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Structure of archetypal oxide melts in Earth's interior: Insights from inelastic x-ray scattering & NMR

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Despite their fundamental geophysical-geochemical implications, full understanding of atomic arrangements and coordination environments in amorphous oxides varying temperature, pressure, and composition (from single to multi-component) has been a fundamental yet unsolved problem in modern physical sciences partly due to lack of suitable experimental probes. Recent advances in element specific experimental probe of local structures, such as high resolution 2-dimensional solid -state NMR and synchrotron inelastic x-ray scattering with advanced x-ray optics unveils new structural insights into the pressure and temperature-induced changes in the bonding nature of the diverse archetypal amorphous oxide. Here, we report recent progress that we have made using these techniques (e.g. Lee SK et al. Phys. Rev. Lett. 2009, 103, 095501; J. Phys. Chem. B. 2009, 1 13, 5162; Proc. Nat. Aca. Sci. 2008, 105, 7925).

The first high-resolution solid-state NMR results for amorphous alumina reveal that four and five-coordinated species are predominant (~ 95 %), while six coordinated species are minor. Such a species distribution is remarkably similar to what has been predicted theoretically for alumina melts and requires that there exists in the amorphous alumina a significant fraction of triply bonded oxygen. Oxygen, boron, and lithium K-edge spectroscopy using IXS, together with B-11 NMR reveal the nature of electronic bonding changes in diverse amorphous oxides at high pressure up to 40 GPa. The O-Kedge spectra for prototypical Mg-silicate glass suggest the formation of the triply coordinate oxygen above 20 GPa. We also account for these differences with a conceptual model that utilizes pressure rigidity (the resistance to structural changes with increased pressurization) defined by the variance of the ratio of energy difference between high and low pressure states to its pressure gradient. We also report the first multi-nuclear 2-dimensional NMR spectra for quaternary, Ca-Mg and Ca-Na aluminosilicate glasses, a model system for natural melts where multiple densification mechanisms can be established. We finally discuss the effect of these pressure-induced structural changes in silicate melts on their configurational thermodynamics properties, such as element partitioning and solubility of noble gases.

Keywords: Silicate Melts, High-Pressure, Inelastic x-ray scattering, Solid-state NMR, Thermodynamic Properties