Self-Diffusion of Atoms in Forsterite Glass

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Forsterite (Mg_2SiO_4) glass exists as dust grains in interstellar molecular clouds and young stellar objects [1]. In interstellar molecular clouds, elements such as hydrogen, oxygen, carbon, and nitrogen deposit on dust grains, and form various molecules (e.g., H_2O, CO, CO_2, NH_3, CH_4, H_2CO, CH_3OH, and so on) [2]. These molecules undergo chemical evolutions to organic molecules through various processes on the surface of dust grains [2]. The structure of forsterite glass is one of the important factors governing the chemical evolutions in interstellar molecular clouds. However, the structure of forsterite glass is less conclusive, because forsterite glass is easily to crystallize [3]. To investigate the mechanism of phase transition of forsterite glass, molecular dynamics (MD) calculations were performed.

The MD calculations were performed using an atom-atom potential model with MXDORTO program [4]. The potential parameters were empirically determined by constraining the model to reproduce the experimental results of density, thermal expansion coefficient, and bulk modulus [5]. A fundamental orthorhombic cell consisting of 160 Mg_2SiO_4 with three-dimensional periodic boundary conditions was used. The glass structure was prepared by quenching the liquid phase from 3000 K to 10 K with 0.4 K/ps in rate. The quenched glass was warmed to 3000 K with the same rate. The MD code was run with NTP ensemble. The pressure was kept at 0.1 MPa.

The result shows that the increasing rate of enthalpy and the decreasing rate of density during the warming process change at 1567 K. This changing point is assigned as the transition point from glassy state to supercooled liquid state (T_g), because the value is significantly lower than the melting point of forsterite (2418 K for our potential model [5]). To investigate the transition mechanism, the self-diffusions of atoms were analyzed using mean-square displacement (MSD). At temperatures just below T_g, the temporal increase of MSD for magnesium was observed, whereas MSD of silicon was approximately constant. This indicates that the glass-supercooled liquid transition is induced by the self-diffusion of MgO_x unit. From the analysis of spatial distribution of MSD of magnesium at temperatures just below T_g, it was found that the MSD value is inhomogeneous and local regions, which has a larger MSD value, exist in a glass structure. It is concluded that the forsterite glass around T_g is in a state of dynamic heterogeneities.

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