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Viscosity of albite melt at high pressure and high temperature

Abstract The viscosity of albite ($NaAlSi_3O_8$) melt was measured at high pressure by the in situ falling-sphere method using a high-resolution X-ray CCD camera and a large-volume multianvil apparatus installed at SPring-8. This system enabled us to conduct in situ viscosity measurements more accurately than that using the conventional technique at pressures of up to several gigapascals and viscosity in the order of 10^0 Pa s. The viscosity of albite melt is 5.8 Pa s at 2.6 GPa and 2.2 Pa s at 5.3 GPa and 1973 K. Experiments at 1873 and 1973 K show that the decrease in viscosity continues to 5.3 GPa. The activation energy for viscosity is estimated to be 316(8) kJ mol^{-1} at 3.3 GPa. Molecular dynamics simulations suggest that a gradual decrease in viscosity of albite melt at high pressure may be explained by structural changes such as an increase in the coordination number of aluminum in the melt.

Keywords Viscosity · High pressure · Magma · Silicate melt · Molecular simulation

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Introduction

Chemical differentiation in the Earth's interior results from partial melting and magmatic activity. Such differentiation, including formation of the Earth's metallic core, occurred largely during the early history of the Earth, when the upper part of mantle was likely to have been extensively molten (magma ocean, e.g., Abe and Matsui 1986; Sasaki and Nakazawa 1986). It is possible that the fractionation of mantle minerals occurred as a result of differences in density between minerals and silicate liquid in this magma ocean (e.g., Ohtani 1985; Agee and Walker 1993; Suzuki et al. 1995, 1998). Physical properties of silicate melts have, however, not been adequately determined at high pressure. In order to model differentiation in the Earth's interior more realistically, the physical properties of silicate melts must be measured at high pressure. In particular, the viscosity of melts is a key parameter, which controls heat and mass transport.

In order to understand the mobility of magmas in the mantle, the viscosity of silicate melts has been measured using the falling-sphere method in the piston-cylinder apparatus (e.g., Kushiro 1976; Kushiro et al. 1976, 1978; Brearley et al. 1986; Scarfe et al. 1987; Brearley and Montana 1989). In this method, the falling velocity was determined from the position of the sphere in several samples recovered from experiments heated for desired durations and quenched by cutting off the electric power supply. Therefore, a relatively large-volume container sufficient for the sphere to move, and several experiments of different duration were required in this quenching method. Moreover, the maximum measurable pressure was limited to about 2 GPa because of limitations of the piston-cylinder apparatus.

In situ measurements were previously performed using MAX 80, an MA-6 type high-pressure apparatus, in the Photon Factory, a synchrotron radiation facility in Tsukuba, Japan (Kanzaki et al. 1987; Dobson et al. 1996). Using the difference in X-ray

absorption coefficient between metal and silicate melt, a falling metallic sphere was observed in an X-ray shadowgraph. This method can be applied to comparatively lower-viscosity liquids and at higher pressures than conventional quenching experiments in the piston-cylinder apparatus. However, the melting temperature and measured viscosity values of albite melt were considerably lower than those expected from extrapolation of Kushiro's data (Kanzaki et al. 1987). For example, radiography of albite melt at 2 GPa, 1100–1200 °C yielded a viscosity value of 77 Pa s, whereas Kushiro (1978) determined the viscosity to be 1800 Pa s at 2 GPa and 1400 °C in the piston-cylinder apparatus. In the experiments in the Photon Factory, a mixture of amorphous boron and epoxy resin was used as the pressure medium to reduce the attenuation of transmitted X-rays. The sample was loaded in a graphite heater. Therefore, the melting temperature and the viscosity would be decreased by volatile components from pressure media. Moreover, since the pressure was also not determined in situ in these experiments (Kanzaki et al. 1987; Dobson et al. 1996), there must be considerable uncertainty in the precision of the pressure. Therefore, some technical improvements were required for in situ viscometry at high pressure to determine the viscosity of the silicate melts reliably. Recent developments in viscometry have been reviewed by Dingwell (1998).

