Chondrules: Precursors and interactions with the nebular gas

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Abstract–Chondrule compositions suggest either ferroan precursors and evaporation, or magnesian precursors and condensation. Type I chondrule precursors include granoblastic olivine aggregates (planetary or nebular) and fine-grained (dustball) precursors. In carbonaceous chondrites, type I chondrule precursors were S-free, while type II chondrules have higher Fe/Mn than in ordinary chondrites. Many type II chondrules contain diverse forsteritic relics, consistent with polymict dustball precursors. The relationship between finer and coarser grained type I chondrules in ordinary chondrites suggests more evaporation from more highly melted chondrules. Fe metal in type I, and Na and S in type II chondrules indicate high partial pressures in ambient gas, as they are rapidly evaporated at canonical conditions. The occurrence of metal, sulfide, or low-Ca pyroxene on chondrule rims suggests (re)condensation. In Semarkona type II chondrules, Na-rich olivine cores, Na-poor melt inclusions, and Na-rich mesostases suggest evaporation followed by recondensation. Type II chondrules have correlated FeO and MnO, consistent with condensation onto forsteritic precursors, but with different ratios in carbonaceous chondrites and ordinary chondrites, indicating different redox history. The high partial pressures of lithophile elements require large dense clouds, either clumps in the protoplanetary disk, impact plumes, or bow shocks around protoplanets. In ordinary chondrites, clusters of type I and type II chondrules indicate high number densities and their similar oxygen isotopic compositions suggest recycling together. In carbonaceous chondrites, the much less abundant type II chondrules were probably added late to batches of type I chondrules from different O isotopic reservoirs.

INTRODUCTION

Chondrules are silicate particles with igneous textures found in primitive meteorites (chondrites), accreted in the protoplanetary disk. Their study began well over 100 yr ago (e.g., Tschermak 1885) and has been intensive in the 40 yr since the Apollo-Allende era, when meteoritics-cosmochemistry became a mature science. Interpretations of chondrules' origins are still controversial, however, particularly since many chondrules post-date both calcium-aluminum-rich inclusion (CAI) and the earliest differentiated bodies (Kleine et al. 2005, 2009; Halliday and Kleine 2006). The early large-scale migration of protoplanets and non-accretionary collisions recently documented (Asphaug et al. 2006; Bottke et al. 2006; Walsh et al. 2011) place chondrule formation in a more complex environment than a quiescent accretion disk. The abundance of chondrites indicates widespread melting in some environment(s) in the protoplanetary disk, before the accretion of chondrules into their parent bodies. Definition of the formation conditions of chondrules therefore throws light on astrophysical or planetary processes, or both, occurring in the first few million years in the early solar system. In particular, it is unlikely that chondrule melts could behave as closed systems in low pressure nebular gases (e.g., Sears et al. 1996), whereas this is more likely in planetary settings. Here, we consider evidence for the nature of chondrule precursors; how chondrules interacted with ambient gas during melting, i.e., the possible roles of evaporation and condensation during heating and cooling; and how ordinary and carbonaceous chondrites differ in these respects.
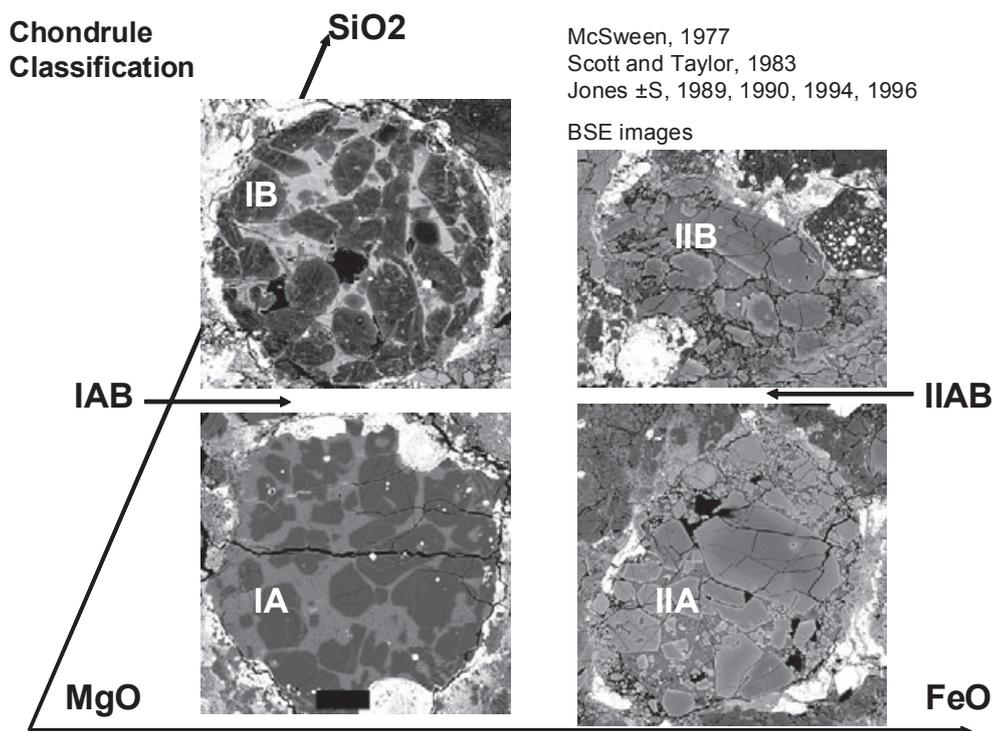
Fig. 1. BSE images of ferromagnesian olivine and pyroxene porphyritic chondrules of types IA (0.7 mm wide), IB (0.7 mm wide), IIA (0.6 mm wide), and IIB (0.8 mm wide) from Semarkona placed in an MgO–SiO$_2$–FeO triangle so as to show their relative Mg, Fe, and Si contents.

CHONDRULE PROPERTIES

Most chondrules are ferromagnesian and are classified as magnesian (type I) or ferroan (type II), with olivine-dominant (A), intermediate (AB), and pyroxene-dominant (B) subtypes (McSween 1977a; Scott and Taylor 1983; Jones and Scott 1989; Jones 1990, 1994, 1996a; Hewins 1997) in each type. We illustrate some typical porphyritic textures in Fig. 1, superimposed on a bulk composition triangle to show their relative Fe/Mg and SiO$_2$ differences. The classification scheme can be extended to other textural types using mineral or bulk compositions (Hewins 1997). The proportions of different types of chondrule and the details of their compositions vary in different chondrite types, with type I dominant in carbonaceous and type II in ordinary chondrites (Zanda et al. 2006). A major question is why we have a dispersion of chondrule bulk compositions in MgO–SiO$_2$–FeO composition space (Fig. 1), as shown by Radomsky and Hewins (1990) and Cohen et al. (2004). Grossman and Wasson (1982) have suggested that this simply reflects a random spectrum of precursor components that originated in condensation processes. However, in the case of a closed system, such precursors would have to be either coarse-grained (and hence result from an anterior high-temperature episode; Hezel and Palme 2007) or

fractionated in a systematic way (such as high T condensates). Alternately, the variability of bulk compositions primarily reflects exchanges with the ambient gas such as evaporation of a chondritic (fayalite-rich) composition (Sears et al. 1996; Cohen et al. 2004), condensation onto a fractionated (forsterite-rich) composition (Tissandier et al. 2002; Villeneuve 2010), or a more complex mixture of processes.

