High-pressure viscometry of polymerized silicate melts and limitations of the Eyring equation

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ABSTRACT

In situ falling-sphere measurements of viscosity have been performed to determine the viscosity of dacite melt (68 wt% SiO2) from 1.5 to 7.1 GPa at temperatures between 1730 and 1950 K, using the T-25 MA8 multianvil apparatus at the GSECARS 13-ID-D beamline at the Advanced Photon Source, Argonne National Lab. The viscosity of dacite melt decreases between 1.5 and 7.1 GPa. At 1.5 GPa and 1825 K the viscosity is 86.6 ± 17.3 Pa s, whereas at 6.6 GPa and 1900 K it is 2.8 ± 0.6 Pa s. The negative pressure dependence of viscosity results in an activation volume of −12.4 ± 1.4 cm3/mol at 1800 K and −5.1 ± 0.9 cm3/mol at 1900 K. These new data are compared with viscosities estimated from the Eyring equation using oxygen self-diffusion data for the same bulk composition at high pressures. The Eyring equation generally predicts viscosities that are greater than measured viscosities. In addition, the Eyring equation predicts a minimum viscosity at 5 GPa, but no minimum was seen in our falling sphere data set. These discrepancies suggest that the mechanisms for viscous flow and self-diffusion of oxygen in polymerized melts may differ at high pressures, thus limiting the utility of the Eyring equation for high-pressure extrapolations. Further development of the Adam-Gibbs theory may provide an alternative for relating self-diffusion and viscosity at high pressures.

INTRODUCTION

Understanding the transport properties of silicate melts at high pressures is critical for modeling igneous processes in the Earth’s mantle. Two methods commonly have been employed to determine melt viscosity at mantle pressures. Traditionally, a high-density sphere is placed at the top of a sample chamber where it may settle through the sample once the sample melts. The settling velocity of the sphere is determined from the differences between the initial and final positions of the sphere and the elapsed settling time. Stokes’ Law is then solved for viscosity, knowing the sphere radius, settling velocity, and the sphere-melt density contrast, and correcting for wall effects in the sample chamber. Falling-sphere measurements of viscosity in quench experiments (hereafter fall-and-quench experiments) largely have been limited to pressures below 2.5 GPa using the piston cylinder device (e.g., Kushiro 1976, 1978), although the technique has been extended to 7 GPa using a multianvil apparatus (Mori et al. 2000). More commonly, at pressures greater than 2.5 GPa, melt viscosity is estimated from the Eyring relationship, using measured or extrapolated oxygen (O) self-diffusion data and assuming a diffusion jump distance of 2.8 Å (e.g., Rubie et al. 1993). However, the validity of the Eyring relationship must be critically evaluated for silicate melts that exhibit maximum (Poe et al. 1997; Tinker and Lesher 2001) or minimum (Reid et al. 2002) oxygen self-diffusion coefficients at elevated pressures.

Experimental efforts to measure melt viscosities directly at high pressures have been advanced greatly in recent years by the installation of large-volume multianvil devices at synchrotron X-ray facilities (e.g., Kanzaki et al. 1987; Rivers et al. 1998). Real-time X-ray radiography offers the opportunity to continuously monitor the motion of an X-ray-opaque marker sphere during its descent (or ascent) through a melt. This capability eliminates many of the problems of accurately determining time-distance relationships in fall-and-quench experiments. Synchrotron X-ray-based experiments have the additional advantage that experimental pressure can be determined by X-ray diffraction of a pressure standard placed within the pressure cell. This technique has been applied to Fe and Fe-sulfide liquids (e.g., Rutter et al. 2002; Secco et al. 2002), and albite (Suzuki et al. 2002; Kanzaki et al. 1987) and diopside (Reid et al. 2003) silicate melts. Viscosities determined in the latter studies are generally consistent with those from fall-and-quench experiments (Kushiro 1976, 1978; Mori et al. 2000, among others).

Previous studies have focused on the relationship between melt polymerization and the pressure dependence of melt viscosity. Kushiro (1976) found that the viscosity of fully polymerized NaAlSi3O8 (jadeite) melt decreased by nearly an order of magnitude between 0.5 and 2.4 GPa (i.e., the viscosity showed a negative pressure dependence). Kushiro (1978) reported a similar decrease in the viscosity of fully polymerized NaAlSi3O8 (albite) melt between 1 atm and 2 GPa. Mori et al. (2000) extended the study of albite melt using the fall-and-quench method in the multianvil apparatus, and found a negative pressure dependence from 3 to 7 GPa that was similar to that reported by Kushiro (1978). In contrast, Scarfe et al. (1987) measured the viscosity of depolymerized CaMgSi2O6 (diopside) melt, and found that melt viscosity increased by a factor of four between atmospheric pressure and 1.5 GPa at 1913 K (i.e., the viscosity showed a positive pressure dependence). Scarfe et al. provided a nice summary of earlier measurements of melt viscosity and showed that the magnitude of the pressure effect on melt vis-