The pressure dependence of viscosity of silicate and aluminosilicate melts depends on the degree of polymerization (Scarfe et al. 1987), which is defined as the ratio of nonbridging oxygens (NBO) to tetrahedrally coordinated cations (Si^{4+} and Al^{3+}) at ambient pressure (Mysen et al. 1980, 1983). Albite melt is fully polymerized, and the structural and physical properties of its glass have been investigated by various methods (Wolf and McMillan 1995). The diffusivity of oxygen and silicon in albite melt were measured to 6 GPa at 2100 K (Poe et al. 1997). Diffusivity was observed to increase to 5 GPa, implying that the viscosity decreases with pressure. Liquids in the $NaAlO_2$–SiO_2 system at high pressure and high temperature have been studied by molecular dynamics simulation (Angell et al. 1982; Dempsey and Kawamura 1984; Stein and Spera 1995, 1996; Bryce et al. 1997, 1999). The simulations of Bryce et al. (1999) showed an increase in coordination number of silicon and aluminum in albite melt with increasing pressure.

In the present paper, we conducted in situ measurements of the viscosity of albite melt using the synchrotron X-ray and a large-volume high-pressure apparatus at temperatures of 1873 and 1973 K in the pressure range of 2.62 to 5.32 GPa. The X-ray radiography falling-sphere method, rather than the quenching method, is especially effective for low-viscosity melt. The capability of the system in SPring-8 and a few of the preliminary results are shown in Funakoshi et al. (2000). In order to extend the pressure range of experiments, a multianvil apparatus was used.

However, the size of the sample container is smaller than that in the conventional falling-sphere method using a piston-cylinder apparatus. Since the size of a sphere also decreases, the resolution of the radiograph is critical in viscometry. Using this system, a sphere with about 100 μm in diameter can be used. Moreover, in situ observation enables us to measure the low-viscosity liquids of the order of 10^{-2} Pa s (Dobson et al. 2000; Urakawa et al. 2001).

Experimental procedures

Pressure was generated using a double-stage multianvil system. An inner Kawai (MA-8)-type apparatus (Kawai and Endo 1970) was compressed by an outer DIA (MA-6)-type cubic guide block driven by a 1500-ton uniaxial press (SPEED-1500) at SPring-8 (Utsumi et al. 1998; Funakoshi et al. 2000). Tungsten carbide cubes with 12 mm TEL were used as second-stage anvils. The cross-section of the furnace assembly is shown in Fig. 1. The whole ceramic assembly was dried at 800 °C for 12 h. The loaded sample container, graphite heater, and pressure marker were stored in an oven at 150 °C. Neither resin nor adhesive was used in the pressure cell assembly. Albite glass was prepared by fusing a stoichiometric mixture of SiO_2, Al_2O_3, and $Na_2Si_2O_5$ in a platinum crucible at 1473 K. Sodium disilicate had been made previously from reagent grade SiO_2 and Na_2CO_3 by a method similar to that of Scheirer and Bowen (1956). A platinum sphere, the diameter of which was measured from an SEM image, was loaded in a molybdenum container with albite glass.

