EXPERIMENTAL CONDENSATION OF CRystALLINE OLIVINE AND PYroxene:
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We reported previously (1,2,3) the results of studies of residues produced by volatilization of basaltic and carbonaceous chondritic materials in a solar furnace. In the case of carbonaceous chondrites, extreme degrees of enrichment of refractory elements relative to volatiles could not be achieved due to production of copious smoke which rose above the sample, coated the Pyrex walls of the sample chamber and attenuated the sunlight, lowering the temperature of the sample. We present here the results of textural and mineralogical studies of the smoke particles in one of our experiments.

In this experiment, an ~1 gm sample of the Murchison meteorite was heated in an aluminum crucible in air. A vacuum gauge placed in the vacuum line near the pump gave a reading of <1 μm prior to and during heating. Almost at the first instant that the beam strikes, smoke appears above the sample but separated from it by ~2 cm of clear gas. The smoke particles rise and stick to the interior of the Pyrex globe surrounding the sample, preferentially on the cooler sides of the chamber rather than the top. After 18 minutes of heating, the smoke coating was so thick that the experiment was interrupted in order to clean the globe. After doing so and gathering spattered material on the floor of the crucible around the main sample bead, heating was resumed for 46 minutes. The material studied here consists of those condensate grains that stuck to the interior walls of the sample chamber during this interval. Although a maximum temperature of 3000°C is possible at the top of the sample bead when the sunlight is focussed to an 8 mm diam. spot on it, the actual time-averaged temperature of the sample during this run must have been considerably lower because of the opacity caused by the smoke particles.

Optical examination with a stereomicroscope revealed that 80% of the material scraped from the walls consists of fragile cylinders which have nearly circular cross-sections 0.1-0.2 mm in diameter and lengths up to 1.5 mm but averaging 0.5 mm. Each cylinder is concentrically zoned in color in the radial direction, the center being brownish-orange, the next zone outward deep orange, the next brownish-black and the outermost light yellow. The cylinders often form planar clusters in which 2 to 6 are stuck together, side-to-side. Most frequently, what is observed is a side view of a cluster split lengthwise. In this case, one sees straight or gently curved strands zoned perpendicularly to their length in such a way that the sequence light yellow, brownish-black, deep orange, brownish-orange, deep orange, brownish-black, light yellow repeats itself. The remaining 20% consists of irregularly-shaped, dark orange to black clumps in which strand structure is seen, interpreted to be simply thick masses of the above material. Also found are rare spherules with metallic or glassy luster, ~0.1-0.5 mm in diameter, similar to those seen among spattered materials on the crucible floor.

Twenty large strands, carefully selected on the basis of freedom from adhering foreign matter, were hand-picked, ground together and mounted in a Debye-Scherrer camera in which they were X-rayed for ~15 hr. The resulting diffraction pattern contained many lines which correspond to a mixture of Fe-rich clinopyroxene, Fe-rich olivine and α-Fe.

A polished thin section was prepared from a scoop of the original bulk sample. The vast majority of the particles seen in the section with the polarizing microscope are mottled orange strands and fragments thereof, many of which show color zonation and none of which show birefringence. A small
number of pale green glass spherules and one or two small chunks recognizable as pieces of bulk Murchison are also present. These may have flown out of the crucible and stuck to the chamber wall during spattering of the sample. SEM study of unpolished strands reveals layers of different electron albedo which are composed of fluffy, 5-20 µm clumps of grains that are ≤ 0.1 µm. The very small sizes of the grains would explain the lack of birefringence. Back-scattered electron images of strands in the polished section show that most of the strands are composed predominantly of material with surprisingly low brightness for the phases indicated by X-ray diffraction, presumably due to the high porosity of such a fine-grained sample. Irregular clumps of much brighter material, 5-20 µm in size, are minor constituents of some strands and are particularly abundant in one.

Energy dispersive analyses were performed with the SEM whose electron beam spot was 1 µm in diam. All analyses of low brightness regions of the strands yielded low analytical sums, 19.2-49.9%, most in the range 25-41%. After subtraction of 0.5-1.1% S as FeS, these analyses are all interpretable as mixtures of Fe-rich olivine and Fe-rich, low-Ca pyroxene. The purest olivine analysis yields the chemical formula Mg_{0.27}Fe_{1.65}Si_{1.92}O_{4} and the purest pyroxene Mg_{0.78}Fe_{1.21}Si_{2.01}O_{5}. High brightness regions have analytical sums in the range 94-97% and element ratios again indicative of olivine-pyroxene mixtures. The correlation of low electron albedo with low analytical sum is consistent with their both being caused by relatively high porosity.

Our interpretation is that crystalline, stoichiometric, fine-grained silicates and metallic iron condensed within seconds from the cooling vapor emitted from a partially volatilized carbonaceous chondrite during the solar furnace experiments. These results differ from those of (4) and (5) in which silicate condensates that formed in milliseconds by cooling of vapor made by laser heating of minerals and meteorites were described as glassy. They also differ from those of (6) and (7) in which silicate condensates produced during millisecond cooling of shock-heated gas mixtures are always amorphous. Finally, they differ from those of (8) and (9) in which silicate condensates made by evaporating SiO or both SiO and Mg from heated crucibles into a lower-temperature, H_2-filled furnace are all amorphous. Whether the differences between these runs products and ours are due to differences in gas composition, partial pressures of condensable species, nucleation mechanism, small differences in cooling rate or some other parameter is unknown.

Results of their experiments led Stephens (7) and Nuth and Donn (10) to conclude that equilibrium thermodynamic calculations are poor predictors of the actual condensation process in astrophysical systems, including the solar nebula, because amorphous, non-stoichiometric solids formed in the experiments rather than thermodynamically predictable crystalline species. Although we have never been convinced of the validity of this conclusion due to the vast difference in cooling times between such experiments and most cosmic clouds, particularly the solar nebula, our experiments clearly show that there exist some conditions under which crystalline, stoichiometric silicates condense from multi-element gas mixtures, even during rapid cooling.