Many chondrules have textures, including the presence of glass, indicating rapid crystal growth at near-liquidus temperatures, and the crystallization of chondrule melts is well understood (Lofgren 1996; Hewins et al. 2005). "Flash" heating and cooling have been proposed for chondrules. However, experiments with very rapid heating and cooling produce large temperature gradients, insufficient crystal growth, and agglutinate-like objects (fig. 13 in Hewins et al. 2005). Here, we show a type IIA chondrule with skeletal hopper olivine crystals (Fig. 2a), very similar texturally to Apollo basalt 12009 (Fig. 2b). Thus, the cooling conditions of this chondrule were not very different from those of some lunar basalts, bearing in mind the different crystal growth rates in liquids of different compositions. The cooling rates estimated for most type II chondrules, based on experiments matching textures and limited olivine zoning data, are 10–1000 °C (Lofgren 1996; Hewins et al. 2005).
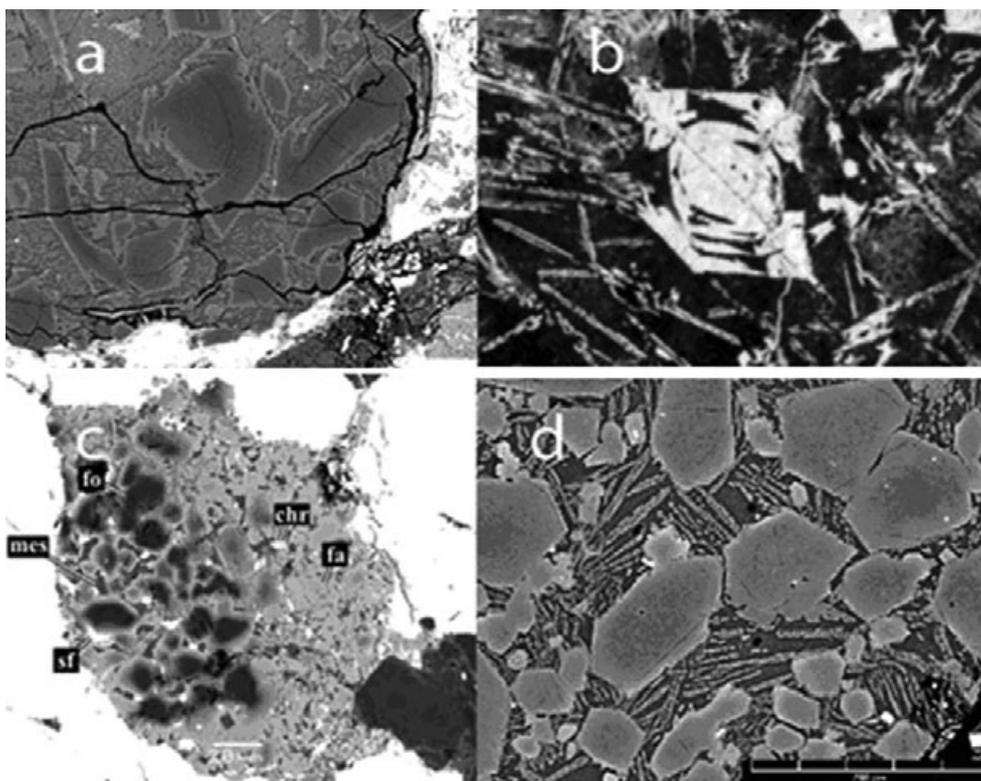
Fig. 2. Comparison of textures. (a) Type IIA porphyritic chondrule from Semarkona with hopper phenocrysts (BSE), which resemble those in lunar olivine vitrophyres. (b) Hopper olivine phenocryst and pyroxene dendrites in Apollo basalt 12009 (transmitted light). (c) Type IIA chondrule containing a large cluster of forsteritic olivine resembling type IA material (BSE), in Isheyevo CH∕CBb chondrite, from Krot et al. (2007). (d) Semarkona type IIA chondrule with olivine phenocrysts and pyroxene dendrites in glass (BSE). Image widths are 0.6, 2.5, 0.25, and 0.4 mm, respectively.

Diffusion modeling of zoning profiles has enlarged this range for PO down to at least $1 °C h^{-1}$ (Miyamoto et al. 2009). These cooling rates apply from the liquidus (1500–1700 °C) across the crystallization interval (at least 300 °C). Fewer experiments have been performed on type I chondrules, and their forsteritic olivine crystals are almost unzoned, but their cooling rates appear to be similar to, or lower than, those of type II chondrules (Wick 2010; Wick and Jones Forthcoming). Many type I chondrules were heated well below their liquidus temperatures (Whattam and Hewins 2009). Reasonable chondrule thermal histories are produced in the nebular shock wave model for chondrule formation (Desch and Connolly 2002).

Many chondrules date from 1–3 Myr after CAI (Kita et al. 2000, 2005; Amelin et al. 2002; Amelin and Krot 2007; Villeneuve et al. 2009), although recent Pb–Pb chronology (Connelly et al. 2011) suggests that the first chondrules formed at the same time as CAI. Type I chondrule olivines have $\Delta^{17}O$ values in at least six clusters, named groups A–F, consistent with formation in six oxygen isotope reservoirs (Libourel and Chaussidon 2011); there is some overlap between carbonaceous chondrites and ordinary chondrites, but ordinary chondrites have the higher values. Echoes of these reservoirs appear in other work, e.g., Tenner et al. (2011a) reported four clusters of olivine oxygen isotopic compositions in a CO3.0 equivalent to groups B, C, D, and E of Libourel and Chaussidon (2011). Isotopic compositions of olivine relict grains, olivine phenocrysts, and igneous rims on chondrules in many chondrites suggest a reaction during heating and cooling to higher $\Delta^{17}O$ values than those of the precursors (Wasson 2000; Jones et al. 2004; Kunihiro et al. 2005; Krot et al. 2006; Ruzicka et al. 2007), as do pyroxene grains crystallized near chondrule margins (Libourel et al. 2006; Chaussidon et al. 2008). However, the opposite is seen in one type IA chondrule, where the olivine rims are more magnesian and enriched in ^{16}O than the cores (Kita et al. 2010). In general, such reactions may be explained by the assembly of different amounts of solids and gas of different isotopic compositions, and subsequent equilibration (Krot et al. 2010a; Libourel and Chaussidon 2011). In some relatively ^{16}O-rich CV and CR chondrules, increase in $\Delta^{17}O$ is related to the addition of SiO to the chondrules (Chaussidon et al. 2008). However, for some chondrules

in Tagish Lake (Russell et al. 2010), CO chondrites (Tenner et al. 2011a), and CR chondrites (Schrader et al. 2011; Tenner et al. 2011b), even heavier oxygen isotope compositions are associated with increasingly ferran compositions, with two trends corresponding to reservoirs B and C continuing into type II chondrules.

Semarkona has olivine in O isotope groups A, B, and C (Libourel and Chaussidon 2011). For other LL chondrites, Kita et al. (2010) reported olivine oxygen isotopic compositions in both type I and type II chondrules that are similar to those of group B type I chondrules of Libourel and Chaussidon (2011). Olivine in relic-free type II chondrules in CR chondrites (Connolly and Huss 2010; Schrader et al. 2011) have $\Delta^{17}O$ values corresponding to those of groups B and C in both ordinary chondrites and some carbonaceous chondrites and type I chondrules plot in groups B, C, and D (Libourel and Chaussidon 2011). Thus, CR chondrules share the isotopic characteristics of ordinary and other kinds of carbonaceous chondrites.

Krot et al. (2007, 2010a) described unique chondrules with skeletal blebby (barred) olivine (SO) along with cryptocrystalline chondrules (CC) in CBb chondrites, and these have later formation ages (4562.7 $\pm$ 0.5 Myr; Krot et al. 2005a) than other chondrules. The transient heating mechanism generating these chondrules is hypothesized to be a collision of protoplanetary bodies, rather than some nebular process as hypothesized for common chondrules. In this hypothesis, the occurrence of CC chondrules within zoned metal particles and the enrichment in refractory elements of SO chondrules are consistent with an origin as liquid condensates and evaporative residues, respectively, formed in the impact plume (Krot et al. 2005a, 2007, 2010a). Heavier Mg isotopes in SO than CC chondrules (Gounelle et al. 2007) are consistent with this idea.

PRECURSORS

Many chondrules contain material that does not appear to have crystallized from the chondrule melts. These objects indicate at least a part of the material existing at the time of chondrule melting, although the surviving population will be biased toward grains hardest to melt. These might be simply the largest grains of an abundant phase in the precursor, or lithic inclusions, or ''xenocrysts'' of rare precursor phases that are hard to melt, like forsterite in type II chondrules. Textures of some chondrules indicate incomplete melting, and these give the most information on the unprocessed material, both of individual grains and of bulk composition, assuming that open system behavior is less in less melted objects.

There is a spectrum of grain sizes in chondrules accompanied by mineralogical and chemical changes.

Fine-grained chondrules, often with outlines that are not rounded, tend to contain more sulfide and/or metal than coarse-grained droplet chondrules (Campbell et al. 2005). These chondrules are conceivably closer to their precursors than common coarser grained ones. Their textural and chemical differences between ordinary chondrites and carbonaceous chondrites (Zanda 2004) suggest different amounts of troilite in their chondrule precursors, in agreement with the bulk chemical compositions of these chondrites (Zanda et al. 2009), while the sequences within each group suggest evaporative loss during the chondrule-forming events.

In porphyritic type II chondrules, the most abundant relict phase is forsteritic olivine (e.g., fig. 2c, from Krot et al. 2007). Pyroxene (Jones 1996b; Hewins 1999; Hewins et al. 2011) and spinel (Hewins et al. 2011) relics are known but much rarer, in part surely because they dissolve rapidly into type II melts at near liquidus temperatures. The relict olivine has a wide range of compositions and conceivably more than one source. Type I chondrules are an abundant possible source material in the chondrule-forming regions. However, AOA have also been found in type IIs by Yurimoto and Wasson (2002). There is a diversity of relict grain compositions even within a single type II chondrule. For example, we observed that a chondrule in the CM Cold Bokkeveld, with normal olivine core compositions Fa_{18} and higher, has two relict grains of $Fa_{0.7 \pm 0.1}$ (n = 9) and $Fa_{6.3 \pm 0.7}$ (n = 10). This strongly suggests heterogeneous precursors aggregated into a dustball, as advocated by Jones et al. (2005), although modification during the formation of type II chondrules could also be considered (Villeneuve 2010).

