



## **Microscopic origins of thermodynamic and transport properties of silicate melts at ambient and high-pressure**

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Full understanding of atomic arrangement in silicate glasses and the corresponding melts at high pressure is essential to the transport and thermodynamic properties of these systems at high pressure and the relevant magmatic progresses in the Earth's interior. Recent advances in solid-state NMR and theoretical analyses have provided a unique opportunity to explore the distribution of cations and anions in silicate glasses and the corresponding melts at high pressure (Lee et al. *Geophy. Res. Letts.* 2003, 30, p1845; Lee et al. *Geochim. Cosmochim. Acta.* 2004, 68, p4023; Du et al. *J. Non-crystal. Solids.* 2004, 337, p196). Here, we present the recent progress and insights made by 2-dimensional solid state NMR of diverse silicate glasses quenched from melts at high pressures and show how these microscopic, atomic-scale distributions in the melts from NMR and simulations affect the thermodynamic (e.g. activity coefficient of silica & configurational enthalpy) and transport properties (e.g. diffusivity and viscosity).

Most glasses and melts show a tendency for cation ordering in connectivity at both ambient and high pressure. The chemical ordering in framework disorder contributes to the total negative deviation of activity of oxides from ideal solution in silicate melts (reduced activity). While no definite evidence of clustering among non-framework cations (e.g.  $\text{Ca}^{2+}$ ,  $\text{Na}^{+}$ ) was found, these cations tend to form dissimilar pairs upon mixing with other types of network modifying cations.

Modeling of melt properties using quasi-chemical approximations clarified the micro-

scopic origins of configurational thermodynamic properties, such as configurational enthalpy, entropy, and heat capacity, and which showed remarkable similarities with experimental data. Though semi-quantitative, the trend in diffusivity of alkali cations in multi-component silicate melts was similarly reproduced from the distribution of cations and anions obtained from solid state NMR. Pressure dependence of transport properties such as diffusivity and viscosity of melts were directly obtained from experimentally measured atomic-scale disorder, and from variation of non-bridging oxygen fraction with pressure from spectroscopic data: the important control on viscosity stems from both the NBO fraction and its pressure dependence and the viscosity increases sharply upon approaching higher chemical order.