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THE EVOLUTION OF CHONDRULES

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Abstract: The range of olivine compositions forming anomalous inclusions in chondrules suggests fractional condensation, as does the sequence of minerals in fine matrix particles. Chondrule bulk compositions vary in a random way as the precursors (condensates) were assembled heterogeneously. Chondrule heating and cooling were moderately rapid, and at high oxygen fugacities, suggesting processing in a localized particle-rich clump in the nebula. Rims deposited on chondrules show that condensation continued after chondrule formation. The hot particle-rich clump slows radiative cooling of chondrules, allows condensation, provides high oxygen fugacity as a result of evaporation and generates the collisions needed to fragment chondrules. The simultaneous evaporation/condensation and melting/crystallization are explained by pressure gradients in the clump. In the thickest parts of the clump, high vapor pressure allows melting, but at the outside evaporation and condensation would proceed. The observation that round chondrules are more magnesian and fragments are more ferroan is similarly consistent with formation in outer low-pressure reduced regions and the thick clump center, respectively. Turbulent migrations of condensates in and out of such clumps would explain the random nature of precursor compositions, while evaporation and gas dissipation would explain changes in oxygen fugacity which chondrules record plus their shape-redox relationship.

1. Introduction

Most of the innumerable papers on chondrules have concentrated on one aspect of their evolution, such as the preservation of relict grains, the conditions of crystallization, or modification after solidification. Each of these topics gives us some clues about conditions and processes at some stage in the environment in which chondrules formed, inferred by most recent investigators to be the solar nebula. This paper attempts to review the entire evolutionary history of chondrules, assuming that a clearer or fuller picture should emerge than if what happens at each stage is considered separately. It is assumed further that the composition and texture range of chondrite components, such as chondrules, inclusions in chondrules, chondrule rims and unequilibrated chondrite matrix, has experienced similar but somehow different environments or processing. A physical model is constructed to explain the chemical and textural differences and thereby to clarify the nature of the chondrule-forming region.

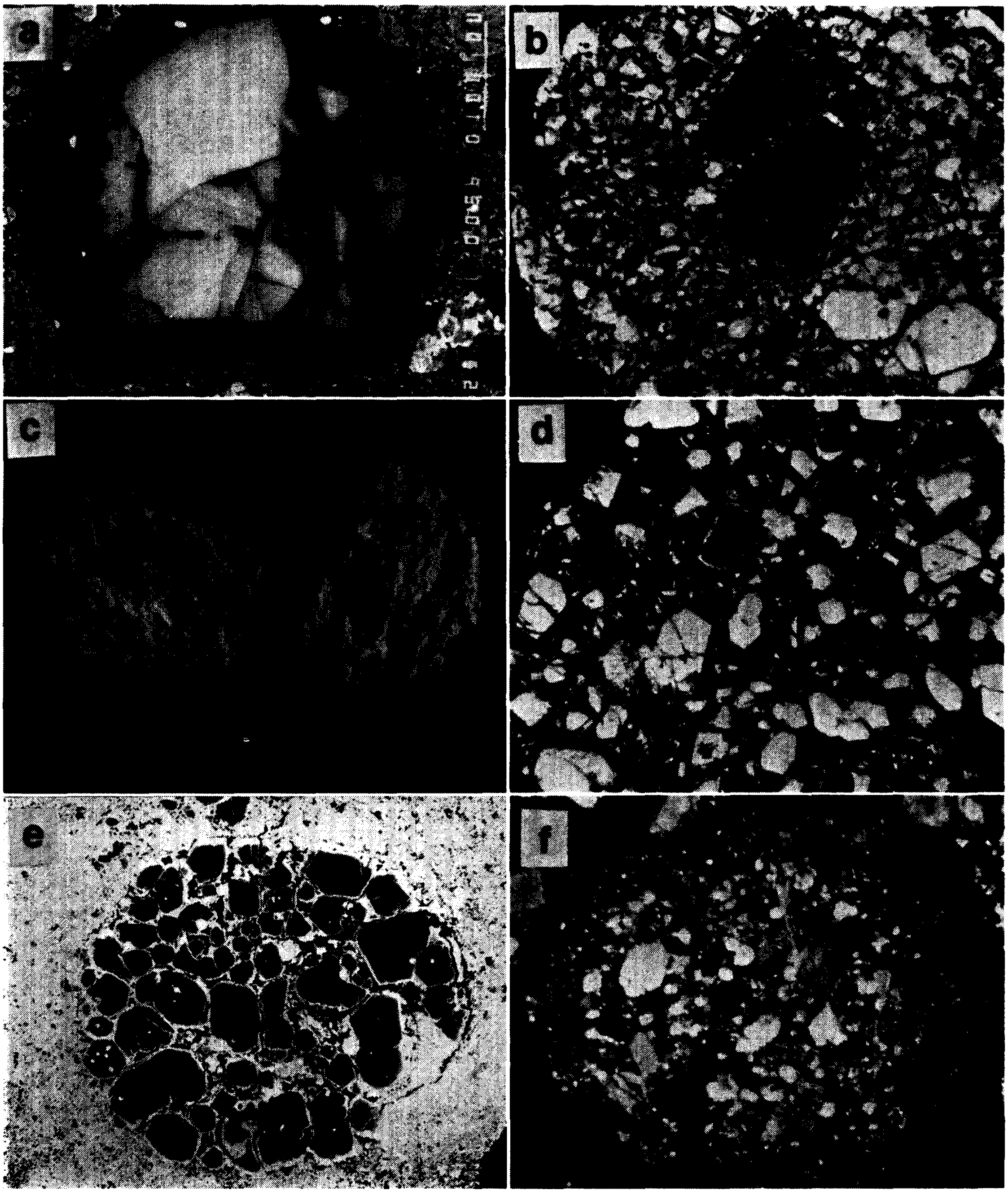

Fig. 1. (*a*) *Single olivine grain in Chainpur in combined BSE and scanning cathodoluminescence image, 0.5 mm long: bright is blue, medium is red and dark is non-luminescing. Similar grains occur inside chondrules. Reprinted with permission, from* STEELE, *Geochim. Cosmochim. Acta,* ***50***, (*b*) *Dusty olivine relict in porphyritic olivine pyroxene chondrule in Chainpur USNM 1251-2, 0.8 mm long.* (*c*) *Cryptocrystalline chondrule from Tieschitz containing dark matrix lump. Picture by J. L.* GOODING, *2.5 mm long, reprinted with permission, from* SCOTT *et al., Geochim. Cosmochim. Acta,* ***48***. (*d*) *Barred olivine chondrule inside porphyritic olivine chondrule in Chainpur USNM 1251-2, 1.6 mm long.* (*e*) *Ferroan olivine precipitated on magnesian olivine porphyry in Allende 3529-33, 1.0 mm long.* (*f*) *Porphyritic olivine chondrule with coarse-grained rim surrounded by fine opaque matrix, Semarkona USNM 1805-4.*

