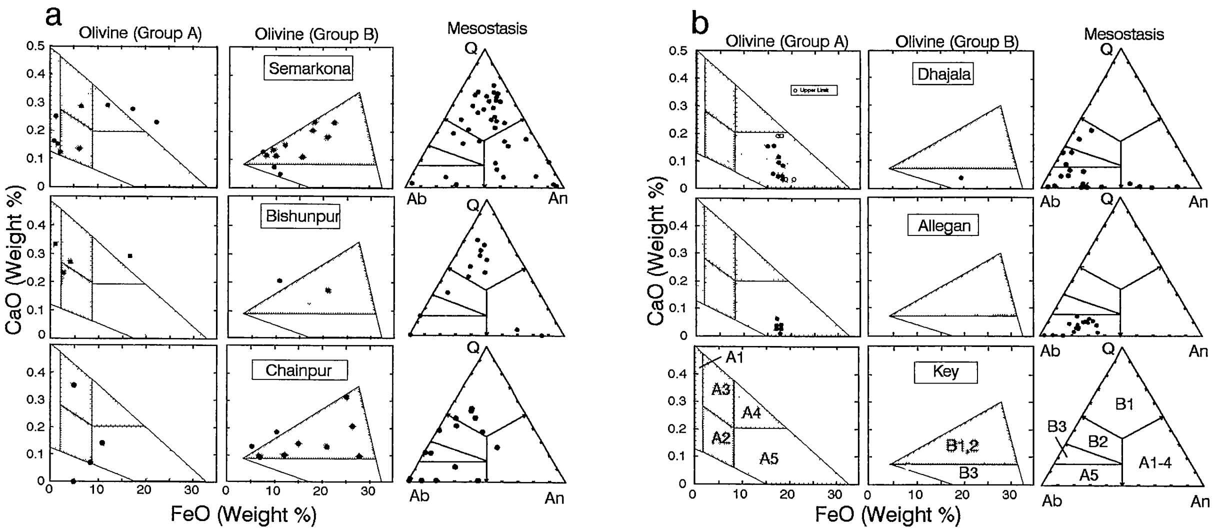

FIG. 2. Plots of the CaO *vs.* FeO contents of chondrule olivines and of the mesostasis compositions (in terms of normative quartz, albite and anorthite) for chondrules from (a) Semarkona, Bishunpur and Chainpur and (b) Dhajala and Allegan. A key to the plots appears in Fig. 2b (after Sears *et al.*, 1992, slightly simplified). The data enable the present chondrules to be assigned to the compositional classes given in Table 1.

TABLE 3. Induced TL data for matrix samples from type 3 ordinary chondrites.*

#	Mass (mg)	TL sens. (Dhaj = 1)	Temp (°C)	Width (°C)
Semarkona				
4	0.783	0.000942	---	---
13	0.362	0.00278	200	100
16	0.846	0.00235	181	150
18	0.287	0.000124	145	75
25	0.186	0.00143	105	60
31	0.190	0.000013	---	---
38	0.0703	0.000526	169	125
40	0.272	0.000474	146	140
43	0.880	0.00042	175	115
61	---	---	117	40
Bishunpur				
1	0.355	0.00147	194	125
6	3.98	0.000756	140	110
14	0.164	0.00798	130	70
16	0.0382	0.000684	---	---
17	0.362	0.00115	118	40
24	---	0.000652	188	150
Chainpur				
7	0.934	0.00976	113	63
	0.823	0.164	113	63
10	1.25	0.224	121	97
14	0.598	0.0799	125	76
17	0.571	0.0371	159	128
19	5.31	0.0719	127	79
27	1.97	0.0656	146	101
28	0.398	0.00723	136	85
35	0.104	0.00982	119	53
38	0.462	0.0237	139	130
47	0.455	0.407	192	155
		0.14	192	155
50	1.45	0.232	130	124
		0.0476	1309	124

* The 2σ uncertainties are ±5 in the last digit.

chondrules we separated from these meteorites are very roughly present in the proportions observed in thin sections (Sears *et al.*, 1992), although handpicking is known to introduce a bias against the small and friable group A chondrules (Sears and DeHart, 1989).

Thermoluminescence Data

Our TL data are shown in Figs. 3–5. Data for more than one chip from a given chondrule are connected by tie-lines and illustrate the difficulties posed by the heterogeneity of these small samples. The TL sensitivities of Semarkona chondrules range over about 3 orders of magnitude, and there is a range of ~100 °C in TL-peak temperature and ~150 °C in TL peak width (Fig. 3). There is no correlation between TL sensitivity and peak temperature and peak width, although seven out of nine group B1 chondrules plot in the lower-left half of the field occupied by the data. All but one of the group A5 and unclassified chondrules, which are often glass-rich microcrystalline chondrules that we did not analyze because their fine texture, plot in the upper-right half of the field.

Figure 4 shows plots of TL sensitivity *vs.* peak temperature for Semarkona, Bishunpur, Chainpur, Dhajala and Allegan chondrules. With increasing petrographic type, the mean TL sensitivity of the chondrules increases. However, peak temperature and width show more complex data. The Bishunpur and Semarkona chondrules show similar scatter in their TL data, but for Chainpur 38 of the 44 chondrules have peak temperatures below 150 °C, and 10 have TL sensitivities >1.0. In the case of Dhajala, more than half the chondrules have peak temperatures >150 °C, and there is a negative correlation between TL sensitivity and peak temperature (Keck *et al.* (1986). The Allegan data cluster tightly with peak temperatures close to 190 °C and TL sensitivities close to 10.0.

