Effect of carbon dioxide on the viscosity of a melt of jadeite composition at high pressure

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Viscosity is one of the important transport properties controlling the migration of magma in the Earth’s interior. Experimental and geochemical studies have shown that magma is generated in the Earth’s deep interior in the presence of CO₂. However, our knowledge of the effect of CO₂ on the viscosity of magma (silicate melt) is limited. In this work, the viscosity of a molten jadeite composition containing 0.5 wt% CO₂ was measured under high pressure. We observed that in the presence of CO₂, the viscosity was one to two orders of magnitude lower than without CO₂.

Keywords: Viscosity, High pressure, Silicate melt, Carbon dioxide, Synchrotron radiation

INTRODUCTION

Carbon dioxide is an important volatile component in the Earth’s interior. Magmas enriched in CO₂, such as kimberlite, are generated in the Earth’s deep interior (e.g., Keshav et al., 2005). The melting temperature of mantle peridotite is reduced by the presence of CO₂ (e.g., Ghosh et al., 2014). The electrical conductivity of the oceanic asthenosphere can be explained by the presence of carbonatitic melt (Gaillard et al., 2008). Because of the important role that CO₂ plays in the Earth’s interior, the physics and chemistry of CO₂-bearing melts are of great interest (e.g., Dasgupta, 2013). Carbon dioxide decreases the density of silicate melt. Based on density measurements at high pressure, the partial molar volume of CO₂ in silicate melt was determined (e.g., Ghosh et al., 2007; Sakamaki et al., 2011). Sakamaki et al. (2011) showed that CO₂ in silicate melt is less compressible than H₂O, and suggested that CO₂ is more effective than H₂O in reducing the density of the melt at high pressure.

Viscosity is one of the important transport properties controlling the migration of magma in the Earth’s interior. The effect of temperature on the viscosity of silicate melts has been extensively investigated at high pressure (Wang et al., 2014). Although it is well known that H₂O strongly reduces the viscosity of silicate melt (e.g., Audétat and Keppler, 2004), our knowledge of the effect of CO₂ on the viscosity of silicate melt is limited. Pure carbonate melts display very low viscosities. Dobson et al. (1996) carried out in situ falling sphere viscometry on carbonate melts at high pressure, and reported viscosities of 0.006 to 0.036 Pa s for molten K₂Mg(CO₃)₂. A recent experimental study by Kono et al. (2014) showed that the viscosities of calcite and dolomite melts up to 6.2 GPa were in the range of 0.006–0.010 Pa s. Brearley and Montana (1989) and White and Montana (1990) reported that CO₂ reduced the viscosity of molten NaAlSi₃O₈ and KAlSi₃O₈, respectively. However, no effect of CO₂ on the viscosity was detected for molten sodium melilite (NaCaAlSi₂O₇) (Brearley and Montana, 1989). Previously, we reported the viscosity of liquid jadeite (NaAlSi₃O₈) (Suzuki et al., 2011). Here, we discuss the effect of CO₂ on the viscosity of liquid jadeite.

EXPERIMENTAL METHODS

A powder of the sample was prepared from the reagents of SiO₂, Al₂O₃, and Na₂CO₃. First, crystalline Na₂Si₂O₆ was synthesized from SiO₂ and Na₂CO₃ (Suzuki et al., 2002). Powders of SiO₂, Al₂O₃, Na₂Si₂O₆, and Na₂CO₃ were mixed in the desired composition of NaAlSi₃O₈ + 0.5 wt% CO₂. The viscosity was measured by the falling sphere method using X-ray radiography. The experimental procedure of in situ X-ray radiography has been described elsewhere (Suzuki et al., 2002, 2011). Experi-
ments were carried out using the MAX–III apparatus installed at the NE7A station of the PF–AR synchrotron radiation facility at KEK (High Energy Accelerator Research Organization), Tsukuba, Japan. The falling of the Pt sphere was observed using an X-ray camera. The density of Pt was calculated using the equation of state of Holmes et al. (1989). The partial molar volume of the NaAlSi2O6 melt at high pressure was calculated using the data from Lange and Carmichael (1987) and Kress and Carmichael (1991). To obtain the densities of the melt with the composition NaAlSi2O6 + 0.5 wt% CO2 at experimental conditions, the partial molar volume of CO2 at high pressure determined by Sakamaki et al. (2011) was used.

RESULTS AND DISCUSSION

The experimental conditions and results are listed in Table 1. The viscosities of the molten NaAlSi2O6(Jd) + 0.5 wt% CO2 composition are plotted in Figure 1. A solid line serves as a guide for the viscosity at 1350 °C. We observed that the viscosity of the CO2–bearing melt decreases with increasing pressure. The data for the molten pure jadeite composition at 1350 °C are also shown (Kushiro, 1976; Suzuki et al., 2011). The result of this study indicates that the viscosity of molten CO2–bearing NaAlSi2O6 was one to two orders of magnitude lower than for the molten pure jadeite composition. The viscosity as a function of reciprocal temperature is shown in Figure 2. The temperature dependence of the viscosity of NaAlSi2O6(Jd) + 0.5 wt% CO2 melt was similar to that of KAlSi3O8(Sa) + 0.5 wt% CO2 melt (White and Montana, 1990), which may imply a similar effect on the viscosity by dissolving CO2. In these melts, the slopes become steeper for composition containing CO2. These changes suggest a structural modification by CO2 in these melts.

