REACTION PRODUCTS OF SYNTHETIC Mg-SILICATES HYDRATED IN A HUMID CHAMBER.

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Introduction: Synthetic silicates have served well for investigating two major events postulated to occur early in solar system formation: vapor-solid condensation [1] and aqueous alteration of proto-minerals [2; 3, 4 and references therein].

Synthetic smokes from the Goddard Space Center Astrochemistry Lab [1] share reflectance spectra with dust around young stars [5, 6] and resemble amorphous materials described in type 3.0 chondrites [7, 8]. These meteorite observations suggest that these materials formed by vapor-solid condensation in the solar nebula with a small component inherited from interstellar sources [8 - 11]. Meteorite studies further suggest that early solar system processes include hydration of amorphous silicate materials [e.g., 10].

Using Goddard synthetic Mg-silicates [1], this project continues work subjecting these amorphous smokes to hydration by exposure to a temperature-controlled, humid N_2 atmosphere. Powder XRD showed that temperatures around 20° C give the most pronounced chemical changes with the most new crystallized material [3]. We now report initial characterization of reaction products by transmission electron microscopy (TEM) and x-ray diffraction with a position-sensitive detector (PSD-XRD).

Experimental: Synthetic Mg-silicate smoke was obtained, stored, pre-treated with flowing dry N_2, and incubated over deoxygenated deionized water as described [3]. Incubation temperatures were 17-27° C.

Two transmission electron microscopes were used: a JEOL 2010F FEG TEM/STEM, for which samples were mixed with a C-based epoxy, cured at room temperature, pressed between two pieces of Si, and ion-milled to electron transparency [9]; a Philips EM420, whose samples were suspended in ethanol and dropped onto a holey carbon grid. EDS on the JEOL was carried out with a 2 nm-beam

XRD analyses were performed in an INEL diffractometer with a curved position-sensitive detector (PSD), an instrument with enhanced capability for phase identification [12]. Powders were mounted without further treatment on single-crystal quartz substrates for analyses using both Cu and Co sources.

Results: Fig. 1a presents a TEM image showing an array of proto-phyllosilicates structures. The hydrated Mg-silicate material is sensitive to beam damage, breaking up almost instantly upon attempts at higher magnification or electron diffraction. The sample in Fig. 1 was hydrated at 27° C, higher than temperatures that later XRD results showed to be more effective [3]. As Fig. 1b shows, Mg:Si ratios in the hydrated material were most consistent with those in sepiolites and serpentines, while anhydrous ("raw") samples ranged from 1:1 to 1:24,.

The TEM image in Fig. 2 presents further evidence for incipient crystallization, this time for a sample hydrated at 19.0° C. Despite the amorphous visual appearance, the small-angle electron diffraction pattern is consistent with crystallinity.

Fig. 3 compares PSD-XRD patterns for a sample incubated at 18.0° C and the unhumidified raw Mg-silicate smoke. Almost all raw material peaks were obliterated by the humidification process, as they were for two other temperature regimes in the study (Table 1). New phases identified include phyllosilicates except for those in the sample incubated at 17° C.

Discussion: The EDS results of Fig. 1b strongly suggest that the protosilicates imaged in Fig. 1a are incipient phyllosilicates. The PSD-XRD patterns summarized in Table 1 serve to confirm the results of both TEM studies. An implication is that some primitive phyllosilicates may form in the nebula before aggregation into protoplanetary bodies, consistent with Brearley's interpretation of CO3 meteorite observations [10].

The optimal temperature for phyllosilicate synthesis must be studied further, but is likely around 18° C. This finding may well provide a useful constraint for modeling of early solar system processes.

An intriguing new question is raised by the fact that synthetic Fe-silicate dust is hardly at all reactive with water [13]. This relative lack of reactivity is opposite to what is suggested in models of parent body rock-fluid interactions [14] and of phyllosilicate formation in the nebula [15]; both suggest that Fe-rich materials are far more easily hydrated. These models are in agreement with modal mineralogy [12] and petrographic studies of hydration reactions [e.g., 16] in meteorites. Thus the reactivity of Fe-silicate dust also calls for further study.

Conclusion: Work with humidification of the Goddard silicate dust is providing experimental support for views of some planetary formation processes and new data for model builders..

