CONDENSATION KINETICS OF METALLIC IRON: AN EXPERIMENTAL STUDY. K. Tatsumi , H. Nagahara, K. Ozawa, and S. Tachibana. Department of Earth and Planetary Science, University of Tokyo, 7-3-1 Hongo, Tokyo 113-0033, Japan. e-mail : chichaneko@eps.s.u-tokyo.ac.jp

Introduction: Metallic elements in gaseous state in a hot interstellar environment condense into dusts during cooling of the gas [1]. In recent IR surveys, existence of magnesium silicates around either evolved stars or young stars have come to evident [2]. Although there has been no observation so far clearly indicating the presence of crystalline Fe, one of the most abundant metallic element in the solar composition next to magnesium and silicon, it is expected to be present as dusts because of its abundance. We have studied condensation kinetics of Fe for the better understanding of condensation processes in the circumstellar environment, such as time scale of condensation, by taking the advantage of its simple crystalline structure and chemistry.

It is unclear how much atoms once collided the already condensed solid surface stick there as new part of solid. The ratio of atoms that incorporate into the crystalline structure to incoming atoms is called a condensation coefficient that is critical to determine the growth rate of solid when considering the 'gas to solid' condensation. In some models which calculated the growth rate of condensate, this value was treated as unity [3, 4]. However, if this coefficient is significantly smaller, result of calculation of the growth rate becomes much lower and the time scale of condensation becomes much longer than before.

In this study, the condensation coefficient of metallic iron is directly estimated experimentally by the measurement of weight loss of evaporation source and weight gain of a target due to condensation.

Note that the first step of condensation is nucleation. To nucleate from gas, high supersaturation state should be achieved to acquire the steady growth state as solid particle. Therefore, the difficulty that an atom can stay on the solid surface once stuck is different in the case of growth state from nucleation state. In this study, growth state is mainly considered.

Method: The condensation experiments were carried out in a vacuum chamber with a high temperature tungsten mesh heater. The chamber is always evacuated by a turbo molecular pump. Gaseous iron was produced by heating a metallic iron plate that was set at the center of the heater. The iron gas condenses on a molybdenum substrate set apart from the gas-source plate. The temperature of the substrate varies depending on the distance from the source. The temperature of substrate is not independently controlled, and therefore, the flux to the substrate surface varies depending on the distance from the source as well as temperature.

The heating temperature of the iron source was 1170°C. The size of the iron source is 12x12mm. The substrate was set either at 54mm or 88mm away from the source and the measured temperatures are 1020°C and 415°C for the distances of 54mm and 88 mm, respectively.. The area of condensation surface was 178 mm². The pressure inside the chamber is at $10^{-5} \sim 10^{-4}$ Pa during the experiment. The experimental duration was 6 to 144 hours at 415°C and 1 to 48 hours at 1020°C.

The weights of the iron source and the substrate were measured before and after experiments. The weight losses of the source and weight gains of the substrate were converted to evaporation and condensation rates, respectively, by dividing by the experimental duration and by the surface area of the source and condensable area of the substrate, respectively. Condensates were observed with a FE-SEM for surface morphology, analyzed with EDS for chemical composition, and with EBSD for the crystallinity and phase identification. Thickness and cross-sectional morphology of the condensates are also examined with a FE-SEM.

Results: The weight loss of the iron plate source increased linearly with time, and the calculated evaporation rate is consistent with that in [5]. The phase and crystallinity of the condensates determined with EDS and EBSD are metallic iron regardless of the experimental conditions, and any other phases were not observed. The weight of the condensates increased linearly with time, and the condensation rate is larger at high condensation temperature (Fig. 1).
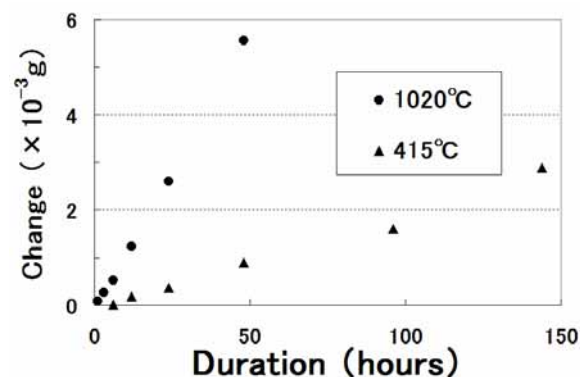

Fig. 1. Weight changes of the substrate (weight of condensates) plotted against experimental duration for substrate temperatures of 415°C and 1020°C.

