NEBULAR FORMATION OF FAYALITIC OLIVINE: INEFFECTIVENESS OF DUST ENRICHMENT.

A. V. Fedkin[1] and L. Grossman[1,2], [1]Department of the Geophysical Sciences, University of Chicago, 5734 S. Ellis Ave., Chicago, IL 60637, [2]Enrico Fermi Institute, University of Chicago.

Introduction: The mean mole fraction fayalite, X_{Fa}, in olivine of unequilibrated ordinary chondrites (UOCs) is 0.18-0.23[1]. Equilibrium thermodynamic calculations predict that X_{Fa}~0 when olivine first condenses from a gas of solar composition at high temperature, and rises with falling temperature. A solar gas is so reducing, however, that X_{Fa} is predicted to reach the level found in UOCs only below 600K, where Fe-Mg interdiffusion rates in olivine are negligible, preventing X_{Fa} from reaching this level in the bulk of condensate olivine, and leaving the problem of how chondrites obtained their FeO contents unsolved. In [2], this situation was reviewed thoroughly and the conclusion reached that chondritic olivine with non-negligible X_{Fa} formed at higher temperature, and thus in a nebular region more oxidized than solar gas, presumably due to enrichment in dust prior to its vaporization. Since then, the problem became more severe due to additional data on solar abundances of C and O [3], making the nebula more reducing; and Fe-Mg interdiffusion rates in olivine [4,5], making diffusion slower; as well as recognition of non-ideality in olivine solid solutions, lowering the solubility of Fa in it [6].

Dust Composition and its Effects: When dust condenses from solar gas, the dust contains a larger proportion of the total O than of the C and H, with the degree of fractionation of O from C and H being dependent on the temperature of isolation of dust from gas. This is why vaporization of regions enriched in dust relative to its complementary gas yields systems more oxidizing than a solar gas. If dust enrichment is the means by which nebular regions became sufficiently oxidizing that chondrites condensed from them, the dust itself must have contained chondritic relative proportions of condensable elements. Three dust compositions are investigated: OC dust, having the bulk composition of ordinary chondrites but with a solar S/Si ratio; C1 chondrite dust; and "icy dust", consisting of 10 parts Orgueil and 1 part H_2O by weight, resulting in 25.5 wt% H_2O. The latter material is imagined to have been isolated from the gas at a slightly lower temperature than C1 chondrites, and into which more H_2O but no additional C condensed.

In cosmic gases, metallic Fe, which would otherwise be available for oxidation to form fayalite, is consumed by reaction with H_2S to form troilite, FeS. Because all three dust compositions have the solar S/Si ratio but subsolar O/Si ratios, dust enrichment enhances the abundance of S more than O relative to so-lar composition. Further, compared to other elements, the rate of increase of the condensation temperature of S (as FeS) with respect to its abundance is unusually large. Thus, while dust enrichment has the desired effect of increasing the f_{O2} at high temperature, it has the undesirable effect of making less metallic Fe available for oxidation, which limits the increase in X_{Fa}.

Condensation Calculations: Full equilibrium calculations were performed at $P^{tot}=10^{-3}$ bar with the same input data and condensation program as in [7], except that olivine and orthopyroxene solid solutions were treated non-ideally using data from [6] and the same S abundance was used as in [8], 13.4% less than in [9]. In solar gas, X_{Fa} is insignificant above 800K, but rises steadily below this temperature, reaching 0.11 at 500K, as seen in Fig. 1. In a system enriched in OC dust by a factor of 10^3 relative to solar composition, X_{Fa} is higher at each temperature than in a solar gas, gradually rising to 0.29 by 500K. A system enriched by a factor of 125 in C1 dust has higher f_{O2} below 1000K than the one enriched in OC dust by a factor of 10^3. As a result, X_{Fa} rises more steeply below 1000K in the C1 dust-enriched system, reaching 0.20 at 825K before levelling off at 0.31 below 600K. Although greater dust enrichment increases the f_{O2}, leading to higher X_{Fa} at all temperatures in the system enriched in C1 dust by a factor of 125 than by a factor of 75, note that X_{Fa} is identical in the two systems at 500K. This is due to the aforementioned effect of S in limiting the amount of metallic Fe available for oxidation at higher dust enrichment. A system enriched in icy dust by a factor of 125 is even more oxidizing than the system enriched in C1 dust by the same factor. As a result, X_{Fa} is higher at all temperatures, reaching 0.20 at 900K before levelling off at 0.32 below 600K. An identical calculation was performed for the icy dust-enriched system except that olivine and orthopyroxene were assumed to be ideal solutions. Comparison of the two curves in Fig. 1 illustrates the dramatic reduction in fayalite solubility due to non-ideality in this temperature range.

