INFRARED SPECTRA OF MG-SIO SMOKES: COMPARISON WITH ANALYTICAL ELECTRON MICROSCOPY STUDIES.

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An important component of current models for interstellar and circumstellar evolution is the infrared (IR) spectral data collected from stellar outflows around oxygen-rich stars and from the general interstellar medium [1]. IR spectra from these celestial bodies are usually interpreted as showing the general properties of sub-micron sized silicate grains [2]. Two major features at 10 and 20 microns are reasonably attributed to amorphous olivine or pyroxene (e.g. Mg_2SiO_4 or $MgSiO_3$) on the basis of comparisons with natural standards and vapor condensed silicates [3-6]. In an attempt to define crystallisation rates for spectrally amorphous condensates, Nuth and Donn [5] annealed experimentally produced amorphous magnesium silicate smokes at 1000^oK. On analysing these smokes at various annealing times, Nuth and Donn [5] showed that changes in crystallinity measured by bulk X-ray diffraction occured at longer annealing times (days) than changes measured by IR spectra (a few hours). To better define the onset of crystallinity in these magnesium silicates, we have examined each annealed product using a JEOL 100CX analytical electron microscope (AEM). In addition, the development of chemical diversity with annealing has been monitored using energy dispersive spectroscopy of individual grains from areas <20nm in diameter. Furthermore, the crystallisation kinetics of these smokes under ambient, room temperature conditions have been examined using bulk and fourier transform infrared (FTIR) spectra.

Repeated IR spectroscopy of bulk unannealed and annealed samples shows that spectral features are essentially unchanged after a period of 2 years storage under ambient room temperature conditions. Prominent 3 and 6.2 micron features, corresponding to a significant increase in hydration, are the only additional spectral characteristics noted. FTIR spectra for 100 micron size clusters of the unannealed sample are shown in Figure 1. Figure 1a is from the unannealed Mg-SiO smoke 2 years after production. Figure 1b is from the same sample after heating in air on a KBr substrate at 423^oK for 1 hour in order to remove adsorbed H_2O and CO_2. Figure 1 shows that the overall IR spectra between $400cm^{-1}$ and $1350cm^{-1}$ are identical to previous bulk spectra of these unannealed samples. Broad minima are observed at 10 and 20 microns and a sharp shoulder due to Si_2O_3 occurs at 11.4 microns. However, an additional sharp feature at $3695cm^{-1}$ is also apparent in the FTIR spectra and suggests the presence of brucite in these unannealed samples. Brucite may form by hydration of MgO or Mg. Thermodynamic calculations for both reactions show that hydrat- ion of metallic Mg is more favorable and support earlier observations by Day and Donn [7].

All samples examined using the AEM show an open, three-dimensional structure. Annealed smokes commonly occur as fluffy clusters of sub-micron sized grains. Some granular clusters contain thin, hexagonal Si-rich plates (20nm-25nm diameter) which increase in abundance with annealing time. The unannealed sample shows a feature-less network of interconnecting ribbons, with ill-defined regions (~150nm in diameter) containing poorly developed crystal faces. The morphology of these unannealed smokes indicates that direct vapor to solid phase nucleation is the dominant condensation mechanism in these experiments [8]. In the unannealed samples, a wide diversity of Mg/Si ratios are observed within and between individual grains. With annealing, the range of Mg/Si ratios decreases, while at 30 hours annealing, the Mg/Si ratios show a bimodal distribution. Selected area electron diffraction (SAED) shows that the dominant minerals in the annealed samples are forsterite and SiO_2 (tridymite?). Enstatite is only observed after the smokes have been annealed for 4 hours. Using SAED, sub-micron sized crystallites as well as amorphous grains are observed in the unannealed smokes.

Results from this work show that chemical and microsctructural development of Mg-SiO smokes after condensation is primarily kinetically controlled. In addition, amorphous materials (at the AEM scale) are formed via condensation from refractory vapors, although micro-crystalline grains also occur. The complexity of structure and chemistry observed in Mg-SiO smokes is strongly dependant on the time-temperature path after initial nucleation and condens- ation. These experiments also indicate that non-stoichiometric, amorphous or poorly crystalline phases may have been the predominant form of early solar system condensates.

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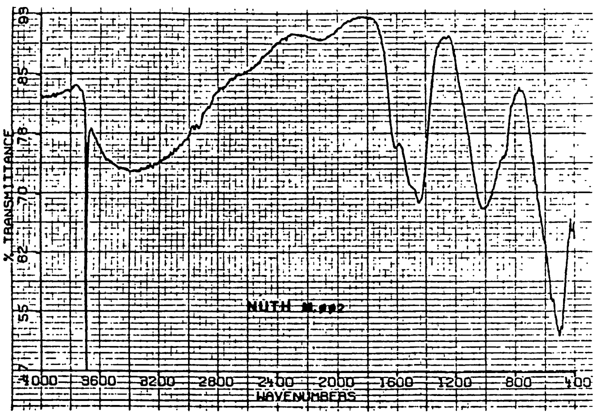

FIGURE 1A

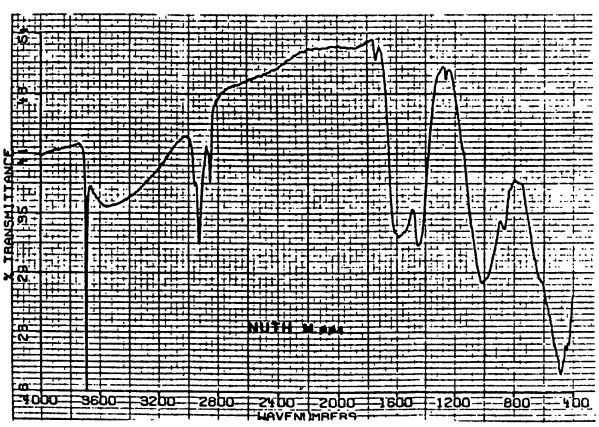

FIGURE 1B