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ABSTRACT

We report an experimental protocol for measuring the frequency dependence of the bulk modulus carried out in a synchrotron x-ray facility based multi-anvil high-pressure apparatus. An oscillating pressure perturbation characterized by x-ray diffraction produces a volume strain measured by imaging. Together, these yield the bulk modulus of the sample. Here, we report data at 3 mHz as an example of the possibility of providing these data for the frequency range of 1 mHz–100 mHz.

The bulk modulus of a material is generally recovered from the slope of the volume–pressure curve or from the acoustic velocities measured with ultrasonic waves[1] or from Brillouin spectroscopy.[2] Normally, these values of the bulk moduli are identical except for contributions from the differences in the thermal path, e.g., isothermal or adiabatic. However, if the time response of the volume to pressure change is not instantaneous, then there will be a frequency dependence of the bulk modulus. Multivariant phase transformations are typically not instantaneous and do affect the volume vs pressure curve, thereby introducing a frequency dependence to the bulk modulus.

Time dependent phenomena have long been studied for the shear modulus where plastic deformation, induced by shear stress, introduces a time dependent shear strain and creates a frequency dependent shear modulus[3,4] or Young's modulus.[5] These time dependent processes also produce attenuation as a function of frequency, particularly in the frequency range associated with the time scale of the relaxation process that produces the modulus change.

In Earth science, it is vital to predict the velocities of longitudinal and shear seismic waves in the seismic frequency band that ranges from Hz to mHz. In contrast, most laboratory measurements rely on ultrasonic velocities and Brillouin spectroscopy in the MHz to GHz frequency band. Our goal has been to develop a methodology to measure the bulk modulus at the frequency of the seismic band.

Here, we report a new experimental protocol to measure the bulk modulus at mHz frequency and pressure and temperature up to 12 GPa and 2000 K using a multi-anvil high pressure apparatus combined with synchrotron x-ray radiation. The overall approach involves three components once the sample has been brought to the desired P and T. The first is to sinusoidally oscillate the pressure without bringing the pressure outside of the zone of interest. Then, the volume strain of the oscillation for the sample needs to be measured. Finally, the pressure of the oscillating field needs to be measured in the sample.

These experiments are conducted at the Advanced Photon Source (APS) at Argonne National Laboratory on a bending magnet beamline 6BMB. The cylindrical sample is about 3 mm long and has 1.3 mm diameter. The sample assembly, illustrated in Fig. 1(a), is located at the center of a boron-epoxy cubic cell with 6.15 mm edge lengths. The sample, located at the center, is illustrated as a gray region and is surrounded by rhenium, illustrated as gold. Rhenium is a 100 μm foil, wrapped around the sample, with two end-caps, one at the top and the other at the bottom. A BN sleeve, illustrated as blue, is outside of rhenium, and it is surrounded by the black graphite furnace. Machinable corundum cylinders, green, are located above and below the sample. Outside of the graphite furnace is an alumina sleeve that contacts the boron-epoxy pressure media. Rhenium serves as an x-ray marker, defining the shape of the sample.

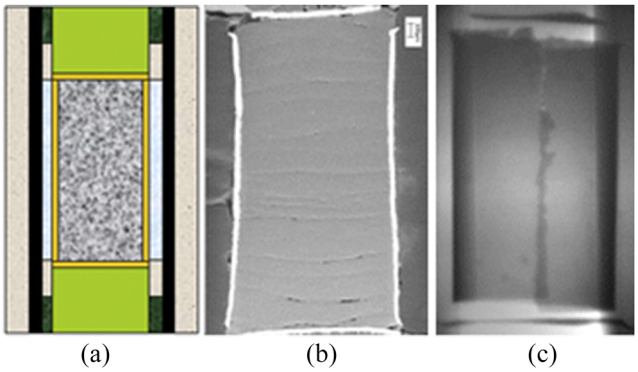

FIG. 1. High pressure sample. (a) illustrates the sample assembly. (b) is an SEM of the recovered sample and (c) is the image of the sample recorded during the experiment at high P and T.

Figure 1(b) illustrates the sample that has been recovered from the experiment. SEM images are used to define the grain size, to identify melt pockets, and to provide chemical compositions of the final phases. The four anvils that compress the sample from the azimuthal direction are sintered diamonds, allowing the x rays to penetrate to the sample and be recorded with the use of a fluorescent screen and with a digital visible light camera. Figure 1(c) illustrates one of these images for the sample at elevated pressure and temperature. The dark line down the center is the overlap of the rhenium foil as it is wrapped around the sample.

