INTRODUCTION

Data on the viscosity of silicate melts are required to understand and to model magmatic processes in the Earth’s crust and in the upper mantle. The system NaAlSi_3O_8-CaMgSi_2O_6 (Ab-Di), as a simple analogue of natural melts from rhyolitic to lherzolite composition, attracts a special interest in viscosity studies. The viscosities of Ab and Di compositions are well known at ambient pressure over a wide range of temperatures (e.g., Urbain et al. 1982; Scarfe et al. 1983; Hummel and Arndt 1985; Tauber and Arndt 1986; Tauber 1987; Neuville and Richet 1991; Taniguchi 1992; Stein and Spera 1993). In an Arrhenius plot, the Ab melt shows a linear relationship between the logarithm of viscosity and the reciprocal temperature, which is typical for “strong” melts (Angell 1985). In contrast, the Di melt is a typical “fragile” melt displaying a strong curvature in the Arrhenius plot. The transition in viscosity behavior from Ab to Di melts was studied by Scarfe and Cronin (1986) at temperatures of 1150–1600 °C using a concentric-cylinder viscometer. At constant temperature, they observed a non-linear variation in the viscosity with the Di content of the melt. Fitting the viscosity data by an Arrhenius relationship, Scarfe and Cronin (1986) found a continuous decrease of the activation energy for viscous flow from 368 kJ/mol for Ab to 159 kJ/mol for Di.

High-temperature, high-pressure viscosities in the system Ab-Di were measured using the falling sphere method (Kushiro 1978; Scarfe et al. 1979; Brearley et al. 1986; Brearley and Montana 1989; Persikov and Bukhtiyarov 1999; Mori et al. 2000; Suzuki et al. 2002). In contrast to the Di melt, which shows a positive pressure dependence of viscosity, the Ab melt exhibits a negative pressure dependence. In situ falling sphere experiments of Suzuki et al. (2002) indicate that the viscosity of Ab melts decrease with increasing pressure at least up to 5.3 GPa. At higher pressures, the rheological behavior of Ab melts is not well constrained due to experimental difficulties and the observed trends are controversial. Based on falling sphere experiments using diamond crystals, Mori et al. (2000) inferred that the decrease in viscosity continues toward higher pressures up to 7 GPa. On the other hand, Poe et al. (1997) observed Si and O diffusivity in Ab melt at 2100 K decreasing with pressure from 5 to 6 GPa, which implies an increase of viscosity in this pressure range. Molecular dynamics (MD) simulations are consistent with a change of transport properties of albite melts at high pressures. Bryce et al. (1999) found

ABSTRACT

The effect of pressure on melt viscosity was investigated in the system NaAlSi_3O_8-CaMgSi_2O_6 (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 –10^11 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 cm^3/mol at 1020 K to 24.8 ± 6.6 cm^3/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

\[
\log \eta = A_{AG} + \left( \frac{B_{AG} + C_{AG}}{T_c} \right) \cdot P \cdot S_{conf}
\]

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.

Pressure dependence of melt viscosity in the system NaAlSi_3O_8-CaMgSi_2O_6

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