Effects of dust enrichment on oxygen fugacity of cosmic gases

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Abstract–The degree to which dust enrichment enhances the oxygen fugacity (fO$_2$) of a system otherwise solar in composition depends on the dust composition. Equilibrium calculations were performed at 10^{-3} bar in systems enriched by a factor of 10^4 in two fundamentally different types of dust to investigate the iron oxidation state in both cases. One type of dust, called SC for solar condensate, stopped equilibrating with solar gas at too high a temperature for FeO or condensed water to be stabilized in any form, and thus has the composition expected of a nebular condensate. The other has CI chondrite composition, appropriate for a parent body that accreted from SC dust and low-temperature ice. Upon total vaporization at 2300 K, both systems have high fO$_2$, >IW. In the SC dust-enriched system, FeO of the bulk silicate reaches ~10 wt% at 1970 K but decreases to <1 wt% below 1500 K. The FeO undergoes reduction because consumption of gaseous oxygen by silicate recondensation causes a precipitous drop in fO$_2$. Thus, enrichment in dust having the composition of likely nebular condensates cannot yield a sufficiently oxidizing environment to account for the FeO contents of chondrules. The fO$_2$ of the system enriched in water-rich, CI dust, however, remains high throughout condensation, as gaseous water remains uncondensed until very low temperatures. This allows silicate condensates to achieve and maintain FeO contents of 27–35 wt%. Water-rich parent bodies are thus excellent candidate sources of chondrule precursors. Impacts on such bodies may have created the combination of high dust enrichment, total pressure, and fO$_2$ necessary for chondrule formation.
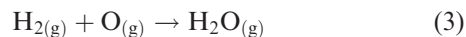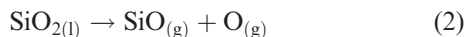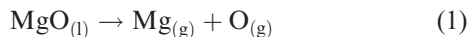
INTRODUCTION

The bulk FeO contents of type I and type II chondrules are <10 wt% and >10 wt%, respectively, in ordinary and carbonaceous chondrites (McSween 1977; Jones 1990, 1994). It has been known for decades that formation of such FeO-rich chondrules requires an environment with O/H ratio much higher than that of solar gas to reach high FeO concentrations in silicates (Wood 1967). High abundances of hydrogen and carbon relative to oxygen (Allende Prieto et al. 2002) make solar gas very reducing at high temperatures, and this causes condensate grains of olivine and pyroxene to contain vanishingly small amounts of FeO. In a system of solar composition, significant amounts of FeO can be stabilized only below 900 K (Grossman 1972; Fedkin and Grossman 2006). Below that temperature, however, diffusion in olivine is too slow for its fayalite content to

reach a significant value in solar nebular cooling times. Even if a system otherwise solar in composition is enriched in water by a factor of 10 and the grain radii are only 0.1 μm, the mean X_{Fa} reaches only 0.019 when the temperature drops to almost 700 K (Grossman et al. 2012). Wood (1967) proposed a local enhancement of the dust-to-gas ratio as a solar nebular mechanism for producing a sufficiently high oxygen fugacity at high temperature to stabilize significant FeO contents of silicate condensates. When chondritic dust condenses from solar gas, it removes a significant fraction of the oxygen atoms from the gas but leaves almost all the hydrogen and carbon behind. Thus, concentrating dust relative to gas creates regions enriched in oxygen relative to hydrogen and carbon compared to solar composition (Grossman et al. 2008). When such a region is heated, the dust melts and undergoes evaporation, during which oxygen is separated from its

parent oxides and reacts with hydrogen to increase the water/hydrogen ratio, and thus the oxygen fugacity, of the dust-enriched region by reactions such as:

$$MgO_{(l)} \rightarrow Mg_{(g)} + O_{(g)} \tag{1}$$

$$SiO_{2(l)} \rightarrow SiO_{(g)} + O_{(g)} \tag{2}$$

$$H_{2(g)} + O_{(g)} \rightarrow H_2O_{(g)} \tag{3}$$

where "l" is liquid and "g" is gaseous.