The sample container was placed in a powdered mixture of MgO and h-BN (vol 1:1), which was compacted into a cylinder. Pressure was determined using the equation of state of MgO (Jamieson et al. 1982). X-ray diffraction spectra were collected with a Ge solid-state detector for 300 to 600 s at the diffraction angle $2\theta = 5.5°$. Temperature was measured using a W97%Re3%–W75%Re25% thermocouple, and no correction was applied for the effect of pressure on the emf. In each experiment, the sample was compressed at room temperature to the desired load, then heated to about 100 K below the melting temperature of albite (Presnall 1995) and held for half a minute to allow the temperature
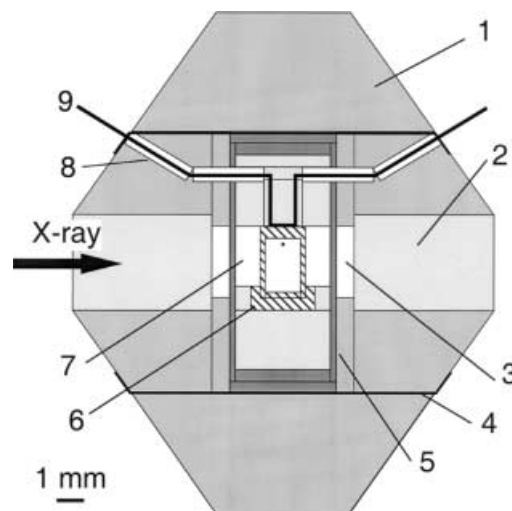

Fig. 1 A cross-section of the pressure cell for the double-stage multianvil system. *1*, zirconia; *2* magnesia; *3* boron nitride; *4* molybdenum electrode; *5* graphite heater; *6* molybdenum sample container; *7* MgO pressure marker; *8* alumina insulator; *9* W97Re3–W75Re25 thermocouple
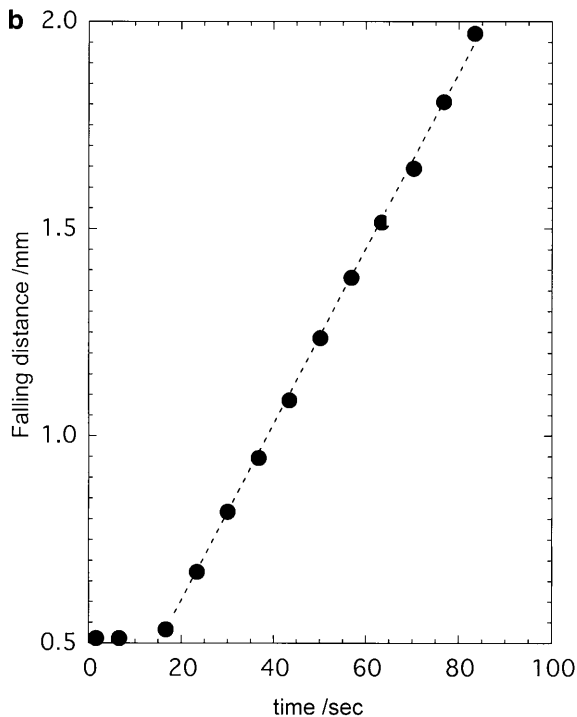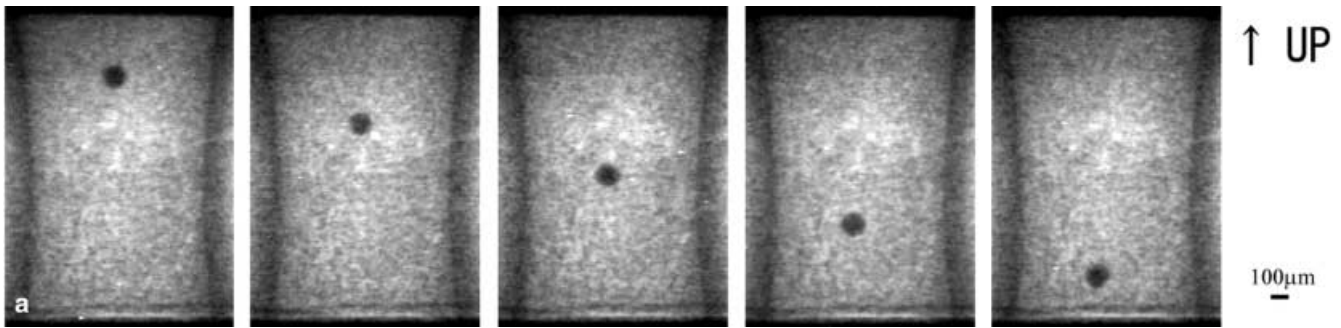
Fig. 2 a An X-ray shadow image of the run S224. A platinum sphere with 140 μm in diameter is falling in albite melt. The time interval between each image is 13.3 s. **b** Time-falling distance diagram of the run S224. The distance is measured *from the top of the picture*. The recording of the image was started at the time of quick elevation of temperature. The sphere reaches the terminal velocity soon after onset of settling

