Formation of the first oxidized iron in the solar system

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Abstract–For fayalite formation times of several thousand years, and systems enriched in water by a factor of ten relative to solar composition, 1 μm radius olivine grains could reach 2 mole% fayalite and 0.1 μm grains 5 mole% by nebular condensation, well short of the values appropriate for precursors of most chondrules and the values found in the matrices of unequilibrated ordinary chondrites. Even 10 μm olivine crystals could reach 30 mole% fayalite above 1100 K in solar gas if condensation of metallic nickel-iron were delayed sufficiently by supersaturation. Consideration of the surface tensions of several phases with equilibrium condensation temperatures above that of metallic iron shows that, even if they were supersaturated, they would still nucleate homogeneously above the equilibrium condensation temperature of metallic iron. This phenomenon would have provided nuclei for heterogeneous nucleation of metallic nickel-iron, thus preventing the latter from supersaturating significantly and preventing olivine from becoming fayalitic. Unless a way is found to make nebular regions far more oxidizing than in existing models, it is unlikely that chondrule precursors or the matrix olivine grains of unequilibrated ordinary chondrites obtained their fayalite contents by condensation processes. Perhaps stabilization of FeO occurred after condensation of water ice and accretion of icy planetesimals, during heating of the planetesimals and/or in hot, dense, water-rich vapor plumes generated by impacts on them. This would imply that FeO is a relatively young feature of nebular materials.

INTRODUCTION

Oxidized iron is an abundant constituent of both chondrules and matrices of primitive chondrites. Olivine grains in the matrices of the least equilibrated ordinary chondrites have a wide range of FeO contents (e.g., Klöck et al. 1989; Brearley and Jones 1998) but may be extremely FeO-rich, with mean fayalite contents from 25 to 50 mole%, as shown by the data from Huss et al. (2006) in Fig. 1. Fedkin et al. (2012) obtained olivine composition histograms by conducting random analytical traverses across individual chondrules. In type I chondrules in primitive chondrites (Fig. 2), the mean fayalite contents are 1 to 4 mole%. In type II chondrules from Semarkona (Fig. 3), the mean fayalite contents are 12 to 20 mole%.

Despite the fact that chondrites are so ancient that they contain inclusions of solar nebular condensate assemblages, no nebular environment has yet been found where the high FeO contents of chondrites can be accounted for by condensation processes. This paper reviews and updates efforts to do so, and suggests alternatives to how and when the first solid form of FeO appeared in the solar system.

HIGH-TEMPERATURE CONDENSATION OF FEO-BEARING LIQUIDS

Ebel and Grossman (2000) showed that a stable condensate assemblage of olivine plus silicate liquid appears at high temperature in a gas of solar composition at extremely high nebular total pressures. In the present work, the same computer program and input data were used as in that effort, a total pressure of 10^{-1} bar was assumed, and sufficient water was added to solar gas composition to stabilize significant amounts of FeO in the condensate at high temperature. The result is shown in Fig. 4. Note that the necessary water enrichments are very
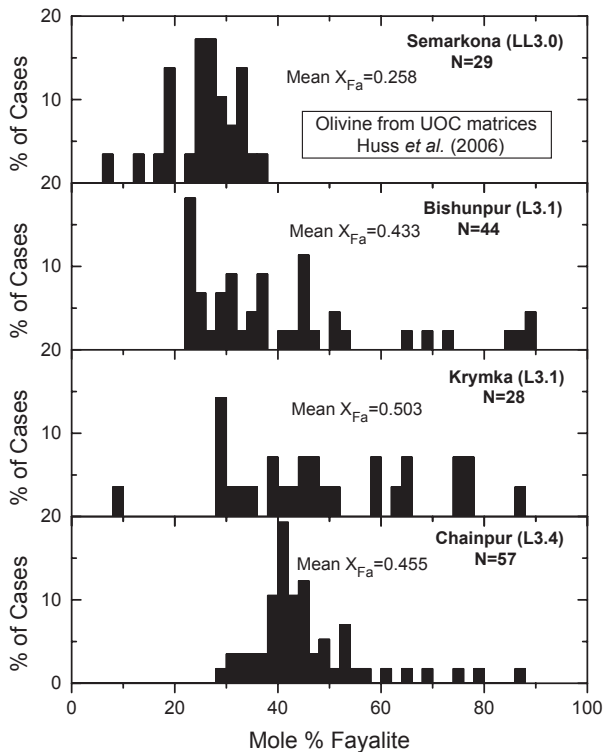
Fig. 1. Huss et al. (2006) found that the mean fayalite content of olivine grains in the matrices of primitive ordinary chondrites ranges from 25 to 50 mole%. X_{Fa}-mole fraction fayalite.

substantial, amounting to several hundred times the water content of a high-temperature gas of solar composition. These water enrichments yield gases with fo_2 between IW-1 and IW-2, i.e., oxygen fugacities between 1 and 2 log units below the iron-wüstite buffer, where metallic iron is in equilibrium with pure FeO. The difference in water enrichment needed, however, to stabilize the fayalite content of olivine typical of a type I chondrule versus that of a type II is quite small. For example, at 1600 K, the equilibrium fayalite mole fraction for 240× water enrichment is 0.07, a type I value, but for 470×, only a factor of two more, the fayalite mole fraction is 0.16, a type II value. Note also that, for a constant water enrichment of 470×, the equilibrium fayalite content varies from type I values at temperatures above ~1675 K to type II values below this temperature.