2. Observations and Experiments

2.1. Solid inclusions in chondrules

Although most chondrules were largely melted, some of them contain anomalous inclusions which convey information about the solid precursors of chondrules, about the melting process, and/or about earlier (nebular) processes. Such inclusions are illustrated in Fig. 1. Some chondrules contain subhedral forsteritic olivine (FeO 0.25–1.0%) showing blue cathodoluminescence and overgrown by more ferroan olivine (STEELE, 1986). This forsterite impresses by its large grainsize (100–500 μm), its high concentrations of refractory elements and its complex zonation. Half the chondrules examined in one study (KRING, 1986) contain luminescing forsterite relicts. Many Type I granular or microporphyritic chondrules are composed mainly of olivine which is almost as magnesian as the luminescing forsterite (MCSWEEN, 1977a; SCOTT and TAYLOR, 1983) but has slightly lower concentrations of refractory elements (Ca, Al, Ti) at the same FeO content (JONES and SCOTT, 1989). Even more calcic forsterite occurs in some CAI (WARK *et al.*, 1987).

Ferroan olivine is also found as relicts in chondrules and the contrast between the relict olivine, with reverse Fe-Mg zoning and dusty inclusions of Fe metal, and the other olivine crystals, with normal zoning and a clear appearance strongly suggests that it survived the melting process (NAGAHARA, 1981; RAMBALDI, 1981). Such grains are generally interpreted as relicts, either of primitive precursor materials or of earlier chondrules, although KRACHER *et al.* (1984) caution that some dusty olivine grains could be the result of reduction of chondrules as they passed into regions of gas with lower oxygen fugacity. Pyroxene also occurs as relict grains, but less commonly than olivine (RAMBALDI, 1981; KRACHER *et al.*, 1984). TEM studies of the dusty cores of relict olivine have shown higher dislocation densities than for associated clear olivine, and this deformation is attributed to impact before chondrule formation (WATANABE *et al.*, 1984; RUZICKA, 1989). It has been shown experimentally that the porphyritic class of chondrule textures cannot be produced if the charge is totally melted, implying that some small fraction of ordinary olivine may be relict (NAGAHARA, 1983; HEWINS, 1988).

Chondrules containing smaller chondrules have also been found, in chondrites such as Dhurmsala (TSCHERMAK, 1885) and Chainpur USNM 1251-3 (SCOTT and TAYLOR, 1983), as well as in the present study in Chainpur USNM 1251-2 and Allende USNM 5492. Incorporation of a chondrule requires that the host be mostly liquid whereas, in the better known case of compound chondrules, the chondrules were nearly solid when sintered. Remelting of a small enclosed chondrule could destroy evidence of the capture process, and this may explain clusters of relict grains in a chondrule (KRACHER *et al.*, 1984). WARK and LOVERING (1982) reported textural evidence of incorporation of CAI material into liquid droplets which became type B CAI. Liquid droplet coalescence is a process for which there should be little or no evidence (although it might explain some anomalous grains) which was probably important in the formation of macrochondrules and large type B CAI. Chondrules in chondrules show that some chondrules formed before others, but this could be a matter of minutes in the same chondrule-forming event, or much longer if the chondrule-forming process was repeated.

Lumps of fine-grained opaque chondrule matrix have been reported by SCOTT *et al.* (1984) to occur inside chondrules of all kinds. The compositions of included lumps tend to be the same as those of the chondrule rims but not of the chondrules themselves. The lumps are composed of ferroan olivine, etc., like rims and matrix. These lumps appear to have been incorporated by the chondrules before they had crystallized but have not influenced chondrule crystallization. It is not clear if the matrix-type material could have been deposited in shrinkage cavities within the chondrule, at the time when rims were formed.

2.2. Melting conditions indicated by experiments

Dynamic crystallization experiments on chondrule analogue compositions are generally interpreted in terms of conditions of chondrule cooling, but they yield equally important information on chondrule heating. Chondrule textures can be reasonably reproduced by heating to near-liquidus temperatures, with nonporphyritic textures (radial, barred, cryptocrystalline, glassy) requiring complete melting and porphyritic texture incomplete melting (*e.g.*, LOFGREN and RUSSELL, 1986; LOFGREN, 1989; RADOMSKY and HEWINS, 1987; RADOMSKY, 1988). The heating time during which melting was taking place in experiments has varied from 2 min (TSUCHIYAMA and NAGAHARA, 1981) to 17 h (LOFGREN and RUSSELL, 1986). Longer heating times were used because in phase equilibrium studies they yield more reproducible results; shorter heating times were used because chondrule origins may very well involve a flash heating mechanism. In our laboratory, we have found that 30 min heating at the initial temperature is required to get a reasonable match to natural chondrule textures using finely powdered starting material (RADOMSKY and HEWINS, 1987; BELL, 1986; CONNOLLY *et al.*, 1988). CONNOLLY *et al.* (1988) showed that if the heating time is reduced to 12 min, barred olivine and glassy chondrule analogues are replaced by spheres with transitional porphyritic/"barred" textures, and good porphyritic textures are replaced by finer-grained, more granular textures. With two minute heating times (TSUCHIYAMA and NAGAHARA, 1981), good porphyritic olivine chondrule textures are produced only when initial temperatures are just below the liquidus, and with lower initial temperatures fine granular olivine appears. HEWINS (1988) therefore made the firm conclusion that heating times of 30 min are required to produce the range of textures of natural chondrules, but this conclusion depends on the assumption that chondrule precursors were fine-grained dust. Conditions which might allow very short duration "flash" heating to produce chondrule textures, such as large precursor crystals (like relict grains) are discussed below.

TSUCHIYAMA *et al.* (1981) studied volatile loss from chondrule analogues and it depends on temperature, pressure, oxygen fugacity and liquid composition. They performed isothermal experiments on totally melted droplets and measured Na loss as a function of oxygen fugacity. Results for 1500°C show that more than half the Na_2O is lost in an hour at 10^{-10} (a little above the fayalite-quartz-iron buffer) but essentially none is lost at 10^{-5}. This suggests that chondrules formed under canonical nebular reducing conditions should be free of Na, but one must remember that the results quoted are for isothermal superheated melts. At the liquidus and 1/2 log unit below the iron-wustite buffer, a pyroxene chondrule composition lost only 15% Na in two hours (HEWINS *et al.*, 1981) but an olivine chondrule composition lost more, 47% Na in 1/2 h (RADOMSKY, 1988), because higher liquidus temperatures promote greater Na loss. Partially melted chondrules cooling rapidly would tend to suffer less from volatile loss, and short melting times suggested below would tend to alleviate the problem of preservation of Na in chondrules. The Na contents of porphyritic chondrules are slightly higher than those of nonporphyritic (GOODING *et al.*, 1980), suggesting that degree of melting (initial temperature) does have some affect. Smaller chondrules should lose more Na than large ones, but there is no simple correlation between size and Na content (DODD, 1978a, b; HEWINS, 1983), suggesting that chondrule Na losses are generally minor. Total gas pressure, which also influences volatilization, is unknown during the chondrule-forming process. Oxygen fugacities were considerably above nebular, from the generally high levels of FeO in all kinds of chondrules, as discussed below, but not enough alone to keep Na/Al at their bulk rock levels (GROSSMAN *et al.*, 1988). Short times at peak temperatures, partly aided by moderately high oxygen fugacities, are the most obvious way in which to reduce Na loss relative to experiments. High Na or NaCl pressure (IKEDA and KIMURA, 1985) would also prevent Na loss.