Dark-zoned chondrules are very fine-grained and enriched in opaque minerals (mostly sulfides), especially toward the outside (Dodd 1971; fig. 1c in Hewins et al. 1997; fig. 1b in Hewins 1997). As they are not highly melted, they are potential precursors for PO chondrules in OCs. However, ultra fine-grained precursors, such as nebular condensates, are incapable of producing the dominantly porphyritic textures of chondrules, producing almost equally fine-grained olivine crystals in experimental charges (Hewins and Fox 2004). They also cannot reproduce the compositional heterogeneity of chondrules in the case of a closed system (McSween 1977a; Hezel and Palme 2007) unless they are fractionated themselves in a systematic way from the beginning. However, in dark-zoned chondrules, besides patches of fine-grained cryptoporphyritic material, which suggests that the precursor was largely fine dust (Hewins et al. 1997), there is a great diversity of sizes of angular relics. Chondrule precursors were probably a mixture of dust and large crystals in many cases. Other chondrules with low degrees of melting are aggregates mostly of coarse crystals and sometimes chondrule fragments
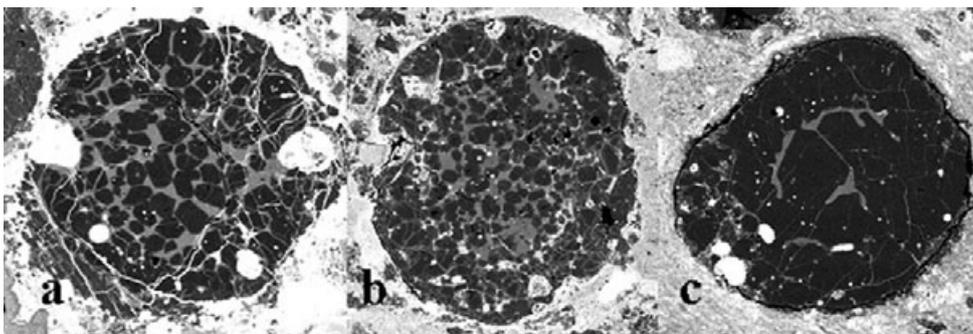
Fig. 3. BSE images of type I chondrules with outer zones of compact olivine (black), possibly formed by liquid phase sintering, surrounding glass-rich cores (medium gray): this polygonal olivine with triple junctions resembles GOA, suggesting that the latter might not be formed by planetary processes. (a) in NWA 4290 LL3.10 (a.k.a. Kelb Ellouz, courtesy B. Devouard). (b) in Vigarano CV3. (c) in Kaba CV3; all three chondrules are approximately 0.6 mm wide.

(Weisberg and Prinz 1996; Nettles et al. 2002), suggesting that many chondrules especially in ordinary chondrites are derived from chondrule debris, as advocated by Alexander (1996).

The dominant relict material in type I chondrules, especially in CV chondrites, is granoblastic olivine aggregate (GOA) (Libourel and Krot 2007). GOA must be either fragments of planetary rocks or highly annealed nebular material. Their granoblastic textures are characteristic of planetary materials (Libourel and Krot 2007), but somewhat similar textures appear as the outer shells of type I chondrules (Fig. 3) and appear to be of nebular origin. Liquid phase sintering would promote the growth of such textures, although there is little evidence of melt in GOA. Wasson and Rubin (2010) have suggested that granoblastic textures in type I chondrules in CR chondrites are due to long thermal pulses. Whether planetary or nebular, GOA produce porphyritic textures by disruption during melting (Libourel and Krot 2007; Whattam and Hewins 2009). Many chondrules appear to be disrupted GOA material, such that the "phenocrysts" are really relict grains, and GOA are important precursors for IA PO chondrules. However, many type I chondrules, although not as fine-grained as dark-zoned chondrules, are too fine-grained to be derived from GOA precursors (Hewins et al. 1997; Campbell et al. 2005; Whattam and Hewins 2009) and could have formed by dustballs. The olivine in these chondrules might be derived from a primitive component, like AOA, or from debris from other type I chondrules. In addition, the occurrence of ferroan olivine that experienced reduction to form dusty forsterite in type I chondrules (Fredricksson 1969) suggests that recycled type II chondrule material was among the precursors of some type I chondrules.

Some CAI were also incorporated into chondrules (Krot et al. 2005b), in one case with a ≥2 Myr time gap. Such occurrences of CAI in chondrules are quite rare, which could be explained by the ease of dissolving CAI with low-temperature igneous phases within forsteritic melts. The dissolution of CAI material within a chondrule would generate a melt on the Mg-poor side of the thermal divide (forsterite-anorthite cotectic), such that plagioclase is stable (MacPherson and Huss 2005; Tronche et al. 2007). This process is likely to have been significant for chondrules as testified by oxygen isotopes in the case of CR chondrites (Krot et al. 2006; Fig. 3) and also by their chondritic Al∕Si, Ti∕Si, and Ca∕Si ratios (based on data from Alexander et al. [2008] for ordinary chondrites and also from McSween [1977b] for carbonaceous chondrites using appropriate correction factors for broad beam EMP analyses). These chondritic ratios imply that either condensation happened everywhere and CAI were converted to chondrules in some regions, or that some chondrites represent regions where evaporation was incomplete and Al, Ti, and Ca remained with forsterite in annealed residues. Both early primitive material (e.g., refractory inclusions) and late nebular material (chondrule debris) were thus available as type I chondrule precursors.

The properties of SO and CC chondrules in CH chondrites and the presence of GOA as well as fine-grained material within the precursors of various chondrules show that chondrules are unlikely to have formed by a unique mechanism and from a single type of precursor. Some chondrules in CV chondrites (Wood 1996) and most chondrules in CR chondrites (Zanda et al. 2002) may be interpreted as having formed from a spray of droplets in a dust-rich environment (Fig. 4). In addition, as discussed above, it is likely that the precursors of chondrules in ordinary chondrites contained S, whereas most of those of carbonaceous chondrites did not. This implies an effect of evaporation or—more likely—of fractional condensation on the composition of chondrule precursors, independently of any chemical changes that may subsequently have resulted from the chondrule-forming event itself.
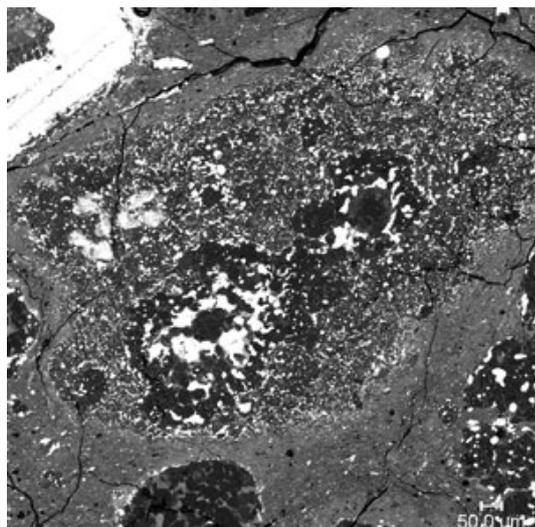
Fig. 4. Composite type I chondrule in Renazzo CR2 chondrite, with coarse-grained well-melted metal-bearing chondrules within finer grained less heated chondrule material set in fine-grained matrix, suggesting formation of a spray of droplets in a dust-rich environment. White—metal; black—magnesian silicates in type I chondrules.
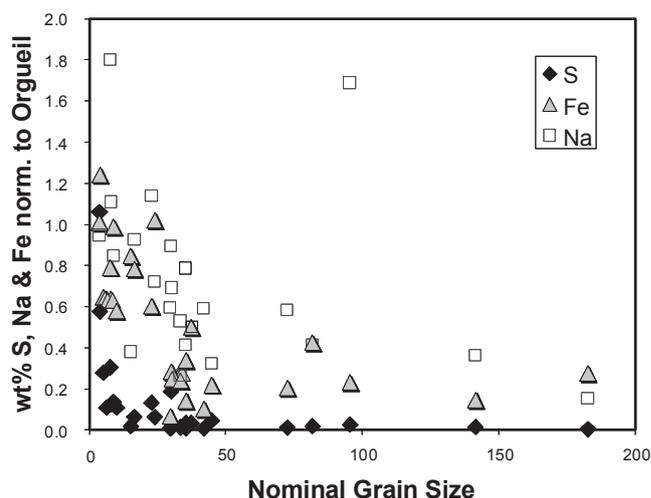

Fig. 5. Concentrations of moderately volatile elements in chondrules in Semarkona LL3.0 show a negative correlation with nominal grain size, an indicator of extent of melting. This suggests that the more highly melted chondrules experienced more evaporation.

