Viscoelastic relaxation measurements in the system SiO₂-NaAlSiO₄ by photon correlation spectroscopy

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Abstract

Relaxation times of longitudinal strain for five supercooled liquids along the join SiO₂ (qz)-NaAlSiO₄ (ne) including the compositions of albite (ab), $ab_{s_0}id_{s_0}$, jadeite (jd), $jd_{s_0}ne_{s_0}$, and $jd_{33}ne_{66}$ were measured by photon correlation spectroscopy within the temperature and relaxation time ranges 711–1116 °C and $35-2 \times 10^{-4}$ s, respectively. The measured time correlation functions are fitted to the Kohlrausch-Williams-Watts equation, $\Phi^{\mu}(t) = \exp(-t/\tau_{\mu\tau})^{\beta}$, yielding isothermal relaxation functions for longitudinal strain at constant stress. The temperature dependence of the longitudinal strain relaxation times obeys an Arrhenian type equation. The relaxation of the longitudinal strain cannot be described by a single relaxation time but by a relaxation time distribution. The width of the relaxation time distribution, which is represented by the parameter β , is independent over a large temperature range, indicating thermorheological simplicity. The decrease of the parameter β corresponds to an increase of the excess (configurational) heat capacity with increasing (Na+Al) content, together indicating an increasing fragility of the supercooled liquids toward the composition of NaAlSiO₄. Increasing fragility is also shown from the activation energy obtained from longitudinal strain relaxation and shear viscosity measurements, which passes a minimum near 40 mol% NaAlSiO₄ at low temperatures but decreases toward NaAlSiO₄ composition at higher temperatures. A comparison between strain, stress, and enthalpy relaxation times shows clearly that the relaxation process resulting from a thermal pertubation is slower than from a mechanical perturbation.