THE FAYALITE CONTENT OF CONDENSATE OLIVINE CRYSTALS

Previous Work

Shown in Fig. 5 is a plot of fo_2 versus temperature for solar system objects. The iron-wüstite curve, labeled IW, is near the top of the diagram. Had chondrules equilibrated with a gas that was solar in composition except for enrichment in water, their FeO contents imply that the oxygen fugacity was only one or two log units below IW. The maximum difference in oxygen fugacity implied by the different fayalite contents of olivine in types I and II chondrules is only one log unit which, on this scale, is very small. A much more serious problem is that a gas of solar composition, marked by the dark shaded band in Fig. 5, lies five log units lower in oxygen fugacity than this. The fact that melilite-, spinel-bearing, calcium-, aluminum-rich inclusions (CAIs) whose fassaitic clinopyroxene has $Ti^{3+}/(Ti^{3+} + Ti^{4+})$ ratios of about 0.5 formed at this oxygen fugacity is proof that some components of chondrites formed in a gas of solar composition (Grossman et al. 2008). In such a gas, nearly all magnesium and silicon condense as forsterite and enstatite, and iron condenses virtually totally as the metal phase, all at temperatures just below the condensation temperatures of the minerals of CAIs, according to equilibrium thermodynamic calculations (Grossman 2010). High abundances of hydrogen and carbon relative to oxygen make solar gas very reducing at high temperature, and this causes the olivine and pyroxene to contain only vanishingly small amounts of FeO. The reason why so much water has to be added to solar gas to account for the fayalite contents of chondrule olivine is to make up the huge difference in oxygen fugacity between the solar gas curve and the chondrule field in Fig. 5.

As seen in Fig. 6a, even solar gas becomes slightly more oxidizing below 800 K, and this stabilizes some FeO at the expense of metallic iron, allowing the equilibrium fayalite content of olivine to become significant (Fig. 6b). Because all the Mg and Si already condensed at a much higher temperature, however, this would have had to take place by diffusion of Fe^{2+} into the pre-existing forsterite grains. The problem with this is that the Fe-Mg interdiffusion rate in olivine is very low at these temperatures, so the equilibrium fayalite contents shown in Fig. 6b cannot be achieved. A way has to be found to stabilize FeO by increasing the fo_2 at a higher temperature, where the diffusion coefficient is higher. One way of doing this is by increasing the amount of oxygen in the system relative to hydrogen and carbon by enriching a region of the nebula in silicate dust. As reviewed by Fedkin and Grossman (2006), however, this is an intrinsically inefficient way of increasing the fayalite content of the olivine because almost all of the free oxygen added to the gas by vaporization of the extra dust is recondensed into the same silicate minerals at temperatures above those where the fayalite content would otherwise become significant. A better way of increasing the fo_2 was proposed by Ciesla and Cuzzi (2006), who suggested transport of
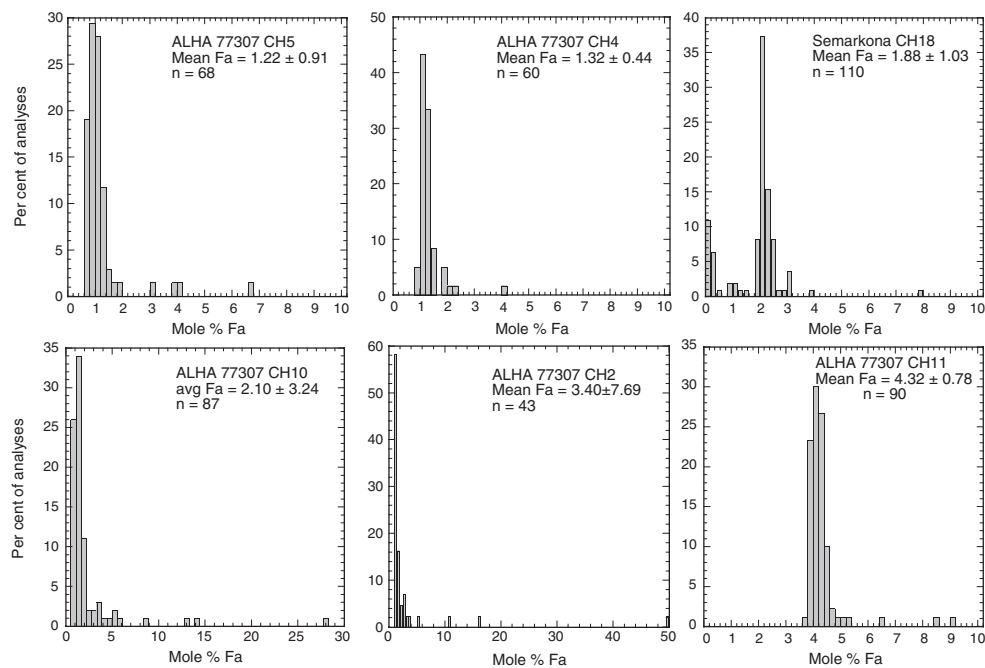
Fig. 2. The mean fayalite content of olivine grains in individual type I chondrules ranges from 1 to 4 mole% in primitive meteorites, such as ALHA77307, a CO 3.0 chondrite, and Semarkona, an LL 3.0 chondrite. From Fedkin et al. (2012).
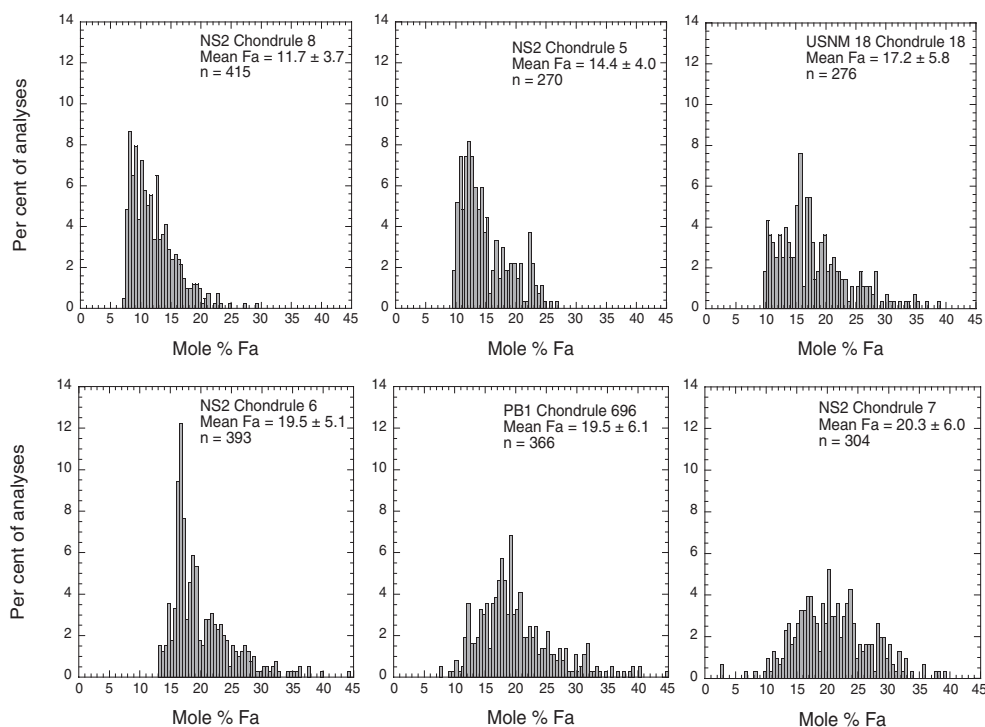