Precursors that have experienced fractional condensation, or even fractional crystallization in early parent bodies, might explain why the concentrations of moderately volatile elements Si, Fe, Na, and S vary significantly in chondrules. However, exchanges with the ambient gas during the chondrule-forming event are likely to be superimposed on precursor composition effects. For example, many OC type II chondrules are not depleted in Na (Jones 1990; Hewins 1991) and they retained on the average 0.34 wt% S (Jones 1990). This might be better explained by heating in a low-pressure environment as suggested by the chondrule textural sequences described in Campbell et al. (2005), followed by recondensation upon cooling. We search below for evidence of the evaporation and condensation during heating at low pressure predicted by experiment (e.g., Tissandier et al. 2002; Cohen et al. 2004) and theory (Ebel and Grossman 2000; Alexander 2004; Davis et al. 2005) in particular types of chondrules. The broader question is why there is the dispersion of MgO–SiO$_2$–FeO compositions (Fig. 1): chondrules could be made either by modifying a ferran precursor primarily by evaporation or reduction or a magnesian precursor primarily by condensation. If primitive solar system dust resembled anhydrous CI chondrite, i.e., minerals assumed to form from nebular condensates in which fayalitic olivine formed by reaction of FeO with enstatite, then evaporation and/or reduction during melting converted them into chondrules (Connolly et al. 1994; Sears et al. 1996; Cohen et al. 2004). Alternatively,

as suggested above, they were derived from magnesian nebular condensates, subsequently processed or not, with lower temperature components added during chondrule formation.

EVIDENCE FOR LIMITED EVAPORATION

We interpreted very finest-grained Semarkona type I chondrules as those with initial properties least disturbed by melting, resembling original dustballs rather than GOA precursors, and noted that they contained sulfide rather than metal. We compared their compositions with successively coarser grained, more extensively melted chondrules (Fig. 5): the moderately volatile elements S, Na, and Fe in bulk analyses decrease with increasing grain size, consistent (Hewins et al. 1997) with an evaporation trend, and the most forsteritic olivine grains have the lowest Mn concentrations. Fe is lost dominantly from silicate. As discussed in the next section, this idea is supported by the frequent presence of FeS-FeNi veneers on chondrules presumably formed by the recondensation of the evaporated elements. An alternative explanation for the volatile enrichment of the very fine-grained chondrules is that these objects formed by aggregation of partly melted particles that scavenged volatiles with an efficiency that depended on their grain size (Scott and Krot 2005). However, the textures of metal-sulfide associations in type II chondrules and in the finer-grained type I chondrules in ordinary chondrites demonstrate that the S was already present within these chondrules when they melted. Thus, for ordinary chondrites, we deduce a mildly ferran dustball precursor for many type I

chondrules, and evaporation as a dominant process. Iron evaporated from silicate melt into hydrogen-rich gas would recondense as metal. Metal and silicate compositions in type I chondrules from CR chondrites were also interpreted as evidence for Fe evaporation during melting followed by its recondensation onto chondrule surfaces along with the other moderately volatile siderophile elements (Connolly et al. 2001; Humayun et al. 2002, 2010; Zanda et al. 2002; Campbell et al. 2005).

Evaporation experiments at P_{H2} of 1.3×10^{-5} bars (Cohen and Hewins 2004; Cohen et al. 2004) have produced some but not all properties of type I chondrules. At temperatures that produce extensive melting within chondrules at total pressures of less than 1 bar, the stable form of most elements including Fe and S is vapor (e.g., Wood and Hashimoto 1993). In free evaporation experiments at 1350 and 1580 °C (Cohen and Hewins 2004), Fe is lost rapidly from melted troilite, and residual Fe metal, and more slowly from silicates. Sears et al. (1996) proposed that chondrule compositions evolved by evaporation first of Fe then of Si from silicates to produce the trend from IIA to IB to IA chondrules in Fig. 1. The loss of S and Fe from different type I chondrules with more extensive melting has been documented (Hewins et al. 1997; Zanda et al. 2002). Evaporation experiments (Cohen et al. 2004) reproduce Ca-rich bulk and olivine compositions similar to those of type I chondrules (Jones and Scott 1989). The Ca-rich forsterite is due to equilibrium partitioning from the liquid (Libourel 1999), no matter how that liquid composition is achieved. If type I chondrules had been formed by evaporation of a ferroan precursor at nebular pressures, this would have required long heating times. However, while these aspects of natural type I chondrule might be related to evaporation, the experiments failed to reproduce some of their major features, such as pyroxene near the rim, possibly formed by condensation (see below) and kamacite globules, which require non-canonical conditions (see below).

Silicate charges containing troilite heated at 1350 °C and P_{H2} of 1.3×10^{-5} bars (< IW-4.8 but charge self-buffered), took between 2 and 10 min to lose all their sulfur (Cohen and Hewins 2004). This suggests that all the S present within chondrule precursors should have been lost. Accordingly, the most plausible circumstance to explain the presence of a few tens of a percent S within porphyritic type II chondrules (Jones 1990) is high ambient sulfur pressure, although some fine-grained chondrules may not have been sufficiently heated. Similarly, Cohen and Hewins (2004) examined the fate of metallic iron in 4 mm synthetic chondrules under different pressure conditions. In the 1350 °C experiments, metal survived for 30 min, while at 1580 °C with the same P_{H2}

of 1.3×10^{-5} bar, Fe metal was never observed (Cohen and Hewins 2004; Cohen et al. 2004). The survival of Fe-metal in chondrules means less drastic conditions. For example, it survives for 6 h at 1580 °C at 1 atm H_2-CO_2 (IW-3), before evaporating (Cohen and Hewins 2004). The combination of Fe-metal and forsteritic olivine in many type I chondrules is thus incompatible with evaporation of any ferroan precursor, such as anhydrous CI precursor, if the ambient pressure is low. The presence of Fe metal in chondrules requires some combination of high total pressure P_{tot}, high P_{Fe}, and/or relatively short heating time.

Isotopic data also argue in favor of limited evaporation from chondrule melts or back reaction from the gas: isotopic mass fractionation is the signature of free evaporation and if chondrules formed at nebular pressures, the absence of mass fractionation in chondrules (e.g., Alexander et al. 2000) must be explained. A potential solution is melting in solid-enriched nebular environments (Hewins 1989, 1991), where isotopic exchange between gas and liquid is possible (Alexander 2004; Cuzzi and Alexander 2006). Kinetic modeling of chondrule formation by shock wave shows extensive evaporation and recondensation of alkalis (Fedkin et al. 2011, 2012). We discuss below evidence for (re)condensation and the particular case of ordinary chondrite type II chondrules, which carry evidence for both loss and gain of Na during their formation.

EVIDENCE FOR CONDENSATION

Type II chondrules are chondritic in alkalis (Hewins 1991) and even at 1 atm, Na could not be retained at cooling rates required for the production of chondrule textures (Yu and Hewins 1998). Chondrules should be devoid of alkalis, as well as of sulfide and metal, if they experienced any evaporation of FeO (Yu et al. 2003; Cohen and Hewins 2004; Cohen et al. 2004), yet even type I chondrules contain moderate concentrations of Na and metal may be abundant. K isotopes are fractionated in experiments, but not in chondrules (Yu et al. 2003). Enrichments in Na (Matsunami et al. 1993) and silica (Tissandier et al. 2002; Libourel et al. 2006; Chaussidon et al. 2008) toward the edge of type Is are consistent with condensation from the ambient gas, and so is the silica enrichment trend observed in the mesostases of type I chondrules (Libourel et al. 2006).

Such considerations have led to a series of melting experiments under high partial pressures of the lithophile elements K (Georges et al. 2000), Si (Tissandier et al. 2002), and Na (Mathieu 2009). These experiments determined the solubilities of alkalis at equilibrium. The duplication of the textures of type IA chondrules, with

pyroxene near the rims as a result of SiO condensation, suggests that IAB chondrules could be a result of this process (Tissandier et al. 2002). The IA–IAB–IB textural association is found in both ordinary chondrites and carbonaceous chondrites (Scott and Taylor 1983), indicating that SiO condensation into chondrules is a general process (Libourel et al. 2006).

Fe–Mn Fractionation in Type II Chondrules: Evidence for Condensation in the Chondrule-Forming Event

Villeneuve (2010) has proposed that type II chondrules were formed from type I chondrule material, partly by oxidation of metal (which he demonstrated experimentally), and by addition of species such as FeO and SiO from the gas during chondrule formation. The common occurrence of relict forsterite of type I composition strongly supports this.

Berlin et al. (2011) have shown that olivine in type IIA chondrules in ordinary chondrites, CR, and CO chondrites has distinctly different Fe/Mn ratios. We illustrate a similar difference between OC and CM chondrites in Fig. 6 with our analyses of IIA olivine in Semarkona and Cold Bokkeveld, Mighei, and Murchison. The different average Fe/Mn ratios of the olivines indicate that the two groups of chondrules were formed in different reservoirs, with different redox conditions and possibly extent of condensation into the chondrule melts or precursors. The question of interest is why the chondrule olivine (and bulk) compositions are so dispersed and more so in carbonaceous chondrites than ordinary chondrites.

