CONDENSATION OF FORSTERITE UNDER CONTROLLED PROTOPLANETARY DISK CONDITIONS.

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Introduction: Meteoritic evidence indicates that dust condensation occurred in the early stage of solar system evolution. For instance, amoeboid olivine aggregates, fine-grained aggregates of forsteritic olivine, have been considered to be primary nebular condensates [e.g., 1]. Equilibrium condensation models predict stable phases at given physical and chemical conditions, but cannot predict neither but neither grain size and number density of dust nor duration of processes cannot be predicted, which should be discussed in the context of dust formation kinetics.

Condensation experiments of dust analogue materials under conditions as close as possible to circumstellar environments are crucial because dust formation kinetics could be different under different physical and chemical conditions [e.g., 2, 3].

In this study, we succeeded in performing condensation experiments of forsterite under controlled protoplanetary-disk conditions, which will make significant contribution to understanding silicate formation and chemical fractionation in protoplanetary disks.

Experiments: Condensation experiments were carried out in the system of Mg_2SiO_4-H_2-H_2O. An infrared vacuum furnace was used in this study, consisting of a silica glass tube (~300 mm in length and 38 mm in diameter) connected to a pumping system and two infrared heating systems. The furnace was continuously evacuated during experiments. A mixed gas of H_2 and H_2O was flowed into the system at a controlled rate to keep a pressure constant. Synthetic forsterite powder in an It crucible was heated as a gas source. A part of evaporated gases were condensed on a substrate of Pt mesh located at a cooler region in the chamber (15-25 mm from the gas source). The substrate temperature was monitored by a type-R thermocouple.

The pressure and temperature conditions were close to those of protoplanetary disks. The total pressure of the system was ~5.5 Pa (5.5×10^{-5} bar), and the substrate temperature ranged from 1320 to 1160 K with ±5-10 K fluctuation. The H_2O/H_2 ratio was set at 0.015, which was ~15 times larger than the solar ratio. The SiO/H_2 ratio was evaluated to be ~0.7-2 % of the solar ratio from the weight loss rate of the gas-source forsterite. Note that the SiO/H_2 ratio can be smaller than these estimates because a part of SiO and Mg may have condensed on the wall of the silica glass tube. Experimental duration ranged from 6 to 237 hours.

Condensates were observed with a field-emission scanning electron microscope (SEM) and their chemical composition and crystallinity were analyzed with energy dispersive X-ray spectroscopy (EDS), electron backscattered diffraction (EBSD), and Fourier transform infrared spectroscopy (FTIR). Transmission electron microscope (TEM) observation was also made for focused ion beam (FIB) lift-out sections of condensates obtained at 1160 K for 93 h.

Results: Sub-micron to micron-sized condensates covered with Pt substrates at 1160 and 1275 K (Fig. 1), but no condensates were found at 1320 K. The typical size of condensates at 1160 K was less than 1 μm irrespective of experimental duration and no effective growth of each condensed grain was observed. Condensates at 1275 K for >40 hours partly had several micron-sized flat regions.

EDS analyses showed that chemical compositions of condensates were consistent with the stoichiometry of forsterite, and their EBSD patterns were well fitted with the patterns from crystalline forsterite (Fig. 2). Coincident EBSD patterns were obtained from the flat region of condensates at 1275 K, suggesting that the area was covered with a single crystal. TEM observation of condensates at 1160 K also found that the condensates were polycrystalline forsterite with a thickness of ~30-150 nm (Fig. 3), and infrared absorption spectra of condensates show clear 10-μm absorption features resembling those of crystalline forsterite (Fig. 4). These evidence indicates that polycrystalline forsterite condensed at 1275 and 1160 K.

Discussion: The mean free path of gas molecules under the present experimental conditions is less than 1 mm, and the evaporated forsteritic gas and the ambient H_2-H_2O gas are expected to be well mixed. Supersaturation ratios (S) for experiments at 1320, 1275, and 1160 K are thus estimated to be <1.2, <10, and <1000-2000. These supersaturation ratios correspond to the supercooling of <5, <60 and <170 K, respectively.

No condensates were found at 1320 K because the degree of supersaturation was too small for nucleation of forsterite or even the vapor was not saturated with forsterite (S<1). The condensates at the supercooling of <170 K (1160 K) imply that heterogeneous nucleation of new grains occurred successively on preexisting grains. On the other hand, with the supercooling of <60 K (1275 K), some grains seem to have grown up to several microns, and some seem to have newly nucleated on preexisting grains, suggesting that both nucleation and growth of each condensate occurred.

These differences would result in a structural difference in forsterite dust condensed in protoplanetary disks. Fluffy aggregates of sub-micron sized fine particle would form with a supersaturation of >1000, while aggregates of micron-sized grains would form with a supersaturation of ~10 that could be an analogue of amoeboid olivine aggregates in chondrites.