Fig. 3. The mean fayalite content of olivine grains in individual type II chondrules ranges from 12 to 20 mole% in the Semarkona LL 3.0 chondrite. From Fedkin et al. (2012).

water ice-bearing planetesimals from cold, outer regions of the solar nebula to the inner, hot parts where silicates were condensing and chondrules were forming. In this model, however, the maximum water enrichment of the inner solar nebula is only a factor of ten. As seen by the curves for a water enrichment of a factor of ten in Fig. 6,
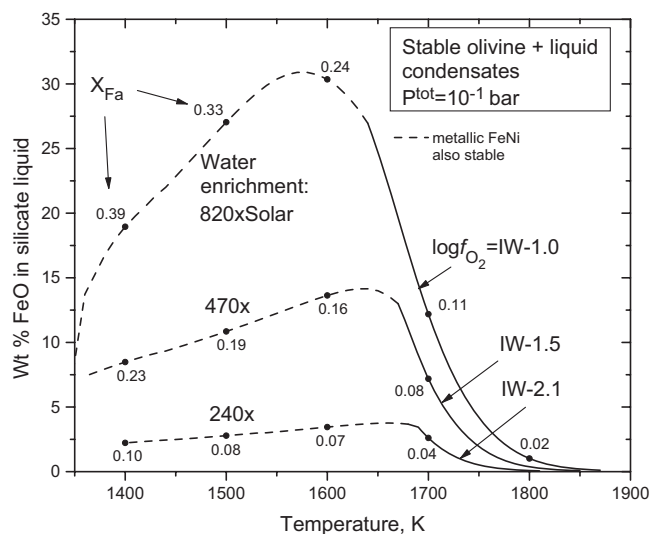
Fig. 4. Equilibrium condensation at high nebular total pressure, P^{tot}, in water-enriched systems that are otherwise solar in composition yields FeO-bearing olivine + silicate liquid. When the water enrichment is several hundred times that of a high-temperature solar gas, the equilibrium fayalite content of olivine varies from type I values to type II values with falling temperature at constant oxygen fugacity, and with an increase in oxygen fugacity of only 0.5 log units at constant temperature.

Fig. 5. The oxygen fugacity of a condensing gas of solar composition (curve labeled C/O = 0.5 within shaded band between error limits on measured O/H and C/H ratios) lies ~7 log units below the iron-wüstite buffer, IW, at a P^{tot} of 10^{-1} atm. The point for Allende inclusions is based on experimental calibration of redox equilibria involving Ti^{3+}-bearing fassaite at the equilibrium solidus temperature of CAIs (see Grossman et al. 2008). The "Chondrules" field, taken from Fig. 4, lies ~5 log units higher in oxygen fugacity.

the oxygen fugacity becomes 2 or 3 log units higher this way, and the equilibrium fayalite content of condensate olivine crystals begins to rise at ~900 K, a somewhat higher temperature than in solar gas.

