Pilot Study of PVT Equations of State of Fluids in a Diamond Anvil Cell by X-ray Radiography

Q. Guo,[1] H. Mao,[1] J. Hu,[1] R. J. Hemley,[1] G. Shen[2]
[1]Geophysical Laboratory, Carnegie Institution of Washington, Washington, DC, U.S.A.
[2]The University of Chicago, Chicago, IL, U.S.A.

Introduction

Knowledge of the densities and specific volumes of fluids, melts, and amorphous solids at high pressures and temperatures is a key for understanding geochemical and cosmochemical processes. Unlike density measurements of crystalline solids, which can be obtained by x-ray diffraction of a very small sample at multimegabar pressures, density measurements of fluids and amorphous materials are limited to relatively low pressures and temperatures. Indirect methods, such as Brillouin scattering spectroscopy [1], yield high-pressure density only for purely elastic systems, but not if the system goes through structure change [2], liquid-liquid phase transition [3], or dissolution-precipitation, which often occur at high pressures [4]. Direct volume measurement of amorphous materials is indispensable.

Methods and Materials

Use of the 13-BM beamline and the x-ray microradiographic technique developed by the GeoSoilEnviro Consortium for Advanced Radiation Sources (GSECARS) is ideal for measuring the sample volume in diamond anvil cells (DACs), because the sample between two parallel diamond anvil surfaces can be treated as 2-D and because the volume can be determined by transmission radiography without rotating the sample.

We loaded a H_2O sample in a Re gasket. The x-ray radiograph or differential radiograph above and below the Re K absorption edge (71.6 keV) provides a measurement of the thickness, inner diameter, and edge features of the Re gasket and the sample cavity. We heated the sample in a DAC with an external resistive heater [4]. Pressures were determined by *in situ* x-ray diffraction of a gold chip (~10 μm) within the water.

Results

Figure 1 shows the x-ray microradiographic images of a water sample in a Re gasket with x-ray energies of 37.4, 71.3, and 71.7 keV. The calculated gasket thickness between the two diamond anvil surfaces and the volume of the sample cavity are listed in Table 1. Our preliminary results from the water pressure-volume-temperature (PVT) experiments demonstrate the feasibility of using the microradiographic technique for determining density in DACs for the amorphous materials.

Discussion

Table 1 shows the calculated volume of a water sample at room temperature and 590K and pressures up to 7.5 GPa. The uncertainties of the experimental data are about 5%. Efforts are being made to improve the measurement precision and accuracy.

Figure 2 shows the x-ray 2-D radiograph of a water sample in a Re gasket with 37.4 KeV x-ray energy. This image shows a crack in the gasket, and the condition of mass conservation of the fluid sample may no longer be met. Moreover, the further pressure-temperature increment may lead to sample leakage.

Table 1. The gasket thickness and volume of a water sample cavity at room temperature and at 590K at various pressures.

Temperature (K)	296	585	593	583	593	593	596	594
Pressure (GPa)	1.27	2.79	3.05	3.88	4.1	4.2	4.81	7.48
Thickness scan (μm)[a]	88.4	87.5	87.1	81.7	76.9	83.9	75.9	63.6
Thickness image (μm)[b]	89.7	89.4	84.5	86.4	75.9	81.1	72.4	63.5
V scan (mm^3)[a]	0.003868	0.006818	0.006925	0.00641	0.00675	0.007000	0.007084	0.007088
V image (mm^3)[b]	0.003925	0.006967	0.006718	0.006778	0.006662	0.006766	0.006757	0.007077

[a]Measurement from the absorption of a single beam 50×50 μm for comparison with the x-ray radiography technique.
[b]Results of x-ray radiography.

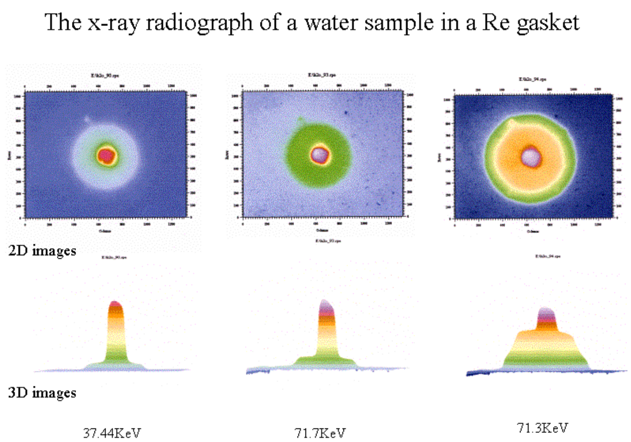

FIG. 1. X-ray radiograph of a water sample in a Re gasket with different x-ray energies.

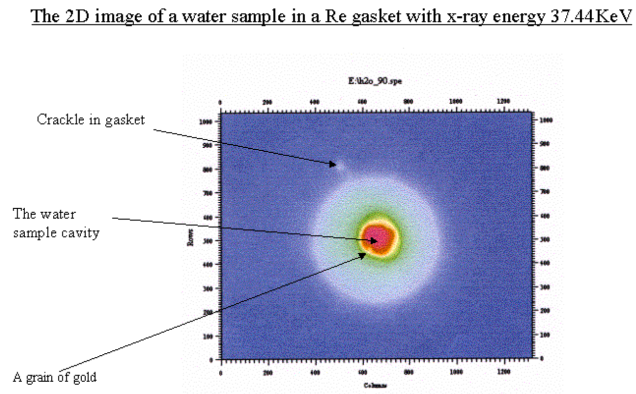

FIG. 2. 2-D x-ray radiograph of a water sample.

The successful H_2O experiment has opened a field for studying other volatile components, including those soluble in H_2O under the mantle conditions. The study may lead to future projects involving He-H_2 mixtures and other planetary gases and ices.

Acknowledgments

Use of the APS was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. W-31-109-ENG-38. This research is funded by the Carnegie Institute of Washington and National Science Foundation.

References

[1] C.-S. Zha, R. J. Hemley, H. K. Mao, T. S. Duffy, and C. Meade, "Acoustic velocities and refractive index of SiO_2 glass to 57.5 GPa by Brillouin scattering," Phys. Rev. B **50**, 13105-13112 (1994).

[2] C. Meade, R. J. Hemley, and H. K. Mao, "High pressure x-ray diffraction of SiO_2 glass," Phys. Rev. Lett. **69**, 1387-1390 (1992).

[3] E. Schwegler, G. Galli, and F. Gygi, "Water under pressure," Phys. Rev. Lett. **84**, 2429-2432 (2000).

[4] Y. Fei and H. K. Mao, "In situ determination of the NiAs phase of FeO at high pressure and high temperature," Science **266**, 1678-1680 (1994).