The TL-peak temperature and peak width data for the chondrules are compared with the two fields defined by bulk samples of type 3 ordinary chondrites (Sears *et al.*, 1991) in Fig. 5. The lower and upper fields correspond to chondrites of petrographic types 3.3–3.5 and 3.5–3.9, respectively. The lower field is occupied by samples with feldspar in the low-temperature (ordered) form, while the upper field corresponds to samples with feldspar in the high-temperature (disordered) form (Guimon *et al.*,

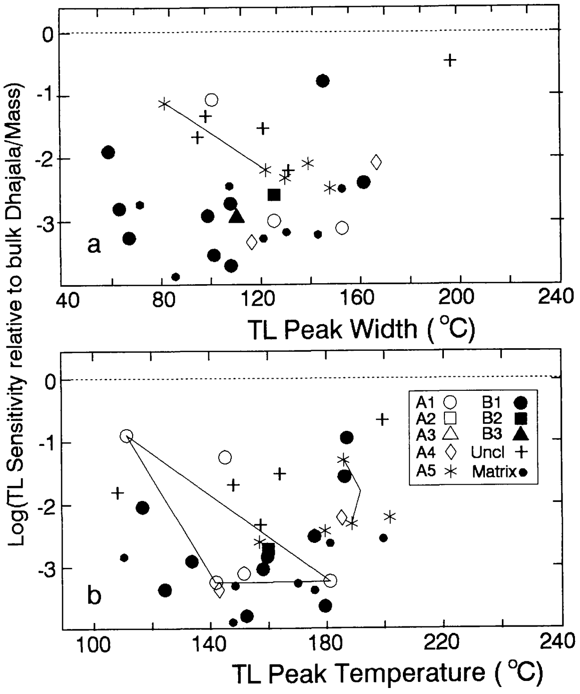

FIG. 3. Plots of TL sensitivity against peak width and peak temperature for chondrules and matrix samples from the Semarkona (type 3.0) chondrite. While there appear to be no correlations between TL sensitivity and TL-peak temperature and peak width, group B1 chondrules tend to plot in the bottom-left half of the distribution, while group A5 and unclassified chondrules (which are largely glass-rich micro-crystalline chondrules) plot in the upper-right half of the field. This is consistent with group A chondrules being better crystallized and crystallizing at higher temperatures than group B chondrules. Matrix samples plot in the lower half of the TL sensitivity range but spread over the full range of TL peak temperatures and widths. Tie-lines connect chips from a single chondrule and illustrate the heterogeneity of these small samples.

1985). Chondrules from the type 3.0–3.1 chondrites straddle the two fields, while Chainpur chondrules mainly occupy the lower field or plot between the fields. Dhajala chondrules plot in a field slightly displaced to the right of the type 3.5–3.9 field or between the fields. Allegan chondrules (with two exceptions) plot in a tight cluster in the upper field.

Matrix samples from Semarkona, Bishunpur and Chainpur are also shown in Figs. 3 and 4. The TL data for the matrix of a given meteorite show similar spread to that of chondrules, except that in the case of Semarkona matrix, TL sensitivities are in the lower half of the chondrule range.

Hydrogen Isotopes and Water Content

Previous stepped pyrolysis measurements for Semarkona yield water with roughly terrestrial D/H at temperatures <200 °C but with considerable D enrichments (up to 4500‰) in the water released at 600–1000 °C (McNaughton *et al.*, 1981, 1982; Yang and Epstein, 1983; Hinton *et al.*, 1983). A fall in D/H ratio for water released in the highest temperature step may indicate a third component. The present six-step pyrolysis of the 20-mg Semarkona sample produced results in good agreement with literature data, even though earlier workers used ~150-mg samples (Fig. 6a–d). Chondrule masses are too small for six-step pyrolysis. However, two-step pyrolysis (≤200 °C and 200–1000 °C) of a 1-mg sample (Fig. 6e) is apparently sufficient to measure the quantity and isotopic value of the deuterium-rich component.

Our water content and D/H data are shown in Table 4 and Fig. 7. As with whole-rock data, most of the water was released in the low-temperature step and had approximately terrestrial D/H values. The high-temperature steps released relatively small amounts of water (<0.2 wt%) with a wide range of δD values. The average water content of the chondrules and δD values during the high-temperature release from the chondrules were 0.12 ± 0.02 wt% and 3890 ± 500‰, respectively (arithmetic means $\pm 1\sigma$). In five cases, the amount of water released was below our 0.017 μmol detection limit.

