

Viscosity of Liquid Fe at High Pressure

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Introduction

The importance and relatively unknown nature of the effect of pressure on liquid Fe viscosity, to both the geophysical and the broad materials science communities, has sparked many studies to elucidate this subject. Very recently, through the development of a novel technique, we have measured the effect of pressure on an Fe alloy with a low S content (8.5 wt%) and found constant viscosity behavior along the pressure-dependent melting temperature [1]. This provided the first experimental verification that the effect of pressure on the viscosity of Fe-FeS alloys can be quantified through its effect on the pressure-dependent melting boundary. In order to understand the complex behavior of Fe and Fe alloys under pressure, a detailed study of the pure liquid Fe end member is required. Here we present measurements of the viscosity of pure liquid Fe at pressures up to 5.5 GPa and isothermally at 2050K by using synchrotron x-ray radiography.

Methods and Materials

High-pressure and high-temperature experiments were carried out in a 250-ton, DIA-type, large volume press (LVP) on the GeoSoilEnviro Consortium for Advanced Radiation Sources (GSECARS) 13-BM beamline at the APS. Solid Fe samples (99.9% purity) were machined to a cylinder and contained in a hexagonal boron nitride (hBN) sample container. A small pit was drilled in the base of the Fe sample to accept the probe sphere, which was constructed from the Pt core in a ruby mantle to produce a composite sphere [2]. Pressure was determined from an MgO pressure marker by using the known equation of state of MgO [3]. X-ray radiography was used to image *in situ* a probe sphere rising through the liquid Fe sample at high pressure. Radiographic images of the vertical flight of the probe sphere were collected on standard VHS videotape at 30 frames/s and subsequently digitized. All radiographic experiments were carried out by using incident x-rays in the energy range 20-120 keV.

Results and Discussion

A series of images of the ascent of the probe sphere through the liquid Fe sample is illustrated in Figure 1. The Pt core of the probe sphere is clearly seen as the dark circular region rising along the length of the liquid sample

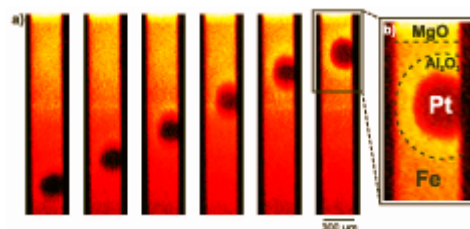


FIG. 1. (a) X-ray radiographic sequence of images of the vertical flight of a probe sphere through liquid Fe at 2050K and 4.5 GPa. (b) Enhanced view.

as the frames advance. The time between frames is ~70 milliseconds (ms), and the Pt core is seen as the dark circular region that rises as the frames advance. The total elapsed time during the vertical flight of the probe sphere is <1 s. From the radiographic images of the sphere's ascent through the liquid sample, the velocity of the sphere can be extracted. The displacement and velocity vs. time for the probe sphere's flight are given in Figure 2. To successfully measure the viscosity of the liquid Fe sample, it is necessary for the velocity of the probe sphere to be terminal (i.e., constant velocity vs. time). The sigmoidal nature of the displacement curve is indicative of the acceleration of the probe sphere as it is released from the base of the sample after the Fe sample has melted, the terminal velocity of the sphere during the bulk of its passage through the melt, and the deceleration as it approaches the container lid. The

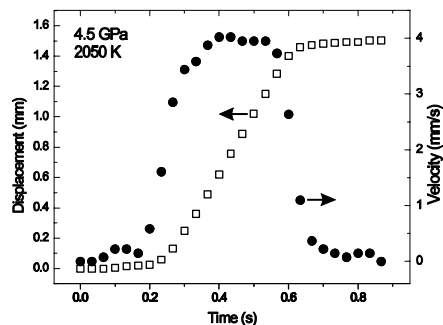


FIG. 2. Displacement and velocity vs. time curves for the flight of a probe sphere in liquid Fe at 4.5 GPa and 2050K.

terminal velocity of the sphere is further reinforced in the large plateau of the velocity curve.

By using a modified form of Stokes' law [1], the sample viscosity can be determined from the terminal velocity of the probe sphere. To accurately determine the density of the probe sphere, measurements of the volumes of the Pt and ruby components of the sphere were made at ambient conditions on the quenched samples by x-ray computed microtomography (CMT) [4]. The recorded image was then virtually sliced along each plane at successive depth intervals, and the areas of the Pt and ruby components were measured. Figure 3 illustrates an example of a CMT image sliced perpendicular [Fig. 3(a)] and parallel [Fig. 3(b)] to the long axis of the cylindrical Fe sample. The Pt core of the probe sphere appears as a bright yellow circular region, and the ruby mantle is the dark rim bracketing the Pt core from the Fe sample (red). The continuity of the ruby mantle can be verified in 3-D, confirming it acts as a successful barrier between the Pt core and the liquid Fe. Plotted in Fig. 3(c) are the measured areas of the Pt core and ruby mantle as the sample was sliced. The density of the probe sphere at the experimental pressure and temperature was calculated with the high-temperature third-order Birch-Murnaghan equation of state and the thermoelastic parameters for Pt and ruby [5-7].

