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Molecular dynamics simulations of sodium tri-silicate melt under high pressure

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To understand volcanic process, genesis of magma, its ascent, and terrestrial magma ocean, the properties of silicate melts under high pressure must be investigated. The properties of silicate melts show peculiar behaviors under high pressures. It is reported that viscosity of acidic silicate melts decreases with increasing pressure (Scarfe et. al 1979). Moreover, it is reported that self diffusion coefficients of O and Si increases with increasing pressure (Rubie et al. 1993). These show that silicate melts soften under high pressure. In this work, the relationship between the structure and properties of Na_2O3SiO_2 melt was investigated by molecular dynamics (MD) simulations. In MD simulations, it is possible to obtain changing configuration of every atom as a function of time. It is also possible it identify the individual atoms or groups of atoms which play a deciding role in diffusion processes.

MD simulations were performed with NPT ensemble (N = 8002) using MXDORTO. Pressure range is 0.1 MPa to 6 GPa (Every 1 GPa). Temperature range is 1473K to 2073K (Every 200K). Time step of equations of motion of atoms is 0.5 fs. The inter-atomic potential model implicitly includes Coulomb potential, short-range repulsion, van der Waals force and covalent terms. Starting from a random coordination and velocities, all the atoms are moved in Newtonian equation maintaining isobaric and isothermal through 2 ns as structural relaxation. After relaxation, statistical averages of physical properties were obtained through 200ps.

In 0 to 2 GPa, the density increases rapidly and Q^4 species increases with increasing pressure. This might means, the polymerization affect to the melt density in that pressure range. Shrinking of Si-O network is composed from increasing large membered rings and decreasing Si-O-Si angles. Small membered rings have small degree of freedom of deformation and large membered rings have large degree of freedom of deformation. Consequently, large membered rings might be easy to deform. In the MD simulations, the number of small membered rings decreases with increasing pressure and the number of large membered rings increases with increasing pressure. Adding to that, Si-O-Si angle decreases with increasing pressure. Consequently, this might be said that degree of freedom of deformation of Si-O network increases under high pressure and Si-O network is collapsed. Densification of Na_2O3SiO_2 melt is occur as a result of that.

In MD simulation results, the self diffusion coefficient of oxygen decreases up to 1 GPa, then increases with increasing pressure. Using Einstein-Stokes relation, viscosity increases up to 1 GPa, then decreases with increasing pressure since self diffusion coefficient of oxygen controls the viscosity of silicate melts. The anomalous pressure dependence might be caused by distortion of Si-O network. One obvious structural distortion is decreasing of Si-O-Si angle. Si-O-Si angle become smaller than stable angle when Si-O-Si angle decreases under high pressure. And then, bridging oxygen becomes instable. Adding to that, distance between SiO_4 tetrahedra shorten. This structural change might activate diffusion of oxygen and SiO_4 tetrahedra. The other obvious structural distortion is decreasing of O-Si-O angle. The average O-Si-O angle decreases from the angle of regular tetrahedra with increasing pressure in MD simulations. Oxygen becomes instable when tetrahedra distorts because of repulsion between oxygen and distortion of sp^3 orbital.

Keywords: Molecular dynamics simulation, silicate melt, high pressure