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Crystallization processes by heating from amorphous silicate nanoparticles with enstatite and forsterite compositions

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Circumstellar and interstellar dust has been investigated by comparing infrared spectra of astronomical observations and laboratory measurements, and crystalline silicates (e.g., olivine ($(Mg;Fe)_2SiO_4$), and pyroxene($(Mg;Fe)_2SiO_3$)) were found in circumstellar environments around oxygen rich young and evolved stars(e.g., [1,2]). In contrast, it is believed that interstellar silicate dust is almost completely amorphous [3]. Therefore, a precursor material for the crystalline silicates in circumstellar regions of young stars is considered to be interstellar amorphous silicate dust. In circumstellar regions around evolved stars, it is considered that amorphous silicates condense from out flow gas and are partially crystallized by heating (e.g., [4]). In order to reveal the conditions of circumstellar environments, it is important to understand crystallization process of the silicates.

In order to understand crystallization process of amorphous silicates, we synthesized amorphous silicates with the enstatite(En) composition (Mg/Si =1) and the forsterite (Fo) composition (Mg/Si =2) using the radio frequency thermal plasma processing at Nisshin Engineering Co. Ltd. TEM observation of these amorphous materials showed that grains of the both samples are spherical in shapes and about 50-200 nm in diameters. The amorphous samples were heated at 650-850℃ for 10 min-240 hrs to investigate the temperature and time dependence of the crystallization. Clinoenstatite ($MgSiO_3$) and forsterite (Mg_2SiO_4) were crystallized from the starting amorphous materials of the Mg/Si ratio of 1 and 2, respectively. A lot of stacking faults were observed in the clinoenstatite crystals under TEM.

The infrared absorption spectra of the heated samples were measured, and the degrees of crystallization were estimated by fitting the infrared spectra of the heated samples. Then, we evaluated a time constant of the crystallization, t, and a kinetic parameter, n, at constant temperatures using the Johnson-Mehl-Avrami equation, and finally estimated a frequency factor, v_0, and the activation energy of crystallization, Ea, by Arrhenius plot. The value of Ea/k for enstatite was 8×10^4(K), while the value of Ea/k for forsterite was lower than that for enstatite. In terms of the kinetic parameter, the values of n of enstatite and forsterite were about 2.4 and 1.4, respectively. This result suggests that the crystallization process of enstatite from the amorphous silicate is different from that of forsterite.

We made Time-Temperature-Transformation (T-T-T) diagrams using Ea and $v_0$obtained in the present experiments and compared the T-T-T diagrams with previous experimental studies, where amorphous silicates were synthesized by sol-gel method and crystallized by heating [4,5]. It is suggested that Ea (En) is higher than Ea (Fo) and t(En) is longer than t(Fo) at the same temperature. The value of kinetic parameter for enstatite, n(En), is close to 2.5, while that of forsterite is close to 1.5. These values suggests that enstatite crystallization occurs as nucleation and three-dimensional diffusion-controlled growth, while forstarite crystallization occurs as three-dimensional diffusion-controlled growth without nucleation.

[1] Waelkens, C., et al. 1996, A&A 315, L245.
[2] Waters, L. B. F. M., et al., 1996, A&A 315, L361
[3] Kemper, F., et al. 2004, ApJ 609, 826.
[4] Murata et al., 2009b, ApJ., 697, 836
[5] Murata et al., 2007, ApJ., 668, 285,