

CONDENSATION OF MAGNESIUM SILICATES IN THE SYSTEM OF $Mg_2SiO_4-H_2O-H_2$: DEVELOPMENT OF LOW-PRESSURE INFRARED FURNACE. S. Tachibana¹ and A. Takigawa², ¹Dept. Natural History Sci., Hokkaido Univ., N10W8, Sapporo 060-0810, Japan (tachi@ep.sci.hokudai.ac.jp), ²Carnegie Institution of Washington, DTM, 5241 Broad Branch Road NW, Washington DC, 20015, USA.

Introduction: Condensation from vapor is a fundamental process responsible for dust formation in protoplanetary disks and in outflows from evolved stars. The kinetic aspect of dust formation should be quantitatively understood to evaluate the size, the number density, and the chemical variety of dust particles, which are the properties necessary to discuss elemental fractionations recorded among chemical groups of chondrites and to interpret infrared spectral features in circumstellar environments.

We have focused on growth kinetics of major minerals and have experimentally determined the growth kinetics of metallic iron [1] and corundum [2]. The condensation coefficient, a dimensionless parameter ranging from 0 to 1 to represent kinetic hindrances for condensation, is ~ 1 for metallic iron [1] and < 0.1 for corundum [2]. However, the growth kinetics of magnesium silicates, one of the most abundant solid materials in the system of solar abundance, has not yet been fully understood due to experimental difficulties in controlling growth parameters [3-5]. We conducted condensation experiments of magnesium silicates in the system of $Mg_2SiO_4-H_2O-H_2$ using an infrared vacuum furnace, and showed that the condensation of magnesium silicates might not occur as efficiently as metallic iron [6]. However, it has not yet been fully possible to make quantitative discussion on kinetics because there are still some experimental difficulties.

Here we report our recent improvements of the low-pressure infrared furnace and experimental techniques for condensation of magnesium silicates under protoplanetary disk conditions.

Concept of experiments: The concept of condensation experiment in this study is the same as those for metallic and corundum [1, 2]; a vapor evaporated from a gas source is condensed onto a substrate placed at a cooler region of the chamber. We use synthetic powder of forsterite (Mg_2SiO_4) put in an iridium crucible as a gas source and a synthetic single crystal of forsterite as a substrate. A difference from [1, 2] is to perform experiments in the presence of low-pressure H_2-H_2O gas to simulate the growth of magnesium silicates under controlled protoplanetary disk conditions (temperature, pressure, and gas chemistry).

Experimental Apparatus: The infrared vacuum furnace consists of a silica glass tube (~ 300 mm in length and 38 mm in diameter) connected to a turbo molecular pump and a rotary pump, and two infrared heating systems (Fig. 1) [6]. Another silica glass tube

(25 mm in diameter) is put inside, which can be replaced and cleaned after each run.

The chamber is connected to a gas line to flow a mixed gas of H_2 and H_2O (Fig. 1). The H_2O vapor is supplied by flowing H_2 through a water vapor saturator, in which an opened microtube containing distilled water (or ice) is placed. The temperature of the saturator is controlled in the range from 80 to $-40^\circ C$, and the gas flow rate is kept constant by a mass flow controller.

The gas-source forsterite in the Ir crucible is placed at the focal point of light from two heating systems, and is heated at a constant lamp current.

Experimental Conditions – Requirements and Current Status:

Total pressure. The plausible pressure range of the inner region of a protoplanetary disk is 100-0.1 Pa, of which dominant gas is molecular hydrogen [e.g., 7]. The pressure range of 100-0.1 Pa is fully covered in the present system by controlling a pumping speed of the pump system and the gas flow rate. A turbomolecular pump and a rotary pump are used for the runs at < 1 Pa, while only a rotary pump is used for most of the runs at > 1 Pa. The total pressure in the chamber is well controlled with a fluctuation of less than 5 % and has a spatial gradient of ~ 10 %.

Temperature. The gas source is required to be heated at a constant temperature higher than the substrate temperature in order to make a controlled supersaturated condition at the surface of the substrate. We heat the gas-source forsterite at a constant lamp current, and have found that the evaporation rate of the gas source shows a clear correlation with the lamp current (Fig. 2). The temperature of the gas source at the lamp current of 16.3 A is estimated to be 1900-1950 K by a pyrometer, which is consistent with the estimate based on the measured evaporation rate and the evaporation coefficient [8].

The equilibrium condensation temperature of forsterite is ~ 1450 - 1250 K in the system of solar abundance at a total pressure of 100-0.1 Pa [e.g., 9]. Considering that the condensation temperature also depends on the chemical composition of the system such as H_2O/H_2 and SiO/H_2 ratios, the temperature of the substrate is required to be controlled within the temperature range of ~ 1600 - 1100 K.

Because the substrate is heated by re-radiation from the Ir crucible, the temperature measured by a thermocouple is not exactly the same as that of the forsterite substrate that is more transparent than the thermocou-

ple. The temperature calibration of the substrate is thus done by using an irreversible transition of orthoenstatite (MgSiO_3) to protoenstatite at 1258 K. For instance, when a single crystal of synthetic orthoenstatite [10] is placed at 20 mm from the gas source, the ortho-proto transition occurs within 1-hour for heating of the gas source at the lamp current of >16.3 A. However, because the phase transition is a kinetic process, a time-temperature-transformation diagram is necessary to determine the temperature, which is now being made using a one-atmosphere furnace.

