

Pressure dependence of melt viscosity in the system NaAlSi₃O₈-CaMgSi₂O₆

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ABSTRACT

The effect of pressure on melt viscosity was investigated in the system NaAlSi₃O₈-CaMgSi₂O₆ (Ab-Di) at pressures from 0.1 to 400 MPa using a parallel plate viscometer. The new measurements in the high viscosity range of 10^{8.5}–10¹¹ Pa·s are consistent with previous data obtained at higher temperature (lower viscosity) in that the pressure dependence changes from positive for the polymerized Ab melt to negative for the depolymerized Di melt. However, a pressure independent viscosity is observed at much higher Ab contents at low temperature (70–90 mol%) than at high temperature (<30 mol%, Brearley et al. 1986). Hence, the sign of the pressure dependence of viscosity changes from positive to negative with increasing temperature for intermediate compositions along the join. The apparent activation volume (V_a), which is derived from the pressure derivative of log viscosity using an Arrhenian relationship, decreases systematically with temperature for Di-rich compositions, e.g., for the Di melt from 52.0 ± 2.7 cm³/mol at 1020 K to 24.8 ± 6.6 cm³/mol at 1057 K. To model the P - T dependence of viscosity, we propose a modification of the commonly used equation based on Adam-Gibbs theory as

$$\log \eta = A_{AG} + \frac{(B_{AG} + C_{AG} \cdot P)}{T \cdot S^{\text{conf}}}$$

where A_{AG} , B_{AG} , and C_{AG} are fit parameters and S^{conf} is the configurational entropy. The parameter C_{AG} can be interpreted as the volume change of a structural unit during transition from one stable configuration to another one. The new approach excellently reproduces the experimental viscosity data for Di melts over a wide range of temperature. A modified Vogel-Fulcher-Tamman equation also describes the data with a similar reproducibility, but one additional fit parameter is needed. Experimental viscosity data for Ab melts scatter too much at given P - T to be fitted by a unique model. To calculate viscosities of stoichiometric Ab melt at elevated pressure and temperature, we suggest that the C_{AG} parameter determined in our study be combined with published viscosity equations for ambient pressure.