Oxidation of Fe from metal would dilute MnO and would not create a positive correlation of MnO and FeO. As the two trends in Fig. 6 exhibit positive correlations, oxidation of Fe from metal can only have played a limited role, such as causing the jump from type IA olivine composition to the most magnesian type IIA chondrules. As the chondrules are independent melt droplets, they cannot be related by crystal fractionation. The correlations are thus the signature of volatility-related processes. They could conceivably reflect either the evaporation of Fe and Mn from the molten chondrules or the condensation into the chondrule melts during cooling of Fe and Mn present in the gas, previously derived from chondrules or dust by evaporation. However, the trends converge in the general area of the compositions of type I chondrule olivine and relict forsteritic grains in type II chondrules. This, along with the very presence of the relict forsteritic grains, suggests condensation onto type I material as the dominant process.

Moderately volatile elements lost during the melting event are expected to recondense into the chondrule on cooling (e.g., Fedkin et al. 2008). Lithophile elements
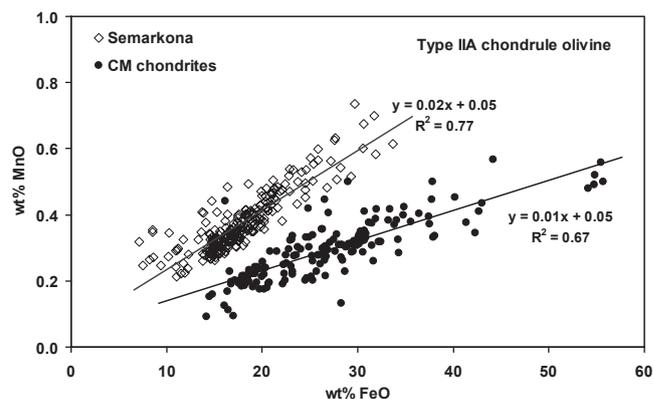

Fig. 6. Olivine in chondrules in ordinary chondrites and CM chondrites (Cold Bokkeveld, Mighei, and Murchison) have different FeO/MnO ratios and enrichment trends, indicating that they formed in reservoirs of different oxidation state. The trends converge in the region of type I chondrule olivine and relict grain compositions. Type II chondrule formation may have involved condensation of FeO and MnO onto type I precursors.

like Na and Si can diffuse through the silicate melt toward the chondrule center. However, S, Fe, and other chalcophile and siderophile elements do not dissolve easily in chondrule melts and therefore build opaque veneers on chondrule surfaces when recondensing.

Opaque Veneers Around Chondrules in CR and Ordinary Chondrites: Evidence for Recondensation Following Evaporation

Metal (and infrequently sulfide) veneers, i.e., concentrations of grains on and near rims, are found on many of the coarse-grained type I chondrules in CR chondrites (Fig. 7a). The distribution of volatile siderophile elements, such as Cu, Pd, and Au, observed by Humayun et al. (2002, 2010) shows that at least one population of chondrule rim metal is depleted in refractory siderophiles and enriched in volatile siderophiles relative to interior metal. This supports the proposal of Connolly et al. (2001) that some of the rim metal can be explained by recondensation of a vapor derived from the evaporation of chondrules onto the chondrule surfaces (Connolly et al. 2001; Humayun et al. 2002; Zanda et al. 2002), an interpretation which is also qualitatively consistent with the zoning of Ni and Co reported by Lee et al. (1992) as pointed out by Campbell et al. (2005).

Type I and type II chondrules in ordinary chondrites also exhibit opaque veneers, but these contain a large amount of S (Fig. 7b). In fact, less than one fourth of the S in Semarkona is contained within its fine-grained matrix and less than 2% within its chondrules. S in Semarkona is mostly found in the opaque veneers and in large metal–
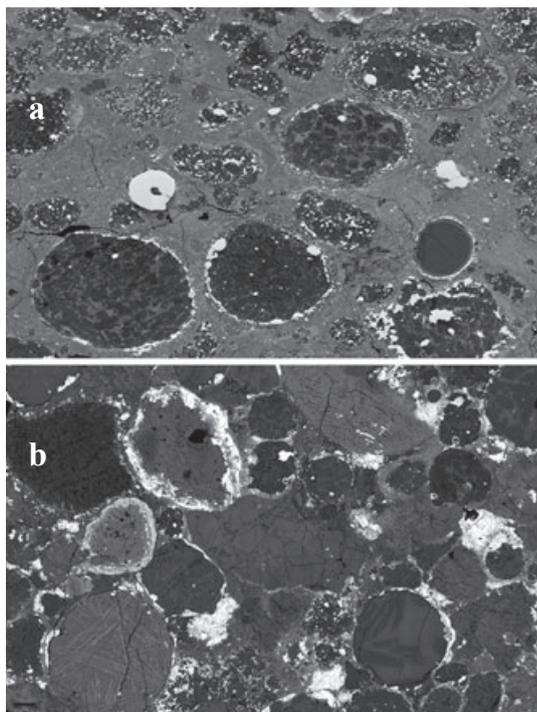
Fig. 7. BSE images of veneers formed by condensation on chondrules. (a) Metal veneers on chondrules in CR2 Renazzo. (b) Sulfide veneers on chondrules in LL3.0 Semarkona. White—metal or sulfide; gray—ferroan silicates in matrix (particularly in a) or type II chondrules (particularly in b); black—magnesian silicates in type I chondrules. This figure also displays the fundamental difference between ordinary and carbonaceous chondrites. Semarkona chondrules are ''clustered'' together, while the chondrules in Renazzo (some of which are ramified and composite-looking) are separated by an abundant fine-grained matrix. The image is 1.2 cm wide for Renazzo (top); 0.5 cm wide for Semarkona (bottom).

sulfide associations, which are likely to have fallen from chondrule surfaces (Zanda and Bourot-Denise, personal communication). The opaque veneers on 3 chondrules often contain intimate troilite-pentlandite intergrowths derived from Ni-rich monosulfide solid solution. Experiments by Lauretta et al. (1997) show that condensate sulfide made from the reaction of gaseous H_2S with kamacite contains significant dissolved Ni. We thus interpret these veneers as the result of S (and Fe) evaporation from chondrules followed by their recondensation on chondrule surfaces. Condensate sulfide is afterward commonly modified by alteration and/or low-temperature metamorphism in chondrites (Zanda et al. 1995).

The difference between type I chondrules in ordinary and carbonaceous chondrites in the nature of their veneers simply reflects the difference in their precursors, which contained no S in carbonaceous chondrites as suggested by Zanda et al. (2009) and discussed above. It also suggests that the chondrules

formed in reservoirs lacking the complement of elements not in the precursors, in particular S for carbonaceous chondrites. These observations on metal and sulfide suggest limited evaporation, and recondensation of the moderately volatile elements, due to high partial pressures, or total pressure. Both evaporation and condensation are expected theoretically, with chondrule melts approaching CMAS liquid compositions temporarily for extreme conditions (Fedkin et al. 2008), but effects due to recondensation may be more obvious than the earlier evaporation. In the next section, we search for evidence of both processes in the same suite of chondrules.

EVAPORATION AND CONDENSATION OF NA IN TYPE II CHONDRULES

The presence of volatiles in chondrules is a potential key to their formation conditions and the ambient gas. Even at 1 atm, Na cannot be retained at cooling rates required for chondrule textures if P_{Na} is low (Yu and Hewins 1998). Most chondrules have higher FeO and Na_2O contents higher than expected for formation in canonical solar nebular gas, suggesting formation of melt droplets at high P_{Na} due to extensive vaporization of accompanying material in a particle-rich clump (Hewins 1989). We estimated the size of these clumps to be about 1000 km (Sahagian and Hewins 1992). Type II chondrules in Semarkona were found to be close to chondritic (Orgueil or Semarkona) in Na/Al ratio (Hewins 1991) as confirmed by more recent analyses: Fig. 8 shows that analyses of type IIA and IIAB chondrules from Semarkona by Jones (1990, 1996a), Alexander et al. (2008), Hewins et al. (2012), and Hewins and Zanda (unpublished data) plot close to the chondritic ratio (Orgueil; Jarosewich 1990). The type IIA chondrules of Jones (1990) have a slightly higher Na/Al ratio, close to 1, suggesting the presence of albite in the precursors (Hewins 1991). The observation of chondritic Na abundances in chondrules led to the idea that the melts became stabilized by the local concentration of dust or ice in the disk (Hewins 1991), so that small amounts of evaporation during heating gave liquid–gas equilibrium (e.g., Wood 1996). In this case, there would be no need for very short duration heating–cooling cycles. Lewis et al. (1993) showed experimentally that Na could be retained in chondrule melts with $P_{Na} \sim 10^{-6}$ atm, whereas Mathieu (2009) has determined Na solubilities for a wide range of melt compositions with a P_{Na} of 1.25×10^{-4} atm. However, such high P_{Na} values, which would have prevented evaporation, seemed to require an implausible level of dust or ice enrichment in the disk (Hewins 1997; Yu and Hewins 1998) and this still presents problems (Fedkin et al. 2008, 2011, 2012).
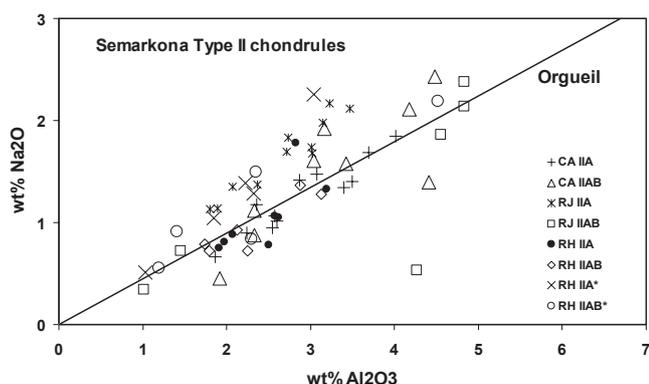
Fig. 8. Al_2O_3 and Na_2O concentrations show that Semarkona type II chondrules have chondritic Na concentrations. Data RJ are from Jones (1990, 1996a), CA from Alexander et al. (2008b), RH from Hewins et al. (2012), and RH* our unpublished data. The reference line is for Orgueil (Jarosewich 1990).

