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Volumetric properties of magnesium silicate glasses and supercooled liquid at high pressure by X-ray microtomography

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ABSTRACT

The volumetric properties of silicate glasses and supercooled liquid are examined at high pressures and temperatures using X-ray computed tomography (CT) and absorption. The high pressure X-ray microtomography (HPXMT) system at the Advanced Photon Source, Argonne National Laboratory (GeoSoilEnvironCARS 13-BM-D beamline) consists of two opposing anvils compressed within an X-ray-transparent containment ring supported by thrust bearings and loaded using a 250-ton hydraulic press. This system permits the pressure cell to rotate under the load, while collecting radiographs through at least 180° of rotation. The 13-BM-D beamline permits convenient switching between monochromatic radiation required for radiography and polychromatic radiation for pressure determination by energy dispersive diffraction. We report initial results on several refractory magnesium silicate glasses synthesized by levitation laser heating. Volume changes during room temperature compression of Mg-silicate glasses with 33 mol% and 38 mol% SiO_2 up to 11.5 GPa give an isothermal bulk moduli of 93–100 GPa for a K′ of 1. These values are consistent with ultrasonic measurements of more silica-rich glasses. The volumetric properties of amorphous $MgSiO_3$ at 2 GPa were examined during annealing up to 1000 °C. We consider the consequences of heating through the glass transition and the implications for thermal expansivity of supercooled liquids at high pressure. Our results illustrate the capabilities of HPXMT for studies of refractory glasses and liquids at high pressure and offer strategies for future studies of liquid densities within the melting interval for magmas in planet interiors.

1. Introduction

Models of the Earth's thermal state, chemical differentiation, and dynamic processes require knowledge of properties of silicate and metallic melts. Determining densities of silicate liquids at high pressures are challenging measurements. The most common approaches are fusion curve analysis (Herzberg, 1987; Agee, 1998; Lange, 2003, 2007), sink–float experiments (e.g., Ohtani et al., 1993, 1998; Agee and Walker, 1993), shockwave studies (e.g., Rigden et al., 1984; Miller et al., 1991) and, more recently, X-ray absorption (Urakawa et al., 2005; Chen et al., 2005). Important constraints on silicate liquid compressibility are also provided by ultrasonic measurements (Rivers and Carmichael, 1987; Lange and Carmichael, 1987; Kress and Carmichael, 1991; Kress et al., 1988; among others). Perhaps the most direct method for determining compressibility and thermal expansion, or their integral properties density and volume, is to measure the physical change in melt volume as pressure and temperature varies, respectively. 2-D dilatometry has been widely used to characterize thermal expansion of silicate liquids at 1-atm (e.g., Lange, 1996), but this approach is not easily adapted to high pressures where the sample of interest is embedded in a confining medium and dimensional changes must be accounted for in 3-D.

Assuming that the high pressure assembly containing the sample can be penetrated by a light source, i.e., X-rays or neutrons, the rendering of the sample in 3-D can be accomplished by combining a series of shadowgraphs (radiographs) of the object as it is rotated through 180° about an axis perpendicular to the incident beam. This imaging technique, known as computed tomography, is widely used in the medical field (i.e., CAT scan) and is becoming more commonplace in the earth sciences for imaging geological samples at ambient conditions (Ketcham and Carlson, 2001; Rivers et al., 2004; Huddlestone-Holmes and Ketcham, 2005). X-ray photons are the most common light source used for imaging geological materials and the highest resolution systems are those utilizing the X-ray brilliance available at synchrotron facilities.

X-ray computed tomography (CT) exploits the absorption contrast described by the Beer–Lambert law:

$$I = I_o \exp\ (-\mu_m \rho x), \qquad (1)$$

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where I is the transmitted beam intensity, I_0 is the incident beam intensity, μ_m is the mass absorption coefficient, ρ is sample density, and x is sample thickness. The technique does not require explicit knowledge of the values of μ_m, ρ, and x, but is of little use if there is no resolvable difference in $\mu_m\rho$ or the sample is too thin or thick. The tomographic reconstruction is fundamentally a 3-D map of $\mu_m\rho$ or density alone, if the composition and mass absorption coefficients are known. The product $\mu_m\rho$ is the linear attenuation coefficient (μ_l) and can be computed from NIST look-up tables for the energy of the incident X-ray beam (Hubbell and Seltzer, 1996).

In this paper we review the principles and practical considerations for computed tomography at high pressures, discuss the capability of the rotating anvil apparatus installed on GeoSoilEnviroCARS 13-BM-D beamline at the Advanced Photon Source, Argonne National Laboratory, and present new data on the compression of refractory magnesium silicate glasses up to 11.5 GPa at room temperature and on heating to 1000 °C at 2 GPa. We compare volumetric properties as determined by direct volume rendering and X-ray adsorption. These experiments demonstrate the utility of high pressure X-ray microtomography (HPXMT) for characterizing glass and supercooled liquid properties at elevated pressures and eventually within the melting interval of compositions relevant to the Earth's crust, mantle and core.