Although high dust enrichments certainly increase oxygen fugacity at high temperature, the dust composition is an important factor in determining how oxidizing the system becomes. Wood and Hashimoto (1993) considered dust compositions containing three chondritic components of increasing volatility, dust, tar, and ice, altogether containing solar relative abundances of oxygen and carbon. Yoneda and Grossman (1995) and later Fedkin and Grossman (2006) used "OC dust," in which elements were assumed to be in solar proportion, as in ordinary chondrites, but whose oxygen abundance is in stoichiometric proportion to all the metals except for metallic Ni, Co, Cr, and the fraction of Fe bound to S. The OC dust composition contains no water or metallic Fe. It corresponds to no known meteorite composition, and there is no known way to make this composition in the solar nebula. In their study of dust enrichment, Ebel and Grossman (2000) calculated a "CI dust" composition from the solar abundances of Anders and Grevesse (1989) for all elements except for H, O, C, and N, and from the mass ratios of the latter elements to Si in Orgueil (Anders and Grevesse 1989). In systems highly enriched in CI dust relative to solar composition, Ebel and Grossman (2000) showed that the FeO content of the bulk silicate increases with falling temperature, and remains at high values until the temperature falls to at least 1200 K. Fedkin et al. (2015) noticed that, in an environment highly enriched in dust of very reduced composition, the oxygen fugacity is high at high temperature, resulting in relatively high FeO contents in silicates, but falls with falling temperature, causing the silicate to undergo reduction. Fedkin et al. (2015) further showed that adding water to the investigated compositions allows the system to maintain its high fO$_2$ at low temperatures, preserving the relatively high FeO content of the silicate.

The purpose of this paper is to show the difference in oxidation state of condensates produced in dust-enriched environments between two cases (1) when the precursor dust composition is that expected for solar nebular condensates and (2) when the dust composition is that of a CI chondrite, representative of a water-rich parent body. A preliminary version of this work was presented by Grossman and Fedkin (2015).

TECHNIQUE

Ebel and Grossman (2000) showed that the oxygen fugacity of cosmic gases is insensitive to total pressure at silicate condensation temperatures. The VAPORS program (Ebel and Grossman 2000) was used to perform equilibrium condensation calculations at a total pressure of 10^{-3} bar. Calculations were started at 2400 K. The Anders and Grevesse (1989) compilation of the relative atomic abundances of the elements in the solar system was used, except for that of S, which was taken from Lodders (2003), and for those of O and C, which were taken from Allende Prieto et al. (2001, 2002), respectively. Two contrasting dust compositions were considered. The first, solar condensate (SC), is the composition of the condensate assemblage that is expected to exist in a nebula of solar composition and is appropriate to condensation calculations relevant to dust-enriched nebular regions. It contains no FeO because of the very slow diffusion rate of FeO in olivine below 900 K (Fedkin and Grossman 2006), the temperature at which FeO becomes stable in solar gas. All iron exists as metal or sulfide (Table 1). Otherwise, SC is assumed to form by equilibration to a low enough temperature that it contains all of the sulfur in solar gas, but no C, N, or water because of their volatility. SC is probably the composition of the nebular condensate assemblage that accreted into the first generation of planetesimals. The other dust has CI chondrite composition. It contains no metallic iron; all iron is present as either sulfide or FeO, and it contains a large amount of water (Table 1). CI chondrites may have formed by geochemical processing of planetesimals made of SC dust and late-accreting water ice. A dust enrichment factor of 10^4 was assumed for both dust compositions in order to ensure that significant amounts of oxidized iron are stabilized even in the system enriched in anhydrous dust. Compositions of systems enriched in dust of these compositions by a factor of 10^4 were calculated by mixing nebular gas complementary in composition to each with $10^4\times$ more dust than in solar composition. The two system compositions are identical except for contents of the volatile elements (Table 2). Note that the system enriched $10^4\times$ in CI dust contains 2.3 times as many oxygen atoms per 10^6 Si atoms as the one enriched by the same factor in SC dust, primarily because of the water content of CI chondrite dust.