2.3. *Melting conditions based on chondrule calculations*

In dynamic crystallization experiments, textures are controlled by the number of heterogeneous nuclei, which is related to the difference between initial heating temperature and true liquidus temperature. With a fixed composition, then, the texture is determined by initial temperature (*e.g.*, RADOMSKY and HEWINS, 1987), but with a fixed initial temperature, the full range of chondrule textures can be obtained by changing the bulk composition, thus varying the liquidus temperature and degree of melting (CONNOLLY *et al.*, 1988). At first sight it would appear that both effects could influence natural chondrules equally, but there are correlations between chondrule textures and composition. The most obvious correlation is that magnesian olivine chondrules form finer-grained, more granular textures (Type I), *i.e.* are less completely melted, compared to ferroan Type II microporphyritic chondrules (MCSWEEN, 1977a; SCOTT and TAYLOR, 1983). This suggests that natural chondrule textures are controlled more by composition changes than by changes in initial or heating temperature.

RADOMSKY and HEWINS (1988) calculated liquidus temperatures for a suite of chondrules for which both bulk compositions and textures were available. Such temperatures would be in error if the chondrules had suffered significant change in bulk composition since they were melted, *e.g.* by Na loss. The influence of Na loss is very small for liquids with low Na/Mg ratios, like chondrules (HERZBERG, 1979). The temperatures, which range from about 1750 to about 1250°C, are a complex function of bulk composition but define a smooth curve when plotted against wt% ($SiO_2+Al_2O_3-MgO$). Nonporphyritic chondrules tend to plot at the low-temperature end of this liquidus, because their compositions are not refractory, and porphyritic chondrules are more abundant towards the high temperature end (RADOMSKY and HEWINS, 1988). This is illustrated in histogram form in Fig. 2, where number of

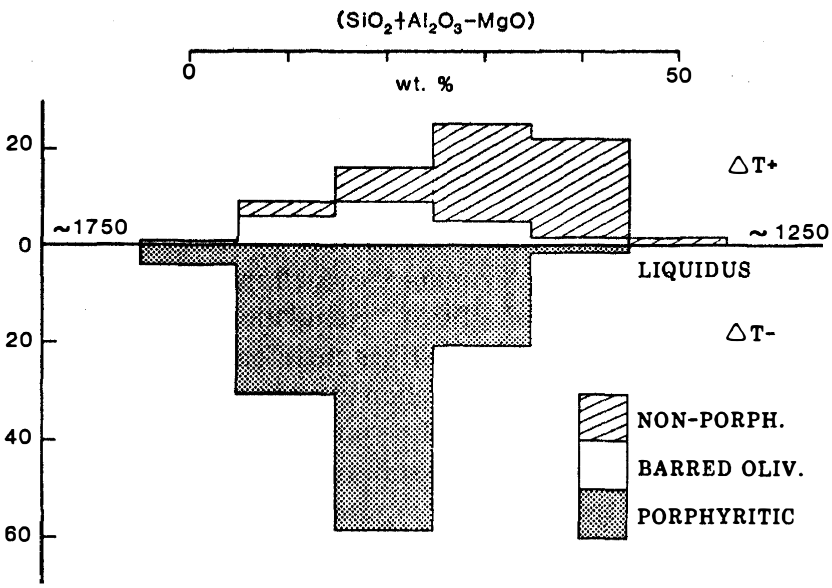

Fig. 2. Histogram showing numbers of porphyritic, barred olivine and non-porphyritic chondrules as a function of wt% ($SiO_2+Al_2O_3-MgO$) which corresponds to a range of liquidus temperatures from about 1750 to 1250°C. Chondrules with high liquidus temperatures tend to be partly melted (porphyritic) but those with low liquidus temperatures tend to be completely melted.

chondrules is plotted against the composition parameter describing the liquidus curve. Totally melted chondrules are plotted above the line, and incompletely melted ones below the line, for clarity. Figure 2 shows clearly that refractory chondrules tend to be incompletely melted, whereas many fewer nonrefractory ones are incompletely melted. RADOMSKY and HEWINS (1988) suggested that as very few chondrules with liquidus temperatures over 1700°C have textures indicative of total melting, 1700°C represents the temperature limit in the chondrule-forming event, and it happens to be a value permitting total melting of non-refractory chondrules but not of the most Mg-rich ones. Although some chondrules reached this temperature clearly not all did. Based on the ease with which superheated chondrule melts produce glass in our laboratory, temperatures approaching 1700°C should cause a much higher fraction of the nonporphyritic chondrules to be glassy than actually observed. If the starting materials were coarser-grained, as discussed below, more nuclei would survive and the textures would tend to be radial or barred rather than glassy. In any case, temperatures for some chondrules reached 1700°C, for a duration say of minutes, but perhaps not as long as 30 min because then more than half the Na would have been lost.

2.4. *Olivine zoning and chondrule crystallization rates*

The textures of chondrules are well described and understood (MCSWEEN, 1977a; SCOTT and TAYLOR, 1983; etc.); despite the presence of some relict solids, most of the olivine and pyroxene crystals grew from melt. Four kinds of olivine zoning behavior have been recognized (MIYAMOTO *et al.*, 1986). In Type A olivine, the zoning is concentric and Ca increases from core to rim with other weakly incompatible elements like Fe, a pattern diagnostic of crystallization from the melt. Type B olivine, with patchy zoning and Fe increasing but Ca decreasing to the rim, has probably been reequilibrated in the solid state. Types C and D olivine, are magnesian and ferroan, respectively, with little zoning and it is not clear whether they were equilibrated during initial cooling or subsequently. In partially equilibrated chondrites it is clear that some chondrule olivine became equilibrated either by slow cooling or was modified by sub-solidus reactions, but type A zoning shows all the characteristics of crystallization from a melt (Miyamoto *et al.*, 1986). Simulation of type A zoning (together with reproduction of appropriate textures) has therefore been the basis of estimation of chondrule cooling rates from crystallization experiments.