2. Methods

2.1. General considerations

To appreciate the challenges one faces acquiring computed tomography data at high pressures it is useful to review the general principles of X-ray computed tomography. To generate a 3-D image of an object of any shape by CT a series of radiographs (2-D representations of transmitted X-ray intensity) are collected as the sample is rotated about an axis perpendicular to the plane of the incident beam. The discrete angular interval between successive radiographs, coupled with the resolution of the imaging system, determines the voxel resolution of the reconstructed image.

For microtomography (objects of interest on the mm scale) radiographs are usually acquired every 0.5° of rotation with a charge couple devise (CCD) camera after the conversion of incident X-rays to visible light by a scintillation screen. In our case, the CCD camera has a 1300 × 1024 pixel photodiode array – each pixel being 6.7 × 6.7 μm. With appropriate microscope objectives and associated optical components, a single pixel of the CCD camera corresponds to a region of about a square micron in the radiograph. After 3-D reconstruction the voxel resolution is a few cubic microns. In practice, the voxel resolution is usually limited by computing power, i.e., data storage and processing capacity. For example, 1300 (horizontal) × 1024 (vertical) pixel images for 359 radiographs yields 3.45×10^8 bits of intensity data. Pre- and post-processing such large datasets are time-consuming on a standard PC, if at all possible.

It is common practice to combine the signals from several pixels (binning) to create an effective pixel containing the sum of signals from each sub-pixel and representing an area equal to the number of sub-pixels squared. In the present work we use a bin setting of 2 × 2 that yields an effective pixel array of 650 × 512 with approximately four times the signal strength. While this reduces our effective pixel resolution to about 3–4 μm, the increased sensitivity achieved by summing the signals permits us to significantly reduce file size and data acquisition time – both advantageous when performing experiments at synchrotron facilities where data storage and beam time are limited. A typical radiograph of our high pressure cell with 2 × 2 binning can be obtained in ∼10 s, requiring about 2 h for a complete data set suitable for tomographic reconstruction.

Several technical obstacles must be overcome to perform X-ray computed tomography on samples under pressure. Firstly, the pressure cell containing the sample must be able to rotate under a load to collect radiographs at discrete angular positions. This requires that the rotating portion of the system carrying the load must be supported by thrust bearings at the top and bottom and rotate smoothly and accurately to within ±0.01°. Secondly, the sample chamber and confining medium must be transparent to X-rays so that the absorption contrast between the sample and surrounding medium can be imaged. This requirement severely limits the type of apparatus and cell parts one can use. Since it is necessary to view the assembly through 360° of rotation, the opposing anvil geometry offers the most convenient configuration, i.e., beveled and truncated cylindrical anvils (Drickamer cell) or toroidal anvils (e.g., Khovostantsev

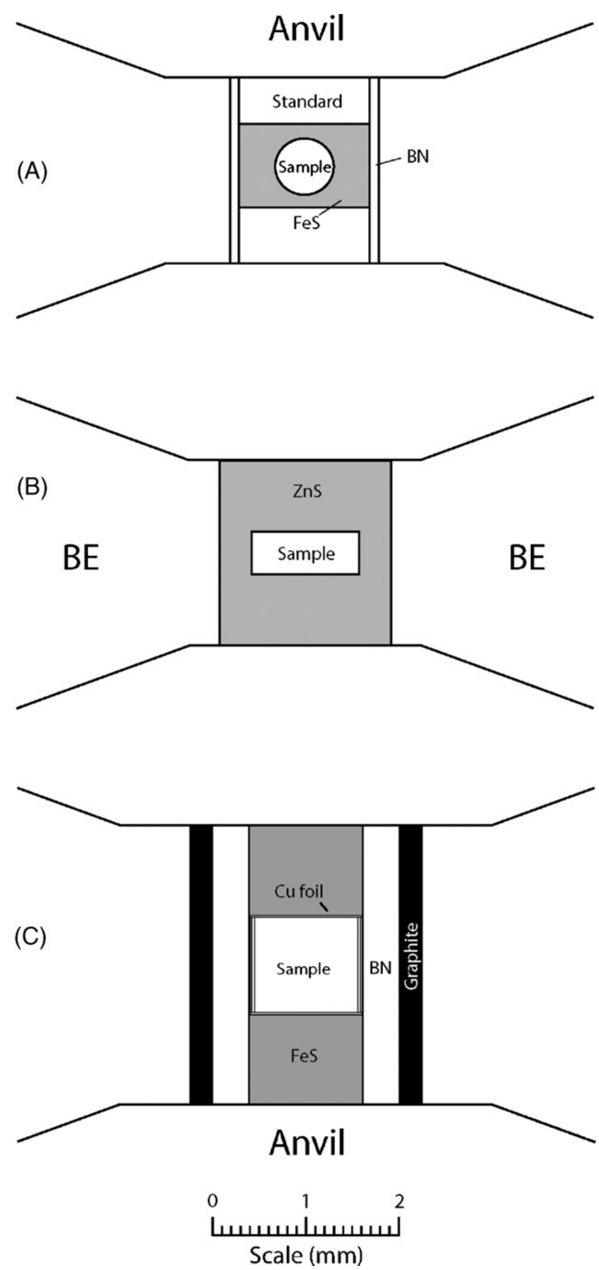

Fig. 1. Scaled drawings of Drickamer cells used in this study. Assemblies A and B are designed for room temperature experiments. Assembly C is used for sample heating. See text for details.