to stabilize. A quick elevation of temperature was then carried out within 10 s from subsolidus to the desired temperature. The shadow image of the charge was observed using a high-resolution CCD (charge coupled device) camera (C4880, Hamamatsu Photonics Co.) and recorded on a videocassette and hard disk. The intense X-ray of the synchrotron radiation enabled us to observe a platinum sphere of about 100 to 140 μm in diameter in albite melt through both the ceramic pressure media and the molybdenum container. The detail of the imaging system has been described by Funakoshi et al. (2000). The terminal velocity of the falling sphere was analyzed from captured images (Fig. 2a). Stokes' equation with the Faxen correction (Faxen 1925) was applied to calculate the viscosity:

$$\eta = \frac{2 \cdot g \cdot r^2 \cdot \Delta\rho}{9 \cdot v} \left\{ 1 - 2.104\left(\frac{r}{r_c}\right) + 2.09\left(\frac{r}{r_c}\right)^3 - 0.95\left(\frac{r}{r_c}\right)^5 \right\} ,$$

where $v =$ terminal velocity, $g =$ acceleration due to gravity, $r =$ sphere radius, $\Delta\rho =$ density difference between the platinum sphere and albite melt, $\eta =$ viscosity, and r_c is the inner radius of container. The density of platinum was calculated from the equation of state of Jamieson et al. (1982). A third-order Birch–Murnaghan equation of state was applied for albite melt. The density was extrapolated from ambient pressure (Lange and Carmichael 1987) using the compressibility data of Kress et al. (1988). The pressure derivative of bulk modulus ($dK/dP = K'$) of albite melt has not previously been reported; however, K' of the anorthite melt

was estimated to be 6.9 (Rigden et al. 1989). We assumed K' to be 4 to 8 for albite melt. Even if such a large uncertainty was considered, the error of the calculated viscosity is less than 0.5%.

Results and discussion

The experimental conditions and results are listed in Table 1. A diagram of time and position of the settling platinum sphere is shown in Fig. 2b. The sphere started settling a few seconds after the temperature was raised to the desired value. The falling velocity does not change to the bottom of the image (Fig. 2a, b). This result suggests that the temperature gradient is negligibly small in the sample container and that there is no thermal convection. The viscosity of albite melt at high pressure and temperature is summarized in Fig. 3. An Arrhenius relationship gives an activation energy of 316(8) kJ mol^{-1} at 3.3 GPa. The pressure dependencies of viscosity at 1873 and 1973 K are similar and negative. A negative activation volume, −5.4(4) cm^3 mol^{-1}, was obtained at 1973 K in the pressure range from 2.62 to 5.32 GPa. The water content in the recovered glass samples was esti-

