

Nebular or parent body alteration of chondritic material: Neither or both?

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(Received 1997 October 8; accepted in revised form 1998 July 2)

(Presented at the Workshop on Parent Body and Nebular Modification of Chondritic Materials
 at the University of Hawaii, Maui, Hawaii, USA 1997 July 16–19)

Abstract—Discussions of meteorite properties often concern whether they are the result of "nebular" or "parent body" processes, but several decades of research have not resolved the issue. Part of the problem is that any gas-phase reaction involving meteoritic materials thought to have occurred is automatically assumed to be nebular, even though the most primitive solar system objects are water- and volatile-rich and could easily generate vapors. Reactions important in meteorite genesis are those involving (1) oxidation of Fe, (2) oxidation of other cations, (3) reduction, (4) olivine–pyroxene equilibria, and (5) hydration of silicates. The P-T-x conditions required are almost invariably incompatible with standard models for the early solar nebula, but clearly many of these reactions occurred prior to final agglomeration. Most of the reactions require high pressures ($>10^{-3}$ atm) or, more importantly, major departures from cosmic composition, even though the final rocks are remarkably cosmic in elemental proportions. We suggest that most of these reactions occurred in a regolith rendered "dynamic" by the flow of copious volatiles. In such a scenario, liquid and gas phase reactions can occur at elevated temperatures and pressures relative to the nebula and with noncosmic gas phase compositions; but the system is "closed" to most components, so that cosmic proportions are essentially preserved.

INTRODUCTION

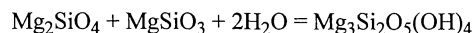
Chondritic meteorites have compositions very similar to that of the solar photosphere, they are the oldest objects in the solar system, and they have unique textures (Kerridge and Matthews, 1988). Their textures are not those of igneous and terrestrial sedimentary processes but have been described as "cosmic sediments," or resembling volcanic tuffs (Urey and Craig, 1953; Ringwood, 1959). They are aggregations of the diverse primitive solid components that formed in the early solar system, yet the rocks as a whole retained essentially solar composition.

Chemical reactions involved in the formation of meteoritic minerals are listed, with literature references, in Table 1. Probably the most frequently discussed reactions, because of their relevance to chondrite origin and classification, are those involving the oxidation of Fe. Most authors favor a nebula scenario for this process (Ikeda and Kimura, 1995; Hua and Buseck, 1995; Nagahara, 1984; Peck and Wood, 1987), but Housley and Cirlin (1983) and Wasson and Krot (1994) have recently discussed parent-body scenarios. A wide variety of oxidation reactions have been attributed also to nebular processes (reactions 7 to 17 in Table 1). Oxidation reactions in an H_2/H_2O -buffered nebula can occur only at very low temperatures (Larimer, 1967, 1968, 1973; Grossman, 1972; Sears, 1978) at which kinetic constraints may become a problem. Either a means of adjusting the nebular H_2/H_2O has to be identified or oxidation must have occurred on or in the parent body.

In contrast, the formation of Si-bearing metal (reaction 18 in Table 1) and several other reductions are favorable in a gas of cosmic composition if the temperature and pressure are high enough (Sears, 1980; Rambaldi *et al.*, 1980). Similarly, the olivine–pyroxene equilibria (such as reactions 19 to 22 in Table 1) are favorable at high temperatures in a gas of cosmic composition (Palme and Fegley, 1990; Hua and Buseck, 1995; Larimer, 1967, 1968; Grossman, 1972). The high temperatures required have led many authors to suggest that some of these reactions accompanied chondrule formation

(Gooding *et al.*, 1980; Huang *et al.*, 1996a), which clearly preceded final agglomeration of the meteorites. Some reactions, like the olivine–pyroxene equilibria, may have occurred also at high temperatures during metamorphism following agglomeration of the meteorites, and these equilibria are sometimes used to estimate metamorphic temperatures (Larimer, 1968).

Until recently, it was universally assumed that the hydration of silicates occurred on the parent body (Krot *et al.*, 1995; Keller and Buseck, 1990; Weisberg *et al.*, 1993; Grimm and McSween, 1989). This was mainly because hydrolysis reactions also require low temperatures and might face kinetic difficulties. Fegley and Prinn (1989) argued that reactions involving solid–solid interaction, such as



that occur at temperatures <400 K are exceptionally slow (10^{23} s at ~ 400 K) even if grain dimensions are only ~ 0.1 μm . However, collision lifetimes between water and forsterite grains are rapid ($<10^3$ s) even for temperatures <300 K (Fig. 1). The feasibility of reaction therefore depends on reaction rates and activation energies. Fegley and Prinn (1989) assumed an activation energy of 70 kJ/mol, which is the value for the hydration of MgO, and argued that hydration was too slow to have occurred in the nebula. However, Wegner and Ernst (1983) found a value of only 32.5 kJ/mol for the hydration of forsterite to serpentine and brucite that corresponds to a reaction time scale of 10^9 s, which is well within the lifetime of the nebula. Of course, larger grain sizes would increase time scales, and the presence of these silicates as amorphous dust rather than sub-micrometer crystals would decrease these time scales. But it seems that there are no kinetic difficulties with the formation of hydrated silicates in the nebula.

Keller and Buseck (1991) recently argued that gas-phase hydration of refractory phases occurred in the nebula. Metzler *et al.* (1992) argued that CM chondrites underwent aqueous alteration in the nebula; but Bischoff (1997)—a co-author of the Metzler *et al.* paper—has argued that it occurred prior to final agglomeration,

TABLE 1. Representative mineralogical reactions in meteorites, proposed locations, conditions and references.