Grossman et al. (2008) investigated how high the fayalite content could become in 0.1 μm-radius condensate olivine crystals immersed in a gas that is enriched in water by a factor of ten but is otherwise solar in composition, and the result is shown in Fig. 7. The Fe-Mg interdiffusion coefficients used in this calculation were those measured by Chakraborty (1997) as a function of temperature and fayalite mole fraction, but were modified to include the $1/6$ f_{O_2}-dependence seen by other workers, e.g., Buening and Buseck (1973). A nebular cooling time of 10^6 yr was assumed. A finite-difference technique was used in which the grain was divided into thin shells. At each temperature, the surface shell was assumed to have the fayalite content that was in equilibrium with the f_{O_2} of the gas. The diffusion coefficient in each shell was determined by its fayalite content, the temperature, and the f_{O_2}. By 715 K, where diffusion stops, the mean fayalite content of the grain still reaches only 1.9 mole%. Thus, even for an extremely long, 1 Myr, nebular cooling time, the maximum water enrichment from theoretical models and very tiny grains, the fayalite content is trivial. The question is how so much chondritic olivine became so fayalitic, or put
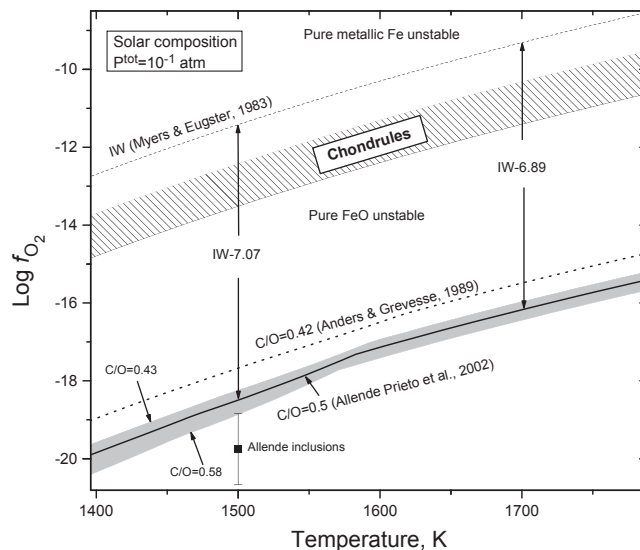
another way, how the first oxidized iron formed in the solar system.

New Diffusion Coefficients

A newer study of the variation of Fe-Mg interdiffusion coefficients in olivine with T, fayalite content, and f_{O_2} was presented by Dohmen et al. (2007) and Dohmen and Chakraborty (2007). They found that the known oxygen fugacity-dependence to the diffusion coefficients exists only for values of f_{O_2} greater than a threshold value; there is no such dependence when the oxygen fugacity is less than this. The threshold was estimated to be ~10^{-15} bar for mantle olivine with 10 mole% fayalite but may vary slightly with olivine composition. Nevertheless, because the f_{O_2} of solar gas, with or without water enrichment, is always many log units below this value in the temperature range of interest, the f_{O_2}-dependence was dropped in the present work. Figure 8 was constructed from various workers' measurements of the dependence of the diffusion coefficient on T, fayalite content, and f_{O_2}. In it, the diffusion coefficient plotted at each temperature is the one corresponding to the f_{O_2} and equilibrium fayalite content for that temperature in a solar gas enriched in water by a factor of ten, i.e., the diffusion coefficient that would actually be used for the surface shell in the finite-difference
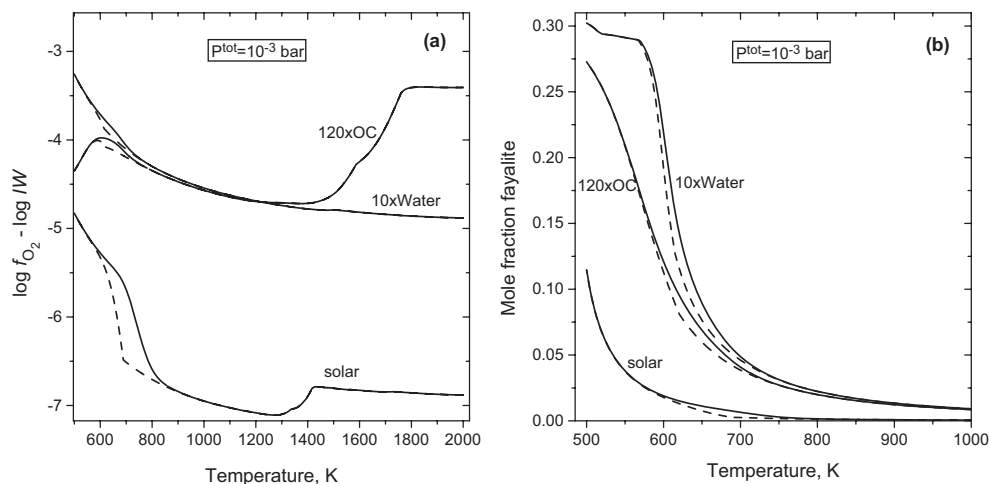
Fig. 6. a) Variation of oxygen fugacity with temperature, and b) the resulting equilibrium mole fraction of fayalite in condensate olivine crystals, calculated for various cosmic systems. A solar gas becomes oxidizing enough for the fayalite content of olivine to become significant only below 800 K, but if solar gas is enriched in water by a factor of ten, the fayalite content begins to become significant above 1000 K. When solar gas is enriched in anhydrous chondritic dust, OC, by a factor of 120, the increase in oxygen fugacity that would otherwise occur is mitigated by recondensation of the added oxygen into silicates between 1800 and 1400 K. The dashed portion of each curve shows departure due to breakdown of gas phase equilibrium. After Grossman et al. (2008).
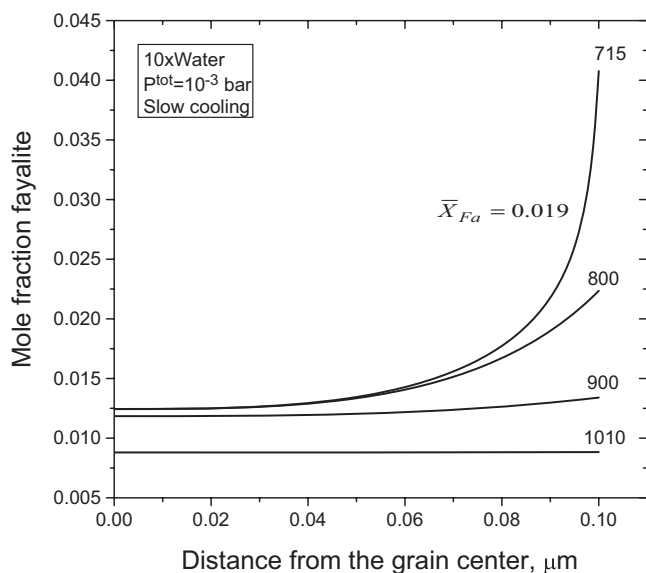

