EXPERIMENTAL CONDENSATION OF CRYSTALLINE MAGNESIUM-RICH SILICATES. R. Ogawa, H. Nagahara, K. Ozawa and S. Tachibana. Department of Earth and Planetary Science, University of Tokyo, 7-3-1 Hongo, Tokyo113-0033, Japan. e-mail : riereir@eps.s.u-tokyo.ac.jp

Introduction: Silicate dusts are abundant components in the astrophysical environments, which are thought to be directly condensed from gas. Infrared space observatory (ISO) discovered the existence of crystalline Mg-rich silicates (forsterite and enstatite) in addition to amorphous silicate around discs or stellar shells of young and in the outflow from evolved stars [1]. The phase, size, shape, crystallinity and chemical compositions of the dust grains are important factors that control the IR spectrum.

In this study, in order to investigate phase and crystallinity of condensates as a function of temperature and gas flux (degree of supersatuation), we carried out condensation experiments of magnesium-rich silicates in a vacuum chamber. Though nucleation is an important process for condensation from gas, we only consider the grain growth process.

Experiments and analysis: Condensation experiments were conducted in a vacuum chamber made of stainless steel with a high temperature tungsten mesh heater. The chamber is evacuated by a rotary pump and a turbo molecular pump. Single crystal of forsterite for the starting material was cut into rectangular parallelepipeds (about $1 \times 2 \times 3$mm size), of which largest area was vertical to the a-axis. The sample was heated at 1600°C to evaporate in an aluminum tube, and condensates were obtained on a molybdenum substrate, of which temperatures ranged from 1250°C to 900°C. The gas flux was calculated from the weight loss of the starting material and conductance in the tube. Forsterite evaporates congruently all through the experiments, and therefore, the gas should have a composition of Mg_2SiO_4. Pressure in the chamber during experiments is 10^{-4} to 10^{-3}Pa. The experimental duration ranged from 8 hours to 60 hours.

Condensates on the molybdenum substrate were observed with an FE-SEM, the chemical composition were quantitatively obtained with EDS, and the crystallinity and phase identification were done with electron back scatter diffraction patterns (EBSD). .

Results: Condensates were observed on the Mo wire along a temperature gradient from about 1250°C to 900°C regardless of experiment duration (Fig. 1). The size of the condensates becomes larger with increasing experimental duration. Condensates at high temperature (~1250°C) were irregular in shape and the size of grains was about 10_m.They homogeneously covered the molybdenum substrate. Condensates at lower temperatures were an intergrowth with molybdenum grain which was probably from the substrate.

The chemical composition of the condensates, which was obtained by averaging the about 10 point analyses with EDS, decreases with decreasing temperature (Fig.3). Mg/Si ratio of the highest temperature condensation was about 3, which becomes lower with decreasing temperature down to 1.5 at the lowest temperature.

The condensates are mostly crystalline forsterite and much lesser amount of clinoenstatite was observed at lower temperatures in short experiments. At the lowest temperatures amorphous silicates were observed. The amount of clinoenstatite is larger in longer experiments (40 and 60 hours).

Discussion: It is worth noting that Mg/Si ratio of the bulk composition of the condensates decreases with lowering temperature, which varies from almost 3 at the highest temperature to about 1.5 at the lowest temperature (Fig. 3). Furthermore, the value changes systematically with experimental duration. Longer duration experiments (warm color symbols in Fig. 3) tend to have lower Mg/Si ratios, whereas, shorter experiments (cool color in Fig. 3) have higher ratios. These results show that the bulk composition of the condensates varied with temperature and experimental duration. The change of bulk composition with temperature can be explained either by fractionation of gas within the tube or kinetic barrier for condensation, where condensation of Si have larger kinetic barrier than Mg at higher temperatures. On the other hand, the change of bulk composition with time is explained either by the change of gas composition with time or the progress of reaction of gas and condensates with time. The latter is possible if Si has larger kinetic barrier for condensation than Mg. In summary, the temperature and time dependence of the bulk composition of the condensates are explained by the larger kinetic barrier of Si than Mg.