Recent discussions (Hewins, 1988; Lofgren, 1989; Radomsky, 1988) of chondrule cooling rates, based on dynamic crystallization studies, are in reasonable agreement and their arguments are therefore only summarized briefly here. Low cooling rates are ruled out for Type II porphyritic olivine chondrules which show strong normal zoning, type A zoning of Miyamoto *et al.* (1986). The textures of these chondrules can be reproduced at low cooling rates (10°/h), but not the zoning or their apparently anomalous Fe-Mg partition coefficients. At cooling rates of 100°/h, and higher, zoning is present and the zoning near the olivine rim is too steep to be resolved with the electron microprobe, resulting in apparent partition coefficients less than the equilibrium value (Radomsky, 1988) as observed in chondrules. For most chondrules with igneous composition patterns, cooling rates were within the range 100–2000°/h. Textures ranging from glassy to granular can all be produced at the same cooling rate (Connolly *et al.*, 1988; Radomsky, 1988). Cooling rates producing the most satisfactory barred olivine textures are 500–1000°C/h (Radomsky, 1988).

2.5. *Chondrule modification before accretion*

Chondrules suffered a number of physical and chemical changes, such as fragmentation and Fe-Mg exchange, before they were accreted into a parent body. Droplets may have splashed into microchondrules, coated or engulfed other chondrules, or coalesced. There are suggestions that other modifications began during chondrule crystallization. Asymmetrical Fe-Mg zoning in olivine in chondrules in Chainpur (LL3.4) has been explained by metasomatism, exchange of Mg by Fe, during crystallization (Ruzicka, 1988). This requires more oxidized material (gas or solid) to react with the droplets. Crystallization of very magnesian pyroxene after the formation of Fe-bearing olivine suggested to Kring (1986) that some UOC and C3 chondrules experienced more reducing environments late in their solidification history. This might also be explained as more rapid diffusion of Fe into olivine than pyroxene, although Ruzicka (1988) showed that olivine enclosed by pyroxene tends to be protected from modification. Watanabe *et al.* (1986) reported oscillatory Fe-Mg zoning in L3 chondrule pyroxene, despite continuously increasing Ca, and interpreted it as due to fluctuations in the redox conditions.

Several kinds of olivine-rich rims have formed on chondrules (Fig. 1). Both very fine-grained opaque matrix-like material and a similar but slightly coarser-grained variety apparently formed by heating such material occur as chondrule rims, sometimes with the fine material coating the coarser rim (Rubin, 1984). The main minerals in these rims are olivine, pyroxene, metal and sulfide. Ferroan olivine also forms rims on Allende chondrules, but these are overgrowths continuous with the chondrule olivine crystals and are marked with local high concentrations of refractory elements such as Cr and Al (Peck and Wood, 1987).

Abrasion and fracturing of chondrules changed the shape of chondrules both before (Rubin, 1984) and after rim formation (Kitamura and Watanabe, 1986; Peck and Wood, 1987). Dodd (1971) suggested a correlation between chondrule shape and composition for Sharps (H-3) with the roundest particles being the most magnesian. Wlotzka (1983) confirmed this relationship, which he interpreted as enrichment in FeO before agglomeration and during fragmentation, either in the nebula or on the parent body surface. Some chondrules also experienced Na metasomatism (Ikeda and Kimura, 1985) although this did not occur generally (Grossman and Wasson, 1983). Changes to chondrules must be discussed in the context of the rapid nature of melting and cooling, and the variety of anomalous inclusions. This is attempted in the next section, which leads to new conclusions about the relation of the chondrule-forming process to the nebular environment.

3. Discussion of the Chondrule-forming Environment

3.1. *Evidence for condensation*

Chondrules contain a great diversity of inclusions, as described above: Mg- and Fe-rich (olivine), fine-grained and coarse-grained (matrix lumps, olivine), solid and melted (forsterite, chondrules). The anomalous inclusions may represent chondrule precursor material, at least in part, but their diversity indicates that a variety of events must have taken place before chondrule formation, although the sequence of events is not recorded in the inclusions themselves.

There are two types of process, oxidation-reduction and evaporation-condensation, which might be responsible for the existence of both forsterite and ferroan olivine. Both reduction, in the form of metal-bearing relict olivine grains, and (late) oxidation, in the form of addition of fayalite component to chondrules (*e.g.*, Wlotzka, 1983; Peck and Wood, 1987; Ruzicka, 1988) have been observed. However, differences in minor element concentrations (*e.g.* Ca *vs.* Mn) show that the forsterite is not simply the reduced equivalent of chondrule ferroan olivine. A condensation origin for the forsterite, as argued by Steele (1986) on the basis of refractory minor element concentrations, is therefore supported. It is clear that most particles in chondrites are not condensates, but rapidly melted and cooled solids, although their precursors, *e.g.* bulk chondrule or CAI material, or specific relicts such as forsterite, could be condensates. Forsterite relicts might alternatively be evaporation residues, in which case they might be the carriers of oxygen isotope anomalies.

The condensation of forsterite is unlikely to have occurred as in the canonical equilibrium condensation model of Grossman (1972). Only very recently has an astrophysical model appeared (Boss, 1988) which produces temperatures nearly high enough for extensive evaporation. This work assesses the effect of transport of angular momentum by gravitational torques through the nebular disk. The heating is due to compressional heating infalling gas and is nonaxisymmetric. As yet, the temperatures do not appear high enough to produce phases condensing at higher temperatures than forsterite and the preservation of isotopic anomalies in CAI suggests they are evaporative residues rather than condensates. Chondrules and CAI have such high cooling rates (Hewins, 1988) that their origin appears local rather than nebula-wide. However, in the Boss model, particles can achieve high cooling rates by moving rapidly relative to the gas and moving laterally or vertically away from hot dense regions. Current nebular and condensation models also involve lower pressures and higher oxygen fugacities than the canonical conditions (*e.g.*, Wood and Morfill, 1988; Wood and Hashimoto, 1988), as do recent condensation experiments (*e.g.*, Mysen and Kushiro, 1988; Nagahara *et al.*, 1988). The model of Boss (1988) would permit both low and high pressures relatively close, suitable for evaporation-condensation and preservation of Na in melt droplets, respectively.

Calculations for $P=10^{-5}$ atm and the solid/gas ratio increased so as to give O/H 41X the cosmic ratio (Wood and Hashimoto, 1988) yield higher Fa contents in forsterite at a given temperature than in the classic condensation model (Grossman, 1972). The lowest Fa contents in relict forsterite, which appear to be due to solid-vapor equilibrium rather than reduction of ferroan olivine (Steele, 1986), are close to the value calculated for 1400 K (Wood and Hashimoto, 1988). In equilibrium calculations, the olivine reacts to become increasingly ferroan as temperature falls, during the formation of pyroxene, but condensation experiments have shown fractional crystallization: Nagahara *et al.* (1988) report the sequence forsterite, enstatite, tridymite, intermediate pyroxene, iron-rich olivine and metallic iron. The presence of forsterite relicts in chondrules, whose precursors also presumably contained ferroan olivine, and of fine ferroan olivine crystals in matrix and included lumps (Scott *et al.*, 1984; Peck and Wood, 1987) suggests that fractional condensation was an important process before and/or during chondrule formation.