Table 1. Dust compositions (wt%) employed in this work.

	SC[a]	CI[b]
SiO_2	32.78	24.15
TiO_2	0.10	0.08
Al_2O_3	2.36	1.74
CaO	1.87	1.38
MnO	0.37	0.27
MgO	23.62	17.40
Cr_2O_3	0.56	0.41
Na_2O	0.97	0.71
K_2O	0.10	0.07
P_2O_5	0.40	0.30
FeO[c]	**0.00**	**13.12**
FeS	**21.38**	**15.75**
Fe	**13.84**	**0.00**
Ni	1.58	0.00
Co	0.07	0.00
NiO	0.00	1.48
CoO	0.00	0.07
H_2O	**0.00**	**19.10**
C	0.00	3.66
N	0.00	0.34

[a]See text.

[b]The CI dust concentrations are taken from Ebel and Grossman (2000) for all the elements, except for S abundance adopted from Lodders (2003).

[c]Critical composition contrasts highlighted in boldface.

Table 2. System compositions (atoms per 10^6 Si atoms).

	$10^4 \times$ SC	$10^4 \times$ CI
Si	1.000E+06	1.000E+06
Ti	2.400E+03	2.400E+03
Al	8.490E+04	8.490E+04
Ca	6.110E+04	6.110E+04
Mn	9.550E+03	9.550E+03
Mg	1.074E+06	1.074E+06
Cr	1.350E+04	1.350E+04
Na	5.740E+04	5.740E+04
K	3.770E+03	3.770E+03
Fe	9.000E+05	9.000E+05
Ni	4.930E+04	4.930E+04
Co	2.250E+03	2.250E+03
S	4.458E+05	4.458E+05
H	2.790E+06	8.069E+06
He	2.720E+05	2.720E+05
C	6.849E+02	7.566E+05
N	3.130E+02	6.011E+04
P	1.040E+04	1.040E+04
O	3.355E+06	7.631E+06

RESULTS

In the system enriched in CI dust, a ferromagnesian silicate liquid (MELTS; Ghiorso and Sack 1995) becomes stable at 2300 K. This phase accounts for 85% of the total iron when the temperature falls to 1880 K (Fig. 1a), where olivine (Fa_9) begins to crystallize from the liquid. Metallic nickel-iron begins to condense at 1800 K. Spinel forms at 1700 K and clinopyroxene containing ~6 wt% Al_2O_3 and ~7 wt% FeO at 1440 K, together consuming less than 3% of the total Fe. The spinel composition varies between Al_2O_3 ~31, Fe_2O_3 ~3, Cr_2O_3 ~37, FeO ~11, and MgO ~17 wt% at 1700 K to Al_2O_3 ~49, Fe_2O_3 ~2, Cr_2O_3 ~16, FeO ~16, and MgO ~16 wt% at 1400 K. Below 1575 K, after all the iron has condensed, the ratio of oxidized to reduced iron gradually increases with falling temperature. The FeO content of the liquid increases continuously with falling temperature, as Fe condenses as FeO under such oxidizing conditions, eventually reaching over 60 wt% at 1400 K (Fig. 2). The mole fraction fayalite in the first-crystallizing olivine is 0.09, and rises to over 0.25 with falling temperature. The FeO content of the bulk silicate is ~33 wt % at 1880 K, and reaches ~35 wt% at 1400 K.

