

## 学位論文の要旨

### Abstract of Thesis

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学位論文題目 Title of Thesis (学位論文題目が英語の場合は和訳を付記)

Silicate liquid at the Conditions of the Earth's Interior : From Atomic Scale

原子スケールから見る地球内部環境下におけるケイ酸塩液体の物性

学位論文の要旨 Abstract of Thesis

Knowledge of the properties of silicate liquid is essentially important in Earth science, metallurgy and glass industry. The transport properties and densities of silicate liquids at pressure are used for the study of magmatic process and early Earth process. The silicate liquids appears as slag in metallurgy. The silicate liquids appears as raw material of glass forming process. The physical properties such as viscosities, densities, electrical conductivity and more, are reported by various researchers in various fields. The atomic structure, especially near neighbor structure and network structure of silicate liquids have been investigated using various spectroscopic method such as X-ray diffraction, neutron diffraction, nuclear magnetic resonance and Raman scattering, etc. The silicate liquids composed of corner shared SiO<sub>4</sub> tetrahedra and network modifiers such as alkali or alkali earth elements under atmospheric pressure. It means that the silicate liquids has no three-dimensional periodic structure but short-range-ordered structure. The properties of silicate liquids at pressure must be investigated to understand volcanic process and early Earth process. The physical properties of silicate liquids show peculiar behaviors under pressure. The viscosities of low basicity liquids decrease with increasing pressure. These shows the softening of silicate liquids at pressure.

I show the relationship between structure and properties of silicate liquids at pressure, and silicate liquid/ crystal interface in this paper by using force-field molecular dynamics simulations.

In chapter 4, the relationship between structure and properties of  $\text{Na}_2\text{O} \cdot n\text{SiO}_2$  liquids at pressure is investigated by using empirical molecular dynamics simulations. The simulated pressure range is from 0.1 MPa to 10 GPa with NPT ensembles and the potential model taken from my previous work which well reproduces structures various silicate crystals.

In the compression, Si-O and Na-O distance remains constant, however Si-Si distances shorten and coordination numbers of O atoms around M atom increase. Adding to those, 3 and 4 membered rings in -Si-O- network decrease with increasing pressure. Those suggest that densification of silicate liquids consist of increasing of flexibility of -Si-O- network, bending of Si-O-Si and increase of coordination number of M atoms. Up to 6 GPa, the decrease of Si-O-Si angle and distortion of  $\text{SiO}_4$  tetrahedra in acidic silicate liquids ( $n < 3$ ) are obviously confirmed. Decrease of Si-O-Si angle by densification causes distortion of  $\text{SiO}_4$  tetrahedra and weakening of Si-O bonding. These structural change might cause the softening of silicate liquids under high pressure.

In chapter 5, structure and properties of the forsterite- $\text{MgSiO}_3$  liquid interfaces are investigated by using molecular dynamics simulations. It is essential to know the structure and physical properties of forsterite- $\text{MgSiO}_3$  liquid interfaces since forsterite is the liquidus mineral of primordial magmas.

By these simulations, characteristic structures in the forsterite- $\text{MgSiO}_3$  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  $\text{MgSiO}_3$  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.