Condensation obviously occurred before chondrule formation, to supply relict forsterite, but may also have been occurring during and after chondrule formation, based on chondrule modification and matrix formation. Rims on chondrules, enclosing matrix and included matrix lumps are similar and may all have formed in part from condensation. The sequence of minerals in composite fine matrix particles is consistent with the matrix forming by condensation (Nagahara, 1984; Nagahara, *et al.*, 1988). Some rims may result from fine condensate grains, which might otherwise have become matrix, being sintered to chondrules at high temperatures. However, as some chondrules were cooled and fractured before the rims were deposited (Rubin, 1984), either chondrules migrated between hot zones or the heating was episodic. A different style of interaction with nebular gases has apparently affected type I chondrules in Allende. Ferroan olivine formed in and on magnesian olivine by diffusion along cracks and by condensation on the outside (Peck and Wood, 1987; Hua *et al.*, 1988). Kring and Wood (1987) argued that some chondrules experienced repeated high temperature conditions during which rim minerals condensed out of the gas. Ferroan olivine is an important mineral in chondrule rims, as it is in chondrite matrix, suggesting a high oxygen fugacity for condensation (Kring and Wood, 1987) although rare outer magnesian rims suggest a possible return to more reducing conditions.

3.2. Precursor history

Composition data for chondrules show no simple patterns for variations in elements such as Mg, Fe, Si, Na (*e.g.*, DODD, 1978a, b) and chemical differences between chondrules cannot be explained as differing extents of equilibrium condensation or a simple fractional condensation process recorded by their precursors. Chondrule precursors appear to have contained random proportions of nebular components (GROSSMAN and WASSON, 1983). Statistical analysis of chondrule composition data reveals a number of chemical precursor components, which have compositions influenced by volatility but corresponding to common solid phases, and which differ somewhat between chondrite groups (GROSSMAN, 1988).

How were the several components combined in random abundances for individual chondrules? Although some olivine in chondrites has been modified, the compositions of relict forsterite and unequilibrated matrix grains appear primary (NAGAHARA, 1984; STEELE, 1986). The sequence of primary olivine compositions (Mg-rich and Fe-rich) found in chondrites suggests fractional condensation, as achieved in hours on a laboratory-scale by NAGAHARA *et al.* (1988) rather than nebula-wide equilibrium condensation. The general evidence of high oxidation states also suggests a localized environment, such as a particle-rich zone or clump (*cf.*, WOOD, 1984, 1986). In laboratory experiments, fractional condensation takes place in a steep temperature gradient along the support wire as vapor escapes from the capsule (NAGAHARA *et al.*, 1988). In nature, condensates would initially nucleate on refractory residues (or possibly at some distance on little heated material). In a nebular clump condensation might occur over time to produce concentrically zoned uniform particles. Turbulence or lateral motion is expected in recent nebular models (WOOD, 1986; WOOD and MORFILL, 1988; BOSS, 1988) which could permit transport of early condensates in a haphazard way so that they intermittently became substrates for continued condensation (precipitation or reaction) and/or aggregation of randomly sized particles formed at different stages in the condensation sequence. A somewhat similar environment has been postulated for condensation and accretion of fine-grained chondrite matrix (NAGAHARA, 1984). Whether this was a single or repeated process, the nature of fractional condensation indicates that chondrule precursors formed in this way would inevitably be chemically heterogeneous.

Some data sets show hints of systematic variation in chondrule bulk chemistry. Chondrule bulk compositions (MCSWEEN, 1977b) suggest two processes modifying initial forsterite-rich chondrules: types I and III define a trend suggesting addition of SiO_2 and types I and II suggest addition of Fa to forsterite (Fig. 3). It is not clear to what extent the FeO or Fa addition was early, affecting precursor compositions, or late, affecting chondrules (WLOTZKA, 1983; PECK and WOOD, 1987; RUZICKA, 1988), but the fact that types I and II PO chondrules can be distinguished by texture (MCSWEEN, 1977a; SCOTT and TAYLOR, 1983) suggests that at least part of the process was early. This data set contains some ordinary chondrite chondrules but is dominated by carbonaceous chondrite chondrules. For UOC chondrules alone, the trends are less well defined. The existence of the two trends suggests that fractional

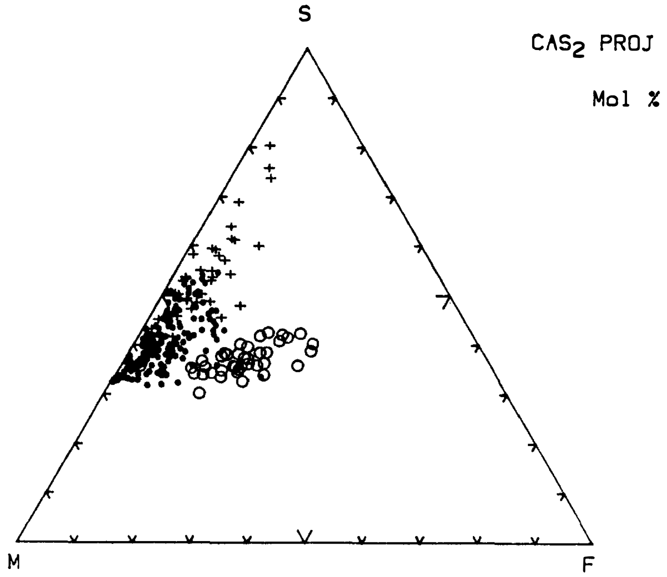

Fig. 3. Chondrule compositions projected from anorthite onto the plane SiO_2-MgO-FeO: type I (dots), II (circles) and III (plus signs) chondrules of McSween *(1977). The trends from forsterite towards silica and towards fayalite suggest precursors formed by condensation at low and high oxygen fugacity, respectively.*

condensation at least at the CC locations occurred at both high and low oxygen fugacities, with forsterite primarily reacting to enstatite at low fO_2 (as at the EC location) but primarily to ferroan olivine at high fO_2. Some support for this argument can be found in the magnesian porphyritic olivine-pyroxene chondrules if the pyroxene crystallized later and at lower oxygen fugacities than the olivine as proposed by Kring (1986). This suggests that reducing conditions were intensified after chondrule melting. Any detailed model for condensation must explain the range of redox conditions: this is attempted in section 3.4.

