

Molecular Dynamics Study of Soret Effect in Silicate Liquids

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There are various transportation phenomena driven a potential gradient. The Soret effect is diffusion driven by a temperature gradient (Soret, 1879; Ludwig, 1856). The inverse effect is known as Dufour effect. The Soret effect is sometime used in explanation of fractionation phenomena in geology (Leshner, 1986; Dominguez et al., 2011). Recently Kishi et al. (2018) reports a distribution anomaly of composition in calcium aluminosilicate glass made by migration of high-temperature metal sphere induced by laser irradiation (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 coefficient (D_T/D , or $\partial \log \rho / \partial T$) is difficult because the coefficient might be affected by not only atomic mass and inter-atomic interaction that affect activation energy but also geometric/structural factor.

I applied the molecular dynamics (MD) simulations for pseudo binary and aluminosilicate liquids to investigate the mechanism of fractionation by temperature gradient. The MD simulation 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 sodium/calcium-silicate/aluminosilicate liquids were performed using MXDORTO code (Sakuma & Kawamura, 2009). The simulation conditions are as follows: The inter-atomic potential model was taken from Feuston & Garofalini (1988). Systems of approximately 30000 particles in rod-shaped (approximately 5 x 5 x 17 nm) simulation cell in periodic boundary condition were firstly annealed for 2 ns at 2000 K from randomly generated structure. Then we started the simulations in temperature gradient. The temperatures in sliced region (approximately 0.35 nm thickness) perpendicular to the longest axis at the end and the center of simulation cell were maintained 2000 and 3500 K using the scaling procedure. 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 and Na_2O in that part slightly decreases. Soret coefficients for each cation has linear relation with the half width of potential energy distribution in binary silicates. The existence of Al_2O_3 content impedes fractionation in sodium-aluminosilicate whereas it enhances in calcium-aluminosilicate. This behavior might be caused by the charge compensation of 4-fold aluminum cation.

Keywords: Silicate Liquids, Molecular Dynamics Simulation, Soret Effect