Fig. 7. Core-to-rim profiles of fayalite content in 0.1 μm olivine crystals calculated for a solar gas enriched in water by a factor of ten during nebular cooling on a 10^6 yr time-scale. Curves are labeled with temperature in K. When the Fe-Mg interdiffusion coefficients of Chakraborty (1997), modified by an oxygen fugacity-dependence, are used, diffusion essentially ceases at 715 K, where the mean fayalite content of the grain is only 1.9 mole%. After Grossman et al. (2008).
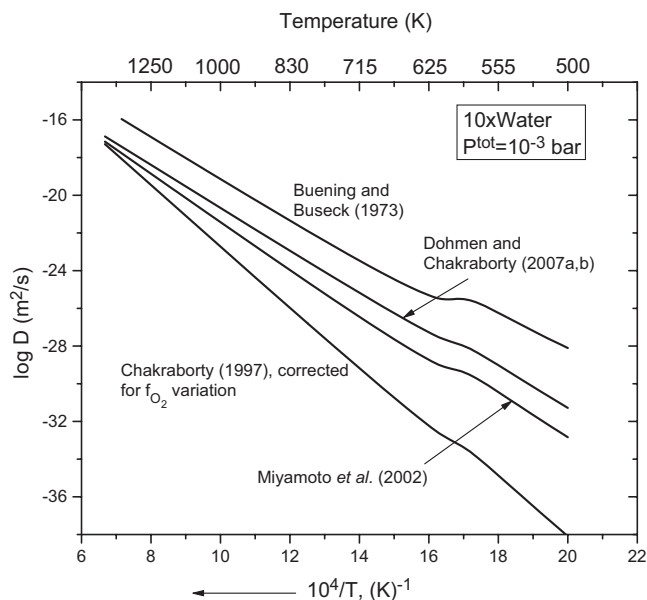

Fig. 8. The variation of the Fe-Mg interdiffusion coefficient in olivine with temperature at the f_{O_2} and equilibrium fayalite content for that temperature in a solar gas enriched in water by a factor of ten, using data from various authors. The diffusion coefficients of Dohmen and Chakraborty (2007) are several log units higher than those used by Grossman et al. (2008).

computation. It is seen that the diffusion coefficients based on the Dohmen and Chakraborty (2007) data are considerably larger than those based on the data of Chakraborty (1997) that had been modified for f_{O_2}-dependence by Grossman et al. (2008).

The calculation of the fayalite content that can be reached by an olivine grain in a solar gas enriched by a factor of ten in water was repeated using the diffusion coefficients of Dohmen and Chakraborty (2007), assuming nebular cooling times of 10^4, 10^5, and 10^6 yrs, the cooling curves for which are shown in Fig. 9. Also
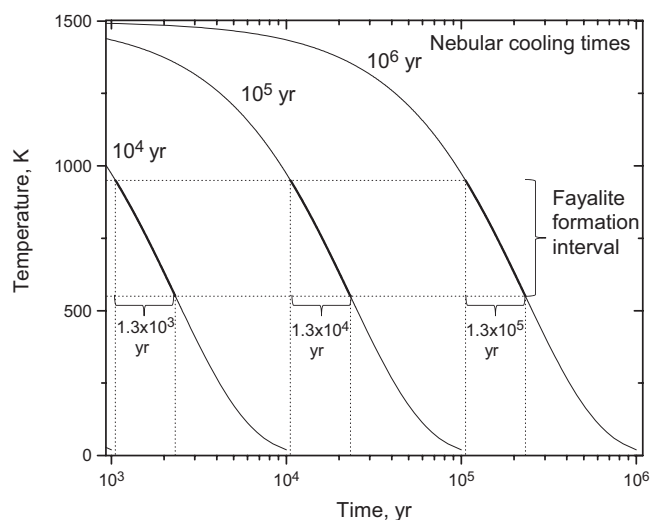
Fig. 9. Exponential temperature-time curves assumed for three cooling time-scales of the solar nebula from 1500 to 20K. For each is shown a fayalite formation interval, defined as the time required to cool from 950 to 550 K, the temperature interval over which the equilibrium fayalite content rises from 1 mole% to its maximum value in a solar gas enriched in water by a factor of ten.
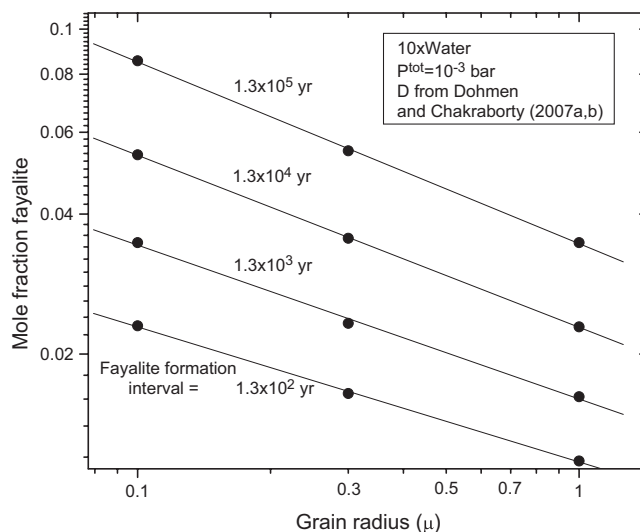