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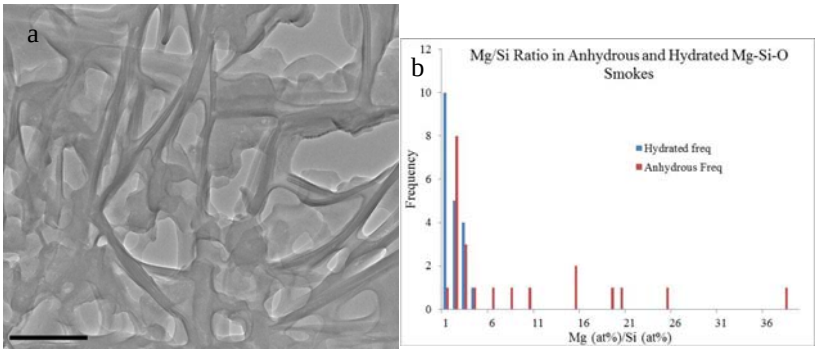

Fig. 1. TEM of Mg-silicate dust hydrated at 27° C for 21 days (JEOL 2010F FEG TEM/STEM) **a.** Tangle of putative phyllosilicates. The scale bar is 200 nm. **b.** EDS Histogram of Mg:Si ratios in 23 anhydrous (red) and 21 regions in smokes hydrated at 27˚C (blue) .Compared with anhydrous smokes, hydrated smokes show a narrow range of compositions, consistent with sepiolite to serpentine.

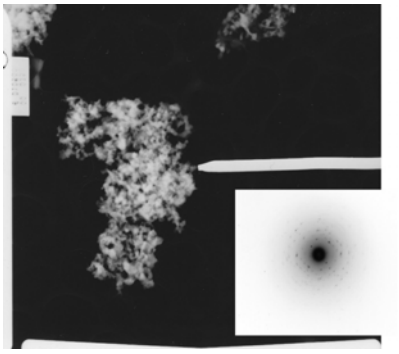

Fig. 2. Cluster of amorphous and crystalline particles from a 19.0° C preparation. Needle indicates location of crystallite in the inset. Inset: Diffraction pattern of small crystalline particle. (Philips EM420)

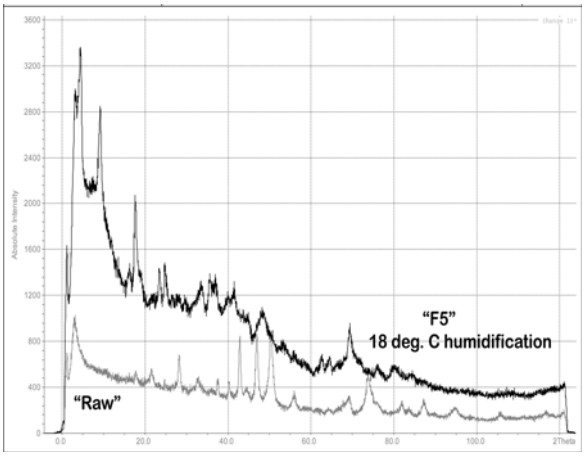

Fig. 3. X-ray diffraction pattern for synthetic Mg-silicate dust exposed to humid N_2 at 18.0° C for 3 weeks ("F5") and for unhumidified starting material ("Raw"). Phases identified are listed in the last and first columns of Table 1, respectively. (PSD-XRD)

Raw	**F1 (17-18 ºC)**	**F4 (17ºC)**	**F5 (18ºC)**
Mg_2Si (magnesium silicide)	Mixed layer phyllosilicate		Mixed layer phyllosilicate
Mg (magnesium)	SiO_2 (zeolite like)	SiO_2 (zeolite like)	SiO_2 (zeolite like)
MgO (periclase)	SiO_2 (hydrous)	SiO_2 (hydrous)	SiO_2 (hydrous)
SiO_2 (μ-porous variety)	various minor SiO_2 (μ-porous variety)	SiO_2 (μ-porous variety)	Minor SiO_2 (μ-porous variety)
SiO_2 (tridymite)	SiO_2 (tridymite)		SiO_2 (tridymite)

Table 1 Phases identified by PSD-X-ray diffraction in raw (unhydrated) and hydrated synthetic Mg-silicate dust (patterns for first and last columns in Fig. 3, adjoining).