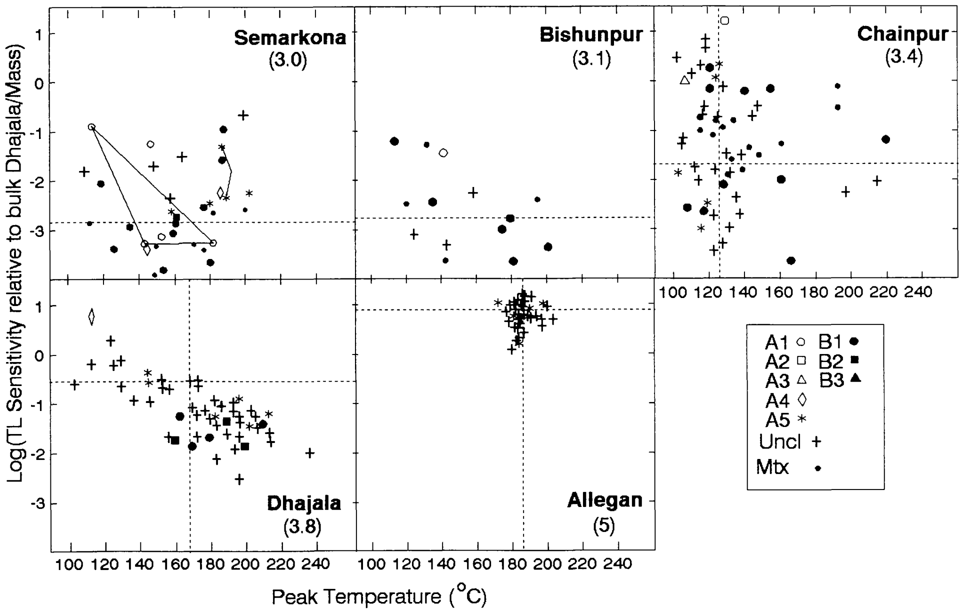

FIG. 4. Plots of TL sensitivity against TL-peak temperature for chondrules from five ordinary chondrites of a range of petrographic type. The cross-hairs refer to the values for the bulk sample. (Because of low signal and sample heterogeneity, it is not meaningful to assign peak temperatures to the bulk Semarkona and Bishunpur). The mean TL sensitivities of the chondrules increase with petrographic type, and there are systematic changes in peak temperatures. The data for Semarkona and Bishunpur scatter fairly randomly, but Chainpur chondrules tend to have peak temperatures <150 °C. More than half the Dhajala chondrules have peak temperatures >150 °C, and Dhajala chondrules show a negative correlation between TL sensitivity and peak temperature. Allegan chondrules form a tight cluster with peak temperatures of about 190 °C and TL sensitivities close to 10.0. Matrix data are also plotted for Semarkona, Bishunpur and Dhajala. These data are consistent with metamorphism being the major factor in determining the TL trends shown by type 3 ordinary chondrites.

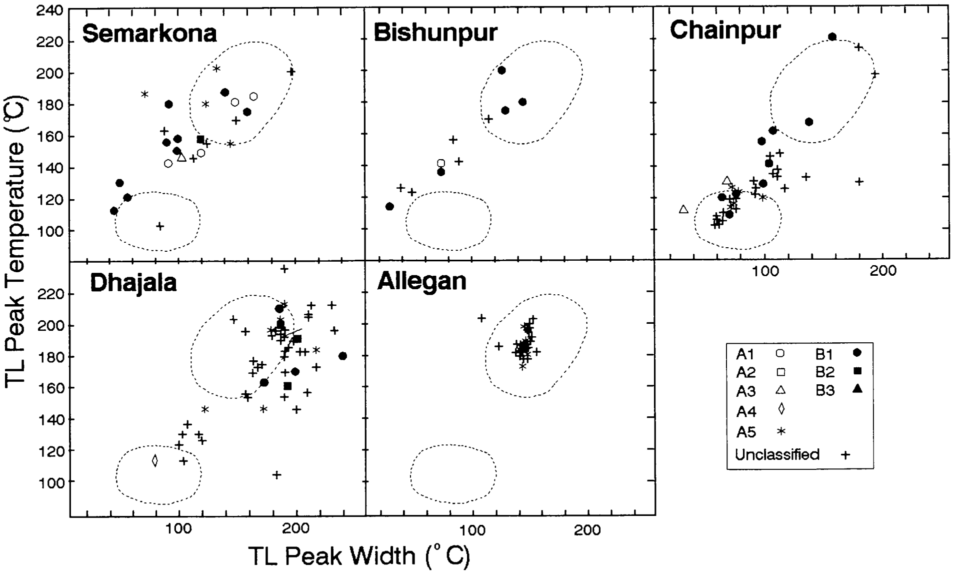

FIG. 5. Plots of TL-peak temperature *vs.* peak width for chondrules from ordinary chondrites. The two fields refer to type 3 ordinary chondrites and samples annealed in the laboratory (Sears *et al.,* 1991; Guimon *et al.,* 1985) and are thought to be due to low- and high-temperature feldspar in the lower-left and upper-right fields, respectively. Semarkona and Bishunpur chondrules show a correlation between these parameters but do not plot preferentially in either of the two fields. On the other hand, Chainpur chondrules plot preferentially in the low field or between the fields, and Dhajala chondrules plot in the upper field (displaced slightly to the right) or between the fields. Allegan chondrules plot in a tight cluster (two chondrules aside) in the upper field. These trends are also consistent with metamorphism being the major factor in determining the TL properties of chondrules in type 3 ordinary chondrites.

All four matrix samples contained very large amounts of water (>0.25 wt%) but only moderate D/H ratios (≤+3000‰). The high D/H observed for the chondrules cannot, therefore, be attributed to contamination with the present matrix. It is possible that chondrule SC17, with conspicuously high water content but moderate D/H value, might have contained adhering matrix although no matrix was present on the corresponding petrographic sections.

Volatile/Mobile Trace Elements

Our RNAA data appear in Fig. 8. The elements are sorted by cosmochemical group and within each group are plotted in order of increasing element mobility as determined from laboratory heating experiments on Krymka (Ikramuddin *et al.*, 1977). In view of the generally low abundance and heterogeneity of these elements, the patterns obtained are surprisingly reproducible from chondrule-to-chondrule regardless of chondrule group, although not all the groups are adequately represented. Lithophile-element abundances are higher than siderophiles, which are higher than chalcophiles. Lithophile-element abundances decrease with increasing element mobility, whereas chalcophile-element abundance increases with increasing element mobility. Siderophile elements are either independent of element mobility or, in some cases, increase with mobility.

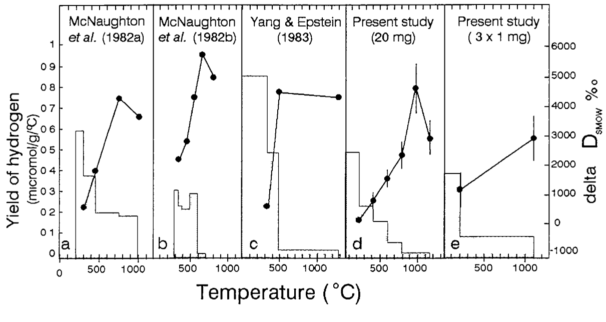

FIG. 6. A comparison of the abundance and isotopic composition of H released during stepped pyrolysis of bulk Semarkona samples. Blocks a-c summarize literature data while blocks d and e summarize data obtained in the present study. Data for six-step pyrolysis of a 20-mg sample are shown in Fig. 6d, and the mean of three 1-mg samples analyzed in two steps are shown in Fig. 6e. All data indicate the presence of large amounts of a component with terrestrial composition, which is released <400 °C, and a D-rich component, which is released >400 °C.