Table 1 Experimental conditions and results

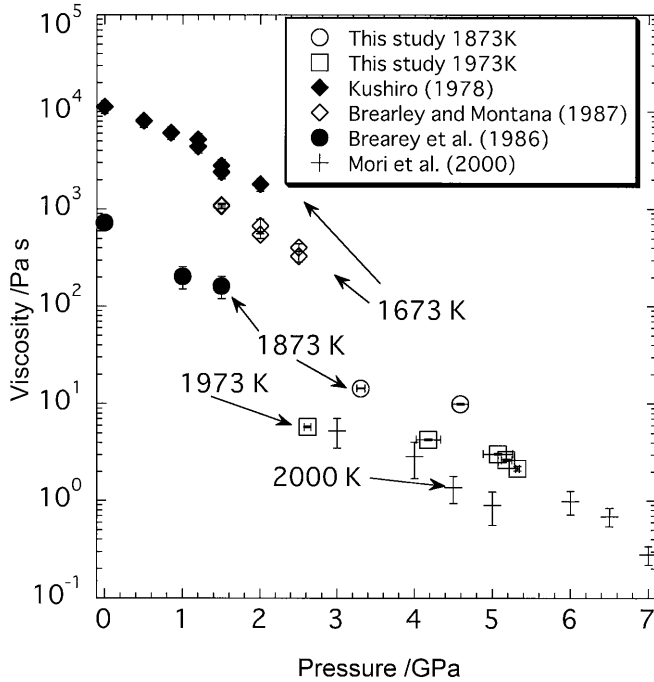| Run	Pressure (GPa)	Temperature (K)	Viscosity (Pa s)
S182	3.30 (5)	1873	14.4 (2)
S171	4.6 (2)	1873	9.9 (2)
S224	2.62 (4)	1973	5.79 (9)
S186	4.2 (2)	1973	4.30 (7)
S232	5.1 (2)	1973	3.06 (5)
S227	5.18 (6)	1973	2.66 (8)
S229	5.32 (2)	1973	2.2 (1)

Fig. 3 Pressure–viscosity diagram of albite melt. The results from previous studies (Kushiro et al. 1978; Brearley et al. 1986; Brearley and Montana 1989; Mori et al. 2000) are also shown

mated from FTIR spectra. Using the integration of the absorption peak corresponding to OH vibration, we estimate that the water content of the glasses was 0.5 to 0.9 wt%. According to a recent study by Mori et al. (2000), the viscosity of albite melt decreases to 5 GPa. Although their method had some ambiguities caused by the nonspherical shape of their diamond falling material, their result is consistent with our in situ measurement using the platinum sphere (Fig. 3). The present experiment shows that the decrease in viscosity with increasing pressure continues at least up to 5.3 GPa.

The decrease in viscosity with increasing pressure was first observed in silicate melts with jadeite ($NaAlSi_2O_6$), basalt, and andesite compositions (Kushiro 1976; Kushiro et al. 1976). Kushiro (1976) estimated the structural change of Al^{3+} species from four to six coordination with increasing pressure based on the prediction by Waff (1975). However, Raman spectra of the quenched glass did not indicate an increase in aluminum coordination to 3.8 GPa (Mysen et al. 1980, 1983). Local structure

surrounding silicon and aluminum has also been investigated by NMR spectroscopy (Ohtani et al. 1985; Stebbins and Sykes 1990; Sykes et al. 1993). There are some discrepancies in their results; however, five- and sixfold aluminum appeared over 6 GPa. The coordination change of Al^{3+} is also suggested by K_β X-ray emission from albite glass quenched at the pressure range of 2 to 6 GPa (Ohtani and Suzuki 1989). Poe et al. (1997) measured the diffusivity of silicon and oxygen in albite melt at 2100 K and estimated the viscosity of the melt from the diffusivity using the Eyring equation. According to this work, the viscosity decreases up to 5 GPa and a further slight increase follows up to 6 GPa. Though the temperatures in our present experiments were different from those of Poe et al. (1997), our results support the validity of the Eyring relation and indicate that the viscosity decreases at least up to 5.3 GPa. Molecular dynamics simulation in the $NaAlO_2$–SiO_2 system (Bryce et al. 1999) shows that the diffusivity of aluminum, silicon, and oxygen increases with increasing pressure to about 20 GPa in albite melt and then gradually decreases at higher pressure. It is also interesting to note that the concentration of higher-coordinated Al^{3+} and Si^{4+} increases with increasing pressure. Higher-coordinated aluminum has been detected in glasses from the albite(Ab)-sodium tetrasilicate (NTS) system. NMR spectra of Ab50/NTS50 glass quenched from a high-pressure melt show an increase in [5]Al and [6]Al species over 6 GPa (Yarger et al. 1995). The amount of [5]Al increases to 8 GPa and then decreases to 12 GPa, although [6]Al increases to 12 GPa. The diffusivity of oxygen increases up to 8 GPa in Ab50/NTS50 melt (Poe et al. 1997). In quenched jadeite ($NaAlSi_2O_6$) glass, which was prepared at high pressure, about 6% of [5]Al and [6]Al were detected from aluminum K-edge XANES spectra (Li et al. 1995). Because the higher-coordinated aluminum exists in some aluminosilicate glasses quenched at high pressure (Ohtani 1985; Ohtani and Suzuki 1989; Li et al. 1995; Yarger et al. 1995), continuous decrease in viscosity of albite melt up to 5.3 GPa (Fig. 3) might indicate a gradual coordination change of Al^{3+} ions.

