CONDENSATION EXPERIMENTS OF MG-RICH CRYSTALLINE AND AMORPHOUS SILICATES IN VACUUM.

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Introduction: Magnesian silicates such as forsterite (Mg_2SiO_4) and enstatite ($MgSiO_3$) are major minerals in universe including disks around young stars [e.g., 1], shells around evolved stars, in particular, oxygen rich stars [e.g., 2, 3] and so on. It is thus important 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. We have been carrying out condensation experiments of silicates under non-equilibrium conditions in vacuum.

In this study, we will present experimental study on condensation of silicates to understand formation of silicate dusts in space. We describe the concept of our experiments and experimental techniques, and experimental results, and discuss the interpretation of the results for dust formation in universe, especially around AGB stars.

Experimental: Condensation of dusts proceeds through nucleation and growth. Nucleation is hardly observed and hardly controlled in experiments, and we perform condensation experiments focusing on a growth process of Mg-silicates to know the condensing phases under highly kinetic conditions and critical conditions for condensation, that is, the relationship between condensation coefficient and the supersaturation ratio. Two types of experiments have been carried out: one is "quench" experiments, where gas generated at high temperature collides with a substrate at much lower temperature [4]. No further surface process takes place, and condensing phases as a function of pressure or temperature are examined.

The second type of experiment is condensation in the environment where gas atoms and molecules interact in a cooling condition, which we call "cooling" experiments [5]. The gas source is synthesized single crystal of forsterite, which was heated to generate Mg, SiO, and O gas molecules with the ratio of 2: 1: 3. Condensation took place on a substrate.

Results: Quench experiments: Equilibrium condensation temperature of forsterite in the quench experiments is about 1200°C, and it should be the only stable phase at lower temperature as far as the bulk gas composition is kept stoichiometry of forsterite. No condensation occurred on the substrate at 1145 and 1040°C except for small amounts of platinum, which was from the thermocouple, as Mo-Pt alloy. No silicate condensed at 860°C, either, but silicon condensed as Mo-Si alloy. Amorphous silicates condensed at

temperatures of 570 and 480°C. There may be MgO in condensates at 570°C.

Such condensates in quench experiments, summarized in Fig.1, are different from those formed in the Mg-Si-O system in previous studies for cooling experiments [e.g., 5, 6], where crystalline forsterite and enstatite and amorphous silicate condensed on substrates depending on temperatures.

The condensate of metallic Si is not an expected condensate, because gas species is SiO. It means that reduction of SiO to Si happened at the surface of the substrate. The condition is within the stability filed of Si if we ignore the forsterite stoichiometry of the gas, which suggests that the atmosphere within the tube is quite out of equilibrium due to the absence of interaction among gas species and that individual gas species behave according to its own system.

Amorphous magnesian silicates condensed at lower temperatures, which could be because condensation of SiO was allowed and Mg began to condense once amorphous silicates formed. The condensed amorphous has the composition of Mg:Si~1:1, which means larger kinetic barrier for condensation is larger for Mg (MgO) than Si (SiO or SiO_2).

The condensation behaviors of silicates in quench experiments could be explained by smaller incoming fluxed of Mg, SiO, and O onto the substrate than those in previous studies with condensation of crystalline silicates (cooling experiments).

Cooling experiments: The condensates showed a systematic change with temperature in terms of phase and composition (Fig.1). The highest temperature (T~1180°C) condensate is platy crystalline forsterite, and clinoenstatite starts to condense at ~1150°C along with forsterite. The relative abundance of forsterite to clinoenstatite decreases with lowering temperature, and all the condensates become clinoenstatite at ~1090°C. The condensates below 1050°C are amorphous. All the condensates are silicates and they are what we have expected to appear under kinetic conditions. A significant difference from the quench experiment is that the gas pressure is much higher with larger condensation flux. Condensation of forsterite from a gas with Mg/Si~1 is not unusual, because forsterite is the phase condenses at first from gas in a wide range of Mg/Si of gas in the Mg-Si-O system. The condensation temperature of ~1180°C is lower than the equilibrium condensation temperature of ~1250°C, which corresponds to a supersaturation ratio for condensation of forsterite, S ($=P/P_{eq}$), of 10 - 50.

Discussion: The experimental results suggest an importance of gas kinematics for condensation of silicate dusts that are products from multiple gas species such as Mg, SiO, O (or O_2) and H_2O. If there is little or no interaction among gas species, gas atoms or molecules do not necessarily know the net equilibrium condition, and they behave as if they are in independent systems. For example, reduction of SiO occurred in quench experiments even in an oxidizing condition as a whole because SiO is not an energetically stable species in the very localized Si-O system. On the other hand, amorphous silicate was formed at a similar condition in terms of total pressure, temperature, gas composition and constituting gas species but with a different gas motion. Possibility of condensation of metastable phases in a stellar wind was discussed by [7] that amorphous silicate and MgO condense at around 1000K whereas SiO condenses at much lower temperature (> 600K), none of which is a thermodynamically stable phase. Our experiment, however, suggests that amorphous Mg-silicate condenses at several hundreds K but that SiO or Si might condense before the amorphous silicate as far as condensation did proceed heterogeneously.

The gas density of winds of AGB stars is low, which corresponds to 10^{-9}-10^{-11}bar for Mg-silicates [e.g., 8]. If the gas motion is unidirectional at such a low-density region, condensation of silicate dusts is difficult. It in turn suggests the importance of turbulence or even the presence of higher density regions due to heterogeneity of the dust shell. Pulsation of AGB stars and the observed clumpy structures would be important for dust formation, although they have been thought to be results of dust formation [e.g., 7].

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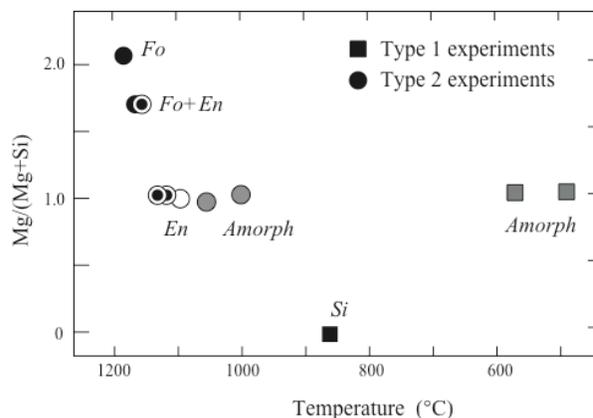

Figure 1. Condensates in quench (Type 1) and cooling (Type 2) experiments. The Mg/(Mg+Si) of condensates is plotted against experimental temperature. Fo; forsterite, En; enstatite, Amorph; amorphous silicate.