

Structural modification in amorphous MgSiO_3 with heat treatments: a role of water

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Anhydrous silicate dust such as enstatite (MgSiO_3) or forsterite (Mg_2SiO_4) are observed around the circumstellar environment [1]. In contrast the interstellar dust is almost amorphous [2]. They were incorporated into the circumstellar disk of young stars, and crystalline silicates could form by heating. In order to understand the formation and evolution of the circumstellar dust, crystallization of amorphous silicates have been performed (e.g., [3-6]). Imai [3] carried out heating experiments of amorphous MgSiO_3 prepared by an induction thermal plasma (ITP) technique. In powder X-ray diffraction (XRD) profile of the run product heated for 3 hrs. at 800°C , some broad and weak peaks appeared in addition to an original halo peak of the starting amorphous material. After 6 hrs. heating, their weak peaks disappeared, and 12 hrs. heating, enstatite crystallized. An infrared (IR) absorption feature of the amorphous silicate was observed at around 18 μm . After 3 hrs. heating at 800°C , a 22 μm shoulder peak grew. In the spectrum of heating for 6 hrs., the 18 μm feature grew again while the 22 μm shoulder peak became unclear. These XRD and IR features may be caused by structural changes of SiO_4 network and/or Mg^{2+} site in the amorphous silicates. The 18-22 μm features of the IR spectra may also affect the estimation of silicate dust species around the circumstellar regions. Thus, we measured high energy X-ray diffraction (HEXRD) data and X-ray absorption fine structure (XAFS) spectrum of the amorphous samples to investigate bond distance and coordination numbers. Previous heating experiments of amorphous Mg-silicates synthesized by sol-gel method showed the similar weak and broad peaks in their starting materials by XRD and these peaks disappeared after heating [4-6]. The sol-gel method is performed with water, suggesting the peak transition may be related to the effect of water. We examined the effect of hydration to the XRD and IR features too.

Both total correlation functions obtained in the HEXRD of the starting material and that of the run products of the heating experiments show that Mg-O distance of the run product of heating for 3 hrs. is slightly longer (~ 0.205 nm) than that of the starting material and heating for 6 hrs. (~ 0.200 nm). This feature is also supported by the XAFS data, suggesting the XRD and IR features are related to the changes of the Mg-O distance. The XRD pattern of the run product on the hydrothermal experiment at 150°C for 3 weeks with a water/rock ratio of 0.1 by weight has some weak peaks and the features are very similar to those of the sample heated at 800°C for 3 hrs. These results suggest that the weak peaks correspond to hydrous phyllosilicates and should relate to the mixed-layer mineral of serpentine and stevensite [7], for example. In the ITP method, H_2O or OH molecules (or H and O separately) which were originated by decomposition of $\text{Mg}(\text{OH})_2$ of the starting material, should be incorporated into the amorphous MgSiO_3 during the rapid condensation. In this case, a possible scenario is follows: (1) phyllosilicate minerals crystallized from the O and H bearing amorphous silicate by heating for 3 hrs., (2) they were dehydrated and became amorphous for 6 hrs., and finally (3) anhydrous silicate (enstatite) crystallized by heating for 12 hrs.

The present results suggest that H_2O (or H and O) should be included in amorphous silicates condensed in circumstellar regions and hydrous silicates may form by mild heating, such as at ~ 1000 K for a few hours, although more kinetic experiments are needed.

References: [1] Tielens et al. (1997) *Ap&SS*, 255, 415-426 [2] Kemper et al. (2004) *ApJ*, 609, 826-837 [3] Imai (2012) PhD thesis [4] Murata et al. (2009) *ApJ*, 697, 836-842 [5] Matsuno et al. (2012) *ApJ*, 753, 141-147 [6] Thompson et al. (2012) *A&A*, 545, A60 [7] Takahashi et al. (2013) *JpGU Meeting*, this volume

Keywords: dust, amorphous silicate, laboratory experiment