Reaction	Temperature (K)	Pressure	Location	Comments	References
Oxidation of Iron					
(1) $\text{Fe} + 0.5\text{O}_2(\text{g}) + \text{MgSiO}_3 = (\text{Fe,Mg})_2\text{SiO}_4$	<500	–	neb, MPB	"Substantial pressure of many gases"	1,2,20
(2) $2\text{FeS} + \text{SiO}(\text{g}) + 3\text{H}_2\text{O}(\text{g}) = 3\text{Fe}_2\text{SiO}_4 + \text{H}_2(\text{g}) + 2\text{H}_2\text{S}(\text{g})$	<1300 290–410 350–500 440–650 640–900 1100–1230	– 10^{-3} atm $\text{H}_2\text{O}/\text{H}_2 = 0.001$ $\text{H}_2\text{O}/\text{H}_2 = 0.01$ $\text{H}_2\text{O}/\text{H}_2 = 0.1$ $\text{H}_2\text{O}/\text{H}_2 = 1$ $\text{H}_2\text{O}/\text{H}_2 = 10$	neb		4
(3) $2\text{Fe} + \text{SiO}_2 + \text{O}_2 = \text{Fe}_2\text{SiO}_4$	290–410	–	neb		16
(4) $4\text{Fe} + \text{NaAlSi}_3\text{O}_8 + 2\text{O}_2 = \text{NaAlSiO}_4 + 2\text{Fe}_2\text{SiO}_4$	–	–	neb	"...low temperatures, oxidizing conditions"	16
(5) $\text{Fe} + \text{CaSiO}_3 + \text{SiO}_2 + 1/2\text{O}_2 = \text{CaFeSi}_2\text{O}_6$	–	–	neb		16
(6) $\text{Fe}(\text{s}) + \text{Si}_{1/2}\text{O}(\text{s}) + \text{H}_2\text{O}(\text{g}) = \text{FeSi}_{1/2}\text{O}_2(\text{s}) + \text{H}_2(\text{g})$	<1500	<1	MPB		21
Other Oxidation Reactions					
(7) $2\text{Fe}_3\text{O}_4 + 3\text{SiO}(\text{g}) + \text{H}_2\text{O}(\text{g}) = 3\text{Fe}_2\text{SiO}_4 + \text{H}_2(\text{g})$	–	–	neb	Same as reaction 2	4
(8) $\text{W} + \text{Ca}(\text{g}) + 2\text{O}_2(\text{g}) = \text{CaWO}_4$	1600 1400	$\log p\text{O}_2 = -12.86$ $\log p\text{O}_2 = -16.32$	neb neb	"P(O ₂) are 10^3 – $10^4 \times$ solar"	11
(9) $\text{W} + \text{CaTiO}_3 + 3/2\text{O}_2(\text{g}) = \text{CaWO}_4 + \text{TiO}_2$	1600 1400	$\log p\text{O}_2 = -10.87$ $\log p\text{O}_2 = -13.75$	neb neb	Same as reaction 8	11
(10) $\text{W} + \text{Ca}_2\text{Al}_2\text{SiO}_7 + 3/2\text{O}_2(\text{g}) = \text{CaWO}_4 + \text{CaO} + \text{SiO}_2 + \text{Al}_2\text{O}_3$	1600 1400	$\log p\text{O}_2 = -8.34$ $\log p\text{O}_2 = -10.87$	neb neb	Same as reaction 8	11
(11) $4\text{S} + 3\text{H}_2\text{O} = 2\text{H}_2\text{S} + \text{S}_2\text{O}_3^{2-} + 2\text{H}^+$	<373	–		"Oxygen-free atmosphere"	12,13
(12) $5\text{H}_2\text{O}(\text{g}) + \text{SiO}(\text{g}) + 2\text{Mg}(\text{g}) + \text{Si}(\text{ss}) = 2\text{MgSiO}_3(\text{s}) + 5\text{H}_2(\text{g})$	<1300 <1430	10^{-4} atm 10^{-3} atm	neb neb		14
(13) $\text{Ca}(\text{g}) + \text{TiO}(\text{g}) + 2\text{H}_2\text{O}(\text{g}) = \text{CaTiO}_3(\text{s}) + 2\text{H}_2(\text{g})$	<1658	10^{-3} atm	neb	"Cosmic gas"	15
(14) $2\text{ScO}(\text{g}) + \text{H}_2\text{O}(\text{g}) = \text{Sc}_2\text{O}_3(\text{s}) + \text{H}_2(\text{g})$	<1673	10^{-3} atm	neb	"Cosmic gas"	15
(15) $\text{LaO}(\text{g}) + 1/2\text{H}_2\text{O}(\text{g}) = \text{LaO}_{1.5}(\text{ss}) + 1/2\text{H}_2(\text{g})$	<1598	10^{-3} atm	neb	"Cosmic gas"	15
(16) $\text{SiC}(\text{s}) + 3\text{H}_2\text{O} = \text{SiO}_2(\text{s}) + \text{CO} + 3\text{H}_2$	–	–	neb		17
(17) $\text{SiO}_2(\text{s}) + 2\text{Mg} + 2\text{H}_2\text{O} = \text{Mg}_2\text{SiO}_4(\text{s}) + 2\text{H}_2$	–	–	neb		17
Reduction Reactions					
(18) $\text{SiO}(\text{g}) + \text{H}_2(\text{g}) = \text{Si}(\text{ss}) + \text{H}_2\text{O}$	1430	10^{-2} atm	neb	Read from Fig. 3	14
Olivine–Pyroxene Equilibria					
(19) $\text{MgSiO}_3 + \text{FeOH}(\text{g}) = \text{MgFeSiO}_4 + 0.5\text{H}_2(\text{g})$	–	–	neb	"high temperatures, enhanced P(O ₂)"	3
(20) $\text{MgSiO}_3 + \text{Fe}(\text{OH})_2(\text{g}) = \text{MgFeSiO}_4 + \text{H}_2\text{O}(\text{g})$	–	–	neb	Same as reaction 19	3
(21) $2\text{MgSiO}_3 + \text{H}_2(\text{g}) = \text{Mg}_2\text{SiO}_4 + \text{SiO}(\text{g}) + \text{H}_2\text{O}(\text{g})$	–	–	neb	Same as reaction 2	4
(22) $2\text{MgSiO}_3 + 2\text{H}_2\text{O}(\text{g}) + 2\text{Fe} = \text{Fe}_2\text{SiO}_4 + \text{Mg}_2\text{SiO}_4 + 2\text{H}_2(\text{g})$	500	$\text{H}_2\text{O}/\text{H}_2 = 0.0015$	neb	32 mol% Fa	8,9,10
Silicate Hydration Reactions					
(23) $2\text{Ca}_3\text{Al}_2\text{Si}_3\text{O}_{12} + 2\text{MgAl}_2\text{O}_4 + 2\text{Fe} + 3\text{H}_2\text{O}(\text{g}) =$ $\text{Ca}(\text{Mg}_2\text{Al})(\text{Al}_3\text{Si})\text{O}_{10}(\text{OH})_2 + 2\text{FeAl}_2\text{O}_4 +$ $5\text{SiO}_2 + 2\text{H}_2(\text{g}) + 5\text{CaO}$	600–1250	–	neb		5
(24) $3\text{MgSiO}_3 + 0.5\text{NaAlSiO}_4 + (\text{n} + 1)\text{H}_2\text{O} =$ $(\text{Na}_{0.5})\text{Mg}_3(\text{Si}_{3.5}\text{Al}_{0.5})\text{O}_{10}(\text{OH})_2\text{nH}_2\text{O}$	<373	–	MPB		6,7
(25) $2\text{CaAl}_2\text{Si}_2\text{O}_8 + 2\text{MgSiO}_2 + 8\text{Fe} + 8\text{H}_2\text{O} + 2\text{CO}_2 + 4\text{O}_2 =$ $((\text{Fe,Mg})_{10}\text{Al}_2)(\text{Si}_6\text{Al}_2)\text{O}_{20}(\text{OH})_{16} + 2\text{CaCO}_3$	<423	–	MPB		18
(26) $4\text{MgSiO}_3 + 2\text{Fe} + 4\text{H}_2\text{O} + \text{O}_2 = 2(\text{Mg,Fe})_3\text{Si}_2\text{O}_5(\text{OH})_4$	<423	–	MPB		18
(27) $2\text{Mg}_2\text{SiO}_4 + 3\text{H}_2\text{O} = \text{Mg}_2\text{Si}_2\text{O}_7(\text{OH})_2 + \text{Mg}(\text{OH})_2$	<573	–	MPB		19

Neb = nebula; MPB = meteorite parent body.⁵

References: 1 = Housley and Cirlin (1983); 2 = Ikeda and Kimura (1995); 3 = Palme and Fegley (1990); 4 = Hua and Buseck (1995); 5 = Keller and Buseck (1991); 6 = Krot *et al.* (1995); 7 = Keller and Buseck (1990); 8 = Larimer (1967); 9 = Larimer (1968); 10 = Grossman (1972); 11 = Bischoff and Palme (1987) (also corresponding reactions for Mo); 12 = Monster *et al.* (1965); 13 = Bunch and Chang (1980); 14 = Sears (1980); 15 = Sears *et al.* (1983); 16 = Nagahara (1984); 17 = Fegley (1988); 18 = Weisberg *et al.* (1993); 19 = Grimm and McSween (1989); 20 = Peck and Wood (1987); 21 = Wasson and Krot (1994).

leaving open the question of whether it was nebular or parent body. The hydration of refractory calcium-aluminum-rich inclusions (CAIs) in Allende and other CV chondrites with the associated introduction of alkalis, Fe, halogens, and CO₂, is also considered a nebular process that occurred 2 Ma after their formation (MacPherson *et al.*, 1988). The reactions are not listed in Table 1

but mainly involve alteration of melilite to a wide variety of products in a variety of environments, some of them oxidizing.