DISCUSSION

We are primarily interested in the extent to which aqueous alteration and metamorphism, separately or together, have affected the type 3.0–3.3 ordinary chondrites. We will first consider evidence for metamorphism, then primary variations and finally aqueous alteration.

Metamorphic Properties

Metamorphism caused three major changes to the chondrules. (1) Mineral and phase compositions homogenized in the manner described by Dodd (1969), Dodd *et al.* (1967), Jones and Scott (1989), Jones (1990), McCoy *et al.* (1991), DeHart *et al.* (1992) and others. (2) Chondrules did not act as closed systems but underwent major compositional changes during metamorphism. As a result, there was eventually almost complete chondrule-to-chondrule uniformity of bulk and mineral composition (Sears *et al.*, 1992). Chondrules, whose mesostases originally ranged from quartz- to anorthite-normative, became oligoclase-normative with metamorphism. Similarly, chondrule olivines lost Ca and gained Fe during metamorphism. (3) Metamorphism produced new feldspar by crystallization of chondrule glass (Van Schmus and Wood, 1967). This form of "secondary" feldspar was ordered or disordered depending on the temperature of crystallization; production of ordered-feldspar from disordered-feldspar is kinetically difficult (McKie and McConnell, 1963).

The TL sensitivity of our samples is governed primarily by the amount of feldspar present, while the temperature and width of the induced TL peak are determined by the relative abundances of high- and low-temperature forms of feldspar (disordered and ordered, respectively; Guimon *et al.*, 1985, 1986, 1988). Composition has a major effect on TL sensitivity by affecting the spectrum of the light (Batchelor and Sears, 1989) but apparently only has a minor effect on TL-peak temperature and width data. Plagioclase-normative mesostases produce TL peaks at

TABLE 4. Water released, and its H-isotopic composition, during two-step pyrolysis of chondrules and matrix samples from the Semarkona type 3.0 ordinary chondrite.

#	Mass (mg)	200 °C H_2O* (wt%)	200 °C $\delta D \pm 2\sigma$ (‰)	1100 °C H_2O* (wt%)	1100 °C $\delta D \pm 2\sigma$ (‰)
Chondrules					
1	1.3731	<0.022	–600 ± 200	<0.022	+4250 ± 1200
2	1.0690	<0.028	+3200 ± 650	0.122	+1800 ± 600
6	1.0118	<0.030	+1120 ± 500	0.247	+1800 ± 600
9	0.0789	<0.382	–780 ± 200	<0.382	+740 ± 800
15	0.8274	<0.036	+750 ± 350	<0.036	+2650 ± 1200
17	0.0869	1.266	+480 ± 300	1.036	+3600 ± 900
19	0.0426	<0.704	+650 ± 300	<0.704	+380 ± 400
20	1.5291	<0.020	–400 ± 300	<0.020	+3200 ± 1400
28	0.3522	<0.085	+150 ± 300	0.142	+4320 ± 1000
34	0.1739	0.345	+900 ± 400	<0.173	+2800 ± 1600
36	0.4820	<0.062	+550 ± 400	<0.062	+5500 ± 1000
40	0.3570	0.252	+480 ± 300	0.3641	+4150 ± 800
42	0.4902	<0.061	+100 ± 300	<0.061	+6200 ± 1300
46	0.5300	0.189	---	0.208	+2000 ± 1300
47	0.3367	<0.089	+1300 ± 400	<0.089	+5000 ± 900
49	0.0591	<0.507	+550 ± 300	<0.507	+850 ± 400
54	1.9223	0.052	–620 ± 200	0.073	+5100 ± 1100
58	0.3143	0.095	+3300 ± 700	0.095	+6620 ± 1500
65	0.9666	0.103	+2650 ± 600	0.103	+7380 ± 1200
70	0.2698	0.556	+1850 ± 450	0.556	+5680 ± 1300
71	0.0952	<0.315	–300 ± 250	<0.315	+280 ± 300
72	1.2479	0.031	+1700 ± 500	0.165	+6050 ± 1000
Chondrule Mean					
	0.6317	*0.083*	*+840 ± 130*	*0.121*	*+3890 ± 500*
Matrix Samples					
4	0.0900	<0.333	0 ± 300	0.330	+1300 ± 1300
16	0.6208	0.097	+1100 ± 400	<0.048	+3000 ± 900
18	0.4488	0.624	+750 ± 350	0.758	+2250 ± 600
40	0.2082	1.057	+1450 ± 400	2.546	+3000 ± 800
Matrix mean					
	0.3430	*0.43*	*+1010 ± 160*	*0.68*	*+2670 ± 520*

* The 2σ uncertainties on the water are ~ ±10% of the quoted amount.

temperatures of ~100–300 °C when anorthitic (group A chondrules), and ~100 °C when albitic in composition (group B chondrules) (Ninagawa *et al.*, 1991, 1992; Matsunami *et al.*, 1992a,b). Thermoluminescence glow curves of the sort attributed by Matsunami *et al.* (1992a) to cristobalite—produced by mesostases containing 84–99 wt% SiO_2—were only rarely observed during the present work. With these generalizations in mind, it is helpful to discuss the TL trends as a function of petrographic type starting with the highest types.

The chondrules in Allegan are compositionally uniform, and their mesostases well crystallized. The TL sensitivity of Allegan chondrules is relatively high, and peak temperature and widths plot in a tight cluster between the fields corresponding to high and low feldspar (Figs. 3–5). We, therefore, conclude that all the Allegan chondrules contain the same mixture of high and low feldspar, consistent with the chondrules experiencing the same thermal history (involving high levels of metamorphism) and responding uniformly during cooling. This requires uniform composition of all the chondrules.

