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Energy barriers to viscous flow and the prediction of glass transition temperatures of molten silicates

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ABSTRACT

Within the framework of the Adam-Gibbs (configurational entropy) theory of viscosity, it is shown that for a given composition, the ratio of parameters B_e (a temperature independent constant) to $S_c(T_g)$ (the configurational entropy at the glass transition) is proportional to the height of the average potential energy barrier to viscous flow ($\Delta\mu$) and the size of the rearranging domains at the glass transition [$z^*(T_g)$]. The ratio $B_e/S_c(T_g)$ is evaluated for several silicate and aluminosilicate compositions of variable polymerization. It is found that the ratio $B_e/S_c(T_g)$ shows simple compositional variations that correspond closely to those that may be expected qualitatively for the height of the potential energy barrier to viscous flow. Assuming that $z^*(T_g)$ is a constant for all compositions, the available data for $B_e/S_c(T_g)$ are parameterized as a function of $\Delta\mu$. The physical basis of this parameterization will therefore allow extension to more complex systems as additional data become available. The A_e term in the Adam-Gibbs equation (another temperature independent constant) shows only minor compositional variation (-2.6 ± 1), but the variation that does exist is found to be a linear function of B_e/tetrahedron. The proposed parameterizations of $B_e/S_c(T_g)$ and A_e are shown to be sufficient for estimating the glass transition temperature to within 15–20 K. Calculated glass transition temperatures may be combined with existing models for viscosities in the range 10–10^5 Pa·s. Interpolation provides the whole viscosity curve and thus also an estimate of the departure from Arrhenian behavior. Although further work is necessary to verify and extend the parameterizations to compositions of direct geological relevance, this work represents a step toward a fully generalizable predictive model of silicate melt viscosity based within a physical framework.