In situ viscosity measurements of albite melt under high pressure

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Abstract
The viscosities of albite ($NaAlSi_3O_8$) melt under high pressures have been
measured using an x-ray radiography falling sphere method with synchrotron
radiation. This method has enabled us to determine the precise sinking velocity
directly. Recent experiments of albite melt showed the presence of a viscosity
minimum around 5 GPa (Poe *et al* 1997 *Science* **276** 1245, Mori *et al* 2000
Earth Planet. Sci. Lett. **175** 87). We present the results for albite melt up to
5.2 GPa at 1600 and 1700 °C. The viscosity minimum is clearly observed to be
around 4.5 GPa, and it might be explained not by the change of the compression
mechanism in albite melt but by change of the phase itself.

1. Introduction

The viscosity of silicate melts is an important property for understanding the processes in
magma and volcanic eruptions. A variety of silicate melts have been investigated, leading to
the conclusion that the viscosity of highly polymerized silicate melts decreases with increasing
pressure, in sharp contrast to the case for normal liquids [1–4]. Albite, one such highly
polymerized silicate melt, is a major component of magma. Thus investigating the pressure
dependence of its viscosity is very important for understanding the magma processes in the
Earth's deep crust and upper mantle. It has been reported that viscosity of albite melt, as well as
other highly polymerized silicate melts, has negative pressure dependence [5, 6]. In addition,
recent measurements have shown the presence of a viscosity minimum at a pressure around
5 GPa, suggesting a certain change in the compression mechanism [7, 8]. These previous
works have been carried out using a quench falling sphere method, where the terminal sinking
velocity is determined by changing the quench rate. In this method, however, the determination
of terminal velocity involves uncertainties due to of the limitation of the sinking distance and
the quench rate. The use of synchrotron radiation has enabled us to observe the sphere sinking
process directly and to determine the precise terminal sinking velocity by implementing an
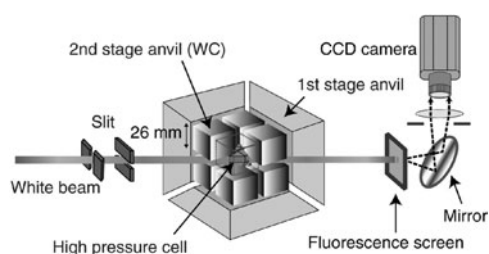
Figure 1. Schematic drawing of the high-pressure x-ray radiography system on the BL04B1 beamline at SPring-8.
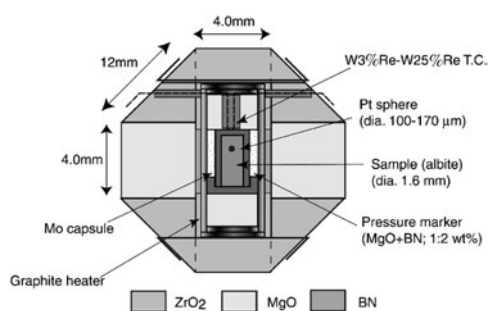

Figure 2. Schematic drawing of the sample cell for the x-ray radiographic experiment.

x-ray radiography technique [9, 10]. Here, we report *in situ* viscosity measurements of albite melt under high pressure using an x-ray radiography falling sphere method.

2. Experimental details

We have set up an *in situ* viscosity measurement system installed on a large-volume multi-anvil apparatus (SPEED-1500) at the BL04B1 beamline at SPring-8 [11]. Pressure is generated by a Kawai (double-stage) system with tungsten carbide cubes with a truncation edge length of 12 mm. The incident white x-ray from the bending magnet irradiates the sample cell through the anvil gap, and an image of the sample is projected on the fluorescence screen. This image is then magnified and detected by a high-speed CCD camera (figure 1). Real-time images of the sample are captured in a personal computer and recorded every 30 ms.

A schematic drawing of the high-pressure cell appears in figure 2. The starting fine powdered albite glass was synthesized and contained in a cylindrical Mo capsule (1.6 mm in inner diameter and 2.0 mm in length), which was located at the centre of the cylindrical graphite heater. A Pt sphere with a diameter of 100–170 μm was embedded in the upper part of the albite sample. A fine powdered mixture of MgO and BN (2:1 wt%) was filled in to surround the sample capsule as the inner pressure marker, and the pressure was calculated from the observed lattice constant of MgO. The temperature was measured using a W3% Re–W25% Re thermocouple with a diameter of 0.15 mm.

The sample was first compressed at room temperature, followed by heating at a constant applied load. To avoid the differentiation effect and partial melting, the compressed sample was first annealed at 1000 °C, and then ramping was performed to reach the target temperatures (1600 and 1700 °C). The heating rate was regulated to be about 200 °C s^{-1}. Once the target temperature was attained, the Pt sphere began to sink in the melt. The observed images from one of the series (4.18 GPa and 1700 °C) are shown in figure 3. Each frame of these images was captured at intervals of 30 ms. The high-speed and high-resolution CCD camera provided very good visual contrast between the Pt sphere and albite melt possible in a short exposure time.
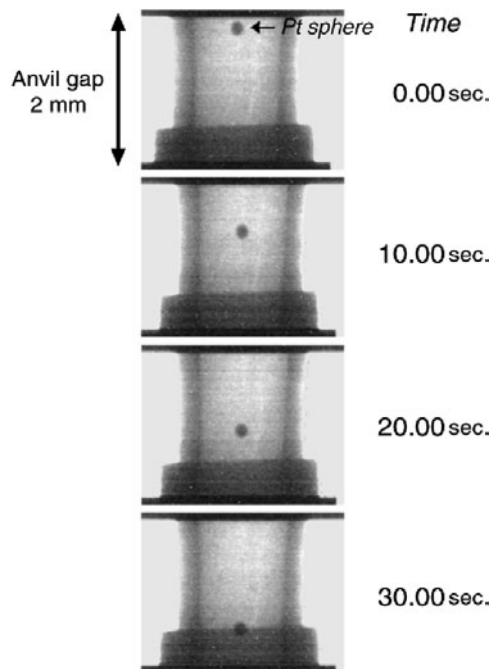
Figure 3. Real-time images of Pt sphere sinking in albite melt at 4.18 GPa and 1700 °C. Each frame of these images was captured at intervals of 30 ms.
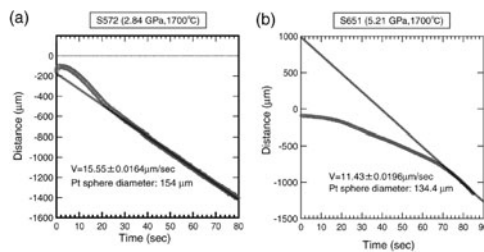