The viscosity of molten silicate is strongly related to the structure (e.g., Wang et al., 2014). The viscosity of polymerized silicate melts decreases with increasing pressure, whereas that of depolymerized melts increases. Suzuki et al. (2011) showed that the viscosity of the jade-

| Run | Pressure (GPa) | Temperature (°C) | Density (10³ kg/m³) | Viscosity (Pa s) |
|-----|----------------|------------------|---------------------|-----------------|
| P220 | 2.08(2) | 1280 | 2.83 | 15.1(8) |
| P216 | 2.11(2) | 1350 | 2.79 | 10.0(3) |
| P217 | 2.36(2) | 1420 | 2.83 | 0.123(2) |
| P219 | 3.48(2) | 1350 | 3.15 | 1.11(2) |
| P218 | 3.52(2) | 1350 | 3.16 | 4.07(5) |

Table 1. Experimental conditions and results

Figure 1. Pressure dependence of the viscosity of melt of composition NaAlSi2O6(Jd) + 0.5 wt% CO2 (solid symbols). The open circles and open squares are the viscosities of the NaAlSi2O6(Jd) composition melt at 1350 °C reported by Suzuki et al. (2011) and Kushiro (1976), respectively.

Figure 2. Viscosities of a melt of composition NaAlSi2O6(Jd) + 0.5 wt% CO2 as a function of reciprocal temperature using data at 2.08–2.36 GPa (solid circles). The data for NaAlSi2O6(Jd) in the pressure range of 2.10–2.36 GPa are from Suzuki et al. (2011). Viscosities of melts of composition KAlSi3O8(Sa) and KAlSi3O8(Sa) + 0.5 wt% CO2 at 2.0 GPa are from White and Montana (1990).
ite composition melt decreases up to 2 GPa, whereas the negative pressure dependence disappears above 2 GPa. An X-ray diffraction study of a molten jadeite composition carried out up to 4.9 GPa showed a decrease in the T–T length (T = Si4+ and Al3+) and T–O–T angle with increasing pressure (Sakamaki et al., 2012).

Fine and Stolper (1985) investigated the carbon species in CO2-bearing sodium aluminosilicate glasses quenched from melts. They synthesized CO2-bearing glasses in the pressure range of 1.5–3.3 GPa. Infrared (IR) absorption spectra showed that the glass of jadeite composition contained molecular CO2 and CO32−. The carbon species in sodium aluminosilicate glasses were also investigated by Brooker et al. (1999) using IR and nuclear magnetic resonance (NMR) spectra. They obtained the ratio of molecular CO2 and ‘network carbonate’ CO32−.

Brearley and Montana (1989) investigated the effect of CO2 on the viscosity of silicate melts at high pressure and showed that CO2 reduces the viscosity of albite melt. However, no detectable change in viscosity was observed in sodium melilitite melt with the addition of 2 wt% CO2. It is known that melts of albite (NaAlSi3O8) and sodium melilitite (CaNaAlSi2O7) compositions have fully polymerized and relatively depolymerized structures, respectively. The study by Brearley and Montana (1989) suggests that the effect of CO2 on the viscosity of silicate melt depends on the composition. In the present study, I observed that the viscosity of the jadeite (NaAlSi2O5) composition melt, which was fully polymerized, decreased with addition of CO2. White and Montana (1990) measured the viscosity of CO2-containing KAlSi3O8 (Sa) melt at high pressure. The KAlSi3O8 (Sa) composition melt also has a fully polymerized structure. The result of this study is consistent with albite and sanidine composition melts by Brearley and Montana (1989) and White and Montana (1990), respectively. It is expected that CO2 in jadeite composition melt modifies the TO4 (T = Si4+ and Al3+) network to decrease the viscosity. It was shown that the solubility of CO2 in silicate melts increases with increasing the NBO/T (nonbridging oxygen to tetrahedrally coordinated cation) ratio (e.g., Brooker et al., 2001). The solution mechanism of CO2 in silicate melts was investigated on quenched glasses formed under high pressure (e.g., Mysen and Virgo, 1980; Brooker et al., 1999; Morizet et al., 2017). Mysen and Virgo (1980) studied the carbon species in glasses with CaMgSi2O6 and NaCaAlSi2O7 compositions by Ramanspectroscopy and suggested the formation of CO32− in those melts. Brooker et al. (1999) carried out a Fourier transformation IR (FTIR), Raman, and 13C MAS NMR study on the C-bearing sodium aluminosilicate glasses quenched under pressure. They showed that both molecular CO2 and carbonate dissolved in jadeite composition glasses. The effect of dissolved CO2 on the viscosity of melt was estimated by Morizet et al. (2007, 2017). Morizet et al. (2007) performed calorimetric measurements of the glass transition temperature (Tg) on CO2-bearing jadeite glass quenched under high pressure and suggested the absence of the effect of CO2 on viscosity. More recently, Morizet et al. (2017) synthesized CO2-bearing silicate glass with melilititic composition at high pressure. They performed NMR spectroscopy and revealed an increase in the degree of polymerization with increasing CO2 content. The positive effect on polymerization and the negative effect on viscosity with increasing CO2 content were explained by the formation of a carbonate sub-network in the melilititic melt. These studies imply a strong correlation between the viscosity and the carbon species in silicate melt. The present study shows that the viscosity of jadeite melts decreases with increasing CO2 content. The present results are inconsistent with the estimation by Morizet et al. (2007) based on Tg. Further investigations are needed to clarify the effect of CO2 on the physical and chemical properties of silicate melts. To clarify the structural modification by CO2, X-ray and/or neutron diffraction studies on jadeite composition melt are necessary.

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