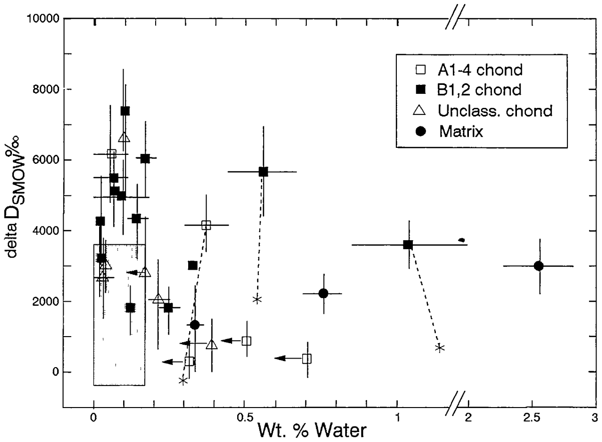

FIG. 7. Plot of δD against water released from chondrules and matrix samples from the Semarkona chondrite. Data for water released <200 °C (thought to be terresrial containation) plots in the shaded field. (Exceptions are three samples whose <200 °C data are plotted as asterisks; tie lines connect these data to their high-temperature counterparts). Arrows indicate upper limits. Data for water released at 200–1000 °C are plotted as squares, triangles (chondrules) or circles (matrix). The very high δD values sometimes observed are not due to the presence of matrix on the chondrules, which has relatively low values of δD. These data suggest that if the high δD component in type 3 ordinary chondrites was originally located in the matrix, it has subsequently been leached by water of terrestrial δD, and only vestiges remain associated with particular chondrules.

This is not the case for meteorites of lower petrographic type. Dhajala chondrules show negative correlations between TL sensitivity and peak temperature and width, demonstrating that some chondrule mesostases are crystallized more extensively than others, and that the chondrules with the most crystallized mesostases contain feldspar in the low form. Apparently, only a few of the chondrules continued to produce feldspar as they cooled through the low-temperature field from peak metamorphic temperatures. Sears *et al.* (1984) showed that this process was compositionally controlled, in part, although chondrule size and other local factors would also affect kinetics of crystallization and feldspar ordering.

Very few Chainpur chondrules contain feldspar in the high-form, and since high-feldspar (unlike low-feldspar) forms very readily, the meteorite could never have been heated into the high-temperature field. The spread in TL sensitivities is very large because the extent of crystallization varies considerably from chondrule to chondrule, reflecting compositional heterogeneity in the meteorite. Sears *et al.* (1992) showed that group A chondrules respond more readily to metamorphism than do group B chondrules.

The TL data for separated chondrules from type 3 ordinary chondrites are therefore readily understood in terms of metamorphic changes occurring after their formation and incorporation into the meteorite. On the other hand, it seems highly unlikely that the mineralogical trends observed in chondrules could be the result of aqueous alteration, and it seems equally unlikely that the TL trends observed above could be the result of aqueous alteration. Guimon *et al.* (1988) performed hydrothermal-alteration experiments on Semarkona and Allan Hills A77214 (3.4) that provide some indication of the TL trends expected from aqueous alteration. Thermoluminescence sensitivities tend to decrease following hydrothermal alteration, but peak temperatures increase in a complex fashion, producing TL peaks not commonly observed in the natural chondrules. This probably reflects preferential destruction of low-temperature peaks, a behavior quite unlike the trends observed among the type 3 ordinary chondrites where low-levels of TL sensitivity is associated with low-temperature peaks.

Primary Properties

The range of TL sensitivities, peak temperatures and peak widths shown by the Semarkona and Bishunpur chondrules give no indication of correlation. This suggests that feldspar is present in varying amounts and in the high- and low-forms, in varying proportions.

Cooling rates at high temperatures during chondrule formation were very rapid, 100–1000 °C/h (Hewins, 1988), so that the cooling phase, which resulted in the formation of feldspar, must have occurred at low temperatures, perhaps during blanketing in dust or regolith in order that significant amounts were in the low form. Since most other chondrules (mainly the unclassified and group A5 chondrules because data for the others are scarce) plot in the upper-right half of the distribution in Fig. 3, compared to group B, these chondrules are more crystallized and contain a greater proportion of high-feldspar. Consistent with this, group B chondrules contain mesostasis compositions, indicating considerable supercooling (Jones, 1990; DeHart *et al.*, 1992).

Like the other group A chondrules, group A5 chondrules avoided supercooling, but they are Na-rich. If these chondrules cooled slowly enough to avoid supercooling, they would, like the other A groups, have lost Na. We suspect that they acquired Na by recondensation. Matsunami *et al.* (1992b) describe a large group A chondrule in which the outer regions of the mesostasis are volatile-rich; they ascribe this to recondensation of volatiles in such an environment. Chondrule 13c of DeHart *et al.* (1992) is another example. In any event, their TL data are consistent with relatively slow cooling with some crystallization in the high-temperature field of feldspar.

The observed TL trends could not result from the effects of aqueous alteration of individual Semarkona or Bishunpur chondrules. As discussed above, Guimon *et al.* (1988) laboratory data indicate that aqueous alteration at low temperatures does not produce low-temperature feldspar from high-temperature feldspar (*i.e.*, decrease the TL peak temperature from 200 to 100 °C), nor does it produce high-temperature feldspar from low-feldspar (which would result in a 100 °C temperature increase).

The most striking aspect of the present data is that none of the present chondrules are significantly depleted in highly mobile chalcophile and lithophile elements; In, Tl, Bi and Cd are remarkably close to CI proportions in all the chondrule groups, including group A. Ikramuddin *et al.* (1977) found that these elements were particularly mobile during laboratory heating and that they have especially low activation energies for evaporative loss. Group A1 chondrules in Semarkona are strongly depleted in volatile elements—Na and K typically by 60%, Mn and Cr by around 40%, and Si by 10–20% (Lu *et al.*, 1990; Lu, 1992).

