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Structure and the extent of disorder in quaternary (Ca-Mg and Ca-Na) aluminosilicate glasses and melts

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ABSTRACT

The structure of multi-component silicate melts and glasses (e.g., Ca-Mg and Ca-Na aluminosilicates) can provide insight into the properties of natural silicate melts and has implications for relevant magmatic processes. In spite of its importance, the atomic and molecular structure of most multi-components (e.g., quaternary) melts and glasses has not been fully described, primarily because of insufficient resolution obtained with conventional spectroscopic and scattering methods; the information obtained by these methods is compromised by severe inhomogeneous peak broadening due to structural complexity. Here we report the first ^{17}O and ^{27}Al 3QMAS NMR spectra for quaternary, Ca-Mg and Ca-Na peralkaline aluminosilicate glasses (i.e., M/Al > 1, M is one monovalent or one-half a divalent cation). These data reveal new details into the molecular structure of multi-component aluminosilicate melts, which include the presence of a substantial fraction of ^{V}Al in the Ca-Mg aluminosilicate glasses and ^{IV}Al-O-^{IV}Al in both glasses at 1 atm. Traditional models of glass structure do not support the presence of such species given these high-silica, peralkaline compositions. These results suggest that Al avoidance is violated in the multi-component peralkaline aluminosilicate glasses, and that the presence of Mg^{2+} in the melts increases the extent of disorder in the melts (compared with Ca^{2+} and Na^{+}). These factors lead to an increase in configurational entropy and the activity coefficients of the oxides, and may provide an explanation for the decrease in viscosity of these complex melts.