

## In situ measurement of viscosity of liquids in the Fe-FeS system at high pressures and temperatures

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### ABSTRACT

The viscosity of liquid FeS and Fe-FeS eutectic was measured at pressures between 0.5 and 5.0 GPa using a synchrotron-based falling sphere technique. We obtain viscosities of  $2 \times 10^{-2}$  to  $4 \times 10^{-3}$  Pa-s in FeS at 1450 to 1700 °C and  $2 \times 10^{-2}$  to  $8 \times 10^{-3}$  Pa-s in Fe-S<sub>eut</sub> at 1150 to 1380 °C. These results are consistent with recent viscosity measurements in Fe-S<sub>eut</sub> at 5 to 7 GPa (Urakawa, in preparation), measured diffusivities (Dobson 2000) and ab initio simulated viscosity (Vočadlo et al. 2000). The results are also similar to the values for pure iron at low pressure (Shimoji and Itami 1986). A systematic increase in viscosity and activation energy is seen with increasing sulfur content. Interpolation between the data presented yields a viscosity of  $1.4 \times 10^{-2}$  Pa-s for an outer core composition with ~10 wt% S at the melting temperature. There is good evidence of homologous behavior for Fe-S liquids which implies that the liquid alloy at the inner core boundary may have a similar viscosity.

### INTRODUCTION

Knowledge of the viscosity of liquid iron alloys at high pressure is important for understanding the chemical, thermal, and geomagnetic evolution of the Earth's outer core. The ill constrained geophysical measurements of outer core viscosity, spanning several orders of magnitude (Poirier 1988; Secco 1995), necessitate its estimation by other means. Experimental measurements of liquid iron alloys are extremely technically demanding and it is only in the last decade that such measurements have been attempted with some surprising results. Brazhkin's (1998) semi-empirical estimation of Fe viscosity, for example, based on quench crystallite size, yields extrapolations to outer core pressures of  $10^{11}$  Pa-s suggesting that the inner core would be in the glassy state. The Fe-FeS system has received considerable experimental attention for two reasons: (1) the low eutectic temperature renders it easily accessible to large volume high pressure devices and (2) because sulfur has been widely suggested as the light element in the cores of the Earth and other planetary bodies. LeBlanc and Secco (1996) used a rising sphere Stokes viscometry method to measure viscosity of Fe-FeS eutectic compositions at 2 to 5 GPa. They found Fe-S<sub>eut</sub> to have a surprisingly high viscosity of  $10^1$  to  $10^2$  Pa-s and a large activation energy, up to 3.5 cm<sup>3</sup>/mol. Secco et al. (1998) extrapolated slightly revised values to Earth's core pressures, to obtain a viscosity of  $10^{-4}$  to  $10^4$  Pa-s. Dingwell (1998) recently reviewed high-pressure melt transport properties.

In contrast, theoretical considerations (Poirier 1988) and ab initio simulations (Vočadlo et al. 2000; Alfé and Gillan 1998) suggest the viscosities of liquid Fe-S alloys are essentially independent of pressure along the melting path, with near-metallic viscosities around  $10^{-2}$  Pa-s at the solidus ( $T_m$ ). This view is supported by recent self-diffusion measurements in eutectic Fe-S at 2 and 5 GPa (Dobson 2000) which are similar to simulations results at outer core pressures (Alfé and Gillan 1998). To resolve these discrepancies, we present results of viscosity measurements of liquid FeS and a near-eutectic composition, Fe<sub>0.61</sub>S<sub>0.39</sub>, using the synchrotron-based Stokes-falling sphere method.