Although heterogeneous nucleation could reduce the growth rate of forsterite, the condensate thickness of ~30-150 nm obtained at 1160 K for 93 hours (Fig. 3) is thinner than the maximum growth thickness predicted by the Hertz-Knudsen equation (~1.2 μm). This implies that there is kinetic hindrance for vapor growth of forsterite as in the case of evaporation (e.g., [3]). A condensation coefficient, which is a dimensionless parameter representing kinetic hindrance for condensation in the Hertz-Knudsen equation [4], is estimated to be 0.025-0.12 at 1160 K.

References: [1] Krot A. N. et al. (2004) *Chem. Erde, 64,* 185–239. [2] Tachibana S. et al. (2011) *ApJ, 736,* doi:10.1088/0004-637X/736/1/16. [3] Takigawa A. et al. (2009) *ApJ, 707,* L97-L101. [4] Hirth J. P. & Pound G. M. (1963) *Condensation and Evaporation, Nucleation and Growth Kinetics.* Pergamon Press.

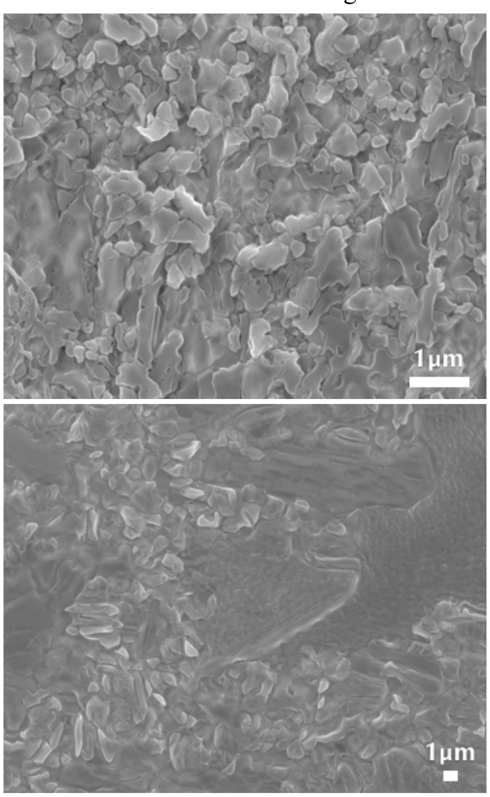

Fig. 1. Condensates formed in the Mg_2SiO_4-H_2-H_2O system at 1160 K for 93 h (top) and at 1275 K for 44.5 h (bottom).

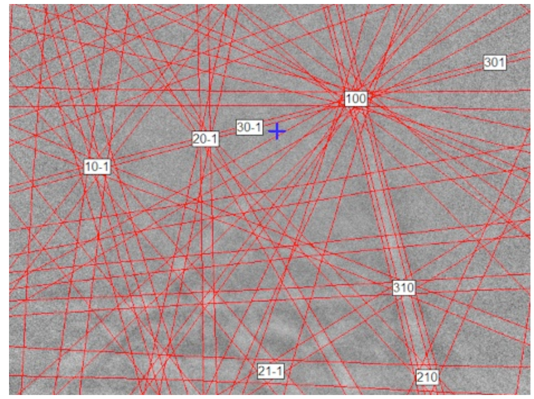

Fig. 2. An example of EBSD pattern obtained from condensates at 1160 K for 93 hours, which can be well reproduced with that of crystalline forsterite.

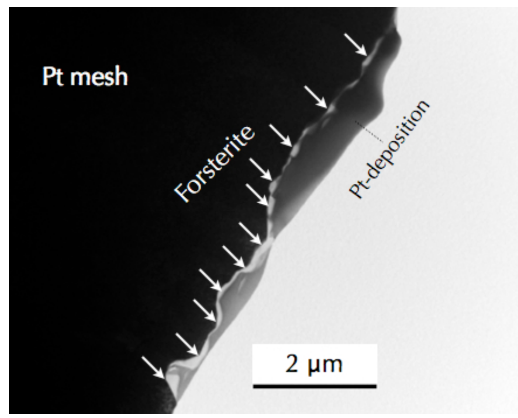

Fig. 3. A bright field TEM image of an FIB section of condensates obtained at 1160 K for 93 hours. The thickness of condensates was <150 nm.

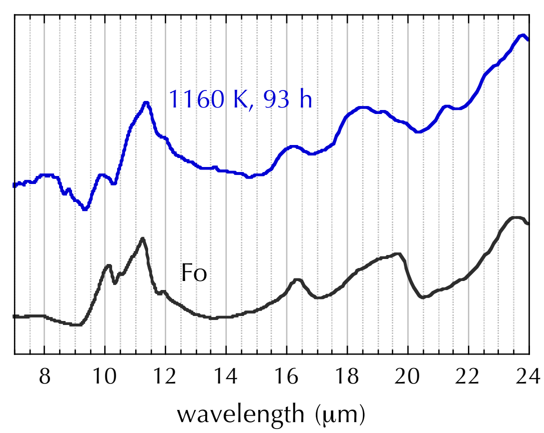

Fig. 4. An infrared absorbance of condensates formed at 1160 K for 93 hours (blue curve) compared with that of crystalline forsterite (black curve). The FTIR analysis was made by pressing the platinum mesh with condensates between KBr plates.