The concentration of Na in minerals such as olivine can potentially indicate if they were in equilibrium with the chondrule melt, in turn giving an idea of P_{Na} when the chondrules melted and cooled. Alexander et al. (2008a) determined Na concentrations in chondrule olivine and melts, and estimated very high P_{Na}. We continued their work (Hewins et al. 2012) using porphyritic Semarkona type II chondrules, adding new approaches: we looked at the Na contents of glass inclusions in olivine, we considered olivine–melt equilibrium using three recent experimental studies on the Na partitioning (Borisov et al. 2008; Kropf 2009; Mathieu 2009), and we compared observed and calculated liquids resulting from fractional crystallization using PETROLOG (Danyushevsky 2001; Danyushevsky and Plechov 2011). The figures and discussion below are based on the results of Hewins et al. (2012).

Figure 9 shows a type IIA chondrule with liquid preserved as glass in various environments. These include primary melt inclusions trapped in olivine, melt channels in olivine sometimes seen to connect to the exterior, open inclusions (Faure and Schiano 2005) or embayments on crystal boundaries, and free mesostasis. The mesostasis in Fig. 9 contains only a few tiny dendrites, but in other chondrules, there are large pyroxene dendrites (Fig. 3d). Figure 10 compares the compositions of these liquids. There is a trend of increasing Si and Al through the various glass compositions, consistent with fractional crystallization of olivine from a liquid with the bulk composition of the chondrule. This is unlike the situation for type I chondrules, where the pattern of glass compositions is inconsistent with olivine crystallization, but shows addition of silica, presumably from ambient gas, to an initial liquid enriched in refractory elements (Tissandier et al. 2002; Krot et al. 2004). The data for
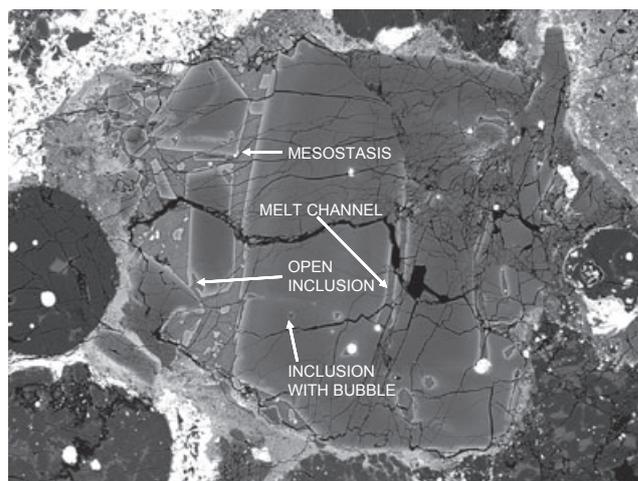

Fig. 9. Distribution of silicate melt in type IIA chondrule 64–65 in Semarkona, showing mesostasis, melt channels, open inclusions, and glass inclusions in olivine.
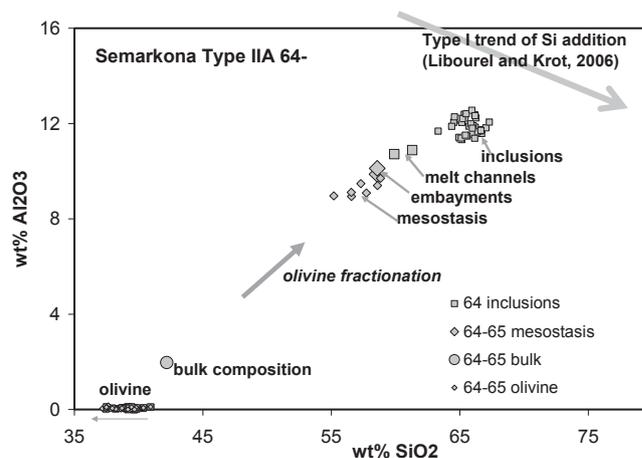

Fig. 10. The correlation of SiO_2 and Al_2O_3 is explained by their enrichment in melt due to olivine crystallization in Semarkona type IIA chondrule 64–65, with no evidence for addition of SiO from the gas.

type II chondrules indicate that they were extensively melted and crystallized olivine, but do not indicate how the chondrule bulk compositions arose.

In Fig. 11, we show the dispersion of mesostasis Na_2O and SiO_2 concentrations due to the effects of initial liquid composition and the crystallization of olivine ± pyroxene. The IIA liquids crystallize olivine only, and are enriched in both Na and Si. The relatively low Si but high Na and Al contents of IIAB mesostases are explained by the crystallization of orthopyroxene with similar Si to the liquids. However, many mesostases have higher Na_2O concentrations than predicted by PETROLOG calculations. Melt inclusions in olivine are Si-rich, because after being enclosed in olivine, they have
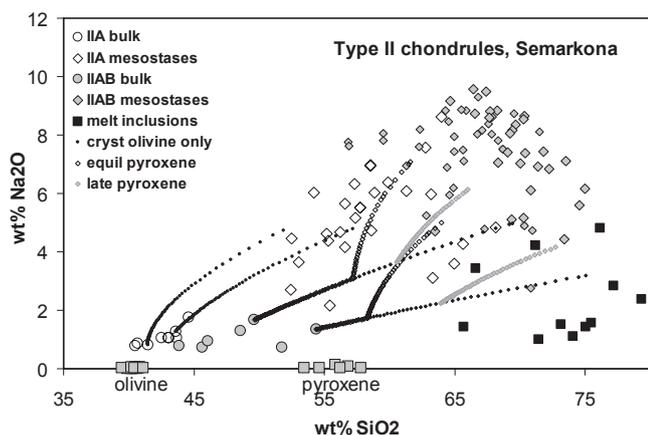
Fig. 11. Compositions of Semarkona IIA and IIAB bulk chondrules, mesostases, and melt inclusions in olivine. Fractionation trends resulting from crystallization of olivine from IIA were calculated using PETROLOG. For type IIAB chondrules, three calculations were made: crystallization of olivine only, olivine followed by equilibrium pyroxene, and olivine followed by late nucleated pyroxene.
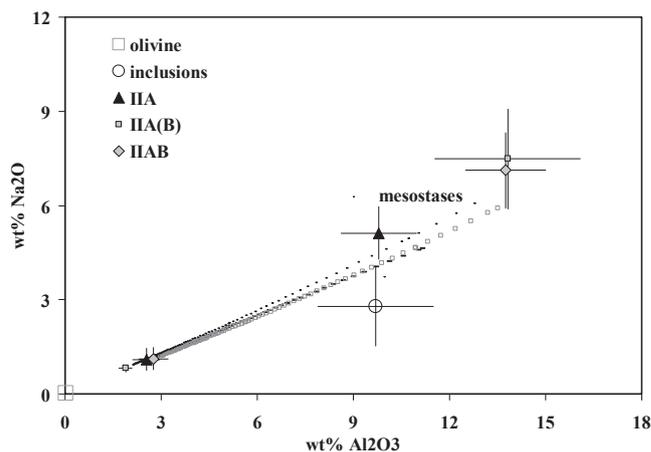

Fig. 12. Average compositions of Semarkona IIA, IIA(B), and IIAB bulk chondrules, mesostases, and melt inclusions in olivine. Compared with daughter liquids calculated for fractional crystallization, melt inclusions have lower Na contents, suggesting evaporation, and mesostases higher Na contents, suggesting recondensation, i.e., open system behavior.

continued to crystallize metastable olivine rather than nucleating pyroxene. Their Na concentrations are much lower than expected for fractional crystallization of olivine from the apparent parent liquid. (Analyses of melt inclusions shown in Fig. 11 are not shown here, because prior exposure to the electron beam compromised the Na data.)