In the SC dust-enriched system (Fig. 1b), olivine begins to precipitate from the ferromagnesian liquid at 2000 K. Oxidized iron accounts for only 17% of the total iron in the system when metallic iron becomes stable at 1960 K. Olivine begins to react with silicate melt to form orthopyroxene at 1760 K, and spinel (not seen in Fig. 1b at this scale) forms at 1730 K. Once the metallic iron condenses, the fraction of the iron in the system that is oxidized falls continuously, reaching only 2% by 1600 K. This behavior is reflected in the compositions of both liquid and solid silicates. The FeO content of the liquid and the fayalite mole fraction reach maxima of 12 wt% and 0.05, respectively, when metallic iron condenses (Fig. 2). Thereafter, the FeO content of the liquid falls to below 1 wt% and the mole fraction fayalite to 0.02 by the time the temperature falls to 1600 K. Despite the very high dust enrichment, reduction of FeO is occurring. The FeO content of the bulk silicate peaks at ~10 wt% at 1970 K, and then falls to ~0.7 wt % at 1400 K.

At 1600 K, metallic FeNi is predicted to begin to react with gaseous S to form pyrrhotite. Because this temperature is well above the minimum melting temperature in the Ni-poor part of the Fe-Ni-S system, a sulfide liquid should be stable instead but its appearance temperature is not rigorously defined due to the absence of a solution model for Fe-Ni-S liquids from the VAPORS code. Thus, in Fig. 1b, a vertical line is drawn at the temperature where sulfur is predicted to begin to condense, illustrating schematically that metallic FeNi and Fe-Ni-S liquid are expected to coexist below that temperature.

Figure 3 shows the oxygen fugacity relative to the iron-wüstite (IW) buffer as a function of temperature in
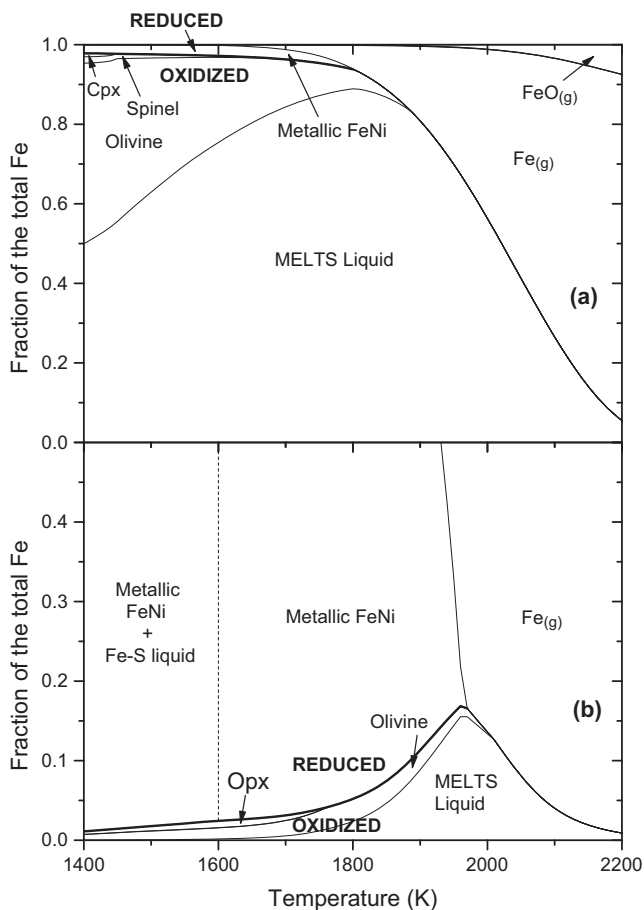
Fig. 1. Equilibrium distribution of iron calculated for a system enriched in (a) CI and (b) SC dust by a factor of 10^4 relative to its complementary solar nebular gas at $P^{tot} = 10^{-3}$ bar. The bold curve separates the fields of oxidized and reduced iron. The dashed vertical line in (b) separating schematically the stability field of metallic FeNi and that of coexisting metallic FeNi and Fe-Ni-S liquid is drawn at 1600 K (see text for details) because the model does not include properties for Fe-Ni-S liquid. MELTS liquid—ferromagnesian silicate liquid (Ghiorso and Sack 1995); Cpx = clinopyroxene; Opx = orthopyroxene.
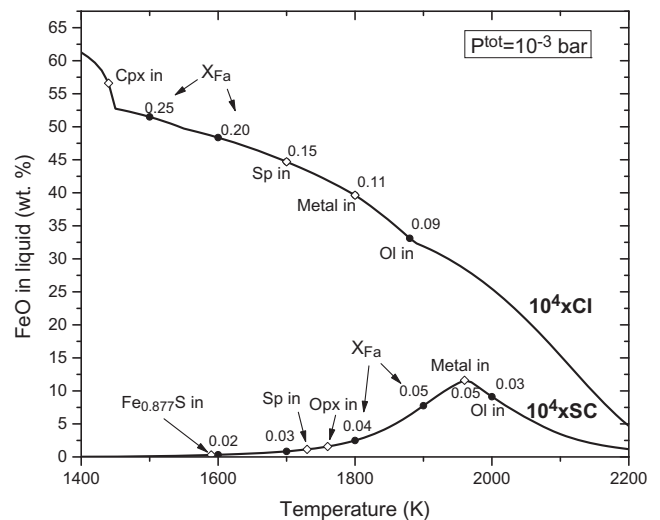