It is generally recognized that the change in viscosity is strongly related to the change in the melt structure. Based on the investigation of Raman and NMR spectra of glasses quenched at high pressure (Mysen et al. 1980, 1983; Sykes et al. 1993), The T–O–T (T = Si^{4+} and Al^{3+}) angle distorts to several gigapascals. No additional peak corresponding to higher-coordinated cation was observed. In order to clarify the cause of the change in viscosity, investigation of the structure of the melt at high pressure and high temperature is required.

Molecular dynamics (MD) simulations have also been performed to investigate the structure of albite melt under the experimental conditions using the program MXDORTO developed by Kawamura (1996). The interatomic potential used here consists of the Coulombic, the exponential-type repulsion, and the van der Waals-type attraction as given by $U(r) = q_i q_j r_{ij}^{-1} + f(B_i + B_j)$

Table 2 Results of molecular dynamics simulation

Pressure (GPa)	Temperature (K)	[4]Si (%)	[5]Si (%)	[6]Si (%)	[4]Al (%)	[5]Al (%)	[6]Al (%)	Density (g cm^{-3})	O–Si–O (deg)	O–Al–O (deg)	Si–O–Si (deg)	Si–O–Al (deg)	Al–O–Al (deg)	Density[a] (g cm^{-3})
0.0 (3)	1973 (18)	98.7	1.3	0.0	88.3	11.0	0.7	2.43 (1)	107	104	144	137	128	2.28 (1)
1.0 (3)	1973 (18)	98.3	1.7	0.0	83.3	16.0	0.7	2.56 (2)	107	103	142	135	127	2.39 (1)
2.0 (3)	1973 (18)	98.3	1.7	0.0	84.7	13.7	1.6	2.65 (2)	107	102	141	134	126	2.48 (2)
3.0 (3)	1973 (18)	98.0	2.0	0.0	82.3	16.7	1.0	2.74 (1)	107	101	139	133	127	2.56 (3)
4.0 (3)	1973 (19)	94.7	5.3	0.0	75.3	20.0	4.7	2.82 (1)	106	100	139	131	125	2.64 (5)
5.0 (3)	1973 (19)	92.7	7.3	0.0	63.7	32.0	4.3	2.88 (1)	106	99	138	130	124	2.70 (6)
6.0 (3)	1973 (19)	92.3	7.3	0.3	60.7	33.3	6.0	2.93 (1)	106	98	138	130	124	2.76 (7)

[a] Calculated densities from high temperature Birch–Murnaghan equation of state using zero-pressure density by Lange and Carmichael (1987) and compressibility by Kress et al. (1988) (see text)
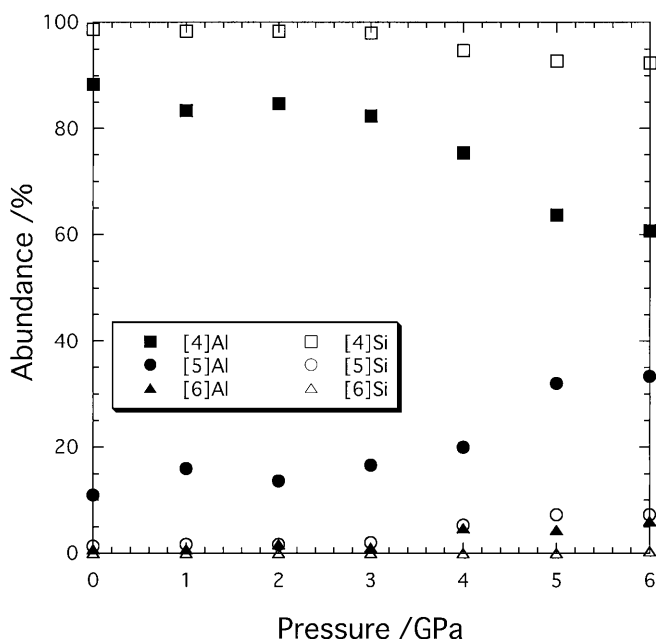