One particular aspect of the anomalous inclusions in chondrules poses a particular challenge to models for their origin in the nebula. Relict ferroan olivine, whether of condensate or early chondrule origin, contains a high density of dislocations due to impact (Watanabe *et al.*, 1984; Ruzicka, 1988). Many arguments have been raised against an impact origin for chondrules, as summarized by Taylor *et al.* (1983), but most of these considered regoliths on asteroid-sized bodies. Some of the physical problems may be overcome, given a small parent body, as 1–10 km weak planetesimals have trivial escape velocities and retain only part of their impact ejecta (Housen *et al.*, 1979). The higher energy ejecta, *i.e.* melt droplets, would tend to travel further and be separated from fragments, so as not to form an agglutinate-rich breccia. Slow crystallization (relative to radiative quenching) and nebular modification of chondrules might take place in a thick enough ejecta cloud, which would be either a transient atmosphere to the target body or a detached nebular domain. Some chemical and isotopic problems, such as explaining oxygen isotope data for chondrules, matrix and CAI, remain. Problems such as these might be overcome in a nebular impact model, but is hard to obtain high relative velocities for small particles. Infall of interstellar dust grains at high velocity has been discussed by Wood (1986) and Cassen and Boss (1988). Impact of these tiny grains on a nebular particle-rich zone could possibly cause the shocked relict olivine, although the energy would probably not be enough to melt chondrule droplets. The formation of the chondrules thus remains a nebular problem and non-impact heating processes remain to be clarified.

3.3. Heating and cooling conditions

Although simulation of chondrule textures has required quite long heating times (about 30 min), this may not rule out transient heating mechanisms, *e.g.* these involving electrical discharge. All dynamic crystallization experiments, except some of Radomsky's (1988), were performed with finely powdered starting materials. This approach is consistent with the widespread assumption that chondrule precursors were dustball aggregates of interstellar grains or fine-grained nebular condensates similar to matrix and rim material, which appears to be partly justified based on the conclusions of Scott *et al.* (1984) and Matsunami (1984). However, relict olivine grains and even other chondrules appear to be more abundant as inclusions in chondrules than matrix lumps. Even allowing for the fact that coarse-grained material is harder to melt than fine-grained, and therefore more likely to survive than matrix lumps, it appears possible that a significant fraction of chondrule precursors consisted of coarse-grained material, either of nebular condensate or early chondrule origin.

Systematic dynamic crystallization experiments have yet to be conducted with coarse-grained (say 100 μm) precursors, but the results may be predicted based on runs described by Radomsky (1988) which were seeded with large olivine grains. Where the liquidus was not exceeded, abundant olivine grew on separate nuclei, not directly on the relict seed, forming a microporphyritic texture; where the liquidus was exceeded and nuclei thoroughly destroyed, new olivine overgrew the unmelted seed olivine forming barred texture. With coarse starting material heated beyond the liquidus for a very short time, it is expected that numerous olivine relicts would be overgrown during cooling to produce a porphyritic texture. Such experiments are currently being carried out. Of course, one cannot predict exactly what combination of temperature, time and grain size will produce satisfactory textural matches to chondrules. It appears, however, that if chondrule precursors were fine-grained, extended heating was required during the melting event but, if the precursors were coarse-grained, chondrules may have been generated by a flash-heating mechanism.

The Wood group has long been concerned with the oxygen fugacities indicated by iron silicate compositions in chondrules. Recent calculations have shown chondrules formed at oxygen fugacities up to the iron-quartz-fayalite buffer, with very few data approaching canonical nebular conditions (Wood, 1984; Johnson, 1986; Kring, 1986). Similar results were obtained by Nagahara (1986). Oxygen fugacities for type II porphyritic olivine chondrules are lower than those used in isothermal Na loss experiments (Tsuchiyama *et al.*, 1981) and similar to those of Radomsky (1988) in runs which lost about half the initial Na content during cooling. Most olivine in rims and matrix is more ferroan, or oxidized, than chondrule olivine. Magnetite-bearing Allende chondrules experienced conditions up to 10 orders of magnitude more oxidizing than a gas of solar composition (McMahon and Haggerty, 1980; Wood, 1984), as did Allende hibonite and melilite (IHINGER and STOLPER, 1986; KOZUL *et al.*, 1988), but during subsolidus alteration rather than melt crystallization. Some chondrules too experienced subsolidus Na metasomatism (IKEDA and KIMURA, 1985). The chondrule environment then was oxidizing both during melting and after solidification, most probably as the result of extensive evaporation in a particle-rich clump (WOOD, 1984; NAGAHARA *et al.*, 1988; DAVIS and MACPHERSON, 1988).

Chondrule crystallization experiments have shown moderately high cooling rates, the significance of which has been emphasized in earlier reports (*e.g.*, HEWINS *et al.*, 1981; HEWINS, 1983). Cooling over a few hours so as to quench glass and retain volatiles requires that the ambient temperature was fairly low and that radiative cooling was retarded by the absorption of heat by other nearby particles, presumably other chondrules and dust (presolar and condensate). These particles must have been hot to retard radiative cooling. Both cooling rate data and petrographic observations (compound chondrules, etc.) suggest chondrule formation in a thick cloud, with many chondrules formed simultaneously and possibly in succession. Only the most magnesian silicates in chondrules indicate conditions approaching those of the canonical nebula (WOOD, 1984; JOHNSON, 1986). This shows generally oxidizing conditions in the cloud, but the crystallization trend towards late magnesian silicates in some chondrules could be due to a change to more reducing conditions, *i.e.* a larger fraction of hydrogen in the gas. Taken together, these observations suggest migration of chondrule droplets during crystallization (a time of about one hour) between regions of different oxygen fugacity, or production of oxygen by local evaporation with subsequent dispersal, or both. This migration is comparable to the turbulent motion required for the precursors to obtain random proportions of condensate components by collisions. If the chondrules are not significantly more resistant than precursor dust to convective motion, they would tend to remain trapped in a particular parcel of gas. Redox change must then be due to H_2-O_2 diffusion.

Chondrules were fragmented, both before and after rim condensation, and spherules are more magnesian than chondrule fragments (WLOTZKA, 1983). If Fe-enrichment occurred on a parent body, one must postulate that some chondrules are resistant to modification (SEARS *et al.*, 1984) but no mechanism has yet been verified. In a nebular setting, one can imagine magnesian chondrules colliding in a warm oxidized gas, produced in a dust-rich clump, in equilibrium with the ferroan olivine deposited in chondrule rims. The surviving magnesian spheres could have been generated very late in the chondrule-forming events (if there were in fact several episodes) or far from the center of the clump where the gas pressures, oxygen fugacities and particle densities were lower. Early or centrally formed chondrules would have had an opportunity to become Fe-rich (oxidized) as they collided in the densest part of the gas. (In impact models, a similar scenario could be invoked in a transient extended atmosphere.) Whether magnesian chondrules originated late in time or far from the center of the clump, it appears possible that accretion was heterogeneous, ending with unequilibrated material with abundant magnesian spheres. Accretion while some particles were still warm would not be ruled out in this model.