Fig. 2. The FeO content of the silicate liquid as a function of temperature in systems enriched in CI and SC dust by a factor of 10^4 relative to its complementary gas at $P^{tot} = 10^{-3}$ bar. Open diamonds indicate the condensation temperatures of the phases; filled circles and numbers next to them are the mole fraction fayalite (X_{Fa}) in the coexisting olivine. Both the FeO content of the liquid and X_{Fa} increase continuously with decreasing temperature in the CI dust-enriched system. In contrast, in the SC dust-enriched system, the FeO content of the liquid and X_{Fa} reach a maximum at 1960 K, the metal condensation temperature, and then both fall as temperature falls.   Sp = spinel;   Ol = olivine;   Cpx = clinopyroxene; Opx = orthopyroxene.

both systems. In the CI dust-enriched system, the oxygen fugacity is about 0.5 log units above IW (IW+0.5) at 2400 K, increases to IW+0.8 at 2280 K, and then falls to IW−0.1 at 1820 K as the silicate liquid and olivine condense. Below 1820 K, log fO_2 relative to log IW gradually rises, reaching IW+0.2 at 1400 K. In the SC dust-enriched system, the oxygen fugacity is IW+0.1 at 2400 K, rises to IW+0.3 at 2320 K, but, in contrast to the CI dust-enriched system, falls steeply with falling temperature, reaching IW−3.9 at 1400 K. In comparison, a system enriched in OC dust (Fedkin and Grossman 2006) is more oxidizing than the SC dust-enriched system at low temperatures, even when OC dust is enriched only 1000× relative to solar

composition. In the latter case, the fO_2 of the system is ~IW−2.7 at 1400 K and keeps rising with falling temperature (Fedkin and Grossman 2006).

DISCUSSION

Oxygen Fugacity and Oxygen Distribution in the $10^4 \times$ CI versus $10^4 \times$ SC Systems

The reason why dust-enriched systems become more oxidizing at high temperature is that free oxygen is released when the dust is evaporated (Grossman et al. 2008). When CI dust is enriched by a factor of 10^4 and then totally evaporated, only 80 gaseous oxygen atoms per 10^6 in the bulk system are from the original solar nebular gas; 65% of them are from evaporation of metal oxides, such as MgO and SiO_2 (Equations 1 and 2, respectively), and 35% of them are from evaporation of water. A large fraction of the total gaseous oxygen that existed when all the dust was evaporated is consumed when the metal oxides recondense into high–temperature, solid and liquid silicates (Fig. 4a). But when this process is nearly complete at 1400 K, 30% of the oxygen atoms are still present in the gas as molecular water. In contrast, when SC dust is enriched
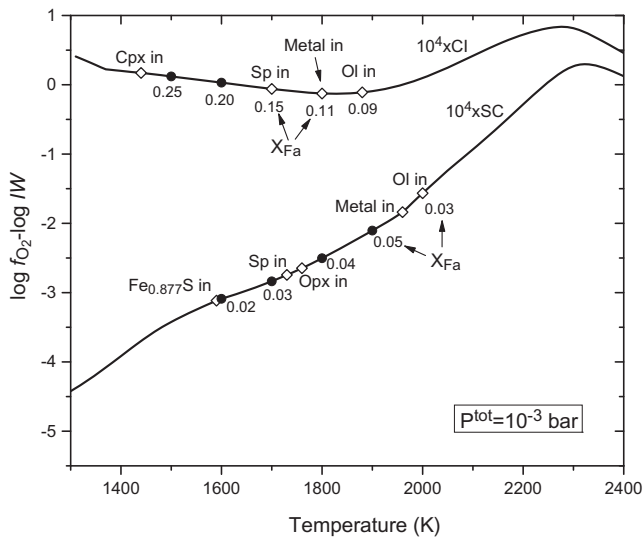
Fig. 3. Logarithm of the oxygen fugacity relative to the iron-wüstite (IW) buffer as a function of temperature in systems enriched in CI and SC dust by a factor of 10^4 relative to their complementary gas at $P^{tot} = 10^{-3}$ bar. Symbols and labels are as in Fig. 2.