With three stable isotopes, and stable gas and solid phases, O provides an independent means of meteorite classification and possibly additional insights into the processes by which they formed. It seems that initial isotopic heterogeneity, and one or more

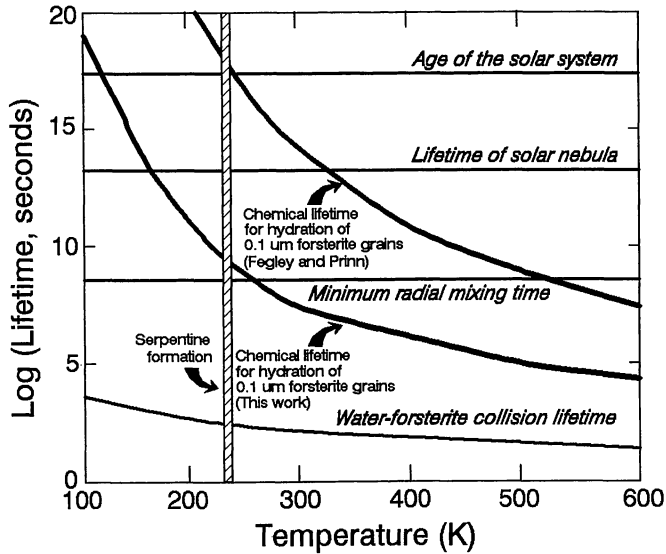


FIG. 1. Chemical and physical lifetimes associated with the hydration of forsterite grains by $\text{H}_2\text{O}(\text{g})$ to form serpentine $\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$ in the solar nebula. All Mg is assumed to be present in forsterite, which is a proxy for diverse suite of Mg-bearing minerals present in the solar nebula at low temperatures. A grain size of $0.2 \mu\text{m}$ is assumed; larger grains, such as those found in meteorites, will react more slowly. The width of the serpentine formation region is governed by whether C is present as CH_4 or CO . The chemical lifetime for forsterite hydration (assuming it is as energetically favorable as MgO hydration) is longer than the lifetime of the solar nebula. Consequently, formation of serpentine (as well as talc and brucite which form at a similar temperature) is very probably kinetically inhibited. (From Prinn and Fegley, 1989, with modifications.)

mass-dependent and mass-independent processes, may be affecting O isotopes. Several proposed scenarios for chondrite formation provide reasonable environments for these processes (Nittler *et al.*, 1994; Thieme, 1996; Clayton and Mayeda, 1984; Clayton *et al.*, 1983, 1985, 1991; Sears and Weeks, 1983).

With such a variety of gas-phase reactions being possible—some involving refractory species at high temperatures, some requiring low temperatures, some involving major silicate minerals in the presence of a gas, and some involving hydration—the environment(s) from which the meteorites came was complex. The age and bulk composition of the chondrites suggests a fairly simple origin in the early solar system. Thus, many of these complexities are sometimes explained in terms of fractional condensation from the nebula gas (Lord, 1965; Larimer, 1967, 1973; Lewis, 1972; Wai and Wasson, 1977), assuming the nebula was initially hot enough to produce an entirely gaseous nebula. Astrophysical arguments now seem to indicate that this was unlikely (Wood and Morfill, 1988), and it is also possible that some or all of these trends reflect heating of solar system materials during chondrule formation (Huang *et al.*, 1996a). It seems clear that there was a nebula of some sort (*e.g.*, Cameron, 1995) and that there were parent bodies (Marti and Graf, 1992). For the chondrite classes, the parent bodies were probably the asteroids, so petrographic observations are often interpreted in terms of "nebular" or "parent-body" processes (Zolensky *et al.*, 1997).

Unfortunately, chondrites are among the least well-understood geological samples because their sources are mostly unknown and, even when probably known, the environment from which they came will remain unfamiliar until manned landings occur on asteroids.

Nevertheless, our understanding of asteroid surfaces has been improved greatly by spacecraft flybys. We now know that small asteroids can have deep regoliths (*e.g.*, Asphaug and Nolan, 1992) and low densities ($\sim 1.5 \text{ g cm}^{-3}$). We also now realize that impact velocities during chondrule formation and chondrite aggregation could have been high ($\sim 5 \text{ km/s}$, Sears *et al.*, 1995). Thus, we will examine the idea that many or most of the reactions in Table 1 occurred in a dynamic volatile-rich regolith on the surface of a CI-like asteroid (Huang *et al.*, 1996b). But first we will review some of the basic properties of chondrites in order to briefly sketch out the scenario we are proposing.

THE CHONDRITE CLASSES, CHONDRULE AND METAL GRAIN SIZES, AND THE UREY-CRAIG DIAGRAM

The chondrite classes are defined on the basis of bulk and mineral compositions, oxidation state of Fe, and O isotopic ratios. However, one of the chief characteristics of the classes is the size distribution of the components and their abundances of chondrules. (For simplicity, we discuss just the best-known classes, but our conclusions can be extended readily to the others.) Most of these fundamental properties are summarized in Figs. 2 and 3. The EH, EL, H, L, and LL chondrites are well resolved on both plots, whereas the metal-bearing CO and CV chondrites are resolved from each other and from the CI and CM chondrites in Fig. 2 (data for Fig. 3 are not available or incomplete). The EH chondrites (with higher than solar Fe/Si ratios) contain metal grains and chondrules that are

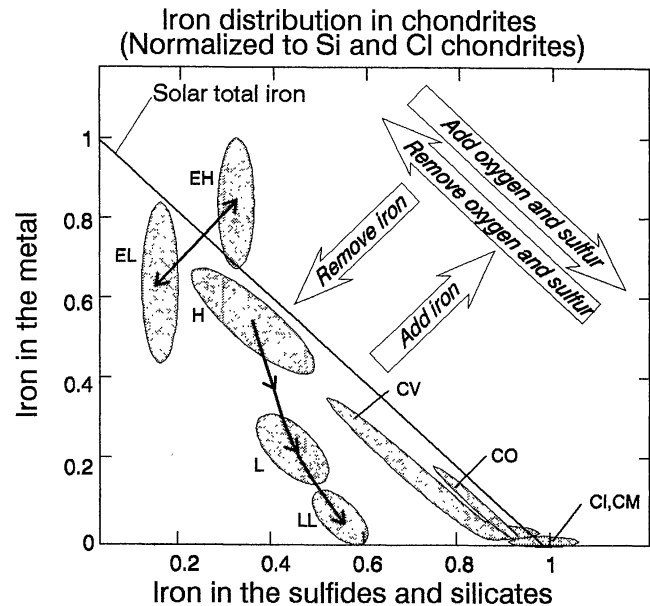


FIG. 2. The Urey-Craig diagram that compares the Fe in the metal state with the Fe in the sulfide and silicate form. The Fe abundance has been divided by Si abundance that, in turn, has been divided by the Fe/Si ratio of CI chondrites. This removes the effects of variations in other quantities, such as water content, and puts the solar (*i.e.*, CI chondrite) value at 1.0. Data plotting on any 45° line with negative slope have the same bulk Fe/Si value, and the addition or removal of S or O moves the data along the diagonal. Most of the chondrite meteorite classes are well resolved on the plot, especially the enstatite and ordinary chondrites. The H, L, and LL chondrite trend has often been ascribed to simultaneous loss of metal and oxidation; but the EH and EL chondrite trend requires the addition of metal and S (relative to solar values) to produce the EH chondrites, whereas the EL chondrites require their removal. (Data from Wiik, 1969, and Jarosewich, 1990.)