Fig. 4 Pressure dependence of coordination number of Al^{3+} and Si^{4+} ions in albite melt at 1973 K calculated by the MD simulation

$\exp[(A_i + A_j - r_{ij})/(B_i + B_j)] - C_iC_jr_{ij}^{-6}$, where r_{ij} is the interatomic distance, f is a constant, and q_i, A_i, B_i, and C_i are energy parameters. The present study used the parameters which were recently optimized for 29 kinds of crystals in the Na_2O–CaO–MgO–Al_2O_2–SiO_2 system by Matsui (1998). Simulations were carried out by the isothermal and isobaric (NTP) ensemble with a particle number of 1300. The system was initially annealed for 100 ps by 50 000 time steps ($\Delta t = 2.0$ fs) at 0 GPa and 3000 K, and then cooled and compressed to the desired conditions for 100 ps. The data were taken from the following 100 ps.

The results of MD simulations are summarized in Table 2. Although calculated densities from MD simulation are always higher than those from the high-temperature Birch–Murnaghan equation of state, the differences in obtained viscosity values are only 0.9(4)%. Coordination numbers are obtained from the number of oxygen surrounding Si and Al up to the cutoff distance, which is the first minimum of the pair correlation function. At 6 GPa, the average coordination number of aluminum is 4.43, whereas that of silicon is still 4.07. Figure 4 shows the ratio of four- five- and six-coordinated cations. Although 92% of silicon remains in four coordination at 6 GPa, 39% of aluminum changes to five and six coordinations. MD simulation of albite melt at high pressure and high temperature was also carried out by Bryce et al. (1999) using the energy parameters optimized by Dempsey and Kawamura (1984). Conditions of simulation were from 4900 to 6130 K and up to 40 GPa for albite melt. Temperatures were very much higher than that of the present simulation; however, an increase in coordination number of network-forming cations (Si^{4+} and Al^{3+}) was also observed. Along with the change in coordination number, O–T–O angles decrease with increasing pressure. Especially, O–Al–O angle distorts from 104° at 0 GPa to 98° at 6 GPa. T–O–T-bridging angles also decrease with increasing pressure. Comparison of MD simulation with experimentally determined viscosity strongly suggests that the increase in Al^{3+} coordination is consistent with the gradual decrease in the viscosity of albite melt.

Conclusion

The in situ falling-sphere method was employed to determine the viscosity of albite melt at pressures up to 5.3 GPa. The settling velocity of a platinum sphere was measured from the X-ray shadowgraph. The advantage of this system is the accuracy of settling velocity and the available pressure range compared with the conventional falling-sphere quenching method. The in situ technique is especially effective for liquid with low viscosity. The pressure dependence of viscosity is negative. The change in viscosity with pressure suggests that the structural change (coordination increase of network-forming cations, Si^{4+} and Al^{3+}, associated with a decrease in the T–O–T-bridging angle) in albite melt continues to 5.3 GPa.

Molecular dynamics simulation shows a gradual change in albite melt structure with pressure. The average coordination number of Si and Al increases with increasing pressure. Particularly the coordination change of Al polyhedra is dominant, and could be

responsible for the observed decrease in viscosity of albite melt.

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