by a factor of 10^4 and then totally evaporated, 99.97% of all the gaseous oxygen atoms in the resulting system come from evaporation of the dust at high temperature; the rest are from the original nebular gas, and none are from evaporation of water. Upon recondensation, the silicate liquid consumes about 80% of the total oxygen by 2010 K, while the rest of the gaseous oxygen condenses virtually entirely into both liquid and solid silicates by 1400 K (Fig. 4b). The field of gaseous oxygen is drastically narrowed between 2400 K and ~1700 K, and then more gradually to 1400 K, at which point only a negligible fraction of the total oxygen is still gaseous.

Figure 5 shows the logarithm of oxygen fugacity relative to IW as a function of the fraction of the oxygen condensed. Both curves show that the oxygen fugacity of the residual gas decreases as the fraction of the oxygen consumed by condensates increases. Because the water in the CI dust-enriched system does not condense over the temperature range where silicates condense, more than 40% of the total oxygen is uncondensed after silicate condensation. The fO_2 thus remains high enough (above IW) at low temperature (<1600 K) not only to preserve the FeO made at high temperature (e.g., ~15 wt% FeO in bulk silicate at 2100 K) but also to increase the FeO content of the condensate. In contrast, in the SC dust-enriched system, only a vanishingly small fraction of the oxygen remains uncondensed when the silicates recondense. In this case, although the oxygen fugacity is high enough to oxidize some iron at high temperature, the fO_2 falls sharply,
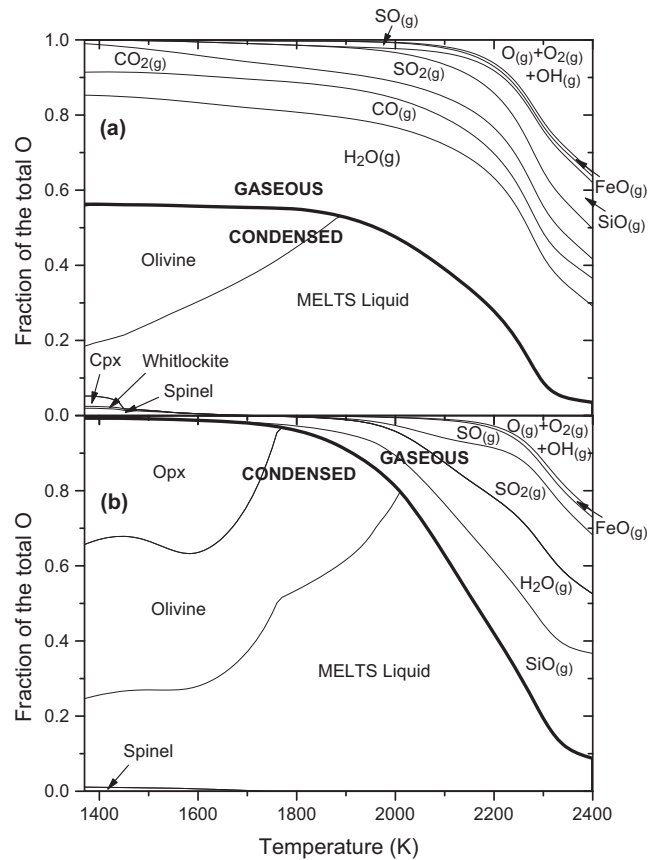

