

Forsterite-MgSiO₃ liquid interface : molecular dynamics perspective

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Knowledge about the viscosity and permeability of partial molten rocks is important to understand the thermal history of the Earth and volcanisms. For understanding those obtained by experiments and estimating the physical properties at extreme conditions those are difficult to reproduce in laboratory experiments, the knowledge about structure and properties of silicate crystal-liquid interfaces is necessary. The properties of melt as sandwiched thin films are considered as being different with ones in bulk melts by the effect of crystal surface. For instance, lateral self-diffusivity of water to crystal surfaces shows different from bulk one in the case of water-brucite surface (Sakuma et al. 2003), water-muscovite mica surface (Sakuma and Kawamura, 2009). The dynamic property anomalies on solid - liquid surfaces affect properties of bulk rock such as permeability (Ichikawa et al. 2001).

In this study, structure and properties of the forsterite-MgSiO₃ liquid interfaces are investigated by using molecular dynamics simulations. It is essential to know the structure and physical properties of forsterite-MgSiO₃ liquid interfaces since forsterite is the liquidus mineral of primordial magmas.

Molecular dynamics simulations were performed with NPT ensemble using MXDORTO code (Sakuma and Kawamura, 2009). The initial structure is a 21440 atom system in which a sheet of MgSiO₃ liquid consist of 8000 atoms (~5 nm) is sandwiched between (010) surfaces of forsterite(Pbnm) and 43440 atom system in which a sheet of MgSiO₃ liquid consist of 30000 atoms (~20 nm) is sandwiched between (010) surfaces of forsterite. Firstly we calculated equilibrated MgSiO₃ liquid film in vacuum starting with a randomly generated structure and randomly generated velocities of atoms through 0.5 ns (1,000,000 steps) at 1973 K and quench to 300 K. Secondly we calculated a bulk forsterite crystal with 13440 atoms (11*5*8 unit cells of forsterite(Pbnm)) starting with a given experimental crystal structure which was obtained by the experiment [5] and with randomly generated velocities of atoms and then cut along (010) surface. Finally we combined forsterite cut along (010) surface and MgSiO₃ liquid film. Under maintaining isobaric and isothermal conditions, we performed the relaxation of 0.5~1.5 ns. Then the statistical averages of the structure and physical properties were obtained from the velocities and coordinates of each atom in the simulations through 500 ps. The function of inter-atomic potential model was same as used in our previous work (Noritake et al. 2012).

By these simulations, characteristic structures in the forsterite-MgSiO₃ liquid interface are observed. The layered structure of alternated crystal surface, Si-rich and Mg-rich layers in the crystal-liquid interface was observed. The layered structure was formed by energy difference between Si-O semi-covalent bonds and Mg-O ionic bonds. Si-O-Si bridging and free oxygen atoms are excessively formed and in the near surface since the energy of Si-O bonding is much lower than that of Mg-O bonding. The difference of layered structure by thickness of MgSiO₃ liquid film might be caused by the difference of the degree of freedom of configuration in liquid film. The two-dimensional diffusivity of oxygen atoms is controlled by two factors. The one is the thickness of liquid film that decreases oxygen diffusivity with decreasing the film thickness because of decrease of degree of freedom of configuration in liquid film. The other is composition of sliced layer where oxygen diffusivity increases with increasing the Mg/Si ratio since Si-O bonding is much stronger than Mg-O ones.

Keywords: Interface, High-Temperature, Silicates, Molecular dynamics simulation