To a first approximation, abundances are governed by mineralogical factors. Metal and sulfides are depleted and silicates enriched in the chondrules. The resulting element-abundance patterns are well known. However, the patterns observed within

TABLE 5. Trace element data for chondrules from type 3 ordinary chondrites.[*]

	Chond Class[‡]	Ag ng/g	Au ng/g	Bi ng/g	Cd ng/g	Co μg/g	Cs ng/g	Ga μg/g	In ng/g	Rb μg/g	Sb ng/g	Se μg/g	Te μg/g	Tl ng/g	U ng/g	Zn μg/g
Semarkona																
39	A5	35	31	8.9	130	110	78	0.59	92	9.0	62	3.5	<0.02	16	16	4.1
40	A5	26	37	22	380	140	2100	1.3	350	5.8	87	2.8	0.049	33	24	13
46	--	80	6.2	410	120	100	430	13	39	33	120	0.58	<0.20	98	15	45
47	A1-4	210	110	<190	3600	500	300	6.6	280	8.1	2800	5.4	1.5	160	250	69
51	A5	8.9	3.6	52	470	86	17	0.46	37	0.35	52	0.17	<0.36	67	140	1.9
60	B1	230	4.9	<260	5400	240	260	1.1	160	5.8	3000	0.36	4.1	220	1400	50
65	A3	12	23	6.3	940	120	190	1.9	60	3.2	300	2.7	0.20	13	14	17
70	B1	89	15	120	1600	140	170	2.1	1700	3.2	530	3.1	0.11	64	260	46
Bishunpur																
3	B1	84	17	37	920	100	590	3.2	13	9.4	270	1.8	1.3	120	150	21
7	--	140	11	520	1500	93	360	2.6	260	2.8	61	1.7	0.067	150	450	24
12	B1	870	130	17	810	540	180	5.5	980	4.2	100	8.3	0.13	990	41	45
18	B1	90	62	<3.2	480	370	230	4.1	230	35	62	8.5	0.14	80	160	30
21	A4	1100	65	<20	7700	350	630	1.2	10000	4.1	630	3.9	1.1	380	250	69
29	A3	230	150	<40	5000	790	350	4.9	15000	15	400	12	0.39	2000	260	56
30	B1,2	2500	99	<18	11000	390	880	3.1	73000	9.9	2300	7.3	<0.84	1400	310	440
Chainpur																
39	--	39	140	330	73	150	390	2.6	64	3.1	70	6.0	0.09	180	170	19
41	B1	3300	95	640	5800	110	790	2.6	390	6.8	1600	0.62	4.8	240	320	38
42	B1,2	240	50	97	920	91	150	1.7	41	3.7	400	1.9	0.45	200	130	23
43	B1	550	25	480	1700	110	280	1.8	140	10	1300	1.8	1.0	52	160	93
44	B1	58	62	33	400	400	87	1.2	21	3.5	230	7.6	0.22	230	170	23
45	A5	110	18	72	43	140	240	3.8	23	9.3	100	2.4	0.26	76	110	21
60	B1	120	15	21	1200	450	400	8.8	300	74	150	4.1	<0.38	140	4100	11

[*] Uncertainties are assumed to be those established from replicate analysis of 5-mg Murchison samples (Zolensky *et al.*, 1992): ≤15% for Ag, Au, Co, Cd, Cs, Rb, Zn; a factor of two for Sb, and 33% for all other elements.

[‡] Electron microprobe and cathodoluminescence data are not available for three chondrules which are therefore unclassified.

the siderophile and lithophile element groups (*i.e.*, the decrease in lithophile abundance and the increase in chalcophile element abundances with increasing mobility) have not previously been observed. The lithophile-element depletion clearly indicates evaporative loss, presumably during chondrule formation. The increase in chalcophile elements, sometimes also observed among the siderophile elements, apparently indicates that the most mobile elements are recondensing on the chondrules or in the chondrule rims. Huang *et al.* (1993) recently proposed that the Fe- and FeS-rich layers in the rims of certain chondrules were the result of evaporation of volatiles from the chondrules and recondensation in the outer portions or rims of the chondrules. Especially noteworthy in the present case is that all chondrules are displaying this effect, although Huang *et al.* found that it was most important for group A chondrules. Group A chondrules, which are those that behaved as open systems during chondrule formation, display this effect most markedly, but apparently the most labile elements show this effect in all groups.

One reviewer invoked an alternative scenario involving multiple heating events. In this scenario, chondrules are partly to completely volatilized in the chondrule-forming event and, after cooling, acquire a fine-grained coating of matrix materials rich in labile chalcophile- and siderophile-trace elements. According to this scenario, during subsequent solid-state reheating, some of the most mobile of these trace elements would be vaporized, while another portion would diffuse into the chondrule. This process could then result in chondrules rimmed by volatile-rich materials.