Fig. 4. Equilibrium distribution of oxygen calculated for a system enriched in (a) CI and (b) SC dust by a factor of 10^4 relative to its complementary gas at $P^{tot} = 10^{-3}$ bar. The bold curves separate the fields of condensed oxygen from gaseous oxygen.

approaching solar gas values, as the silicates recondense. This is why the FeO stabilized at high temperature undergoes reduction with falling temperature in this system.

Conditions for Chondrule Formation

To prevent evaporation of Na from chondrule melts at liquidus temperatures (Alexander et al. 2008; Hewins et al. 2012), Fedkin and Grossman (2013) showed that chondrule formation must have occurred at combinations of dust enrichment and total pressure that were orders of magnitude higher than those found to be possible in dynamical models of a canonical solar nebula. As had been suggested earlier by, for example, Campbell et al. (2002) and Fedkin et al. (2012), Fedkin and Grossman (2013) proposed instead that chondrule formation occurred inside vapor plumes resulting from planetesimal impacts, where the required combination of dust concentrations and total pressures seems more plausible. Although the analytical evidence presented by
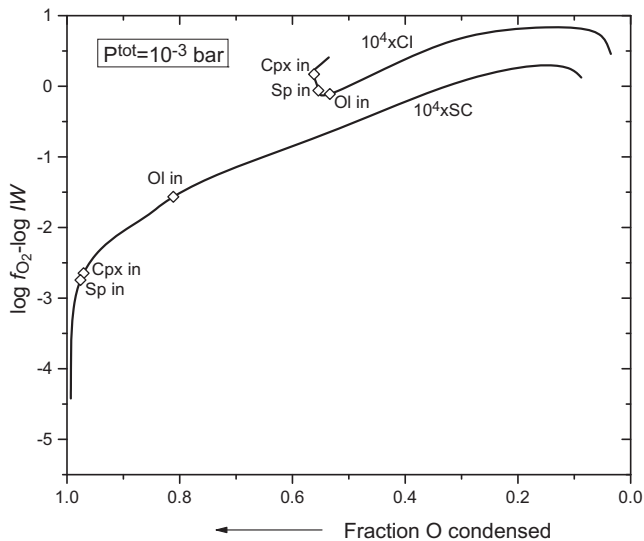
Fig. 5. Logarithm of the oxygen fugacity relative to the iron-wüstite (IW) buffer plotted against the fraction of the oxygen condensed in systems enriched in CI and SC dust by a factor of 10^4 relative to their complementary gas at $P^{tot} = 10^{-3}$ bar. The shown temperature range is 1300–2400 K for both curves. Symbols and labels are as in Fig. 2.

Alexander et al. (2008) for Na retention at the liquidus temperature of type I chondrules is much less definitive than for type II chondrules, it is inconceivable that objects on different sides of an arbitrary division within a continuous chemical series have different origins.

Another property of chondrules that cannot be accommodated in canonical solar nebular models, due to the extremely low oxygen fugacity of solar gas, is their relatively high FeO contents. To address this problem, various means have been hypothesized to increase the oxygen fugacity in a high-temperature gas of otherwise solar composition. As reviewed by Fedkin and Grossman (2006) and Grossman et al. (2012), these involve increasing the concentrations of dust or water in a localized region of the solar nebula by physical means, but the processes that have been investigated that are relevant to conditions in a protoplanetary disk invariably yield dust or water enrichments far smaller than those required for stabilization of observed FeO contents. It should be noted that, when CI dust is enriched by a factor of 120 relative to solar composition, the maximum dust enrichment achieved in gravitational settling models (Cassen 2001), a kinetic treatment analogous to that presented in fig. 10 of Grossman et al. (2012) reveals that fayalite contents of 15 mole% can be reached by condensate olivine grains one micron in radius for nebular cooling times of 10^4 yr. The problem is that there is no known way of making hydrated silicates by solar nebular condensation, and CI composition is widely recognized