The dual nature of ferroan chondrules, types A and D of MIYAMOTO *et al.* (1986), is not considered in this scenario. Type A has igneous zoning and is abundant in the most unequilibrated chondrites, whereas type D was equilibrated either in the nebula or in a parent body. Thus the Fe enrichment of fragmentary chondrules could have happened before or after crystallization. The meteorites studied by Wlotzka (1983) may be partially equilibrated (Sharps, H3.4; Tieschitz, H3.6) and then presumably contain both kinds, as do the 3.6 and 3.7 chondrites studied by Miyamoto (Sears *et al.*, 1982). It is therefore likely that the more ferroan chondrules contain partly (type B) or completely reequilibrated (type D) olivine. However, if unequilibrated type A olivine is dominant, as it might be in Sharps (H3.4), then one would have to argue that the FeO enrichment occurred to the precursors before the chondrules were formed, because of high oxygen fugacity at the center of a dust-rich clump, and that the fragmentation happened later because of the high chondrule density there. This important distinction could readily be resolved by imaging olivine chondrules with back-scattered electrons. Furthermore, if early Fe enrichment were proven, then any associated type B olivine zoning would very likely be nebular rather than parent body. If late Fe enrichment were the rule, this would suggest the chondrules stayed hot after cooling through the crystallization interval.

3.4. Varying nebular conditions to make chondrules

Chondrule models must deal with condensation continuing from before to after chondrule formation, the random nature of precursor compositions, the change of chondrules' environment from reducing to more oxidizing (and sometimes vice-versa), and the general character of the environment rich in volatilized components, such as O_2, FeO, Na_2O, and particles. Turbulent migration of precursor grains and gas movement relative to chondrules seem to be required. Chondrule generation in a thick gas-dust clump has been considered by many others (*e.g.*, Hewins *et al.*, 1981; Wood, 1984, 1986) and above. Let us consider whether this approach will accommodate the apparent simultaneous formation of chondrules and condensation, and redox data.

Heating apparently melted some particles but evaporated others at the same time, since condensation overlapped chondrule formation. Recent evaporation experiments (Mysen and Kushiro, 1988) have revealed the stability relationships

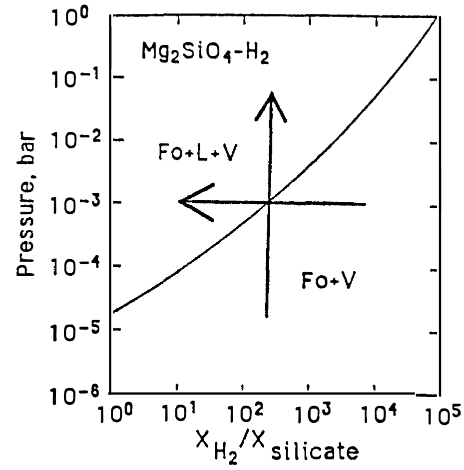

Fig. 4. The system forsterite-hydrogen with stability fields from Mysen and Kushiro (1988). The arrows show that melting can be achieved at constant temperature by increasing the gas pressure or the concentration of silicate in the gas.

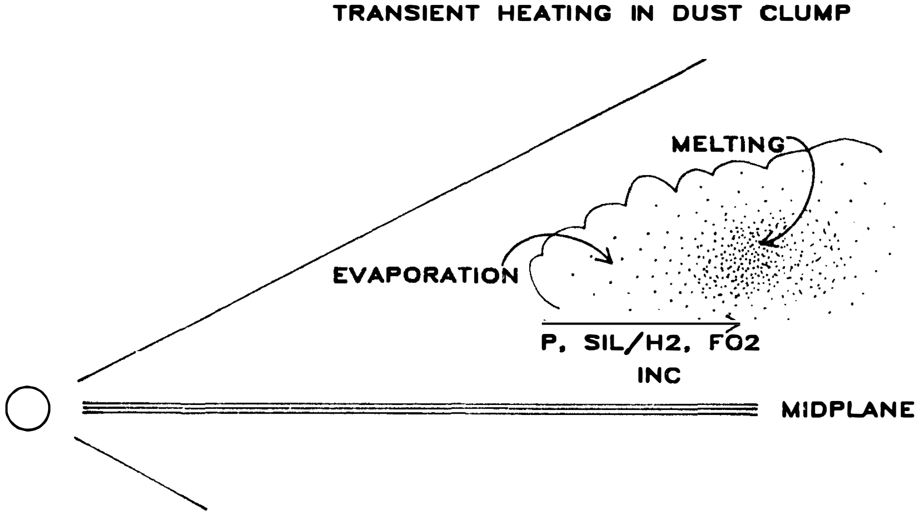

Fig. 5. *Heating of particle-rich clumps would produce gradients in gas pressure, silicate concentration and oxygen fugacity. Hence evaporation and condensation would take place at the edge of a clump and melting and crystallization would occur in the center. Migration due to turbulence and gas diffusion would cause random mixtures of condensates in precursors, and changes in oxygen fugacity recorded by chondrules.*

between solid, vapor and liquid in the system Mg_2SiO_4-H_2. Their pressure-composition (hydrogen/silicate ratio) diagram shows that melting may be achieved in two ways, by raising the total gas pressure or changing the composition, *i.e.* decreasing the silicate fraction in the gas (Fig. 4). In the case of particle-rich zones in the nebula, whether the mid-plane, an infalling clump or a turbulent shear zone beneath the nebula surface, one would expect lowest total pressures and silicate fractions in the gas on the outside where the density of particles, however heated, is lowest. One would therefore expect evaporation-condensation at the outside of a particle-rich zone, and melting-crystallization on the inside, in response to the pressure and composition gradients. This is shown schematically in Fig. 5, for a particle-rich clump arbitrarily located between the midplane and nebular surface. Because the thermal history involves only a few hours, a transient heating event rather than a nebula-wide event is imagined, although Boss (1988) has proposed a nonsymmetric disc with bars or spirals in which particles could move out of hot regions within a few hours. The dense clump could be as small as 1000 km, if high particle velocities relative to the gas are realistic (Boss, 1988).

