

# EXPERIMENTAL STUDY OF CONDENSATION IN THE SYSTEM OF Mg-Si-O BY A NEWLY DEVELOPED INFRARED VACUUM FURNACE. S. Tachibana<sup>1</sup>, S. Tamada<sup>1</sup>, H. Nagahara<sup>1</sup>, and K. Ozawa<sup>1</sup>,

<sup>1</sup>Department of Earth and Planetary Science, University of Tokyo (7-3-1 Hongo, Tokyo 113-0033, Japan; tachibana@eps.s.u-tokyo.ac.jp)

**Introduction:** Magnesian silicates such as forsterite ( $\text{Mg}_2\text{SiO}_4$ ) and enstatite ( $\text{MgSiO}_3$ ) are major minerals in the solar system and in circumstellar environments. It is thus crucial to understand evaporation and condensation behaviors of magnesian silicates, which are major phase-transformation reactions at low pressures, in order to understand evolution of solid in space.

In this study, we developed a new infrared vacuum furnace, which have several advantages over vacuum apparatus used in previous experimental works, and performed condensation experiments in the system of Mg-Si-O using the infrared furnace with changing Mg : Si : O ratio in order to understand condensation behaviors of magnesium silicates at low pressures in a wide range of chemical compositions..

**Infrared Vacuum Furnace:** We designed a vacuum furnace consisting of a silica glass (~300 mm in length and 40 mm in outer diameter) tube connecting to a turbo molecular pumping system, two infrared heating systems, and a quadrupole mass spectrometer (Fig. 1). The ends of a silica glass tube was sealed to Kovar (Fe-Ni-Co alloy) and to vacuum flanges by changing chemical compositions of ends of the tube in order to obtain high vacuum. The vacuum in the furnace reached down to  $\sim 10^{-6}$  Pa. This pressure is much lower than the minimum pressure obtained in an infrared furnace, in which an O-ring and a rubber tube were used for glass sealing [1].

Infrared heating systems consist of halogen lamps and gold mirrors, and light from halogen lamps is focused by gold mirrors to heat a sample located in the silica glass tube. Temperature was controlled by a Pt-PtRh<sub>13</sub> thermocouple closely located to the sample.

There are several advantages in the new furnace: (1) Samples are heated from the outside of the furnace, so that no contamination from heating elements occurs, (2) The structure inside the furnace is simpler than vacuum furnaces with metal heaters and reflectors, which makes it easier to simulate gas flow in the furnace and to introduce reactive gas such as hydrogen and oxygen with a controlled flow rate at low pressures, (3) Due to localized heating and high transparency of silica glass against infrared light, the inside wall of the silica tube is cold enough for evaporated gas species containing metallic elements to condense, which reduces contamination by once-evaporated components.

However, there are a few disadvantages as well: (1) The maximum heating temperature cannot exceed  $\sim 1700^\circ\text{C}$ , (2) Samples highly transparent against infrared light are difficult to be heated.

**Condensation Experiments in the System of Mg-Si-O:** A single crystal of forsterite was used as a gas source of Mg-Si-O system. Because forsterite is transparent against infrared light, it was put in a graphite capsule for effective heating. The heating temperature was controlled at  $\sim 1650^\circ\text{C}$ , but the obtained evaporation rate of forsterite suggests that the actual temperature of forsterite may have been several tens degrees lower than the controlled temperature.

A substrate of molybdenum plate was put at various distances from the gas source to change condensation temperatures from  $1145$ – $480^\circ\text{C}$ .

Condensation experiments were carried out for 24–72 hours at a pressure of  $\sim 10^{-5}$  Pa, which is close to the pressure in a mass-loss wind from an evolved star.

The condensates were observed with a field-emission scanning microscopy (FE-SEM, JSM-7000F), and their chemical compositions and crystallinities were determined by energy-dispersive X-ray spectroscopy (EDS) and electron backscatter diffraction (EBSD) equipped with FE-SEM.

We also analyzed gas compositions inside the furnace during experiments.

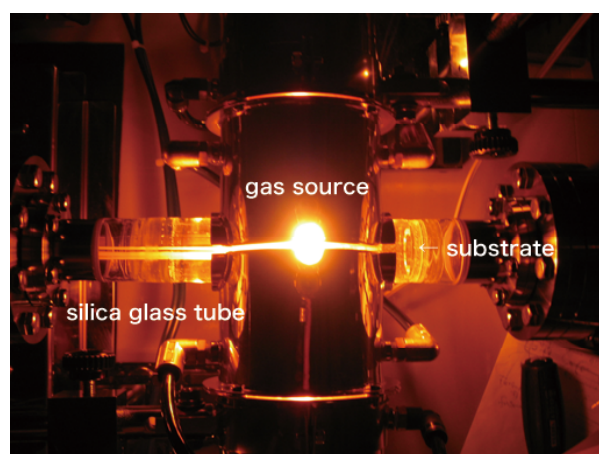
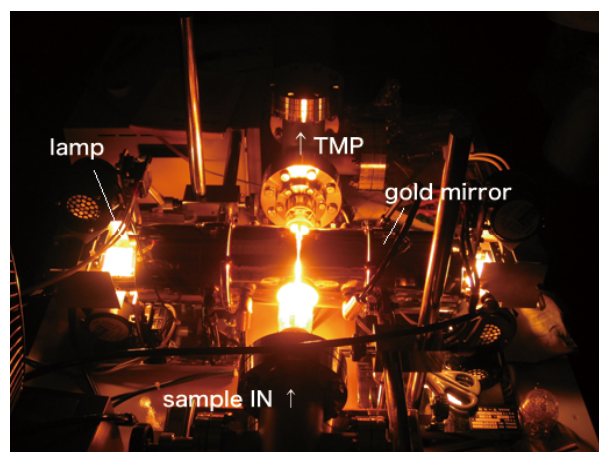
**Results and Discussion:** No condensation occurred on the substrate at  $1145$  and  $1040^\circ\text{C}$  except for small amounts of platinum, which was from the thermocouple, as Mo-Pt alloy. No silicate condensed at  $860^\circ\text{C}$ , either, but silicon condensed as Mo-Si alloy. Amorphous silicates condensed at temperatures of  $570$  and  $480^\circ\text{C}$ . There may be MgO in condensates at  $570^\circ\text{C}$ .