Fig. 10. Mean mole fraction of fayalite reached by olivine grains as a function of their radius in a system of solar composition enriched in water by a factor of ten, for various estimates of fayalite formation interval.

shown is the temperature interval over which the equilibrium fayalite content becomes significant in a gas of this composition, and the time periods required to cool through that interval for each of the nebular cooling times. These range from 1.3×10^3 to 1.3×10^5 yrs.

Results are shown in Fig. 10. For the most reasonable fayalite formation interval, between 1.3×10^3 and 1.3×10^4 yrs, it is seen that a 0.1 μm olivine grain could reach a mean fayalite content of 4 mole%, while a 1 μm grain could reach only 2 mole%. Thus, even at the maximum water enrichment, the achievable fayalite contents are still well below the typical values found in olivine grains in the matrices of primitive chondrites and that would be needed for the precursors of many chondrules. Diopside and enstatite are other nebular condensates that are potential hosts for FeO but Fe-Mg interdiffusion coefficients for these phases (Cherniak and Dimanov 2010) are 1.5 to 3 log units lower than those for olivine at the same temperature, FeO content, and oxygen fugacity. Consequently, pyroxenes formed during nebular condensation are expected to have even lower FeO/(FeO + MgO) ratios than olivine.

SUPERSATURATION OF METALLIC IRON

With the failure of the water enrichment model to produce the fayalite contents observed in chondritic olivine, a completely different idea was investigated for yielding fayalitic olivine during condensation. Blander and Katz (1967) found that, if kinetic barriers to homogeneous nucleation of metallic iron allowed it to become supersaturated in a system of solar composition, olivine that condenses at high temperature would reach high fayalite contents at temperatures much higher than when metallic iron condenses at equilibrium. Using the same computer program and input data as in Fedkin and Grossman (2006), full equilibrium condensation calculations were performed for a gas of solar composition, without allowing either metallic iron or Fe_3C to condense. Shown in Fig. 11 is the resulting fayalite content of olivine as a function of temperature, compared with that computed when metallic iron condenses at equilibrium, for a total pressure of 10^{-3} bar. It is seen that fayalite contents of 30 mole% are readily produced at 1200 K, a temperature far in excess of 800 K, where the fayalite content would first become significant if metallic iron condensed at equilibrium. Furthermore, because the temperature of fayalite formation is so high in this case, diffusion is capable of equilibrating 10 μm olivine grains with the gas even if it cooled from 1400 to 1100 K in only 600 yrs. Thus, if metallic Fe could have been supersaturated significantly during solar nebular cooling, the problem of making oxidized iron during condensation of a gas of solar composition would be solved.

To investigate this phenomenon further, nucleation theory was used to estimate S, the critical supersaturation ratio, which is the factor by which the actual partial pressure must exceed the equilibrium vapor pressure over a phase to allow the latter to nucleate homogeneously. S is related to σ, the surface tension of the condensed phase, through the expression,
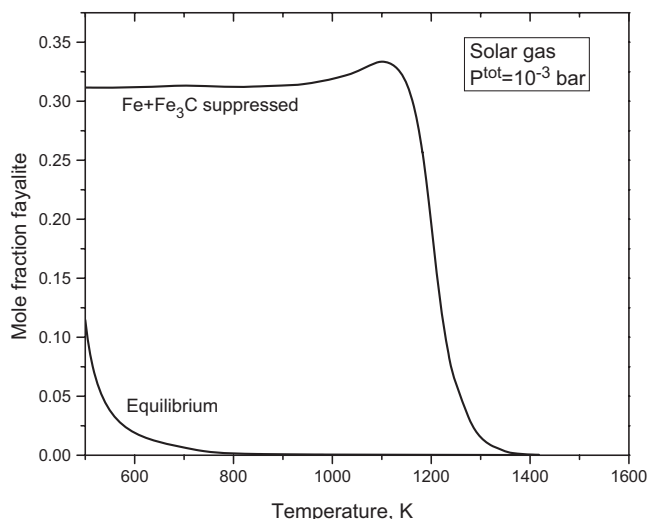
Fig. 11. Equilibrium fayalite content of condensate olivine grains as a function of temperature in a system of solar composition at complete equilibrium and when condensation of metallic iron and Fe_3C are suppressed.
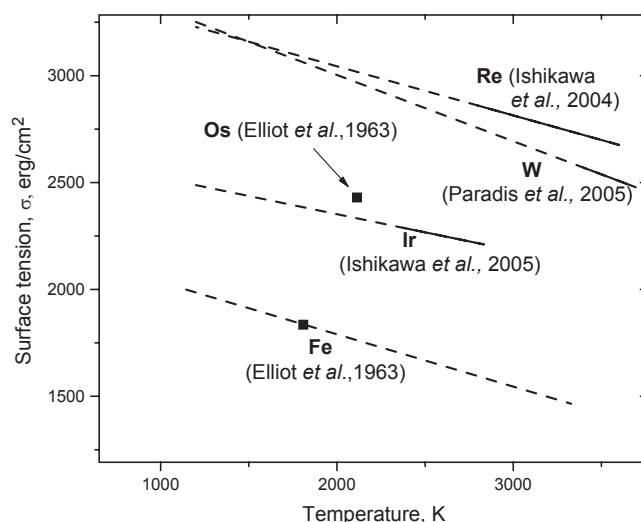

