

## **Viscosity, fragility, and configurational entropy of melts along the join $\text{SiO}_2\text{-NaAlSiO}_4$**

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### **ABSTRACT**

Viscosities of fourteen melts close to the join  $\text{SiO}_2\text{-NaAlO}_2$  were measured in the range  $1\text{--}10^{12}$  Pa·s (700–1650 °C) using a combination of concentric cylinder and micropenetration techniques. These compositions cover five isopleths in silica content from 50 to 82 mol% and vary from mildly peralkaline to mildly peraluminous. Greatly improved constraints on the temperature dependence of viscosity in the system  $\text{SiO}_2\text{-NaAlO}_2$  result because exactly the same compositions were used for both high- and low-temperature measurements, viscosities over an extended range of silica contents were measured at temperatures close the glass transition, and several compositions at constant silica content and variable alkali/Al ratio were measured, allowing interpolation of data to compositions exactly along the join  $\text{SiO}_2\text{-NaAlO}_2$ . At high temperature (1600 °C) viscosity and activation energy are shown to be approximately a linear function of silica content, but large nonlinearities occur at temperatures close to the glass transition range. Defining fragility as the gradient of the viscosity curve at the glass transition temperature ( $T_g$  taken to be the  $10^{12}$  Pa·s isokom) on a reduced temperature scale ( $T_g/T$ ), it is found that the fragility increases in a nonlinear fashion as  $\text{NaAlO}_2$  is substituted for  $\text{SiO}_2$ , with fragility increasing more rapidly at lower  $\text{SiO}_2$  contents. The viscosity data are combined with heat capacity data available in the literature to estimate configurational entropies of albite, jadeite, and nepheline glasses using the Adam-Gibbs theory. Fragility, when defined in terms of the Adam-Gibbs parameters, is shown to increase with configurational heat capacity (difference in heat capacity between the liquid and the glassy states) but to decrease with increasing configurational entropy at the glass transition. In the light of independent phase equilibria and spectroscopic and calorimetric evidence that suggests the Al-Si ordering increases as silica content decreases from  $\text{SiO}_2$  to nepheline, the modeling of configurational entropy in terms of Al-Si mixing suggests the following: (1) Melt configurational entropy has contributions from both cation mixing (chemical contribution), as well as variations in the topology of the O network (topological contribution), of which the latter dominates. (2) The chemical contribution is due to mixing of tetrahedral rather than O sites. (3) At the glass transition ( $10^{12}$  Pa·s isokom) the topological contribution shows little, if any, variation.