Average values for Al_2O_3 and Na_2O of bulk, mesostasis, and inclusion melts are plotted in Fig. 12. We distinguish type IIA(B) chondrules, an olivine-dominant IIAB resembling chondrule C120 of Jones (1996a) here, although they were included with type IIAB in Fig. 11. Error bars are one standard deviation. We show calculated daughter liquids for three typical compositions. Those which crystallize pyroxene (IIA[B] and IIAB) show slight deviations from the olivine-only trend (IIA) toward higher Na, because Al is more compatible in pyroxene than Na. Figure 12 shows that inclusions have lower Na and mesostases higher Na than is consistent with fractional crystallization of the bulk chondrule compositions.

The data in Hewins et al. (2012) may be used to investigate Na partitioning between olivine and melt. We initially make the conventional assumption that olivine cores were in equilibrium with liquids of bulk chondrule composition and calculate apparent partition coefficients $D_{Na}(Ol/Liq)$. We assume that rim olivine crystallized from residual melt in the mesostasis, and used the highest Na analyses, e.g., of the glass in embayments shown in Fig. 10 to calculate D_{Na}.

Olivine partitioning experiments were made by Borisov et al. (2008), Kropf (2009), and Mathieu (2009).

Kropf and Mathieu have almost identical coefficients for Na partitioning in the Fe-free system. Mathieu defined the effect of Fe in olivine on Na partitioning, using data for terrestrial rocks in which evaporative loss is not an issue. We use his expression to calculate the Na content of melts in equilibrium with our olivines. We see that the rims and glasses are very close to being in equilibrium, whereas cores are not. In particular, the IIA bulk compositions have on average only a third of the Na expected for equilibrium partitioning. Thus, we conclude that chondrules lost Na by evaporation during olivine crystallization, leading to the formation of melt inclusions that are less enriched in Na than expected for fractional crystallization. Addition of Na by condensation after inclusions were cut off inside olivine is required to explain the Na-rich mesostasis compositions and the olivine rim compositions.

The record of olivine and glass Na concentrations is thus consistent with evaporation at high temperature followed by condensation at low temperature. This sequence has been predicted for chondrules heated in nebular shocks, e.g., Fedkin et al. (2008). Exchange of Na between melt and gas means that Na in the bulk composition after the chondrule-forming event was not its original value.

CHONDRULE RELATIONSHIPS

The presence of GOA in many type I chondrules, the addition of species such as SiO to their rims, and the abundance of forsteritic relict grains in type II chondrules all suggest that chondrule compositions evolved from magnesian to ferran, particularly in
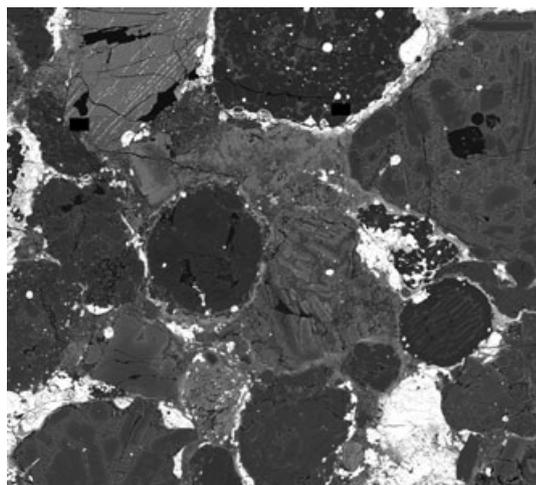
Fig. 13. Type I and type II chondrules in Semarkona are bound together and molded around each other in tight clusters resembling large compound chondrules. Many ordinary chondrites are "cluster chondrites" (Metzler 2011).

carbonaceous chondrites. The survival of Ti^{3+} in pyroxene and olivine in type II chondrules also suggests reduced precursors (Simon et al. 2011). This is consistent with the more ^{16}O-rich isotopic composition of olivine in many type I chondrules (Wasson 2000; Jones et al. 2004; Kunihiro et al. 2005; Krot et al. 2006; Libourel and Chaussidon 2011; Tenner et al. 2011b). Since the return of Genesis, we have learned that the Sun, and hence early nebular gas, had low $\Delta^{17}O$ (McKeegan et al. 2010), and it has been proposed that primordial solids had higher $\Delta^{17}O$ (Krot et al. 2010b). In this interpretation, there were lower dust–gas ratios for regions in which carbonaceous chondrite type I chondrules, or their precursors, formed than where type II chondrules formed, and hence more dust–gas interaction. This is consistent with lower partial pressures of O, Na, and S during the formation of type I chondrules, and with overlapping formation times of type I and type II chondrules (Amelin and Krot 2007; Kurahashi et al. 2008; Villeneuve et al. 2009).

Dusty olivine derived from type II chondrules occurs as relicts in some type I chondrules, which may therefore have formed from dustballs accumulated after the earliest chondrule-forming events. Statistics are lacking, but dusty relicts appear more common in ordinary chondrites than in carbonaceous chondrites, suggesting possibly more recycling in ordinary chondrites than in carbonaceous chondrites, unless this simply results from the absence of type II material in the environment in which carbonaceous chondrites type I chondrules formed.

Type II chondrules have chondritic Na (Hewins 1991) and crystallize Na-bearing olivine. Evaporation

experiments (Yu and Hewins 1998; Yu et al. 2003) showed that rapid cooling was not adequate for reproducing chondrule volatile contents even at high total pressure. The above observations demonstrate that the ambient gas had high partial pressures of elements such as Na, Fe, and S, produced by evaporation from chondrules and accompanying fine dust, and (re-) condensation during chondrule formation would have been normal. Many calculated chondrule silicate melt composition glasses have close to the equilibrium compositions for gas enriched up to 1000× in lithophile elements (Ebel and Grossman 2000; Alexander 2004). Mathieu (2009) used experiments on Na solubility in CMAS liquids to determine P_{Na} of 10^{-8} to 10^{-5} atm during chondrule formation. Hewins (1989) proposed formation of chondrules in a large dusty clump yielding high P_{Na} on heating, an environment already seen as necessary for lowering cooling rates (Hewins et al. 1981), and further justified by the recent work on Na in chondrule olivine (Alexander et al. 2008a; Borisov et al. 2008; Mathieu 2009).

Nebular turbulence tends to concentrate chondrule-sized objects into clumps (Cuzzi et al. 1996) and chondrule cooling rates can be explained in shock wave models if chondrule formation involved clumps of chondrule-sized precursors (Desch and Connolly 2002). If big enough, these clumps are self-gravitating (Cuzzi et al. 2008). Partial pressures $P_{SiO,Fe,K,Na,S}$ were high, as most chondrules contain metal or ferroan olivine as well as troilite and Na-bearing glass and minerals. These partial pressures if generated from evaporating precursors require a high number density of melting chondrules as well as large clumps to preserve the evaporated gas in contact with chondrules. The time chondrules spent in free flight was not always long enough to ensure solidification before collision with other chondrules or accumulation. However, the low number of compound chondrules observed suggests lower number densities than the 10^4 m^{-3} estimated by Alexander et al. (2008). This paradox can be resolved for many ordinary chondrites, because large clusters of agglomerated chondrules are seen in polished sections, e.g., in Semarkona, as reported by Zanda (2004) and illustrated in Figs. 7b and 13. Such meteorites have been called cluster chondrites (Metzler 2011). Thus volatile-rich chondrules were generated in very dense clumps, and some solidified in clusters, being packed together, while some were solid and others still partly molten.

Morris and Desch (2010) have proposed two ways to overcome the chondrule volatile problem. Vapor from the densest core region of a precursor clump, in which all solids are completely evaporated, can move outward, by pressure-driven expansion. In the rest of the clump, melted chondrules cool in contact with this chondrule

vapor from the densest region. An alternative possibility (Morris and Desch 2010) is that chondrules are focused into the clumps, after peak heating, so that they cool in the presence of very high pressures of chondrule vapor that can only arise from a region of higher chondrule concentration. The requisite P_{Na} can be met by evaporating a more reasonable chondrule concentration, possibly attainable by turbulent concentration.

Following the success of nebular shock wave models in reproducing chondrule cooling rates (e.g., Desch and Connolly 2002), Fedkin et al. (2011, 2012) developed a kinetic evaporation model and investigated a range of thermal histories for chondrule formation in gas shocks. They tracked the changing compositions of olivine as it crystallized and isotopic compositions during evaporation and recondensation. They required enrichment by 550× solar in water, to make ferran olivine in chondrules, and 600× solar in dust, to retain Na, uncomfortably large modifications to canonical conditions. The range of Fa contents of olivine in type II chondrules was reproduced, but large isotopic mass fractionations not observed in natural chondrules, particularly in Fe, were also produced. Isotopic heterogeneity could be suppressed only with 100× shorter heating times coupled with dust enrichments $\geq 6 \times 10^4$ and/or $P_{tot} \geq 10^{-2}$ bar. Fedkin and Grossman (2010) and Fedkin et al. (2011, 2012) argued that such conditions could not be achieved in the disk where the gas shocks occurred, so that chondrules were more likely to have formed in collisions of ice-bearing planetesimals.