The temperature of the ambient H_2 - H_2O gas should also be close to the substrate temperature. Because the present furnace system heats only the gas source, in order to heat the ambient gas, we put a multi-layered platinum mesh nearby the Ir crucible (Fig. 1) as in [11]. We confirm that the platinum mesh is heated at a temperature higher than the ortho-proto transition of enstatite at the lamp current of 18 A. The mean free path of hydrogen gas is less than 1 mm at the total pressure of >1 Pa, which is shorter than the diameter of the silica glass tube, so that the temperature of the gases evaporated from the gas source is expected to be homogenized to that of the ambient gas by collisions.

Gas chemistry. The $\text{H}_2\text{O}/\text{H}_2$ ratio of the ambient gas is controlled by the temperature of the water vapor saturator, and can be checked by the weight change of water in the microtube before and after each run. The ratio is well controlled at the ratio expected from the saturation pressure of H_2O at a fixed temperature (Fig. 3), including the solar $\text{H}_2\text{O}/\text{H}_2$ ratio of $\sim 10^{-3}$.

The SiO/H_2 ratio of the gas hitting the substrate is determined from the evaporation rate of the gas source. Because a part of evaporated gases condense onto the wall of the silica tube, its effect should be evaluated. Our preliminary experiment on condensation of metallic iron onto an alumina substrate, of which condensation coefficient is nearly unity [1], under the same experimental condition showed that $\sim 1\%$ of evaporated gas hit the substrate surface [6]. Although further experiments are required to determine this factor more precisely, the SiO/H_2 ratio at the surface of the substrate could be ~ 0.4 x the solar ratio at the total pressure of 5.3 Pa and at the lamp current of 16.3 A.

In summary, the low-pressure infrared furnace has been developed and improved to simulate the dust growth under protoplanetary-disk conditions. We will use the furnace to determine the condensation kinetics of forsterite as a function of temperature, pressure and gas chemistry, which will be reported at the meeting.

References: [1] Tachibana S. et al. (2011) *ApJL*, 736, 16. [2] Takigawa A. et al. (2012) *LPS XXXXIII*, #1875. [3] Nagahara H. et al. (1988) *Nature*, 331, 516. [4] Tsuchiyama A. (1998) *Mineral J.*, 20, 50. [5] Nagahara H. et al. (2009) In

Cosmic Dust - Near and Far, 403. [6] Tachibana S. et al. (2011) *LPS XXXXII*, #2682. [7] Wood J. A. (2000) *Space Sci. Rev.*, 92, 87. [8] Takigawa A. et al. (2009) *ApJL*, 707, L97. [9] Ebel D. (2006) In *Meteorites and the Early Solar System II*, 253. [10] Tachibana S. et al. (2002) *GCA*, 66, 713. [11] Kuroda D. and Hashimoto A. (2002) *Antarct. Met. Res.*, 15, 152.

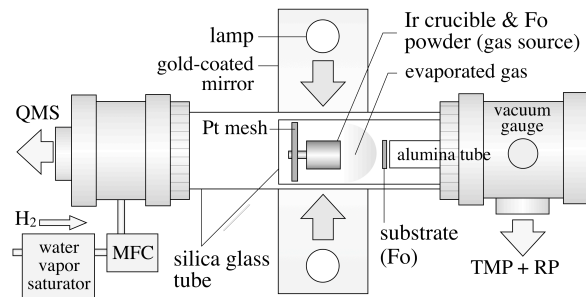


Fig. 1. Schematic illustration of the infrared furnace.

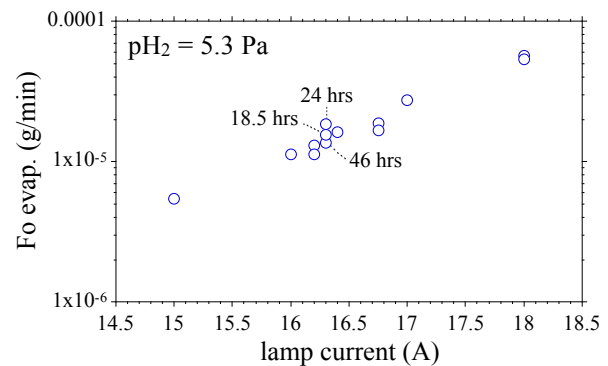


Fig. 2. Evaporation rate of the gas-source forsterite at the H_2 pressure of 5.3 Pa. The heating duration is 1 hour except for three runs at the lamp current of 16.3 A.

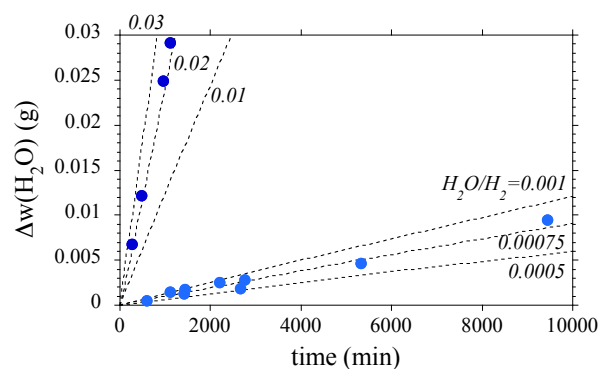


Fig. 3. The weight changes of water in the water vapor saturator at a room temperature (blue solid circles) and at -16°C (light-blue solid circles) as a function of the gas-flowing duration. The pressure of H_2 in the gas line is 2 atm, and the gas flow rate is set at $1.5\text{ cm}^3/\text{min}$. The dashed lines represent the predicted weight losses for different $\text{H}_2\text{O}/\text{H}_2$ molar ratios (shown in italic).