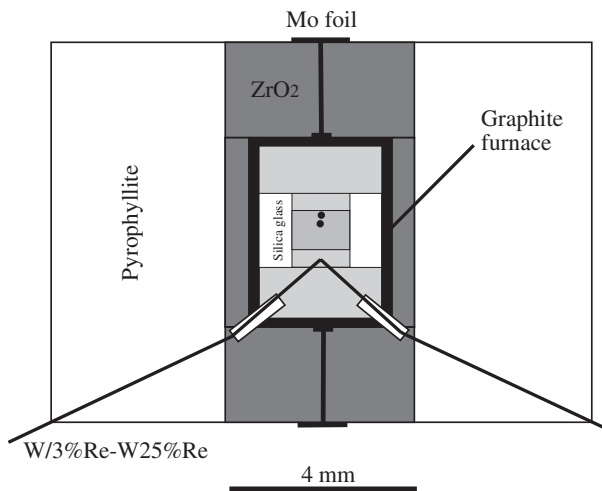
### EXPERIMENTAL METHODS

Samples consisted of synthetic 99.9% pure FeS or Fe-FeS powder mixtures, pre-pressed into dense pellets of 1.6 mm diameter and 1 to 1.5 mm long. Ir and W spheres were produced by passing an electric arc between two wires under nitrogen. Spheres between 40 and 120 μm diameter were placed in the top third of the sample and the whole was then loaded into a silica glass capsule and high pressure cell. The silica glass capsules acted as a barrier to water diffusion from within the pressure medium and helped to minimize oxidation and hydride formation in the sample.

Experiments were performed in the DIA apparatus in the 250 ton hydraulic press installed on bending magnet station 13-BM-D (Wang et al. 2000) at the GeoSoilEnviroCARS sector of the Advanced Photon Source (Rivers et al. 2000), using the high pressure cell design shown in Figure 1. The cubic pyrophyllite pressure medium of 8.0 or 8.5 mm edge length was compressed on each face by 6 mm edge length tungsten carbide anvils. The box-geometry of the graphite furnace and ZrO<sub>2</sub>

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**FIGURE 1.** High pressure cell for in situ high-pressure, high-temperature viscosity measurements.

thermal insulation served to reduce thermal gradients. A second sphere, placed centrally within the sample, which was central to the furnace, was used to detect the onset of melting. The measured temperature at which the central sphere first moved was typically 75–100 °C below the known melting temperature of FeS (Ryzhenko and Kennedy 1973). We can thus estimate that thermal gradients at experimental conditions are no larger than 75 °C/mm in the central 1.5 mm of the furnace, in good agreement with finite difference calculations based on the cell geometry used. Quoted temperatures have been corrected for the discrepancy between published melting temperatures and those measured by the onset of sphere movement. Tungsten-rhenium thermocouples were used with cold-reference junctions at room temperature and pressure; no pressure correction was applied.

Spheres were observed in situ using the synchrotron radiographic technique described by Kanzaki et al. (1987). The detector consisted of a double-polished YAG single crystal wafer placed in the transmitted beam path behind the press (Wang et al. 2000). White X-ray intensity contrasts caused by the differential absorption of spheres and the sample generated optical fluorescence contrast in the YAG crystal which was detected by a video camera. The camera was calibrated by moving the press in 50  $\mu\text{m}$  steps and observing the position of an absorbent marker in the X-ray beam. The spatial resolution of the camera-crystal arrangement depended on the thickness of the crystal and the resolving power of the camera and was approximately 4  $\mu\text{m}$ . Despite their high absorption, spheres smaller than 40  $\mu\text{m}$  diameter could not be reliably detected due to their low contrast to the background. Experiments were performed by compression at room temperature to the desired load. Pressure was calibrated against the equation of state of MgO. The sample was heated slowly ( $\sim 100$  °C/min) until first movement of the central sphere was observed upon which the temperature was rapidly increased by 100 to 200 °C. The central sphere

which detected the onset of melting typically fell approximately 1 second before the top spheres which were used to measure viscosity. The lower sphere fell during the ramp stage of heating, whereas the rapid final heating was achieved before subsequent spheres fell, ensuring their fall through a sample with constant temperature distribution. After the spheres had fallen, the sample was quenched in temperature, recovered and sectioned for electron probe analysis. One high temperature pressure calibration experiment was performed with MgO replacing the sample, but an otherwise identical geometry. The pressure-temperature curve due to cell relaxation and thermal expansion was assumed to be reproducible between experiments. Pressures are quoted to  $\pm 0.5$  GPa.

