Molecular Dynamics Study of Soret Effect in Calcium-Aluminosilicate glass

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There are various transportation phenomena driven by a potential gradient. The Soret effect has been known as the diffusion driven by a temperature gradient (Soret, 1879; Ludwig, 1856). While the inverse one is known as Dufour effect. The Soret effect is sometime used in the explanation of fractionation phenomena in geology (Lesher, 1986; Dominguez et al., 2011). Recently Kishi et al. (2016, conference abstract) reports a distribution anomaly of composition around the trace of the migration of high-temperature metal sphere induced by laser irradiation in calcium aluminosilicate glass(Hidai et al., 2016). The rate of fractionation by temperature gradient is determined by both thermal diffusivity (D_T) and mass diffusivities (D) because the total net flux must be zero in a steady state. Consequently, understanding the Soret effect and determination of Soret coefficient (D_T/D) are difficult because the coefficient is affected by not only atomic mass and inter-atomic interaction that affect activation energy but also geometric/structural factor. Calcium-aluminosilicate system is suitable for the investigation of the Soret effect because three oxides have different characteristics; namely, network former, network modifier, and intermediate oxide.

We applied the molecular dynamics (MD) simulations for this system to investigate the mechanism of fractionation by temperature gradient. The MD simulation is an appropriate method for this study because it gives trajectories of each atoms in the simulation cells and potentials of each atom at any point in simulation time. MD simulations of $Ca_3Al_2Si_6O_{18}$ glass were performed using MXDORTO code (Sakuma & Kawamura, 2009). The simulation conditions are as follows: The inter-atomic potential model was taken from Noritake et al. (2015). System of approximately 30000 particles in rod-shaped (approximately 5 x 5 x 17 nm) simulation cell in periodic boundary condition was firstly annealed for 2 ns at 1873 K from randomly generated structure. Then the liquid was quenched to room temperature at the rate of 10^{12} K/s. Then we started the simulations in temperature gradient. The temperatures in sliced regions (approximately 0.35 nm thickness) perpendicular to the longest axis at the end and the center of simulation cell were maintained 300 and 3500 K using the scaling procedure, respectively. After several tens nano-second simulations, we confirmed the changes in distribution of composition. The concentration of SiO_2 in the high-temperature center part slightly increases as simulation proceeds. In contrast the concentration of CaO in that part slightly decreases. The distribution of concentration of Al_2O_3 does not change apparently. Quantification of coefficient and mechanisms will be discussed in this presentation.

Keywords: Molecular Dynamics, Silicate Glass, Silicate Melt, Soret Effect