Artifacts in the molecular simulation of silicate liquids

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Silicate liquids are important materials in geoscience and technology. It is very challenging to understand the relationship between the properties and atomistic structures from physical perspectives. The molecular dynamics (MD) simulation is a suitable method for investigating the silicate liquids because it allows direct access to the atomic trajectories at desired condition which cannot be obtained in real experiments. MD simulations of silicate liquids have been performed by various researchers up to date[1,2].However, the MD simulation has the Achilles' tendon that it contains a lot of artifacts in the calculation. One famous artifact is the finite size effect. The simulations of silicate liquids were usually performed with systems consisting of several thousands to several tens of thousands of particles[1,2,3,4]. Horbach et al. (1996) points out that the simulation of pure silica liquids/glass needs about 8000 particles to avoid finite size effect. However, it is not clear how many particles are required to avoid the finite size effect in simulations of other systems. In addition, it is unclear: (1) how long should we anneal to obtain the equilibrium liquid (2) how small should we set Δt value for the numerical calculations of the equation of motion. To check these artifacts, I performed the MD simulations of sodium silicate (Na_2O□$nSiO_2$, n = 1, 2, 3, 4) liquids at various conditions. The total number of particles for the simulations were approx. 2000, 4000, 8000, 12000, and 20000. The equations of motion were integrated with the Verlet algorithm with a time step of 0.25, 0.5, 1.0, 1.5, and 2.0 fs. From simulation results, I conclude that: (1) 2 ns-annealing is needed to relax the silicic (n = 3, 4) system whereas basic systems need several hundreds of pico-seconds for relaxation at 1873 K. (2) The integration time, Δt does not affect the physical properties at 1873 K. Note that it affects density and diffusivity at higher temperature (2400 K). (3) To avoid the finite size effect, 8000 particles are needed in the tetra-silicate system and the required number of particles decrease with decreasing SiO_2 content.

[1]Horbach et al.. (2001), Chem. Geol., **174**, 87-101.
[2]Bauchy et al., (2013), Chem. Geol., **346,** 47-56.
[3]Noritake et al., (2012), J. Noncryst. Sol., **358**, 3109-3118.
[4]Wang et al., (2014), Nat. Comm., **5**, 3241.