

Current limitations of molecular dynamic simulations as probes of thermo-physical behavior of silicate melts

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ABSTRACT

Molecular dynamic simulations offer promise as an essential tool, complementary to experiments, for expanding the reach of computational thermodynamics in igneous petrology by evaluating excess thermodynamic properties of multicomponent silicate melts. However, we present evidence suggesting that current practices in simulation may not achieve the precision needed to predict complex phase equilibria relevant to modeling the Earth's interior evolution. We highlight the importance of quantification of the chemical short-range order in terms of cation-cation pairs in the melt and its impact on different kinetic aspects of molecular dynamic simulations. We analyze published molecular dynamic simulation studies of silicate melts to identify specific criteria and best practices for achieving and demonstrating equilibrium and producing accurate results. Finally, we propose a list of experimental and numerical investigations that need to be performed in the future to ensure full consistency between these two approaches to reduce the gap in our fundamental understanding of silicate melts between the atomic level and the macroscopic scale.

Keywords: Molecular dynamics, ab initio calculations, classical thermodynamics, silicate melts