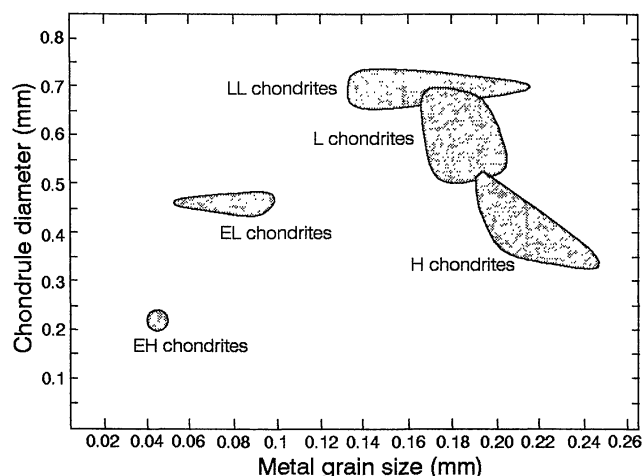


FIG. 3. Chondrule and metal dimension field for ordinary and enstatite chondrites based on fairly meager literature data for 18 samples. Despite problems with these data (chondrule and metal data for a given meteorite are sometimes from different sources; a variety of techniques were used; the point for EL chondrites is an average of only three meteorites—Qingzhen, Kota-Kota and ALHA77156—and data for Parnallee plots outside the fields), the general trends are fairly well established. Each of these major classes occupies an essentially unique field, and although metal grain size decreases as chondrule size increases for the ordinary chondrites, this is not the case for the enstatite chondrites. (The data are from Dodd, 1976; King and King, 1979; Hughes, 1978; Huang *et al.*, 1996a; Rubín and Grossman, 1987; Schneider *et al.* 1998.)

smaller than those of the EL chondrite class (which has lower than solar Fe/Si ratios). In contrast, the H, L, and LL class show a systematic increase in chondrule and metal grain size and a decrease in solar Fe/Si ratios. These variations in bulk Fe/Si are widely interpreted as reflecting a metal–silicate fractionation in the nebula (Donn and Sears, 1963; Orowan, 1969; Larimer and Anders, 1970; Larimer and Wasson, 1988; Skinner and Leenhouts, 1993; Newsom, 1995). Chondrule abundance varies from ~75 vol% in ordinary chondrites, to 30–40 vol% for the enstatite chondrites, CO and CV chondrites, to ~9 vol% for CM chondrites; and essentially zero for CI chondrites (Grossman *et al.*, 1988).

Most of the major chondrite properties are arguably related to the formation of chondrules and the metal–silicate fractionation, but details are controversial (King, 1983; Kerridge and Matthews, 1988; Hewins *et al.*, 1996). Chondrules are clearly the result of momentary heating of milligram amounts of silicate materials to produce complete or partial melting. They were cooled then at rates that were slow for the open nebula (10–1000 °C/h, *c.f.*, 10⁶ °C/h) but fast for igneous or metamorphic processes (~10–100 °C/Ma) (*e.g.*, Huang *et al.*, 1996a). Suggestions for the process are (1) heating nebular dust by shock, lightning or Jupiter-induced shock waves; (2) clumps of interstellar dust that were melted by frictional heating or exothermic reactions as they entered the inner solar system; and (3) impact into a regolith (Boss, 1996; Weidenschilling *et al.*, 1998). Many mechanisms for explaining the metal–silicate fractionation displayed by chondrites have been proposed. They are based on differences in (1) size and density, (2) magnetic properties, (3) ductility, and (4) crystal growth mechanisms of the chondrules and metal grains (Donn and Sears, 1963; Larimer and Anders, 1967, 1970; Skinner and Leenhouts, 1993; Clayton, 1980; Haack and Scott, 1993).

THE DYNAMIC REGOLITH

Spacecraft observations of Gaspra, Ida, Mathilde, Deimos, and Phobos indicate that significant regoliths can accumulate on very small asteroids (Carr *et al.* 1994; Belton *et al.*, 1994), and this suggests possible scenarios for meteorite origins. Their low densities also suggest that asteroids might be also low-strength targets. Hartmann (1985) investigated impact into unconsolidated targets and found that ejecta velocities were much lower than for impact in to solid targets. In fact, under certain circumstances, there was 100% retention of the projectile by the target. Theoretical studies also are consistent with these new observations, because ejecta velocities for such low-strength targets are an order of magnitude lower than for consolidated basalt and 50–95% of impact ejecta could be retained (Housen, 1992; Asphaug and Nolan, 1992; Asphaug and Melosh, 1993).

The limited range of chondrule and metal grain sizes as well as the differences in chondrule size distribution between the classes indicates sorting. Following suggestions by Whipple (1972), Dodd (1976) compared chondrule and metal size distributions for ordinary chondrites and the predictions of aerodynamic drag. If the mean free path of gas molecules is less than grain sizes, then velocities will follow Stoke's law; whereas, if the mean free path of gas molecules is larger than grain sizes, then Epstein's law is followed. Assuming the metal grains and chondrules are experiencing aerodynamic sorting by the same gas, then one can take the ratio of their sizes and calculate the exponent in the relationship:

$$\rho_m s_m^x = \rho_c s_c^x$$

in which ρ_m and ρ_c refer to the densities of the metal and chondrules, and s_m and s_c refer to their sizes. Then $x = 2$ for Stoke's law and $x = 1$ for Epstein's law. Figure 4 shows the Dodd (1976) data. The data do not cluster around a value of 1 or 2 but scatter between, above, and below these values. Dodd (1976) suggested that the Stoke regime gave way to the Epstein regime as the gas diffused away, and that solid-state recrystallization had coarsened some of the metal grains. However, this does not explain why so many chondrites have values below one and above two. We argue that gravitational sorting accompanied aerodynamic sorting and that instead of occurring in the nebula the sorting occurred in a regolith. We make four premises.

Premise I: Primary Parent Bodies Were CI-like in Composition

The basis for this premise is as follows: (1) CI chondrites are the meteorites closest to the solar photosphere in composition (Fig. 5). In fact, comet nuclei are closer to solar in composition than CI chondrites (see below), and these may better resemble the primitive asteroids than even CI chondrites. Several asteroids, including some near-Earth asteroids, have reflectivity spectra resembling that expected for extinct comets. We refer to all these primitive bodies as "CI-like". (2) Main belt and near-Earth asteroids with surface compositions resembling CI, or perhaps CM, compositions are quite common (Fig. 6), whereas asteroids with ordinary chondrite surfaces are very rare. (3) Xenolithic clasts in meteorite breccias are predominantly CI- or CM-like, regardless of the host meteorite (Lipschutz *et al.*, 1989). (4) Fall mechanics shows that carbonaceous chondrite fragmentation rates during atmospheric passage are perhaps as much as 1000× greater than ordinary chondrite fragmentation rates (Fig. 7), so that when corrected for this effect, carbonaceous chondrites are the most abundant meteorites reaching the Earth (Baldwin and Scheffer, 1971).