We do not find this construct to be at all credible, experimentally or theoretically. There is no evidence that highly labile trace elements diffuse into condensed phases in chondrites on heating. Highly labile elements are not concentrated in a single phase (Lipschutz and Woolum, 1988), and data from numerous studies of naturally annealed chondrites indicate that these elements are simply vaporized (Walsh and Lipschutz, 1982; Huston and Lipschutz, 1984; Paul and Lipschutz, 1989). Experimentally determined activation energies for vaporization of the solids are quite low (Lipschutz and Woolum, 1988). Highly labile trace elements in chondrule rims are in an ideal location to be vaporized since they are already at grain boundaries. Finally, there is evidence for recondensation of highly labile trace elements onto

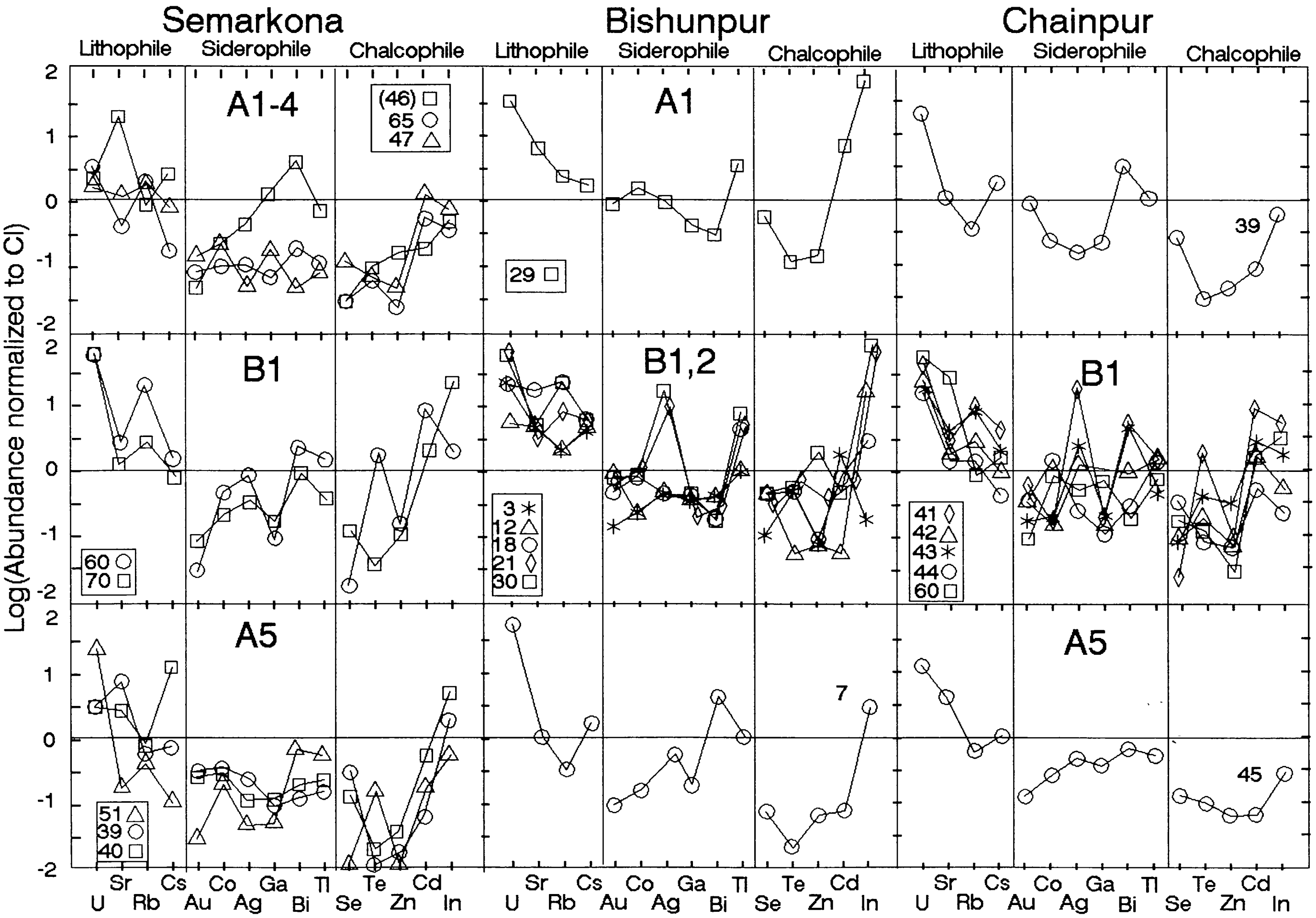

FIG. 8. Plots of the CI-normalized abundances of volatile/mobile elements in chondrules determined by radiochemical neutron activation. The numbers give the chondrule identification. Within the cosmochemical groups, elements are plotted in order of increasing mobility during laboratory heating experiments (Ikrammuddin *et al.*, 1977). In addition to the trend lithophiles > siderophiles > chalcophiles, the lithophiles decrease while the chalcophile (and sometimes siderophile) elements increase in abundance with increasing mobility. The decrease in abundance with volatility shown by the lithophiles reflects evaporative loss. However, the highly labile elements seem to have recondensed, and the chondrules are now enriched in these elements, perhaps associated with the metal- and sulfide-rich rims.

meteorites of lunar origin and eucrites (Kaczaral *et al.*, 1986; Lindstrom *et al.*, 1991; Paul and Lipschutz, 1989).

Thus, it seems that the elemental-abundance patterns for the chondrules are readily interpreted in terms of chondrule formation processes and are difficult or impossible to reconcile with redistribution during aqueous or thermal alteration.

Like previous workers who have analyzed olivine and mesostasis compositions in chondrules of type 3 ordinary chondrites (Tsuchiyama *et al.*, 1980; Taylor and Cirlin, 1986; Jones and Scott, 1989), we found values of K_D ($(FeO/MgO)_{ol}/(FeO/MgO)_{melt}$) well below the value of 0.3 determined in experimental systems by Roeder and Emslie (1970). Taylor and Cirlin (1986) suggested that these low values reflected higher crystallization temperatures for the low-Fe chondrules while Tsuchiyama *et al.* (1980) attributed low K_D values to steep Fe gradients in the olivine. However, we observe that values <0.3 are independent of Fe content (Fig. 9), and we suspect that these low K_D values are the result of the melt having had plagioclase-rich compositions, as suggested by Jones and Scott (1989). While K_D values are <0.3 for chondrites of the lowest petrographic type, they rapidly approach 1.0, and the olivines approach their uniform FeO values with increasing metamorphism. Semarkona appears to lie at one end of the type 3 ordinary chondrite continuum.