Fig. 12. Literature data for surface tensions of pure iron and pure liquid metals having higher solar nebular condensation temperatures than metallic nickel-iron. Solid portions of curves indicate the temperature interval over which the measurements were made; dashed portions are extrapolated.

$$J = [4.5x10^{33}(P/T)\sqrt{\sigma M/\rho}]\exp[-17.56(M/\rho)^2 \times (\sigma/T)^3/(\ln S)^2], \tag{1}$$

where J is the number of nuclei formed per second per mole, T is the equilibrium condensation temperature, P is the partial pressure of the condensing species in atmospheres at the equilibrium condensation temperature, and M and ρ are the molecular weight and density, respectively, of the condensed phase (Blander and Katz 1967). Literature data for surface tensions of liquid metals are plotted as a function of temperature in Fig. 12. The surface tension of metallic nickel-iron was assumed equal to that of pure liquid iron, whose value at 1823 K from Elliot et al. (1963) was extrapolated to higher and lower temperature using the mean slope of the experimental data for Re, W, and Ir. Condensation calculations were performed for solar gas over a range of total pressures from 10^{-1} to 10^{-5} bar without suppressing condensation of metallic nickel-iron. Substituting P and T from the output into Equation 1, S for the metal phase was found to vary from ~ 150 at 10^{-1} bar to 4.6×10^3 at 10^{-5} bar, resulting in undercooling of 250 to 270 K over the range of total pressures of interest. The condensation calculations were then repeated over the same pressure range, this time by suppressing condensation of metallic iron and Fe_3C, to see the effect of metallic iron supersaturation on olivine composition.

The equilibrium condensation temperatures of metallic nickel-iron and forsterite are plotted as a function of total nebular pressure in Fig. 13a. In Fig. 13b,

the homogeneous nucleation temperature of metallic nickel-iron is compared to the equilibrium condensation temperature of olivine as a function of total nebular pressure. Also shown are contours of the fayalite content of olivine, which increases with decreasing temperature at a given total pressure. After nucleation of metal, the system reverts to the equilibrium state, so the fayalite contours are truncated by the homogeneous nucleation curve, at temperatures below which the fayalite content would once again become near-zero. At temperatures above the homogeneous nucleation temperature of metal, however, it is seen that the fayalite content of the olivine reaches ~ 8 mole% at 10^{-2} bar, ~ 14 mole% at 10^{-3} bar, ~ 25 mole% at 10^{-4} bar, and > 30 mole% at 10^{-5} bar. Thus, if heterogeneous nucleation of metallic nickel-iron near its equilibrium condensation temperature did not occur, the fayalite content of olivine grains would become quite significant, particularly at relatively low nebular total pressures. Furthermore, as mentioned above, this would occur at high enough temperatures that the calculated contents could actually be produced in grains many microns in size by diffusion on a nebular cooling time-scale. These fayalite contents are appropriate for precursors of many chondrules. To serve as such, however, the grain compositions would have to be preserved by isolation from further chemical communication with the gas, before nucleation of metallic nickel-iron, which would otherwise form partly at the expense of the fayalite that is, after all, metastable.

There are, however, several ways by which metallic nickel-iron could have been prevented from becoming
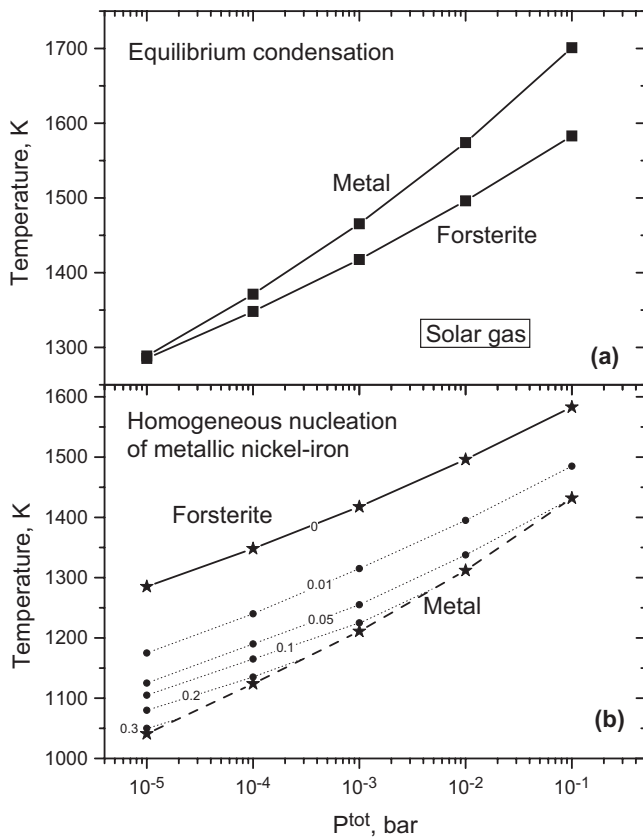
Fig. 13. a) Equilibrium condensation temperatures of metallic nickel-iron and forsterite in a system of solar composition as a function of nebular total pressure, P^{tot}. b) Homogeneous nucleation temperature of metallic nickel-iron in a system of solar composition as a function of P^{tot} after supersaturation of the metal phase by an amount computed via Equation 1. Dashed curves at temperatures above the metal nucleation temperature are contours of the mole fraction of fayalite that can enter the olivine metastably due to supercooling of the metal phase.