## RESULTS

Distance-time graphs for experiments at 1.5 GPa are shown in Figure 2. The plots have clear regions of constant fall velocity. For a solid sphere of density  $\rho_s$  and radius  $r_s$  falling in a liquid of density  $\rho_l$ , the viscosity is given by:

$$\eta = \frac{2gr_s^2(\rho_s - \rho_l)F}{9vK} \quad (1)$$

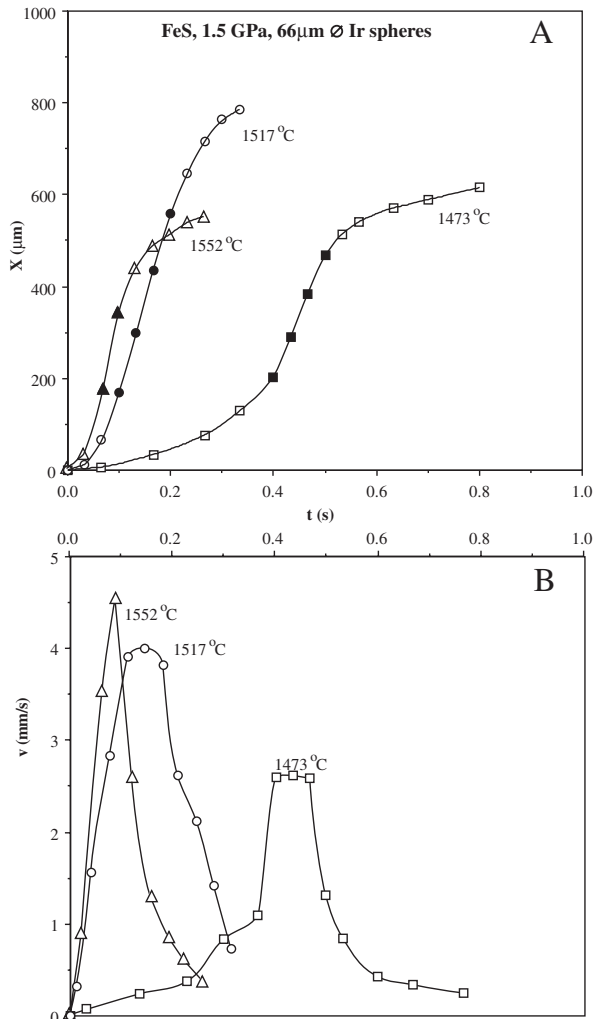
where  $v$  is the terminal velocity,  $F$  is the finite cylinder correction, and  $K$  is the finite length correction (Faxen 1925; Kingery 1959):

$$F = 1 - 2.104(r_s/r_c) + 2.09(r_s/r_c)^3 - 0.95(r_s/r_c)^5 \quad (2)$$

and

$$K = 1 + 3.3(r_s/h) \quad (3)$$

$r_c$  is the sample radius and  $h$  is the sample length, taken to be the entire fall distance. Sphere density and diameter were corrected for  $P/T$  using compression and thermal expansion data of Katsnelson et al. (1997) and Wimber (1976) respectively. The density of liquid Fe-S eutectic was calculated from density measurements of Sanloup et al. (2000). FeS density was estimated from the density of FeS at ambient melting (Lennie et al. 1997), combined with the compressibility data for the most sulfur-rich composition of Sanloup et al. (2000). This procedure slightly overestimates the 1 atmosphere liquid FeS density, but slightly underestimates FeS compressibility. Measured terminal velocities and calculated viscosities are given in Table 1. These viscosity values of  $10^{-2}$  to  $10^{-3}$  Pa·s are in good agreement with those obtained by Urakawa et al. (in preparation). Errors in calculated viscosity arise from uncertainties in density of sphere and melt, sample length and sphere diameter. Errors in sphere and melt density may result in up to 3% error in viscosity. Due to the thermal gradient in the cell, it is likely that the whole sample was not molten when the spheres fell, for measurements near the melting temperature, hence the fall distance provides the best estimate of the effective sample length,  $h$ . Reaction between the melt and sphere material caused a reduction in sphere size during the experiment. We estimate that the sphere diameter changed by less than 25% during the falling part of the experiment which would result in 40% overestimation of viscosity.