as being the result of parent body hydration reactions. The likely composition of dust available to be concentrated by solar nebular processes, SC, is one that resulted from cessation of equilibration with solar gas at high temperature. Enrichment of SC dust by a factor of 120 relative to solar composition is inadequate to make olivine condensate grains that could serve as suitable precursors even for most type I chondrules. The point of the present work is that, even if a way could be found to produce enrichments of such dust great enough to stabilize FeO in silicates recondensed at high temperature, these FeO contents would fall dramatically due to reduction accompanying continued consumption of gaseous oxygen into condensing silicates with falling temperature.

Only if there is abundant water in the precursor dust, as in the CI composition, does the fO_2 of the system stay sufficiently high during silicate recondensation to preserve FeO contents of high-temperature silicates that are high enough to match those of both type I and type II chondrules. In the system enriched by a factor of 10^4 in CI dust at a P^{tot} of 10^{-3} bar, the P_{H_2O}/P_{H_2} ratio varies between 1 and 2, depending on temperature. This is several thousand times higher than in a gas of solar composition at the same total pressure and temperature. Just as there is no known dynamic mechanism for enriching dust by more than a factor of ~120 in a region of the solar protoplanetary disk (Cassen 2001), no way is known for enriching a solar nebular region in water ice by more than a factor of 10 (Ciesla and Cuzzi 2006).

Water ice, however, is a very low-temperature condensate that probably formed well after accretion of SC dust began, and undoubtedly accreted into planetesimals. The existence of asteroids of carbonaceous chondrite composition (Burbine et al. 2008) is *prima facie* evidence that water-rich planetesimals of this composition existed in the early solar system. Impacts on such water-rich bodies would have created plumes highly enriched in dust and water, a highly favorable environment for producing and preserving the relatively high FeO contents of chondrules. To yield the chemical effects discussed herein, it makes no difference whether the water is present as ice or within the crystal structures of hydrated silicates at the time of impact, provided that the water content of the ice-rock mixture is that of CI chondrites. Chondrule formation requires a combination of relatively high total pressures and high dust enrichments, which may be found in impact plumes. The oxidation state of chondrules is another feature that is more likely to have originated in plumes generated by impacts between planetesimals than in processes in the protoplanetary disk.

CONCLUSIONS

The SC dust composition, which contains no water or FeO, represents the likely composition of the pre-accretionary condensate that existed in the solar nebula, and is the appropriate dust composition to use in condensation calculations relevant to dust-enriched nebular regions. Vaporization of systems enriched in such dust relative to the gas compared to solar composition, even by factors of 10^4, yields oxidizing conditions at high temperature, but subsequent condensation leads to significant reduction at lower temperatures. The bulk silicate FeO content reaches 10 wt% at 1970 K but falls to <1 wt% at 1400 K in a system enriched in SC dust by a factor of 10^4 relative to solar composition at $P^{tot} = 10^{-3}$ bar. Vaporization of systems enriched in water-rich CI dust by the same factor yields oxidizing conditions that are maintained throughout cooling and silicate recondensation. Thus, oxidizing conditions in the former case are only transient, and reduction results in a limited range of FeO contents, whereas in the latter case, reduction does not occur, and FeO contents of condensates span the entire range of chondrule values. As CI chondrites probably formed by interaction of cometary ice with SC dust inside planetesimals, the need for abundant water in the precursor dust to chondrules suggests that impacts on such planetesimals had a role in their formation. Such an environment is also suggested by the combination of high total pressures and dust enrichments needed to retain sodium at chondrule liquidus temperatures.

Editorial Handling—Dr. Ed Scott

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