Fig. 14. Equilibrium condensation temperatures of corundum (Al_2O_3) and pure metal phases compared to their homogeneous nucleation temperatures after supersaturation of each, as a function of P^{tot}. Os, W, and corundum are so much more refractory than metallic nickel-iron that, even if they became supersaturated during solar nebular cooling, they would still nucleate homogeneously above the equilibrium condensation temperature of metallic nickel-iron, providing nuclei for heterogeneous nucleation of the latter phase near its equilibrium condensation temperature. The shaded region represents conditions for homogeneous nucleation of Al_2O_3, bounded by upper and lower curves based on the surface tensions of the liquid and solid phases, respectively.

significantly supersaturated during nebular cooling. All involve the presence of other condensed phases at or above the equilibrium condensation temperature of metallic nickel-iron. First, the model requires that, after supercooling occurs, olivine condenses at a higher temperature than the metal. The olivine grains themselves might have provided nuclei for heterogeneous nucleation of metallic iron, causing the latter phase to condense nearer its equilibrium condensation temperature, removing a large fraction of the metastable iron from the gas and once again preventing the olivine from becoming iron-rich. Second, residues of metastable, presolar phases such as graphite and SiC may have provided nuclei for heterogeneous condensation of metallic iron. Third, phases with higher equilibrium condensation temperatures than metallic nickel-iron, such as refractory siderophile alloys, corundum, hibonite, perovskite, melilite, and spinel, could
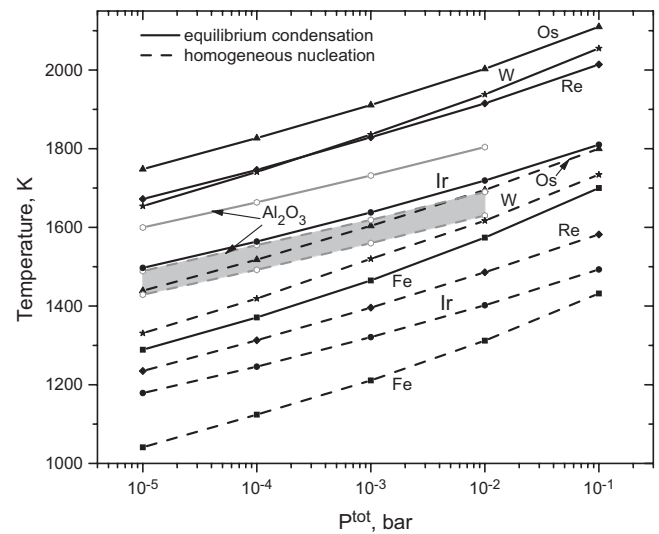
have provided nuclei for heterogeneous condensation of metallic iron, and thereby prevent its supersaturation, if they themselves were able to nucleate homogeneously above the equilibrium condensation temperature of metallic iron. Surface tension data are seen in Fig. 12 for four phases, Os, W, Ir, and Re, whose equilibrium condensation temperatures are higher than that of metallic iron. Also, the surface tension was measured for corundum in both solid and liquid form by Kingery (1954, 1959), respectively.

For each of these five phases, the surface tension data were used in Equation 1 to calculate S, from which the temperature of homogeneous nucleation was computed using condensation calculations. Results are shown in Fig. 14. Even if supersaturation of corundum, Os, and W occurred, each of them would have still nucleated homogeneously at temperatures above the equilibrium condensation temperature of metallic iron. This would have provided heterogeneous nuclei for condensation of metallic iron near its equilibrium condensation temperature, precluding high-temperature condensation of fayalitic olivine.

It is conceivable that, when the primary cooling and condensation stage of the solar nebula ended, there was

not yet any appreciable iron oxide in the inner solar system. A generation of planetesimals may have accreted from CAIs, magnesium silicates, metallic nickel-iron, troilite, and water ice. Formation of the first oxidized iron may not have occurred until liquid water interacted with metallic nickel-iron and magnesium silicates at elevated temperatures inside such planetesimals. The resulting assemblage, then containing FeO-bearing silicates, could have been the source of chondrule precursors. Impacts on icy planetesimals could have generated rapidly heated, relatively high pressure, water-rich vapor plumes containing high concentrations of dust and droplets, environments favorable for formation of types I and II chondrules (Fedkin et al. 2012). Such a sequence of events would be in agreement with radiometric ages indicating that chondrules are ~1–2 Myr younger than refractory inclusions (Amelin et al. 2002). Considerations different from those discussed herein also led Asphaug et al. (2011) to explore chondrule formation in plumes resulting from planetesimal collisions.

CONCLUSIONS

Production of oxidized iron during primary nebular condensation is unlikely. Even in a system of solar composition enriched in water by a factor of ten, the maximum enrichment produced in dynamical models of the solar nebula, diffusion limitations restrict the fayalite content of 0.1 μm olivine grains to ≤5 mole% fayalite even after 10^4 yr. Metastable production of high fayalite contents at elevated temperatures by supersaturation of metallic nickel-iron is unlikely due to the predicted presence of many other phases at its equilibrium condensation temperature, any one of which may have promoted heterogeneous nucleation of the metal phase instead. Formation of oxidized iron may have been delayed until after condensation of water ice and its accretion into planetesimals containing FeO-free silicates. At this point, the first FeO-bearing silicates could have formed at elevated temperature inside the planetesimals by reactions between metal, silicates, and liquid or gaseous water, or by interaction of silicate droplets with water-rich vapor inside impact-generated plumes.

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