**FIGURE 2.** Distance-time (a) and velocity-time (b) graphs for three experiments. The acceleration region and terminal velocity region (solid symbols) are clearly seen. A region of deceleration is seen due in part edge effects and to thermal gradient effects is also seen.

## DISCUSSION

Measured viscosity for liquid  $\text{Fe}_{0.61}\text{S}_{0.39}$  and FeS (Fig. 3) are compared to ab initio simulated viscosities for  $\text{Fe}_{0.61}\text{S}_{0.39}$  (Vočadlo et al. 2000). The measured viscosity of  $\text{Fe}_{0.61}\text{S}_{0.39}$  in this study is consistent with the values predicted from diffusion measurements (Dobson 2000) and the higher activation energy for diffusion is also seen in liquid Fe (Yang et al. 1957; Shimoji and Itami 1986) and other metal systems (Poirier 1988). Previous viscosity measurements (LeBlanc and Secco 1996; Secco et al. 1998) give viscosity values 2 to 3 orders of magnitude higher than the present study and that of Urakawa et al. (in preparation). This discrepancy needs some comment:

(1) In our experiments, convection may contribute underestimation of viscosity. By contrast, LeBlanc and Secco may overestimate of viscosity due to convection. LeBlanc and Secco used a floating sphere technique, whereas we used a falling sphere technique, thus convection which would be downward in the cooler center of the sample and upward at the warmer sample edges would accelerate falling and decelerate rising spheres. Convection is easy to detect using in situ radiographic techniques since the position of the sphere is directly observed, whereas electro detection does not directly image spheres and convection may be missed. It seems unlikely, however that thermal convection is significant in the set-up of LeBlanc and Secco who had small thermal gradients. Likewise, our short sample lengths lead to a low Raleigh number around  $10^{-2}$  which would suggest insignificant convection. Some experiments, in the near-eutectic composition, did show convection and affected results were discarded. Convection in eutectic systems may be related to changing equilibrium melt composition during heating. If this is the case, gravitationally unstable chemical gradients in the melt may ensue, resulting in double-diffusive convection. Electron microprobe analysis of recovered run products demonstrated a small decrease in sulfur content in near-eutectic experiments with increasing pressure (Table 2). The lowest pressure eutectic experiment contained some relict Fe liquidus crystals at one end of the sample, otherwise quench textures indicative of complete melting were observed in all recovered samples. Double-diffusive convection would not affect experiments on FeS and convection was not observed in those experiments.

**TABLE 1.** Experimental conditions and results

$P$ (GPa)	$T$ (°C)	Sample	$r_s$ ( $\mu\text{m}$ )	$v$ (mm/s)	$h$ (mm)*	$F/K$	$\eta$ (mPa-s)
0.5	1450	FeS	33, 33	1.77, 2.30	0.4, 0.3	0.718, 0.596	17.2, 19.5
1.5	1580	FeS	33, 55	6.3, 7.99	0.60	0.773, 0.657	5.2, 9.5†
1.5	1517	FeS	33	3.91	0.80	0.804	8.6
1.5	1473	FeS	33	2.66	0.65	0.782	12.2
2.5	1547	FeS	55	5.50	0.65	0.669	13.9
2.5	1552	FeS	22, 44	2.55, 4.51	0.75	0.859, 0.741	6.2, 11.8†
4.8	1650‡	FeS	28	5.8	0.7	0.818	4.2
5.0	1707	FeS	22	3.84	0.50	0.823	3.8
1.5	1290	Fe-S <sub>eut</sub>	50§	5.5	1.2	0.649	11.7†
2.2	1374	Fe-S <sub>eut</sub>	44	6.95	0.75	0.741	7.4
5.5	1150‡	Fe-S <sub>eut</sub>	250#	15	1.5	0.260	22.0†

\* Total fall distance.

† Terminal velocity not reached.

‡ Temperature estimate based on power.

§ Platinum sphere.