Impacts cause rapid melting and vaporization, and produce clouds of silicate droplets in vapor with enormous enrichments of volatilized rock, and in water if the bodies contained ice. A very good case has been made that late-formed chondrules in CBb and some related chondrites were formed in such an impact plume. There is a very strong evaporation trend of refractory element enrichment in skeletal (blebby barred) olivine chondrules, and cryptocrystalline chondrules interpreted as condensed liquids are intimately connected with condensate metal (Krot et al. 2007). It seems strange that normal chondrules should be so different from CBb chondrules, if both were formed by impact of planetesimals: different target compositions and absence of ice in the CBb case might be an explanation. Differentiation or melting in the target may also be important (Asphaug et al. 2011).

Morris et al. (2012) have investigated another possible chondrule formation mechanism, melting in bow shocks around protoplanets or planetary embryos. Melting of nebular dust particles in bow shocks around asteroid-sized planetesimals is inefficient, but recent work on the dynamical evolution of the disk emphasizes early planetary embryos and perturbations of their orbits. Walsh et al. (2011) show that Mars could be a survivor

of these embryos, whereas Dauphas and Pourmand (2011) find isotopic evidence that Mars was formed very early, when nebular gas and dust were still present. Morris et al. (2012) found that the embryos' eccentric orbits provide strong bow shocks that can process large quantities of chondrules and cool them at appropriate rates. Passage of the chondrules through the upper atmospheres of embryos with magma oceans may inhibit evaporation.

Chondrites contain type I and type II chondrules, both of which contain relict grains probably derived from the other. Thus, their formation times must have overlapped, although type I chondrules in carbonaceous chondrites are more primitive in oxygen isotopes, as well as more reduced. Both are found trapped and deformed in the same clusters (Zanda 2004; Metzler 2011) in ordinary chondrites, where they have similar $\Delta^{17}O$ (Kita et al. 2010), although their formation conditions differed. This suggests turbulent mixing of hotter and colder regions, which may be hard to explain with a very large-scale nebular shock, or mixing from domains with different P_{H2O} but the same isotopic composition (Connolly and Huss 2010; Kita et al. 2010).

In general, chondrule $\Delta^{17}O$ values are explained by the equilibration of different amounts of solids and gas of different isotopic compositions (Krot et al. 2010a; Libourel and Chaussidon 2011). High P_{H2O} is considered responsible for fayalite enrichment in chondrite olivine (Fedkin and Grossman 2006) and transport of ^{16}O-depleted ice from outside the snowline is considered the cause of nebular gas of heavier isotopic composition (Krot et al. 2006). Connolly and Huss (2010) combined these ideas and proposed that type II chondrules in CR chondrites experienced both oxidation and depletion in ^{16}O during melting, by isotopic exchange with this water-enriched ^{16}O-poor nebular gas. Type I chondrule olivines have a wide range of $\Delta^{17}O$ values (Libourel and Chaussidon 2011; Tenner et al. 2011a), with nearly constant low olivine Fa contents indicating low P_{H2O}; type II chondrules have a more restricted range of $\Delta^{17}O$ values near and above TF, but a large range of olivine Fa contents. This suggests an abrupt increase in Fa content of olivine in type II chondrules where isotopic compositions in the vicinity of TF were achieved (Russell et al. 2010; Schrader et al. 2011; Tenner et al. 2011b); however, they may belong to reservoirs B and/or C of Chaussidon et al. (2008). LL type II chondrules (Kita et al. 2010) have lighter oxygen isotopic compositions (reservoir B) than type II chondrules in CC (reservoir C) such as Tagish Lake (Russell et al. 2010), CO chondrites (Tenner et al. 2011a), and some CR type II chondrules (Connolly and Huss 2010). In addition, type I chondrules with similar isotopic compositions (reservoir B) are present in LL chondrites. Thus, oxidation and

enrichment in heavy isotopes may be decoupled, if, for example, SiO and H_2O molecules equilibrate in the gas (Javoy et al. 2012). The question remains whether the necessary water pressures can be achieved on a large scale, or whether local disruption of icy planetesimals is required (Fedkin et al. 2012).

Some type I chondrules are zoned, with evidence of Si gain and Na gain, whereas type II are close to being in equilibrium with an Na-rich gas. As both type I and type II chondrules entered clusters in the same ordinary chondrite event, they may have been processed separately in domains that mixed into the same clump during cooling. Alternatively, both types of chondrules may have been heated simultaneously with the type I chondrules acquiring elements evaporated from the type II chondrules.

Carbonaceous chondrites are very different from ordinary chondrites, lacking their clusters of closely packed chondrules. Their chondrules were less processed and, as discussed above, had precursors devoid of S (Zanda 2004; Zanda et al. 2009). They accreted type I chondrules with abundant matrix and type II chondrules are in the minority. Although type II chondrules must have been formed under similar circumstances for ordinary chondrites and carbonaceous chondrites, except for differences in fO_2, those in carbonaceous chondrites appear to have been removed from their dense clumps and transported, like refractory inclusions, to join a batch of type I chondrules, or rather several batches of type I chondrules from several reservoirs (Libourel and Chaussidon 2011). Carbonaceous chondrite type II chondrules may have been derived from a source region different from that of the type I chondrules as well as from that of ordinary chondrite type II chondrules, based on their Fe–Mn trends (Fig. 6). More ice in this dust-rich source region would explain the higher fO_2 recorded in the type II chondrules of carbonaceous chondrites. Type I and type II chondrules were mixed without reheating in domains rich in dust, which became carbonaceous chondrite matrix.

CONCLUSIONS

Chondrules exhibit large variations in isotopic, chemical, mineralogical, and textural properties and differ in a systematic way between ordinary chondrites and carbonaceous chondrites. Type I (magnesian) and type II (ferran) chondrules, which require different formation conditions, are not distributed evenly between the two groups: type II chondrules represent only a few percent of carbonaceous chondrites, but most of ordinary chondrite chondrule material.

This variability of chondrule properties can, in part, be ascribed to precursor effects. This is in particular the case for S, which was present in ordinary chondrite chondrule precursors, but not in those of carbonaceous chondrite chondrules (with the possible exception of the rare type II). Relict grains in incompletely melted chondrules coming from AOA, GOA, or older generations of chondrules, and the variability of chondrule compositions, make it unlikely that chondrule precursors consisted simply of CI dust.

To a large extent, the variability of chondrule properties may be ascribed to open system behavior, i.e., chemical and physical interaction between chondrules and ambient gas, including evaporation and condensation, for which there is evidence in both type I and type II chondrules. Textural sequences suggest evaporation, as well as the low S content even of type II chondrules, but the extent of evaporation is difficult to evaluate due to the superimposition of (re-) condensation upon cooling.

Non-lithophile elements such as Fe and S did not dissolve back into the chondrule melt on recondensing and formed opaque veneers. The compositions of these opaque veneers in carbonaceous chondrites and ordinary chondrites match those of their chondrule precursors from which the elements evaporated: metal veneers decorate carbonaceous chondrite chondrules, while ordinary chondrite chondrule veneers consist of sulfide with relict metal grains. The survival of some S within ordinary chondrite chondrules, and the presence of metallic Fe and forsteritic olivines side by side, suggest evaporation to have been limited by the ambient gas partial pressures.

Lithophile species show evidence of condensation from the gas: Si, Na, Mn, and Fe as FeO, but, as these will dissolve into the chondrule melt, it is more difficult to decide whether these came from the evaporation of the chondrules themselves or from that of adjoining dust. The dominance of forsterite as relict material suggests an evolution from magnesian to more siliceous or ferroan compositions with more processing. This evolution is supported by oxygen isotope data and could indeed result from a net addition of material from the ambient gas. Type II chondrules can be explained by condensation of FeO and MnO onto type I precursors under more oxidizing conditions for carbonaceous chondrites than for ordinary chondrites.

The sequence of Na-rich olivine, Na-poor melt inclusions, and Na-rich mesostases in type II chondrules in Semarkona records the whole history of evaporation followed by recondensation. This sequence could be explained by the formation of ordinary chondrite chondrules in domains with large number densities and high partial pressures of the rock-forming elements. These might be dense clumps in the disk heated by gas shock waves, impact plumes formed by collisions of planetesimals, or the bow shocks of protoplanets. As

chondrule clusters in ordinary chondrites associate type I and type II chondrules, it seems likely that these were recycled simultaneously and reacted with the same gas reservoir. Carbonaceous chondrite type II chondrules were added late to batches of type I chondrules and accreted with abundant matrix dust

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