The temperature of substrate greatly affects morphology of condensates. Loosely grown dendritic Fe

metal covered the substrate at 415°C, but compactly packed Fe metal grew at 1020°C (Fig. 2). In 415°C experiment, in which crystal size can be easily measured, crystal size increases with experimental time (Fig. 3), although the size change cannot be determined at 1020°C experiment due to the compact surface.
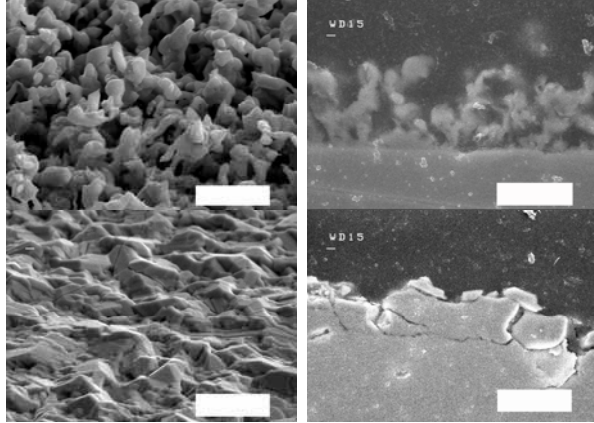

Fig. 2. FE-SEM images of surface of condensates (left panels) and those of cross sections (right panels) obtained in experiments at 415°C for 96 hours (upper panels) and at 1020°C for 24 hours (bottom panels). In the cross section images, solid lower part in the upper right panel and lower 2/3 below fracture in the lower right are molybdenum substrate. The scales for upper panels are both 3μm, for lower left panel 6μm, and for lower right panel 5μm.
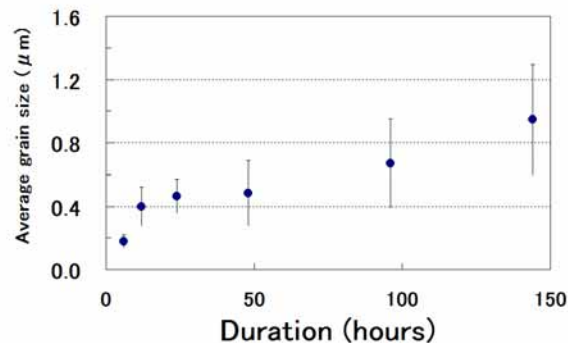

Fig. 3. Change of average crystal size condensed at 415°C. Vertical bars represent mean standard deviations.

Discussion: The velocity of Fe atoms evaporating from the source is ~800 m/s, and the mean free path is greater than 100 m., indicating a molecular condition is fulfilled in the experimental chamber with outer diameter of 30 cm. The surface microstructure of condensates gives some implications about the growth process. In the 415°C experiment, Fe atoms are inferred to have condensed to cause growth of already existing crystals with very minor formation of nuclei, because the average size increases with experimental duration. The loosely packed dendritic structure of condensates suggests a high condensation coefficient

at this temperature because the high sticking probability inhibit for Fe atoms to go deep inside the dendrite. On the other hand, at 1020°C, the condensates are likely to be annealed due to the higher temperature of substrate, although a lower condensation coefficient might facilitate the compact structure.

In order to evaluate the efficiency of condensation, the incident flux of Fe atoms onto the substrate should be precisely determined. There may be two sources for the incident flux: one is the flux that comes directly from the Fe metal source and the other is ambient pressure build inside the heating chamber. The former is estimated by assuming the Fe plate as a point source to be 1.8% of the total evaporation flux when the substrate is set at 54 mm away from the source for the substrate temperature of 1020°C. The latter flux is estimated by an experiment, in which the direct flux was shielded. The wait gain of this experimental configuration results in reduction by 80%, which determines the absolute flux came from the ambient Fe gas. Based on these experiments, the condensation coefficient is estimated to be 0.5~0.6 at 1020°C. Similar calculation for the experiment at 415°C substrate gives the condensation coefficient of ~0.8.

The observed temperature dependence of the condensation coefficient for 1020 and 415°C is different form that for the evaporation coefficient for the temperature range of 1074-1445°C, where the evaporation coefficient is larger at higher temperatures [5]. The latter is because it is probably easy for atoms in a crystalline structure or in a half-crystal position to break bonds with adjacent atoms at higher temperatures and evaporate. On the other hand, the larger condensation coefficients at lower temperatures may be explained as follows. The incoming Fe atom exchanges its thermal energy efficiently with the surface of substrate. If this thermal accommodation is efficient for iron gas and solid (i.e., a large thermal accommodation coefficient), it is difficult for the atom stuck on the surface to re-evaporate especially at lower temperatures. Those stuck atoms may incorporate into the crystalline structure without moving on the substrate surface, resulting in efficient growth of condensates. More detailed and systematic experiments are needed for more quantitative discussion.

References: [1] Larimer, J. W. (1967) *GCA, 31,* 1215-1238. [2] Molster, F. J. et al. (2001) *Astron. & Astrophys., 366,* 923-929. [3] J. R. Burke, J. R. and Hollenbach, D. J. (1983) *Ap.J., 265,* 223-234. [4] Kozasa, T. and Hasegawa, H. (1987) *Prog. Theor. Phys., 77,* 1402-1410. [5] Tachibana, S. et al. (2001) *LPS XXXII,* abstract #1767.