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Correlation between Fragility and Configurational Heat Capacity in Calcium

Aluminosilicate Glasses

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Abstract: Enabling accurate prediction of the properties of aluminosilicate glasses and glass-forming

liquids is important for the development of new glass compositions for high-tech applications. In this study,

we use a combined topological and thermodynamic approach to connect the configurational heat capacity

 $(C_{p,conf})$ with the liquid fragility (m) and glass transition temperature (T_g) of calcium aluminosilicate glasses.

To obtain glasses with different structural and dynamical features, we study two glass series; one at the

tectosilicate join with varying SiO₂ content and one with constant CaO content but varying Al₂O₃/SiO₂ ratio.

 $C_{
m p,conf}$ is determined using differential scanning calorimetry (DSC), while m and $T_{
m g}$ are determined through

both DSC and direct viscosity measurements. The $C_{p,conf}$ model is found to generally predict the measured

data well, but deviations between modelled and measured $C_{\text{p.conf}}$ values appear for the strongest glasses in the

tectosilicate series and for the most peraluminous glasses in the constant CaO series. We discuss the origins

of these model-data discrepancies based on the structural evolution in the glasses as determined through

Raman spectroscopy measurements.

Keywords: calcium aluminosilicate glasses, configurational heat capacity, fragility, glass transition,

network structure.