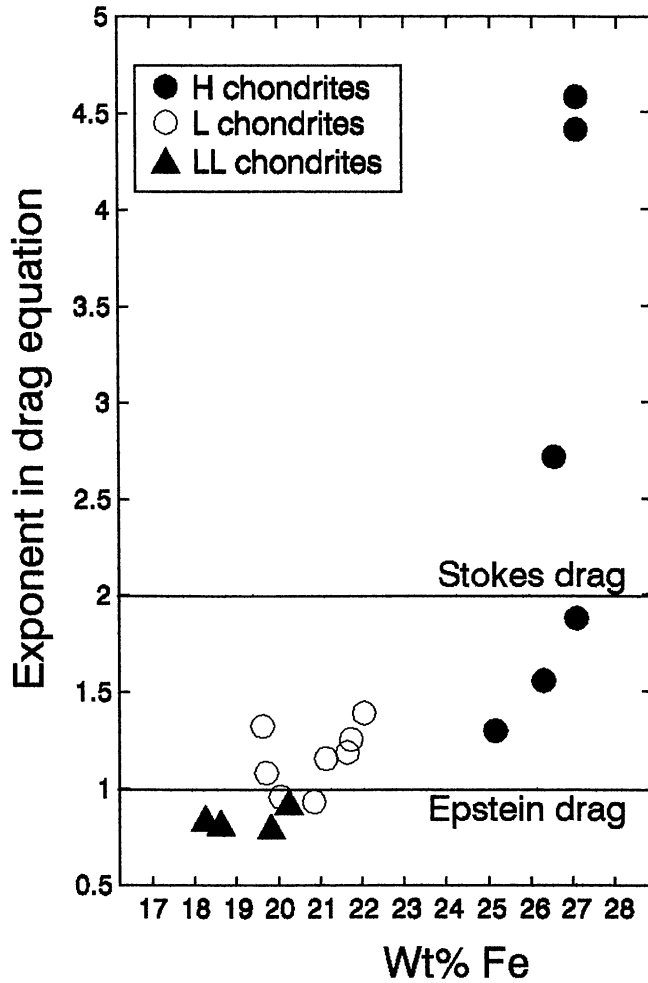


FIG. 4. Plot of the exponent in the expression $\rho_m s_m^x = \rho_s s_s^x$ for coexisting metal and silicates in ordinary chondrites. When the mean path length of the gas molecules is less than the size of the particles, Stoke's law applies and $x = 2$; but when the path length is greater than the particle size, Epstein's law applies and $x = 1$. Not only do the data not cluster around values of one or two, they spread above and below these values. This suggests that forces other than simple aerodynamic drag were involved in determining the sizes of coexisting metal and silicates. Not all of Dodd's silicate particles were complete chondrules, but using more recent chondrule data makes little difference to the plot. (Reproduced from Dodd, 1976.)

Premise II: Even Modest Amounts of Radioactive or Impact Heating Would Cause the Regolith to Dry Out and Become "Dynamic"

The mobilization of a surface dust layer by the passage of gases from below is the basis of the industrially important fluidization process (Kunii and Levenspiel, 1991), and it is an important aspect of pyroclastic volcanism (Wilson, 1980) and impact (Newsom *et al.*, 1986). The process also has been observed during experimental impact experiments into unconsolidated targets where an explosive eruption of gas-charged powders was observed (Hartmann, 1993). By equating the aerodynamic drag to the gravitational attraction, it can be shown that very modest flow velocities will mobilize the dust layers on asteroid-sized objects (Huang *et al.*, 1996b). Furthermore, CI or CM abundances of water will permit degassing to last for months to years at these flow rates, depending on the permeability of the dust layer. In fact, segregation occurs in the laboratory and

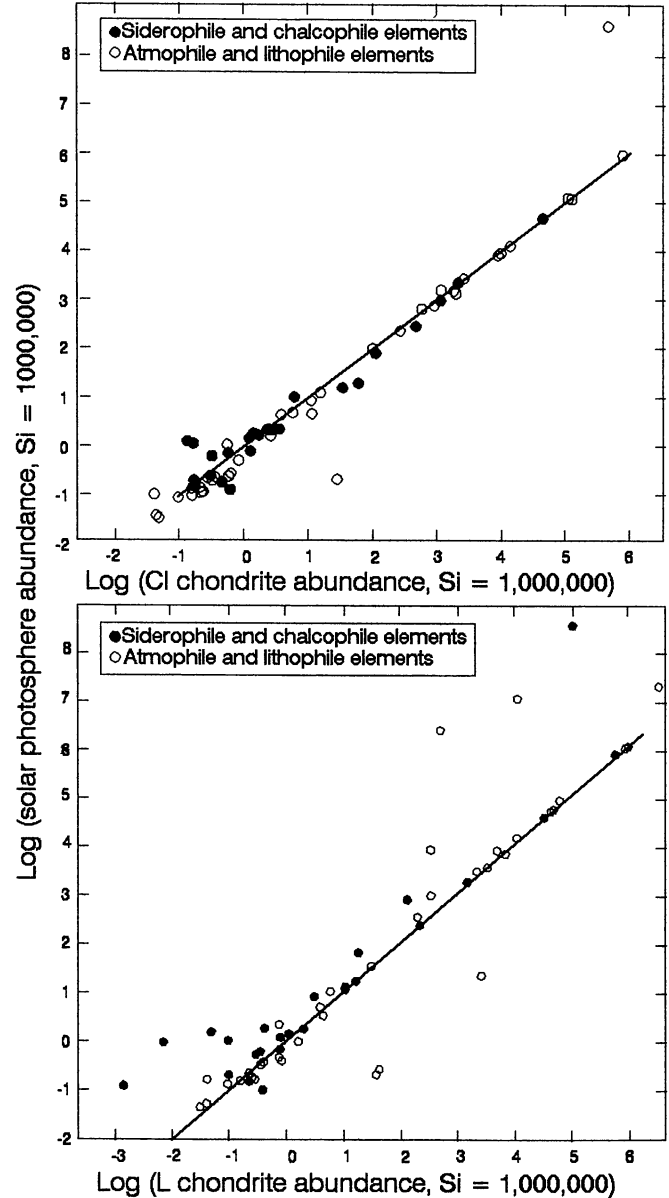


FIG. 5. Plots of elemental abundances in the solar photosphere against CI chondrites (upper) and L chondrites (lower). Both CI chondrites and L chondrites have very similar abundances to the solar photosphere but the L chondrites are characterized by slight systematic depletions of siderophile elements. (Meteorite data from Mason, 1979; solar data from Ross and Aller, 1976.)

during pyroclastic volcanism on the timescale of seconds to hours. Impact and radioactive decay can both generate enough heat to drive the process (Huang *et al.*, 1996b).