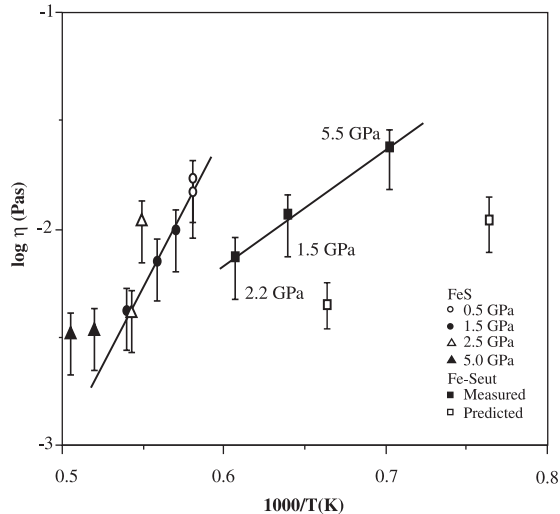
# Tungsten carbide sphere.

|| Sphere fell capsule length in one 0.1 s frame.

**TABLE 2.** Compositions of recovered samples

$P$ (GPa)	Near-eutectic			FeS		
	1.5	2.2	5.5	1.5	2.2	5.0
Fe	$57.5 \pm 0.5$	$59.8 \pm 0.6$	$60.2 \pm 0.7$	$50.8 \pm 0.9$	$51.5 \pm 0.6$	$51.6 \pm 1.1$
S	$39.9 \pm 1.0$	$39.7 \pm 0.5$	$38.8 \pm 0.5$	$46.5 \pm 0.7$	$46.7 \pm 1.6$	$46.1 \pm 1.0$
O	$2.7 \pm 1.2$	$0.5 \pm 0.5$	$0.9 \pm 0.8$	$2.7 \pm 1.2$	$1.9 \pm 1.9$	$2.2 \pm 0.7$
S/(S+Fe)	0.41	0.40	0.39	0.48	0.48	0.47

Notes: Standards were pure metal, natural FeS<sub>2</sub> and natural fayalite for Fe, S, and O, respectively. Analyses were performed by rastering the beam over a 1600  $\mu\text{m}^2$  area to average quench textures.



**FIGURE 3.** Arrhenius plot for viscosity in FeS and Fe<sub>0.61</sub>S<sub>0.39</sub>. The data for FeS at 0.5 to 2.5 GPa fall along the same curve, as do those for Fe<sub>0.61</sub>S<sub>0.39</sub> at 1.5 to 5.5 GPa, suggesting the effect of pressure is small. The error in the data is too large to estimate activation volumes over such a small pressure range. The Fe<sub>0.61</sub>S<sub>0.39</sub> viscosity is similar to that predicted from ab initio calculations on Fe<sub>0.61</sub>S<sub>0.39</sub> at 5 GPa (Vočadlo et al. 2000).

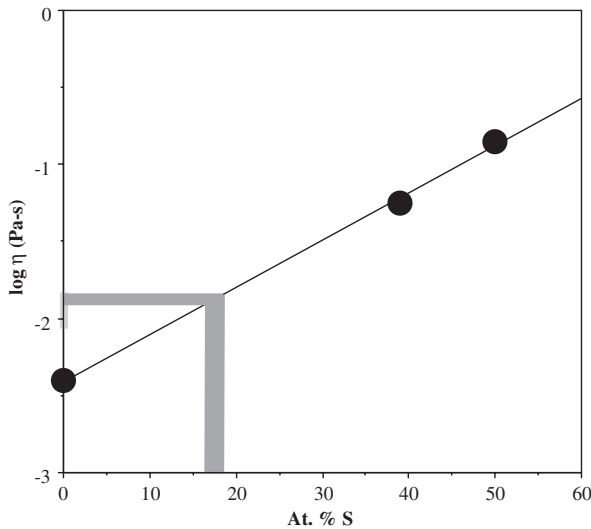
(2) Our early experiments at the APS on Fe<sub>0.61</sub>S<sub>0.39</sub> mixtures produced confusing results with high apparent viscosities. It was found that if heating rates were too slow, the sphere fell through a liquid-crystal mixture, even at temperatures significantly above the solidus. This can be explained if the density of the eutectic melt is intermediate to the densities of the FeS and Fe solid phases. In this case, during slow heating, the solid phases can segregate, with the lighter FeS floating to the top and the denser Fe sinking to the bottom of the sample. Once this density separation has occurred, the local composition may be a long way from the eutectic composition and so liquidus crystals persist for a long time. We decided to test this by performing experiments on end-member FeS and saw no experiments in this composition with anomalously high measured viscosities.

