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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 ($C_{p,\text{conf}}$) 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 SiO_2 content and one with constant CaO content but varying $\text{Al}_2\text{O}_3/\text{SiO}_2$ ratio. $C_{p,\text{conf}}$ is determined using differential scanning calorimetry (DSC), while m and T_g are determined through both DSC and direct viscosity measurements. The $C_{p,\text{conf}}$ model is found to generally predict the measured data well, but deviations between modelled and measured $C_{p,\text{conf}}$ 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.