Premise III: This Would Inevitably Cause (1) Chondrules and Metal to be Sorted by Gravity and Aerodynamic Drag and (2) Loss of Matrix

Because each chondrite class has a characteristic distribution of chondrules and metal grains, the consequence of aerodynamic and gravitational sorting would differ for each class. Laboratory experiments and theoretical treatments show that usually a gas flow rate sufficient to support the chondrules would drag the metal grains

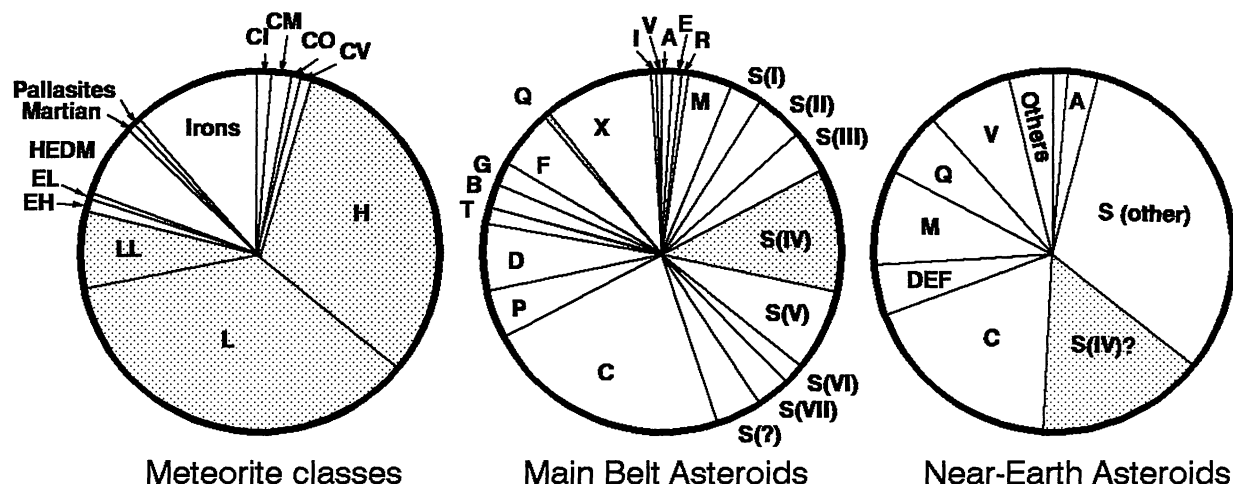


FIG. 6. Pie charts showing the relative abundance (in terms of numbers) of the asteroid spectral reflectivity classes and meteorite classes. Data for asteroids are from Tholen (1989), Gaffey *et al.* (1996), and McFadden *et al.* (1989); whereas, data for the meteorites are from Graham *et al.* (1985). Ordinary chondrites are more heavily represented in the meteorites than their closest equivalents are among the asteroids (Q asteroids and some fraction of the S(IV) asteroids). Similarly, iron meteorites and HEDM (howardite–eucrite–diogenite–mesosiderite) meteorites are more abundant than their asteroid look-alikes, the M and V asteroids, respectively.

through the layer and concentrate them near the surface. Only if the metal and chondrules were comparable in size would the denser metal grains fall in the regolith (Chiba *et al.*, 1980; Huang *et al.*, 1996b).

If the flow rate required to suspend the large components exceeds twice the terminal flow velocity of one of the finer components (*e.g.*, the fine-grained matrix), then the fine material will be lifted out of the column entirely ("elutriated"), much like dust is lifted from the surface of comet nuclei (Grün and Jessberger, 1990). This might explain why there is an inverse correlation between the extent to which this process is thought to have occurred (EH, EL > H, L, LL > CV, CO > CI, CM; see below) and the abundance of fine-grained matrix.

Premise IV: Chondrules Were Formed by Impact into a Regolith and the Oxidation State of the Iron Was Determined by Reduction During Chondrule Formation

One of the main difficulties of producing chondrules by impact is putting enough energy into the surface, but this is not the case when the target is an unconsolidated regolith. The amount of melt produced by impact increases from ~5 to 60 vol% as the porosity increases from essentially zero to ~35% (Schaal *et al.*, 1979). The abundance of agglutinates and impact-melt spherules on the Moon indicates that impact melting on the milligram scale is common place on airless planetary surfaces. Crystalline lunar spherules, with physical properties similar to meteoritic chondrules but with highland compositions, are common at the Apollo 14 site that is thought to contain ejecta from the Imbrium impact. Thus, impact spherules take on the structural properties of chondrules only when the impact is sufficiently large in relation to the size of the target body and flight times are long enough for extensive crystallization (Symes *et al.*, 1998). Only on asteroids that have experienced impacts large enough to produce craters with radii comparable to the radius of the asteroid would one expect the proportion of chondrules to melt spherules and agglutinates to be high. Maybe one should expect all the impact melt on asteroids to be in the form of chondrules because melts would be dispersed readily in the low gravitational field and take a relatively long time to settle.

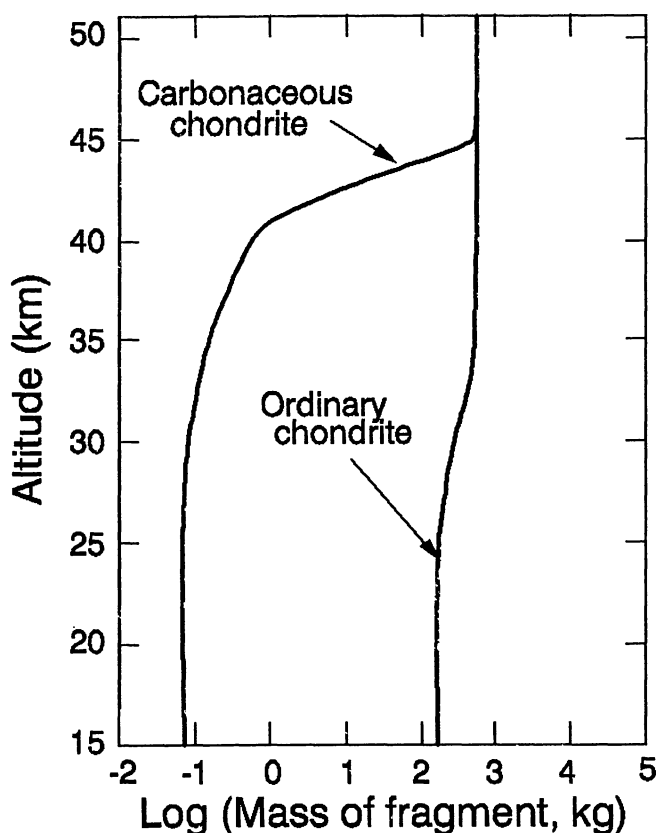


FIG. 7. Plot of fragment size against remaining mass for a carbonaceous chondrite with CI or CM physical properties and an ordinary chondrite entering the atmosphere with an initial mass of 500 kg, velocity of 14.2 km/s and angle of entry of 52°. Because of their lower tensile strength, carbonaceous chondrites suffer 1000-fold greater fragmentation than ordinary chondrites. Because the likelihood of recovery is proportional to mass, it seems that 1000× as many carbonaceous chondrites enter the atmosphere than are recovered on the ground, which means that ~96% of the meteorites entering the upper atmosphere are carbonaceous chondrites. (From Baldwin and Sheaffer, 1971.)