(3) Electron probe analysis of sectioned samples after experiments revealed the presence of up to 4 at% oxygen in our experiments, although the value was typically around one to two at% (Table 2). Oxygen may act to reduce viscosity in S-polymerized melts as discussed by Dobson (2000), and has been suggested as reducing viscosity in liquid Fe: a liquid contain-

ing 33 at% O was calculated to have a viscosity of  $4.5 \times 10^{-3}$  Pa-s under inner core boundary conditions (Alfè et al. 1999). This halving of viscosity of pure iron with 1/3 of the atoms replaced by oxygen strongly mitigates against the small amount of oxygen contamination in the present experiments being the cause of the low viscosity.

The present viscosity measurements in liquid FeS and Fe<sub>0.61</sub>S<sub>0.39</sub> are consistent with ab initio simulations values (Vočadlo et al. 2000), independent falling sphere measurements (Urakawa, in preparation) and values derived from diffusivity measurements using the Stokes-Einstein relation (Dobson, 2000; Majewski and Walker 1998). They are also close to measured values for iron and FeS at atmospheric pressure. Sulfur content has a large positive effect on activation energy with  $\Delta Q_{\text{Fe}} = 41.4$  kJ/mol (Shimoji and Itami 1986),  $\Delta Q_{\text{Fe}_{0.61}\text{S}_{0.39}} = 100$  (5) kJ/mol and  $\Delta Q_{\text{FeS}} = 255$  (18) kJ/mol, consistent with other studies (Secco 1999a, 1999b). Sulfur is known to have a large effect on compressibility in the liquid Fe-S system (Nasch et al. 1994, 1997; Sanloup et al. 2000), which may suggest that the large activation energy for viscosity is related to a large thermal expansion, although this has not been measured. It might be reasonable to expect that the pressure dependencies of viscosity in the Fe-S system would also be strongly dependent on sulfur content. The present study covered too small a pressure range and errors were too large to resolve the activation volume for viscosity in liquid FeS and Fe<sub>0.61</sub>S<sub>0.39</sub>. Other studies are currently underway to delineate this question (Urakawa, in preparation).

The remarkable agreement between the measured viscosity in this study, that of Urakawa et al. (in prep) at 7 GPa, and the value obtained from simulations results at 15 and 330 GPa (Vočadlo et al. 2000; Alfè and Gillan 1998) strongly implies that Fe-FeS compositions display homologous viscosity behavior. As previously noted by Poirier (1988) such homologous behavior implies that the viscosity of the liquid iron alloy at the inner core boundary is the same as the ambient pressure  $T_m$  value. We have extrapolated our current viscosity measurements, and those for pure iron (Shimoji and Itami 1986), to the melting temperature, assuming the Arrhenius temperature dependence does not change between the measured and extrapolated range. Interpolation between these extrapolated  $T_m$  viscosity values to the 18 at% sulfur value required to fulfill outer core density requirements (Fig. 4) (assuming the light element is entirely sulfur; Alfè and Gillan 1998), we obtain a  $T_m$  viscosity value of  $1.4 (\pm 0.2) \times 10^{-2}$  Pa-s, in remarkable agreement with ab initio simulations results at inner core boundary conditions.



**FIGURE 4.** Extrapolated  $T_m$  viscosity vs. at% S from the present experiments and Shimoji and Itami (1986). The medium grey line shows interpolated viscosity at 18 at% S, which is the suggested outer core content based on density constraints (Alfè and Gillan 1998). The ab initio simulation value under outer core boundary conditions is shown by the pale grey line along the viscosity axis.

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