Diffusion Calculations: Fayalite concentration profiles were calculated from the diffusion equation for spherical geometry using a finite difference technique and bulk Fe-Mg interdiffusion coefficients, D, from [4] whose logs were regressed against log f_{O2}, 1/T and X_{Fa}. From the output of the condensation code at 1K intervals, f_{O2} was used to calculate D and the equilibrium X_{Fa} was assumed to be equal to the fayalite concentration at grain surfaces. Each degree was subdivided into
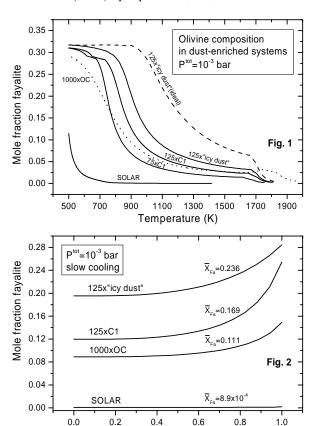
10^4 equal temperature intervals, for each of which the surface X_{Fa} was interpolated, as was f_{O2} for calculation of D. Using cooling times of 10^5 yr (for fast cooling) or 10^6 yr (for slow) from [10] for a dust-laden solar nebular cloud with declining mass accretion rate, cooling rates were assumed to be proportional to temperature and were computed for each temperature interval. In this way, the surface X_{Fa} and D are known for each time step of the diffusion calculation. The time required to cool from 1000 to 800K was $\sim 5 \times 10^3$ and $\sim 5 \times 10^4$ yr for fast and slow cooling, resp. For all grain sizes and gas compositions considered here, a temperature interval exists below the olivine condensation temperature over which diffusion allows homogeneous crystals to persist for both cooling rates. Below this temperature, the diffusion rate slows, and the rate of increase of the surface X_{Fa} becomes greater than that of the interior, causing curvature of concentration profiles near the surface. Profiles are calculated at progressively lower temperatures until a temperature is reached, below which the mean X_{Fa} of the interior becomes virtually invariant. Such final profiles are shown in Fig. 2 for grains of 1 μm radius during slow cooling of systems of various compositions at 10^{-3} bar.

In solar gas, the concentration profile of a 1 μm grain becomes invariant at 780 K, where the central X_{Fa} is 6.2×10^{-4}, the surface $X_{Fa}=1.9 \times 10^{-3}$ and the mean $X_{Fa}=8.9 \times 10^{-4}$. This demonstrates the need for a system more oxidizing than one of solar composition to account for olivine with the mean X_{Fa} found in UOCs. In a system enriched by a factor of 1000 in OC dust, the mean X_{Fa} of a 1 μm grain reaches 0.11. Thus, even when OC dust is enriched by a factor much greater than 125, the maximum dust enrichment produced in the coagulation and settling models of [11], the mean X_{Fa} is well below the values in UOCs. In a system enriched by a factor of 125 in C1 dust, the mean X_{Fa} of a 1 μm grain reaches 0.17, still below the UOC range, and that of a 0.5 μm grain reaches 0.19. Thus, olivine with mean X_{Fa} within the UOC range can be produced in systems enriched in C1 dust, but only for the slowest cooling rates, the maximum dust enrichments and very tiny grains, ≤0.5 μm. A possible solution is enrichment in dust even more oxygen-rich than C1 chondrites, e.g. icy dust. In a system enriched by a factor of 125 in icy dust, the mean X_{Fa} of 3 μm grains reaches 0.16 and of 1 μm grains 0.20, even during fast cooling. During slow cooling, the mean X_{Fa} of 3 μm grains reaches 0.20 and of 1 μm grains 0.24.

Conclusions: Enhancement of nebular f_{O2} by dust enrichment is seen to be ineffective at producing olivine with X_{Fa} as high as ~0.2. Because reasonable degrees of enrichment of dust of plausible compositions

stabilize such fayalite contents only at temperatures where diffusion is very slow, the maximum sizes of olivine grains that can reach such X_{Fa} are ≤0.5-3.0 μm. While supernova grains which condensed in only ~2 yr [12] have such sizes, nebular condensate grains, which grew over thousands of years, would be expected to be much larger. If the mean X_{Fa} of olivine in UOCs resulted from reduction of significantly more oxidized material during chondrule formation, the origin of such a precursor would be an even more difficult problem.

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Fig. 1

Fig. 2