It was thought once that impact velocities were too low for chondrule production and that any impact melt spherules produced would be lost to space. However, it is now at least arguable that chondrules formed several million years after their parent bodies (Hutcheon *et al.*, 1995) and that by this time Jupiter had increased relative velocities in the asteroid belt to 4 or 5 km/s (Davis *et al.*, 1979). The object that is thought to have created the Moon by a giant impact on Earth would have had an escape velocity of 4–5 km s⁻¹ and would have created velocity dispersions of this order. As mentioned above, there is also a strong case to be made now, which could not be made before the spacecraft flybys of the asteroids and Martian satellites, that even these small objects are capable of retaining most of their impact ejecta (Asphaug and Nolan, 1992; Asphaug and Melosh, 1993).

There are many other arguments requiring that chondrules formed in an environment more closely resembling a regolith than the open nebula. These are (1) their high number densities, (2) the existence of compound chondrules, (3) the lack of charged particle tracks in their outer layers, (4) the high O and Na fugacities in which they formed, and (5) the slow cooling rates relative to cooling in a vacuum (Sears *et al.*, 1995). The interpretation of the fine-grained rims in terms of recondensation of volatiles lost during chondrule formation seems more consistent with a regolith than the open nebula (Huang *et al.*, 1996a; Hewins, 1989). Most importantly, the nebula probably did not exist at the time that the chondrules formed. Various authors have shown that the nebula phase of solar system history lasted only ~10⁵ years (Cameron, 1995; Wetherill and Stewart, 1987; Zuckerman *et al.*, 1995); whereas there is growing evidence that chondrules formed ~4 Ma after the CAIs and the parent bodies of the differentiated meteorites (Hutcheon *et al.*, 1994; Hutcheon and Jones, 1995; Russell *et al.*, 1998). Thus, nebula scenarios for chondrule formation all seem very unlikely, including the recent idea of Weidenschilling *et al.* (1998) that chondrules formed in shock waves generated by Jupiter perturbations. However, we have previously suggested that gravitational perturbations by Jupiter might mean that impacts sufficient to create chondrules occur preferentially near resonances (Symes *et al.*, 1998).

There is ample evidence that reduction accompanied chondrule formation. This was reviewed by Huang *et al.* (1996a). Perhaps the strongest evidence is the presence of Ni-poor metal (<5.5 wt% Ni, the cosmic proportion of Ni to Fe) in the refractory, low FeO-chondrules that are thought to have suffered the greatest alteration of their compositions during formation.

PUTTING THE PREMISES TOGETHER: A SCENARIO FOR CHONDRITE FORMATION

Figure 8 is a schematic diagram showing how these processes might occur in a dynamic regolith, using the LL chondrites as an example. Figure 9 shows similar proposals for the other ordinary and enstatite classes. Depending on their relative size and density, the chondrules and metal grains will generally become separated as gases pass through the surface layers. Separation is weakest for the H chondrites because their chondrules and metal grains require similar gas flow rates, and is strongest for the LL chondrites. For most classes, metal-rich surface and chondrule-rich subsurface

layers form. The ordinary chondrites and the EL chondrites are samples of the thick chondrule-rich layer; the EL chondrites are samples of a thick metal-rich layer. Therefore, there should be metal-rich layers on the LL (and L parent bodies) that have either not yet been sampled, or are represented among the numerous anomalous metal-rich chondrites, like ALH 85085. Metallographic cooling rates (Wood, 1979) are consistent with chondrite formation and metamorphism in the regolith and mega-regolith of a Hebe-sized asteroid, because of the significant amounts of insulation caused by a few kilometers or so of regolith (Akridge *et al.*, 1998).

The predicted metal-silicate ratio for this process, assuming ordinary chondrite chondrule and metal size distributions and calculated from empirical equations, is in good agreement with observations when flow velocities are just above the minimum flow rate required to sustain the layer (Huang *et al.*, 1996b). This is because conditions are "frozen in" when the gas flow drops below the minimum required rate.

A SCENARIO FOR MINERALOGICAL REACTIONS INVOLVING A GAS PHASE

Thus, we suggest that impact into a regolith could have produced chondrules and, perhaps associated with the same impact, aerodynamic and size sorting resulted in the chondrite classes. We now wish to explore whether any or all of the gas-phase reactions listed in Table 1 were associated with the same process.

Predicted temperatures in the regolith of asteroids range from ~160 to ~200 K (present-day blackbody temperatures) to melting temperatures (during impact for instance). Heating by decay of ²⁶Al

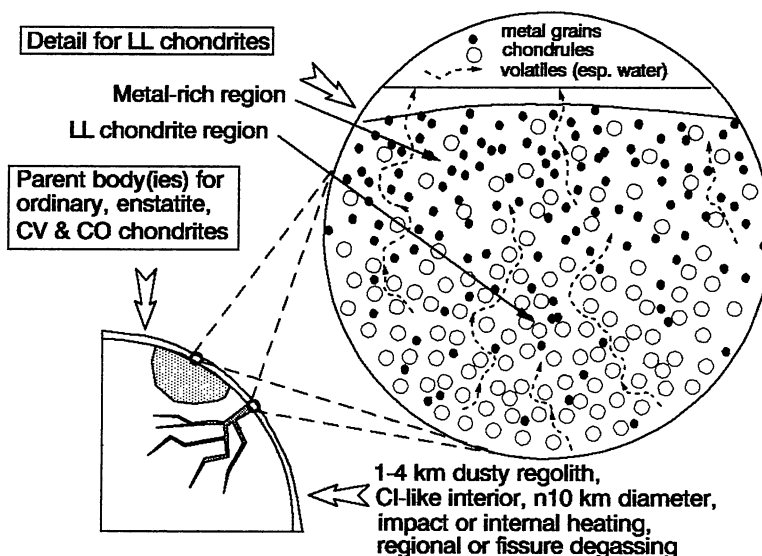


FIG. 8. Generalized schematic diagram describing the way in which metal silicate fractionation could occur in a dynamic regolith of the meteorite parent body; by way of example, the LL chondrite case is shown. The CI-like and, therefore, volatile-rich asteroid is heated by impact or internal heating, and the surface is dried-out. A several kilometer thick regolith forms. The volatiles rising from the interior on a regional basis or through a number of discrete vents mobilize the surface allowing large chondrules to sink through the dust layer and small metal grains to be carried to the surface. The degree of separation and the relative sizes of the metal-rich and chondrule-rich zones depend on the relative sizes of the metal grains and chondrules, and in this way, the extent of metal-silicate fractionation depends on the size ratio and will differ for each chondrite class. The process can be modeled in the laboratory and is similar to the industrially important fluidization process and certain forms of pyroclastic volcanism (Wilson, 1980). If the heat source is impact, the whole process could be linked to one or a few major impacts and to the formation of chondrules as crystallized impact melts.

