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Effect of structural transitions on properties of high-pressure silicate melts: ^{27}Al NMR, glass densities, and melt viscosities

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Abstract

The densities and viscosities of silicate melts depend strongly on pressure, in part because of potentially measurable structural rearrangements. In an attempt to further understand these changes and how they affect macroscopic properties, we have used ^{27}Al MAS NMR to determine the coordination of the Al cations in a series of aluminosilicate glasses quenched from melts at pressures of 2 to 8 GPa, have measured the glass densities, and have applied an in-situ falling sphere method to measure melt viscosities at high pressure. Spectra from these four- and five-component glasses show increasing Al coordination with increasing pressure and with increasing average field strength of the modifier cation, as was previously reported for simpler compositions. These data also indicate that when multiple modifier cations are present (e.g., Ca and K), the Al coordination is lower than what would be expected from linear combinations of the appropriate aluminosilicate end-members.

The viscosity of $Ca_3Al_2Si_6O_{18}$ melts, measured using a falling sphere method that combines multianvil techniques with synchrotron X-ray radiography, may reach a minimum at a pressure below 6 GPa. A quasi-thermodynamic approach using equilibrium constants for the reactions that generate high-coordinated Al suggests that this pressure may be related to a maximum in the concentration of five-coordinated Al. These results further support the concept that pressure-induced network structural transitions have direct implications for the macroscopic properties of high-pressure melts.

Keywords: Calcium aluminosilicates, potassium aluminosilicates, aluminosilicate glasses and melts, glass properties, NMR spectroscopy, high-pressure studies, melt properties