The present results are suggestive of the change of composition of condensates in outflow of evolved stars, where large temperature and pressure gradients are present which is similar to the present experimental conditions. The gas composition of the present experiments has Mg/Si ratio of 2, and the ratio of condensates varied from 3 to 1 with decreasing temperature. If the gas has the ratio of unity (~solar ratio), that of condensates would have Mg/Si from above unity to below unity in highly kinetic conditions. The condensates in such a condition would be forsterite, enstatite, and amorphous material rich in Si with decreasing temperature. The observation by IR that forsterite, enstatite, and amorphous silicate often coexist around evolved stars are well understood as the products of highly kinetic condensation.

References: [1] Molster F. J. et al. (2001) A & A, 366, 923-929. [2] Nagahara H. and Ozawa K. (1996) GCA, 60, 1445-1459. [3] Paule R. C. and Margrave J. L., pp.130-151. Wiley, 1967

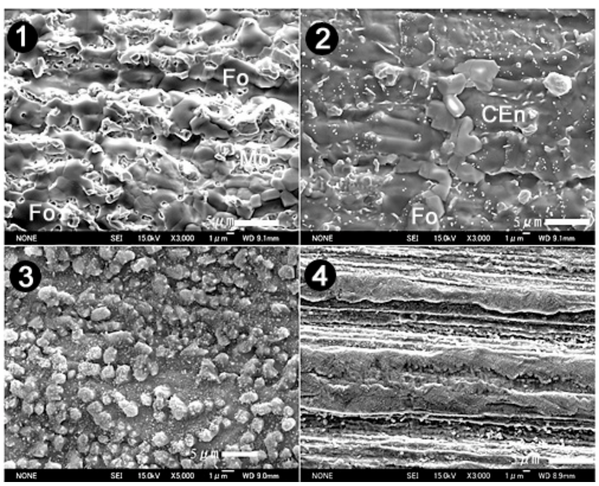

Fig. 1. FE-SEM images of condensates for 60hours. (1) Irregular shaped forsterite at ~1215°C. (2) Condensates at ~1160°C. Grains with brighter contrast are forsterite and those with darker contrast are clinoenstatite. (3) Clinoenstatite, intergrown with molybdenum at ~1110°C. (4) Amorphous silicates condensed at ~1000°C.

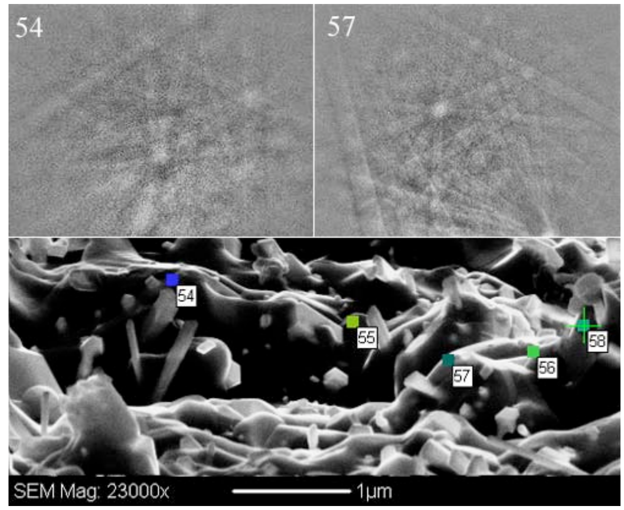

Fig.2. EBSD patterns and an FE-SEM image of condensates at ~1050°C for 60 hours heaitng. The numbers in the upper panels are the analyzed location shown in the lower panel. (54) shows patterns for clinoenstatite, and (57) are those for forsterite.

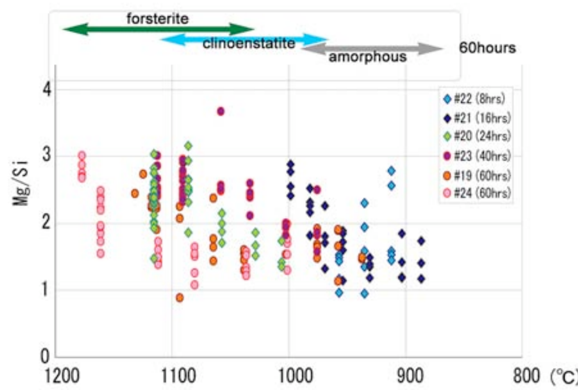

Fig.3. Chemical composition of condensates with different experimental durations. The Mg/Si ratio is plotted against the condensation temperature. Individual data represents a single analysis with focused beam.