Viscosity of Fe-S Liquids at High Pressure

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Introduction

The Earth's outer core is composed mainly of iron, with some light alloying elements.[1-3] The viscosity of the Earth's outer core is an essential parameter necessary to understand and model core processes, such as the geodynamo. However, viscosity estimates of outer core candidate liquid compositions are poorly constrained and span 14 orders of magnitude.[4] Consequently, there is a need to understand the pressure and temperature effects on the viscosity of outer core liquids through experimental studies. We present here the first high pressure and temperature measurements on the viscosity of $FeS_{(8.5\ \mathrm{wt.\%})}$.

Methods and Materials

Due to the experimental challenges in measuring viscosity of liquids under high pressure, current techniques rely on clever modifications of the traditional Stokes' viscometry method. This involves measuring the velocity with which a probe sphere moves through a pressurized melt and employing Stokes' law to determine the melt viscosity. Previous experimental measurements suffered from errors due to the difficulty in resolving sphere velocity (quench and probe, electrodetection) or swift reaction between the probe sphere and the sample (x-ray radiography).[5-7] We employ a synchrotron radiographic method to image, *in situ* and in real time, the velocity of the probe sphere,[8] combined with a novel technique of sphere shielding to eliminate all reaction between sphere and melt.

We chose $FeS_{(8.5\ \mathrm{wt.\%})}$ as a possible candidate outer core composition.[3] In order to isolate the siderophile Pt core from the sample, a technique was developed to produce composite spheres made of a Pt core surrounded by a ruby mantle.[9] In addition to preventing a reaction between the Pt and Fe-S sample, this technique allows the density of the composite sphere to be tailored by adjusting the relative radii of the core (~125 µm) and mantle (250 µm). High-pressure and temperature, *in situ*, viscosity experiments were performed in a 250-ton DIA-type high-pressure apparatus at beamline 13-BM-D, Advanced Photon Source (APS), Argonne National Laboratory.[10] Temperature differences in the sample region were measured to be <50°C and pressure was determined using the equation of state for MgO.[11]

Results

Sample viscosity was calculated from terminal sphere velocity using a corrected form of Stokes' equation.[12] Density of the composite sphere at high pressure and temperature was determined from the initial sphere size and applying the high-temperature 3rd-order Birch- Murnaghan equation of state. The density of the sample was determined from the liquid equation of state and thermal expansion of $FeS_{(8.5\ \mathrm{wt.\%})}$.[13-15] Figure 1 displays the real-time, density contrast sequence of images of a composite sphere rising through the sample melt at 4.5 GPa and 1550°C. Dark vertical bars bracketing the lighter sample region are the tungsten carbide (WC) anvils of the press. The Pt core is clearly visible, and the ruby mantle appears as a light halo surrounding the core. Clearly the ruby mantle is an effective barrier between the melt and the platinum core as it remains intact during length of the experiment. The displacement vs. time curve of the ascent of a composite sphere through the melt, at 4.5 GPa and 1550°C, is given in Fig. 2. The expected sigmoidal shape of the curve is indicative of the acceleration (stage I in Fig. 2), terminal velocity (II) and deceleration (III) phases of sphere motion. The terminal velocity of the sphere, which is used to calculate the viscosity of the samples, was determined by a linear fit to the terminal velocity portion of the displacement vs. time curves. The pressure dependance of viscosity from isothermal experiments at 1550°C

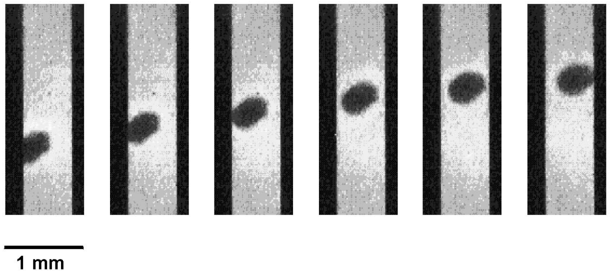

FIG. 1. Real-time density contrast images, at 4.5 GPa and 1550°C, of a composite sphere rising through the $FeS_{(8.5\ \mathrm{wt.\%})}$ melt. Elapsed time between frames is 430 msec. The platinum core is visible as a dark circle that rises as frames advance. The ruby mantle appears as a bright halo surrounding the platinum core. Dark bands bracketing the sample are the WC anvils.