In evaporation experiments, an initial pure hydrogen, low pressure atmosphere is rapidly converted to one where oxygen fugacities are near fayalite-quartz-iron (Nagahara *et al.*, 1988). The center of a particle-rich clump would develop a high-pressure gas rich in volatiles and especially O_2 and FeO as particles were heated. Thus any material processed in a particle-rich clump would tend to become oxidized relative to canonical solar values. Palme and Fegley (1987) performed calculations which showed that forsteritic olivine will tend to form ferroan rims by high temperature oxidization as it reacts with such a high-oxygen-fugacity gas. FeO addition to chondrules is therefore a natural consequence of heating a thick clump of precursor material and analogous oxidations in CAI (Fegley and Palme, 1985; Davis and

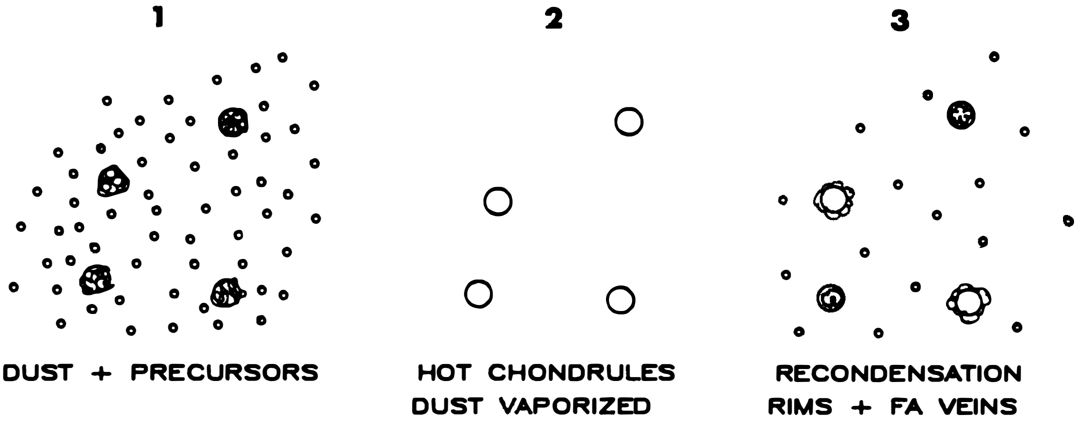

Fig. 6. Chondrule rims might be the result of condensation after the vaporization of dust.

Macpherson, 1988; Kozul *et al.*, 1988) are noted above. Precipitation of other volatile components such as Na is common in altered CAI and happens rarely in chondrules (Ikeda and Kimura, 1985).

The role of gas pressure in allowing either melting or evaporation has implications considering the precursors of chondrules. If the precursors to chondrules were assembled from condensates, the chondrule-forming environment must have changed over time. It was initially at low enough pressures for evaporation-condensation, and more material arrived and was heated until pressures became high enough for melting of randomly assembled precursors. Alternatively if the earliest chondrules were made from fine interstellar dust, the currently observed chondrules with heterogeneous bulk compositions and large relict grains were produced after one or more cycles of migration in and out of the melting environment.

Chondrules clearly collided with each other in the clump before and after the rim condensation process. High particle densities and high Fe-concentrations go naturally together in this scenario but what is the specific source of ferroan olivine? The late condensates could represent material lost when chondrules were heated, but such losses were apparently minor. The condensation process, producing forsterite relicts, could have been rapid but localized, and could have continued after chondrule formation. Alternatively, if the transient heating process operated on isolated precursor aggregates and not on all dust particles, radiation from chondrules might heat and partially evaporate dust. This might be the source of the rim deposits (Fig. 6). The evaporation of such (insulating) dust might control the cooling of chondrules by steadily increasing the transparency of the clump to infrared (Wood, 1986).

Any discussion of nebular processes which generate chondrules must also consider how other nebular particles, such as calcium-aluminum-rich inclusions, formed. CAI in this model represent refractory residues (or condensates) formed by extreme evaporation in dust-poor regions at canonical oxygen fugacities. Depletions in Mo and W (Fegley and Palme, 1985) may represent residual metal nuggets equilibrated in a thick gas which dissipated before CAI droplets produced fassaite. The CAI suffered not only melting, but in some cases recrystallization and/or low temperature alteration. A Vigarano CAI experienced a volatilization event rapid enough for the evolved gas to convert Ti^{3+} to Ti^{4+} in fassaite (Davis and Macpherson, 1988).

Allende CAI, like Allende chondrules, experienced nebular oxidation at moderately high temperatures (KOZUL *et al.*, 1988). The unequal distribution of CAI among chondrite groups suggests that most chondritic material was processed in particle-rich clumps.

4. Conclusion

The above review of chondrule evolution is consistent with a number of conclusions on how and where chondrules may have formed.

(a) Forsterite and ferroan olivine that occurred in chondrule precursors represent at least in part condensates (or annealed condensates) formed by fractional, rather than equilibrium, condensation.

(b) Precursors appear to be random mixtures representing various temperature segments of a fractional condensation sequence, plus in some cases material from other chondrules. Turbulent motion in the gas is indicated.

(c) Precursors were heated on a time scale of minutes, for the most part near the range of liquidus temperatures, up to a limit of 1700°C. Chondrules cooled over a matter of hours. A transient heating mechanism in an opaque cloud (particle-rich clump) is indicated.

(d) For the most part chondrules experienced much higher oxygen fugacities than the canonical solar nebula value, which stabilized the high FeO and Na_2O contents of chondrules. The high oxygen fugacities are a result of extensive evaporation.

(e) The trend from forsterite relicts to magnesian chondrules to ferroan rims suggests generally more oxidizing conditions with passing time, although this is also a natural sequence in fractional condensation. Many chondrules, however, apparently experienced more reducing environments as they crystallized producing late magnesian silicates. Migration out of dense clumps, or dissipation of the gas, would explain this.

(f) Chondrules may have been open to Fe-Mg exchange during crystallization, and certainly experienced late condensation, either direct precipitation or sintering of ferroan olivine onto the chondrule exterior. This reflects the end of the condensation that produced precursors or evaporation of nearby dust when chondrules were formed.

(g) The FeO enrichment of chondrules that appears to have accompanied collision and fragmentation may also have been a condensation/metasomatism phenomenon. Both the chemical and physical changes are natural consequences of cooling in a thick clump.

(h) Chondrule melting closely allied to evaporation and condensation melting can be understood as a function of pressure gradients as gas is evolved in a particle-rich clump. Higher pressure promotes melting rather than evaporation.

(i) Rim and matrix condensation represents either the end of precursor formation or the results of evaporation of dust by heat from the chondrule forming mechanism or from molten chondrules.

(j) The heating mechanism can be better understood if the possible locations of the particle-rich clump or in other words the dust enrichment mechanism can be better defined. These locations are immediately beneath the nebular surface, at the mid-plane or in between.

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I appreciate many discussions with friends of chondrules over the last decade. While the debt to those who have written on chondrule origins should be obvious, it is more important to acknowledge all those who have painstakingly recorded all the awkward facts about chondrules which are so hard to explain in our simple models. I apologize to those whose data are not adequately dealt with, and hope this effort will at least spur further critical thinking about the problem. Two figures were kindly made available by J. L. GOODING and I. M. STEELE. The invaluable critical comments of two anonymous referees led to considerable clarifications in the arguments advanced in the paper. This work was supported by NASA grants NSG-7327 and NAG 9-35.

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