Figure 4. Examples of the settled distance of a Pt sphere in albite melt as a function of time ((a) 2.84 GPa and (b) 5.21 GPa at 1700 °C). We used the linear part of the plot and determined the terminal velocity of the Pt sphere by a linear least-squares calculation.
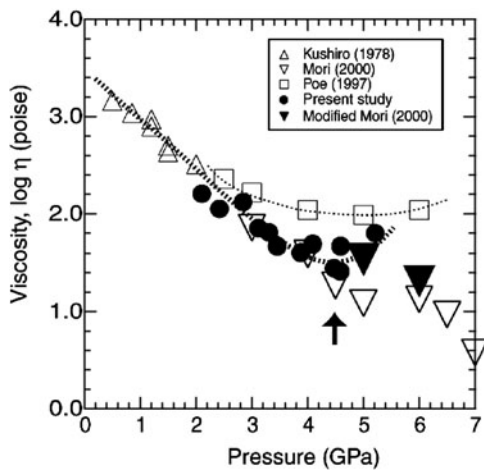

Figure 5. Comparisons of the pressure dependence of albite melt viscosity determined by *in situ* and quench experiments [5, 7, 8]. The solid triangles show the modified viscosity values of Mori *et al* [7], determined by recalculation of the terminal velocity using their raw data. Quench values were extrapolated 1700 °C with activation energy $E_a = 368$ kJ mol^{-1} using an Arrhenius relation [17].

These measurements were carried out at several P–T conditions up to 5.2 GPa at 1600 and 1700 °C. To determine the terminal velocity of the sinking sphere, we analysed the images and

obtained the geometrical centre position of the Pt sphere from each captured frame. Examples of the settled distance are plotted in figure 4 as a function of time. We used the linear part of the plot and determined the terminal velocity of the Pt sphere using a linear least-squares calculation. The viscosity was calculated from this velocity using Stokes's equation, including the Faxen correction for the wall effect:

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

where v is the sinking velocity, η is the viscosity, $\delta\rho$ is the difference in density between the melt and sphere, and r and r_c are the sphere and sample capsule radius, respectively. Here, the density of the melt under high-P–T conditions was estimated from the temperature dependence of the bulk modulus [12] using the density at ambient pressure of Lange and Carmichael [13].

3. Results and discussion

The viscosities are summarized in figure 5, together with the data from previous quench experiments [5, 7, 8]. To compare the pressure dependence at the same temperature, these values were extrapolated to 1700 °C using an Arrhenius relation [14]. In the quench experiments, it is difficult to judge whether the velocity of sphere sinking is constant, and therefore the determination of the terminal velocity may involve large uncertainties. On the other hand, the present x-ray radiography technique enables us to determine the terminal very precisely because the sphere sinking process can be observed directly. The errors of our viscosity values are estimated within 0.01 poise. As shown in figure 5, our values (solid circles) clearly indicate a decrease in the viscosity with increasing pressure, which is consistent with the quench results.

Furthermore, we observed the clear viscosity minimum at 4.5 GPa. Mori *et al* [7] also measured the viscosities of albite melt by the quench method, and the viscosity minimum was observed around 5 GPa (open triangles). Such a viscosity minimum was also observed in the oxygen self-diffusivity measurement by Poe *et al* [8] (open squares), but at slightly higher pressure than that in our results. This difference might be caused by an overestimation of the terminal velocity in their studies. In figure 5, the solid triangles show the values of Mori *et al* determined by recalculation of the terminal velocity using their raw data. We have found that their modified viscosity values at 5 and 6 GPa could increase significantly, and thus the viscosity minimum might shift to lower pressure. Consequently, the variation of their modified viscosity values is in good agreement with the present results.

The existence of such a viscosity minimum should be associated with some structural changes in albite melt. However, it has been reported that no drastic change in the Al coordination number occurred up to 8 GPa [15–19]. Poe *et al* [8] proposed that the compression mechanism of albite melt might change at this pressure. Here, we suggest another possibility, the segregation of melt. As shown in figure 4(b), the sinking velocity at 5.2 GPa is not constant. In our experiments at pressures greater than 5.2 GPa, constant terminal velocities were not obtained. This suggests that the segregation of melt may occur under this condition. Therefore, the viscosity minimum might be explained not by the change of the compression mechanism in albite melt but by a change of the phase itself, such as that resulting from the decomposition of albite into the jadeite phase. Although the previous quench results indicated a continuous viscosity decrease above 5 GPa, it is possible that they measured the viscosity of a partial melting of a different compositional melt from albite. Further *in situ* studies particularly at higher temperatures are necessary to clarify the structural changes in albite melt in this pressure range.

Acknowledgment

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