

非晶質フォルステライトの結晶化における水蒸気圧効果 An experimental study on the effect of water vapor on crystallization of amorphous forsterite

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Infrared spectroscopic observations (e. g. ISO and Spitzer Infrared Telescope) have provided the evidence of existence of crystalline silicate dusts in comets and protoplanetary disks (Henning, 2010 and references therein). Wagstaff and Richards (1966) suggested that water vapor enhances the crystallization rate of SiO₂ glass by breaking [Si-O-Si] bonds and forming hydroxyl groups. Thus, if amorphous silicate dust is exposed to relatively water vapor-rich environments such as the post shock region of shock wave (Ciesla et al., 2003) and impact plumes generated by asteroid impacts (Fedkin & Grossman, 2013), amorphous silicates may crystallize more effectively with the aid of water.

In this study, in order to investigate the effect of water vapor on the crystallization kinetics of amorphous forsterite, crystallization experiments were conducted in vacuum condition ($\sim 10^{-4}$ Pa) using a gold-image vacuum furnace (Thermo-Riko GFA430VN) at 500, 680, 730, 750 °C, and in sealed glass tubes, in which water vapor pressure is kept at 0.65 bar by a Ca(OH)₂ – CaO buffer system, at 500 °C in a box furnace. Amorphous forsterite powder, synthesized by a thermal plasma method, was provided by A. Tsuchiyama, Kyoto University. Temperature of both furnaces was calibrated against the melting points of NaCl, KBr, LiBr and In. Run products were analyzed with FT-IR (KBr pellet method). Quantitative analysis of the degree of crystallization was made with the spectral fitting of run products in the 10 μm band, where the structural evolution of amorphous forsterite can be observed as a change of Si-O stretching features.

The time-dependence of crystallization in vacuum was estimated by the Johnson-Mehl-Avrami equation for each temperature, and the Arrhenius plot of the time constant of crystallization, τ , showed a linear correlation with the reciprocal temperature. The obtained value of E_a/k_B was 4.94×10^4 K, where E_a is activation energy for crystallization and k_B is the Boltzmann constant. Kinetic parameter n in the Johnson-Mehl-Avrami equation obtained at 680, 730, 750 °C in vacuum were ~ 1.5 . Assuming that the crystallization mechanism in vacuum does not change at lower temperatures, we can estimate the timescale of crystallization at 500 °C in vacuum, which is about 430 years for the crystallization degree of 26 %. On the contrary, experiments at $P_{H_2O}=0.65$ bar showed that the degree of crystallization reached about 26 % only for 12 hours. It was also experimentally confirmed that amorphous forsterite remained unchanged by heating at 500 °C in vacuum for 72 hours. This clearly indicates that the crystallization of amorphous forsterite is promoted in the presence of water vapor. Kohara et al. (2004) reported the structure of Mg₂SiO₄-composition glass synthesized by a containerless liquid phase processing technique, and MgO_x units act as a network former and SiO₄ units form polymer and dimer. We proposed that water molecules diffuse into the amorphous structure to break Si-O-Si bonds and MgO bonds by acting as a network modifier and promote the crystallization of amorphous forsterite.

Experiments at lower water vapor pressure conditions are needed for a direct application to the crystallization of amorphous silicates in canonical protoplanetary disks, but the present results imply that the crystallization of amorphous silicates might take place more effectively in the water-enriched regions compared with canonical solar nebula condition.

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