Another important primary property of the chondrules is that a significant number of them display high D/H ratios. There is considerable evidence for changes in isotopic and elemental composition during chondrule formation (Clayton *et al.*, 1983, 1985, 1991; Lu, 1992; Rubin *et al.*, 1990). However, Robert *et al.* (1987) rejected the idea that the high D/H ratios were produced during chondrule formation since unpublished C- and N-isotopic data for chondrules show no evidence for fractionation and because there is no correlation between δD and chondrule abundance. In any event, both D and H would almost certainly have been entirely lost during chondrule formation.

Aqueous Alteration

Both Robert *et al.* (1987) and Yang and Epstein (1983) also suggested that a component with terrestrial D/H was located in the inter-chondrule matrix and associated with phyllosilicates. McNaughton *et al.* (1981, 1982) found that the highest δD is released at 550–650 °C during step-wise heating, which corresponds to the temperature range for release of bound water from smectite (Hutchison *et al.*, 1987). It is possible that the D from an organic-carrier phase exchanged with H in the earliest formed smectite and that the later formed smectite is isotopically "normal." Yang and Epstein (1983) suggested that the present low D/H of the matrix may be due to exchange with water at temperatures of ≥200 °C. Alexander *et al.* (1989a) suggested that aqueous alteration occurred at temperatures <260 °C. We suggest that perhaps the exotic high-D component was originally located in the matrix but, following leaching by water with near-terrestrial D/H, the only vestiges of original high-D component remaining are now located in cracks in chondrules and in rims.

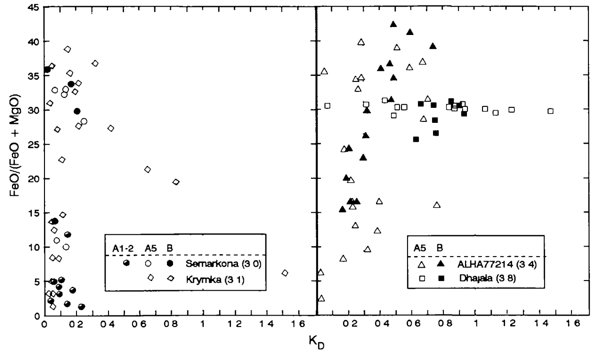

FIG. 9. Plots of Fe-number (FeO/(FeO + MgO)) for olivine *vs.* of K_D ($(FeO/MgO)_{ol}/(FeO/MgO)_{melt}$) for the mesostasis (assumed to be "melt") for chondrules from type 3 ordinary chondrites. The K_D values approach 1.0 with increasing metamorphism, while chondrules from the most unmetamorphosed chondrites have K_D values less than the equilibrium value of 0.33, independent of Fe-number. These data suggest that (1) values of $K_D > 0.3$ are indicative of metamorphic redistribution of Fe and (2) values <0.3 are independent of chondrule Fe, contrary to suggestions that the Fe content of the olivine affects K_D.

Our TL data provide at best only weak evidence for extensive aqueous alteration of the matrix. It seems fairly clear that Semarkona and Krymka contain a great many potential TL phosphors, while the TL of Chainpur matrix samples has peak temperatures and widths similar to Chainpur chondrules, where the feldspar is in the low form. The generally much smaller spread in TL sensitivities displayed by the matrix could reflect the homogenization of TL properties by destruction of feldspar; the aqueous alteration produced in the Guimon *et al.* (1988) experiments also resulted in a small spread in TL sensitivities. Alternatively, the uniform matrix TL sensitivities could reflect thorough mixing of fine grains prior to lithification of the meteorite. Guimon *et al.* experiments also showed that aqueous alteration preferentially destroys the low-temperature TL and thereby increases apparent peak temperature. There is little or no evidence for such a trend in the TL data for Semarkona matrix.

SUMMARY AND CONCLUSIONS

The question of the relative importance of aqueous alteration and metamorphism in the early history of type 3 ordinary chondrites is related to the question of the "primitiveness" of the matrix—a question on which little consensus exists (Scott *et al.*, 1988). Apparently, while some primitive properties may be present, they are difficult to distinguish from metamorphic and aqueous alteration effects, especially since brecciation appears to have been common.

It seems that the chondrules were susceptible to thermal metamorphism but resistant to aqueous alteration. Therefore, they constitute a metamorphic series. However, the matrix history is more complicated. The matrix of Semarkona is unique among the ordinary chondrites and similar to that of the primitive members of other chondrite groups by having abundant uniformly fine-grained forsterite. Ikeda *et al.* (1981), Kurat (1969) and Scott *et al.* (1988) also observed that the matrices of ordinary chondrites resembled those of CM chondrites. We suggest that little nebular dust survives in Semarkona (3.0) and Bishunpur (3.1), although grains of interstellar material (SiC, diamond and, perhaps, organics) are present. On the other hand, it is clear from a wide variety of data that the matrix in meteorites of type >3.3 has undergone significant textural and compositional changes during metamorphism.

Even the most unaltered nebular material (other than the chondrules), namely the matrices of the Krymka and the type 3.2–3.3 chondrites, has experienced significant levels of metamorphism. Nevertheless, they are the most fertile hunting grounds for primitive nebular material. While there are a great many known ordinary chondrites, most type 3.1–3.3 chondrites are finds and have been subjected to (sometimes considerable) terrestrial alteration. Included in this category are a large number of paired type 3.2 chondrites from the Lewis Cliff. Only Ngawi and St. Mary's County are observed falls. Ngawi is a regolith breccia (Sears *et al.*, 1991) with a comminuted coarse matrix enclosing various clasts of low petrographic type material, some of type 3.3. The mineralogy of the 23-g St. Mary's County was described by Noonan *et al.* (1977), and Lu *et al.* (1989) described its CL properties, but its matrix has not yet been studied.

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