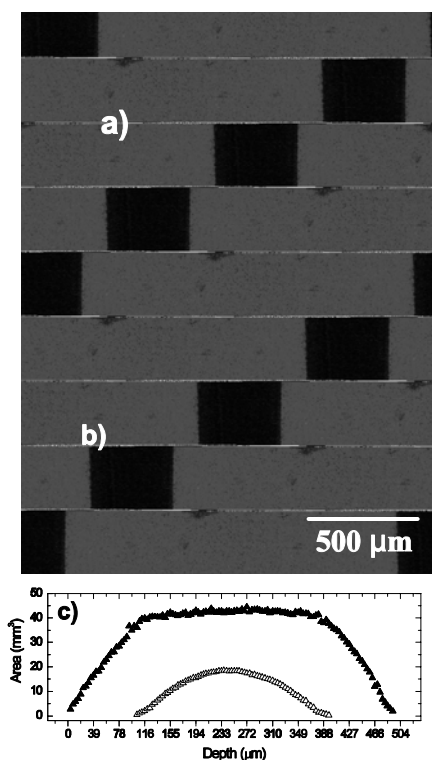


FIG. 3. X-ray CMT image of the unperturbed Fe sample and probe sphere after the experiment.

Viscosity data, derived from individual experiments at 2050K, are plotted as a function of pressure in Fig. 4 (solid diamonds). Upon first inspection of the data, the effect of pressure on the viscosity of liquid Fe is small, and the data agree well with the room pressure experimental (solid circle) [8] and theoretical (open circle) [9] values. This result contrasts sharply with our recent measurements of a liquid Fe-FeS alloy (8.5 wt% S), which indicate a large effect of pressure on viscosity [1]. The semi-empirical theory [10] describing transport properties in liquid metals under pressure, which until now has not been confirmed in pure liquid Fe, is used to fit our data.

When it is assumed that no change in liquid structure occurs in the pressure and temperature region examined, the effect of pressure on viscosity can be quantified through its corresponding effect on the melting curve. By taking the apparent activation volume as being 4% of the atomic volume, the predicted effect of pressure on viscosity is shown in Fig. 4 by a dashed line. The theoretical line successfully fits our data and the room pressure data well. The theoretical fit to our data implies that the viscosity of liquid Fe can be scaled to its melting boundary, allowing for the determination of viscosity in the pressure range for which the melting curve of Fe is known. In addition, the viscosity value determined for Fe by Ref. 11 through a first-principles, molecular-dynamics study, coincides with our experimental data at similar pressures and falls on the theoretical fit to the data (Fig. 4). The high-pressure viscosity datum determined by Ref. 12, which is plotted in Fig. 4, falls outside the viscosity region defined by our experiments and theory. Given the apparent activation energy of viscosity (~ 40 kJ/mol) for liquid Fe at room pressure [13], a correction for the effect of temperature on viscosity to 2050K is insufficient to bring it in line with our data.

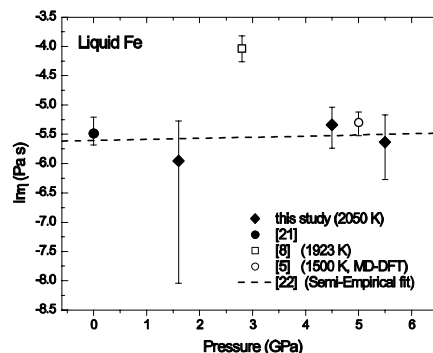


FIG. 4. Effect of pressure on the viscosity of liquid Fe at 2050K. Solid diamonds = data points measured in this study; solid circle = room pressure values taken from Ref. 8; open circle = first principles and molecular dynamics computation value of Ref. 11; open square = experimentally measured value of Ref. 12; and dashed line = semi-empirical fit from Ref. 10.

The importance of the experimental measurements of the viscosity of pure liquid Fe under high pressure is twofold. First, these data show, for the first time, the effect of pressure on liquid Fe viscosity by using a method that does not allow chemical contamination and provides very good resolution. The effect of pressure on viscosity can be used to address the nature and behavior of dynamic liquid properties in liquid Fe and Fe alloys at high pressure. Second, the agreement between the semi-empirical theory and the experimental values confirms constant viscosity at the pressure-dependent melting boundary for pure liquid Fe. This portends well for predicting the viscosity along the high-pressure melting boundary on the basis of the more easily determined 1-atm value of other liquid metals that do not undergo changes in liquid structure.

Acknowledgments

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