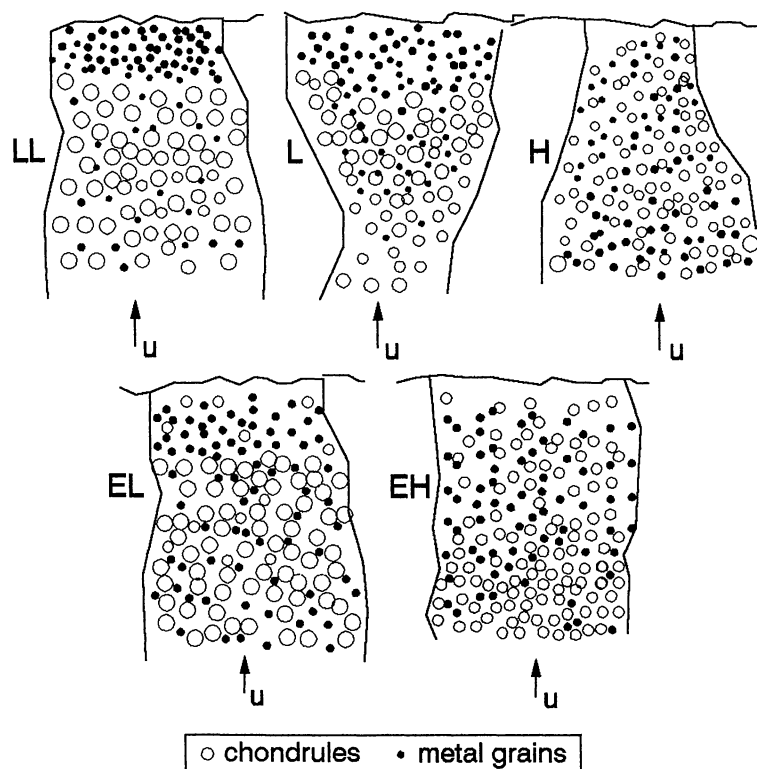


FIG. 9. Schematic diagrams showing how metal-silicate fractionation in a dynamic regolith could have given rise to the enstatite and ordinary chondrites. The "u" and the upward pointing arrow refer to gas flow through the regolith layer. The ordinary chondrite classes show a systematic decrease in the chondrule to metal size ratio so that the metal-silicate separation is largest for the LL chondrites, less for the L chondrites and virtually zero for the H chondrites. Thus, there are zones of LL and L chondrite material below a metal-rich surface layer, whereas the H chondrite regolith is essentially well mixed. Similar mechanisms existed in the vicinity of the enstatite chondrites, but the sampling details are different since EL chondrites come from the chondrule-rich layer beneath the surface; whereas, EH chondrites are samples of the metal-rich surface layers. The various layers not represented by the major classes may have been sampled as various anomalous meteorites, but it is highly unlikely that the Earth's sampling of the asteroids and their various regolith layers is even remotely representative. Not discussed in the text at any length is that CO and CV chondrites show very modest metal-silicate fractionations because they contain relatively little metal; whereas, CI and CM chondrites show even less fractionation and contain no metal and few chondrules. Metamorphic temperatures and cooling rates are consistent with these scenarios.

produces a steep temperature gradient throughout the regolith, so that if the surface is at black body temperature, the base of a 2.5 km thick regolith reaches ~ 1200 K. Impact also produced significant localized heating. Impact-melt clasts have been found in many chondrites (indicating temperatures of >1300 K; *e.g.*, Rubin, 1995), and many anomalous meteorites are thought to be impact-melt rocks (*e.g.*, Zhang *et al.*, 1995; Rubin, 1985, and references therein). Impact heating may have even caused the often-discussed metamorphism of chondrites (Stöffler *et al.*, 1991). More than 50% of the Apollo rocks are impact-melt rocks, whereas 50 vol% of the remainder are localized impact melts (agglutinates and spherules, Hörz *et al.*, 1991).

The flux of water vapor through a thick regolith depends on the nature of the heat source, but heating of asteroids of essentially CI or CM chondrite composition by any mechanism would release considerable quantities of water. The CI and CM chondrites are ~ 17 and $\sim 10\%$ chemically bound water, respectively (Jarosewich, 1990). Significant amounts of CO_2 , sulfur compounds, and other minor components also would have been present. The vapor pressure of

water at 330 and 630 K is 0.17 and 180 bar, respectively (Weast *et al.*, 1986). Thus a dynamic regolith would contain H_2O - and CO_2 -rich vapors at high temperatures and pressures. If, as we suspect, chondrule formation also occurred at this stage of parent body evolution, then elements like S, Na, K, and lesser amounts of Fe and Si also would have been present in the vapor.

Oxygen isotopes also might be fractionated in a volatile-rich regolith. Some original heterogeneity might be expected due to presolar components (Nittler *et al.*, 1994), but these components were probably rare. The mechanisms for fractionating O isotopes are numerous, but all or most of the proposed mechanisms are just as likely to have occurred in a regolith as in the open nebula, perhaps more so because particle and gas densities would be much higher in a regolith. A large number of mechanisms and processes have been discussed. These are (1) nonmass-dependent fractionations (Thiemens, 1996), (2) mass-dependent volatilization (Clayton *et al.*, 1985), (3) mass-dependent reactions with ambient gases (Sears and Weeks, 1983; Clayton *et al.*, 1991), and (4) mass-independent mixing and exchange with ambient fluids and adjacent solids (Clayton *et al.*, 1983, 1991).

Chemical reactions could occur in the regolith at any time, including the present day, although most reactions will probably be associated with ^{26}Al heating or early transient heating events. Most impact heating events would probably have occurred during the active early phase of solar system history, as relative velocities reached several kilometers per second but before the inner solar system was swept of most of its material. Chondrule ages, typically several million years after the first-formed solar system solids, might mark this period of maximum surface activity for asteroids.

One of the most perplexing aspects of the origin of chondrites is that the process occurred in a highly reducing environment to produce metal and reduced chondrules, and yet the environment was oxidizing enough to leave many chondrules and the matrix with high amounts of oxidized Fe. The levels of reduction required to produce metal are consistent with nebula conditions, whereas the chondrules and metal-silicate fractionation are clearly

more consistent with regolith conditions. For an origin of chondrites by surface processes on asteroids, the important question is how can impact of CI-like target material produce the copious amounts of metal observed in, say, H or EH chondrites? Several possibilities exist. The target could be more reduced than CI chondrites, with higher levels of C or H. The CI chondrites are not as close to solar in composition as comet nuclei, and comet nuclei have higher C/O and H/O ratios than CI chondrites (Grün and Jessberger, 1990). Second, the impact process could be highly heterogeneous, with regions of atypically high levels of reduction resulting in H and EH chondrites and regions of atypically high levels of oxidation resulting in CI chondrites.

SUMMARY AND CONCLUSIONS

For many of the proposed nebular reactions involving aqueous alteration, there would seem to be little reason to preclude alteration on a parent body. Phyllosilicates are stable <500 K but theoretical condensation calculations have trouble predicting the observed hydrated phases and abundances in carbonaceous chondrites. The

water pressure required for most of the proposed nebular reactions are not usually discussed; however, they seem to require localized water pressures significantly greater than the solar value (e.g., Hua and Buseck, 1995).

It seems plausible that many gas-phase reactions could have taken place upon accretion of the material to a parent body or subsequently, during parent-body degassing, making it difficult to discern a pure parent body or nebular origin for most meteorite properties. Huang *et al.* (1996b) suggested that degassing of volatiles during accretion by impact or radiogenic heating would create a nebularlike environment in which gases (mainly water) would be enriched relative to their solar values. Wegner and Ernst (1983) studied the hydration and dehydration rates for serpentine as a function of fluid pressure. Hydration at 273 K could occur in $\sim 10^4$ years or for 298 K in only a year. Dehydration rates were reported also to be rapid and may explain the redox conditions for ordinary chondrites if their parent body(ies) were heated more extensively than the carbonaceous parent bodies.

Acknowledgments—We are grateful for constructive reviews by J. S. Lewis, M. E. Lipschutz, and an anonymous reviewer.

Editorial handling: W. K. Hartmann

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