Such condensates found in the present study are different from those formed in the Mg-Si-O system in previous studies [e.g., 2, 3], where crystalline forsterite and enstatite and amorphous silicate condensed on substrates depending on temperatures.

Gas analyses by the quadrupole mass spectrometer showed that the graphite capsule reacted with residual gas in the furnace or oxygen evaporated from forsterite to form CO and CO<sub>2</sub> molecules. If most of oxygen from forsterite reacted with graphite, the Mg : Si : O in the gas phase available for condensation might be 2 :

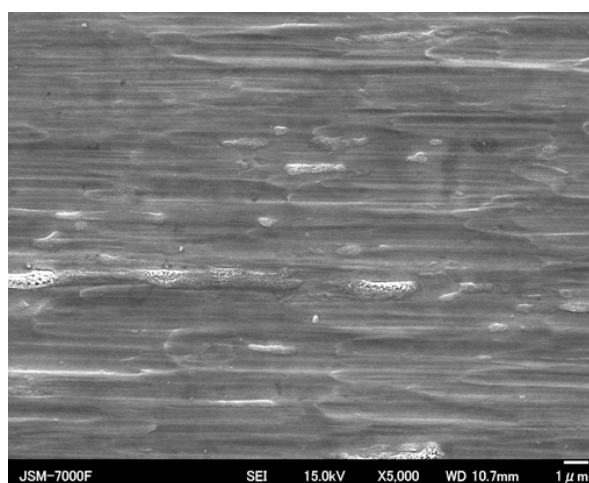
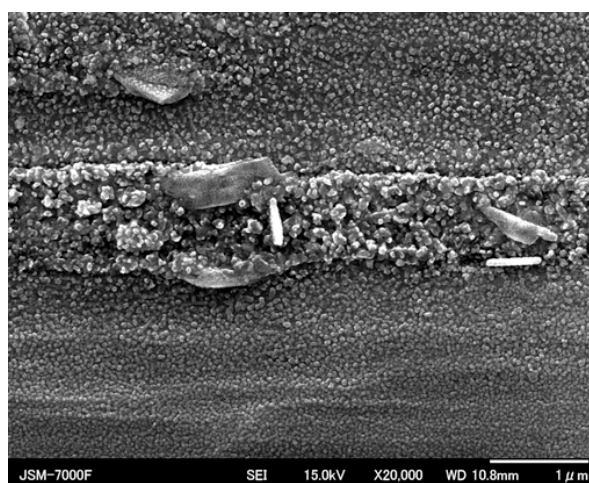
1 : <<4 (or even ~0). If there is little oxygen available for condensation, silicon may not condense as forsterite but an alloy at higher temperatures [e.g., 4]. Neither graphite nor SiC was observed as condensates, implying that the C/O ratio was not larger than unity.

The stability field of forsterite becomes smaller under reducing conditions [e.g., 5, 6], but it can still condense at lower temperatures. No crystalline forsterite was found in this study. This may be because the temperature of the substrate was too low to form crystals and/or because chances of encounter of three kinds of molecules containing Mg, Si, and O, respectively, under O-poor conditions. Although more systematic experiments are needed, the present study indicates that crystalline forsterite cannot condense at ~850 K under reducing conditions, but amorphous silicates condense.



**Figure 1:** A infrared vacuum furnace. A sample located inside a silica glass tube is heated by infrared light from two halogen lamps focusing by gold mirrors at the sample location. In the present study, forsterite was heated to generate gas in the Mg-Si-O system, and condensation of gas occurred on a Mo substrate located at various distances from forsterite.

**References:** [1] Tachibana S. and Tsuchiyama A. (1998) *GCA*, 62, 2005–2022. [2] Tsuchiyama, A. (1998) *Mineral J.*, 20, 50-89. [3] Ogawa R. et al. (2006) *LPS XXXVII*, Abstract #2415. [4] Lodders, K. and Fegley, B. Jr. (1999) in *Asymptotic Giant Branch Stars*, pp. 279-289. [5] Wood, J. A. and Hashimoto, A. (1993) *GCA*, 57, 2377-2388. [6] Lodders, K. and Fegley, B. Jr. (1995) *Meteoritics*, 30, 661-678.



**Figure 2:** FE-SEM images of condensates at 570°C (top) and 480°C (bottom). Scale bars are 1 μm.