

## Crystallization of amorphous enstatite dust in protoplanetary disks

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Infrared spectroscopic observation of astronomical environments showed that silicate dust is almost amorphous in the interstellar medium, while crystalline and amorphous silicate dusts are present in protoplanetary disks (Waters et al., 1996; Kemper et al., 2004). This indicates that amorphous silicate dust is a precursor of dust in protoplanetary disks and transform into crystalline dust due to thermal processes in protoplanetary disks. Moreover, disk observation has shown that the fraction of crystalline enstatite dust relative to crystalline forsterite is larger in the inner disks, (Bouwman et al., 2008). These dust distributions should reflect physical conditions of protoplanetary disks and dust evolution in the disks.

Experimental studies have been performed to understand crystallization of amorphous silicates (e.g., Fabian et al., 2000). Crystallization of amorphous forsterite was found to be promoted by water vapor (Yamamoto and Tachibana, in prep.). However, the effects of ambient gas on crystallization have not yet been fully investigated. In this study, we conducted crystallization experiments of amorphous enstatite in various ambient conditions.

Amorphous enstatite, synthesized by an induced thermal plasma method, was annealed under five different conditions; in air ( $P_{\text{H}_2\text{O}}$  of  $\sim 10^{-3}$  bar), at  $P_{\text{H}_2\text{O}}$  of  $\sim 10^{-5}$  bar, in vacuum ( $P_{\text{H}_2\text{O}}$  of  $\sim 10^{-9}$  bar), at  $P_{\text{H}_2}$  of  $\sim 10^{-4}$  bar, and at a total pressure of  $10^{-4}$  bar of  $\text{H}_2\text{O}-\text{H}_2$  mixed gas ( $\text{H}_2\text{O}/\text{H}_2 \sim 10^{-3}$ ). We focused on the effects of total pressure and partial pressures of water vapor and hydrogen on crystallization kinetics. Heated samples were examined with FT-IR, XRD, SEM, and TEM.

SEM observation of heated samples showed that the aggregates in air had a framboidal rounded shape, while those heated under low-pressure conditions had a blocky irregular shape with sharp edges. This indicates that effective aggregation occurred during annealing under low-pressures probably because of vapor transport under low pressures. The dominant crystalline phase in heated samples was confirmed to be orthoenstatite by XRD and TEM.

We obtained time constants for crystallization and Avrami parameters from the time evolution of infrared spectra using the Johnson–Mehl–Avrami equation. The activation energy for crystallization decreases with increasing the water vapor pressure, indicating that water vapor lowers the kinetic barrier for crystallization. The crystallization rate is slower in air than that at low pressures at the experimental temperature range. Crystallization at  $P_{\text{H}_2\text{O}} \sim 10^{-5}$  bar occurs at the fastest rates among those under all the conditions studied. Crystallization at  $P_{\text{H}_2} \sim 10^{-4}$  bar occurs slower than that in vacuum, which indicates hydrogen does not promote the crystallization. Because the Avrami parameter is almost 2.5 in air, the crystallization mechanism is likely to be 3D diffusion-controlled growth with homogeneous nucleation. At low pressures, selective evaporation of Mg and O may occur from amorphous silicate (Rietmeijer et al., 1986). Heterogeneous nucleation of crystalline enstatite may take place at the grain surface due to effective chain structure formation, which may lead to faster crystallization. TEM observation showed grains having crystalline lattices only near their surfaces. This crystallization mechanism promoted by evaporation may explain the intense aggregation at low pressures and may explain the smaller

crystallization rate at the higher total pressure.

Crystallization behavior of amorphous enstatite in protoplanetary disks was examined based on the crystallization rates in the present study. We found that crystallization of amorphous enstatite dust occurs within the disk lifetime only at  $>900$  K and that crystalline enstatite can be present only in the inner warm part of the disk as suggested by Imai (2012).

Keywords: protoplanetary disks, silicate dust, crystallization, kinetics