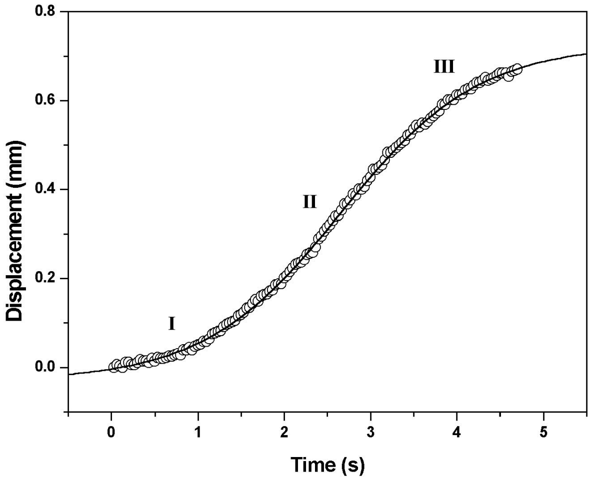

FIG. 2. Displacement vs. time curve for rising composite sphere flight in $FeS_{(8.5\ \mathrm{wt.\%})}$, at 4.5 GPa and 1550°C. Open circles (○) indicate measured displacement from real-time radiographic images and each frame digitized. Solid line is a sigmoidal fit to the data. (I) indicates the acceleration phase of the sphere's flight, (II) indicates the terminal velocity phase, and (III) indicates the deceleration phase.

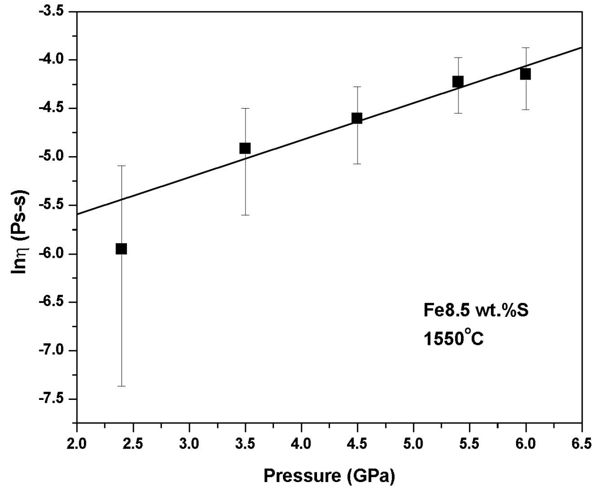

FIG. 3. Arrhenian plot of viscosity as a function of pressure. Solid squares (■) indicate measured viscosity values. Solid line is a linear fit whose slope give activation energy (R=0.95).

is plotted in Fig. 3. Error bars incorporate the error in the calculated density contrast between the composite sphere and the melt, error in the velocity measurement, and error in the size of the composite sphere. The largest source of error results from the uncertainty in the density contrast. This was calculated, based on the error in measurement in Pt core diameter and ruby mantle volume, to be a maximum of 0.2 g/cm^3. Other sources of error have comparatively little effect on the calculated viscosity.

Discussion

Clearly, the effect of pressure on the viscosity of the liquid is substantial and can be quantified through the activation volume of viscosity, defined as:

$$\Delta V_{\eta}^{*} = RT\left(\partial \ln \eta / \partial P\right)_{T} \qquad (1)$$

where R is the universal gas constant, T is temperature in Kelvin, and P is pressure. The error associated with the lowest pressure viscosity value was too large for it to be used to calculate the activation volume and therefore in all fitting procedures it was excluded. Activation volume obtained from a linear fit to the data is 5.81cm^3/mol. These results are significant because they provide a firm base upon which geophysical questions, such as the effect of sulphur on liquid viscosity and the scaling of dynamic fluid properties to the melting boundary, can be addressed.

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