The sample is KLB-1, a natural rock, a spinel lherzolite from the Kilbourne Hole crater in New Mexico. The main mineral in the sample is olivine (60%). X-ray diffraction is gathered from the sample from inside the rhenium capsule. The other minerals are less abundant and difficult to identify in the diffraction patterns.

At a preset pressure and temperature (1.5 GPa and 1430 K), we oscillate the pressure with a sinusoidal variation in the ram pressure of the DDIA high pressure apparatus. The ram load was about 18 tons, and the amplitude of the sinusoidal oscillation was ±1.6 tons. The pressure variation due to the oscillation was about ±0.17 GPa. The temperature is defined by the wattage of the furnace and prior watt–temperature calibrations. Olivine diffraction is used as the pressure marker. The period of the sinusoidal oscillation is 360 s. One image and one diffraction pattern are taken every 30°.

The volume strain is measured from the x-ray radiograph. The sample is a cylindrical rod and contained in a metal foil capsule. The metal foil capsule defines the length and diameter of the sample; the sample volume can be derived from the two-dimensional x-ray image assuming cylindrical symmetry. We calculate the changes in the dimension of the capsule by choosing a reference image and perform a correlation analysis with other images including a strain term to match the intensities of the pixels. We also correct the image for displacement relative to the reference image. Over the course of an experimental run, several reference images may be chosen as the character of the foil may change over long periods of time. The strain is calculated from the difference in the dimension of each frame compared to the reference frame divided by the size of the frame. This value of strain includes the oscillation strain along with secular

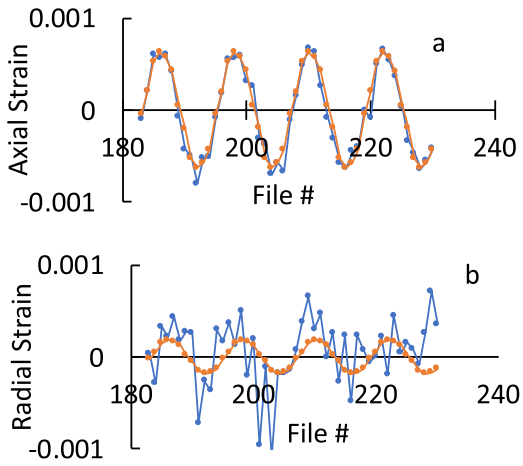

FIG. 2. Radial (a) and axial (b) strain derived from sample images. File # is the sequence number of the images taken as a function of time. The blue dots are the measured points and the red curve is a fitted sinusoidal curve.

variations that reflect long term strains. To remove this latter contribution, we subtract a moving average from the strain. The measure strains are illustrated in Fig. 2 in blue for the radial and axial directions for four cycles at the target P-T condition. A sine wave with a period of 360 s is fitted to the data; the results are indicated in red. The sine wave fitting the axial strain has an amplitude corresponding to a change in the length of 0.69 pixels, while the radial strain amplitude corresponds to 0.088 pixels, which in turn corresponds to length changes of 1.6 μm and 0.2 μm, respectively. The underlying sinusoidal character of the signal allows one to resolve length changes on this scale.

The pressure variation is determined from x-ray diffraction data. The best pressure marker is a material that is inside of the sample capsule. We use olivine that is a major mineral in KLB-1. However, the diffraction data were compromised by absorption from the rhenium capsule and, at high temperatures, by recrystallization of olivine. We use a white beam with ten energy dispersive detectors located at 6.5° of two-theta.[5] Because we want to sample the sinusoidal field, we are limited in the amount of time that we can collect data. We choose 23 s allowing us to collect a full set of diffraction data and image data in 30 s or 30° of the sine wave. We determined that the differential stress in the run was small and did not couple strongly with the oscillating pressure field. Thus,

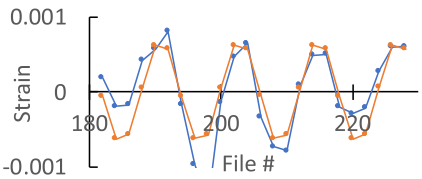

FIG. 3. The strain of the (112) lattice spacing from diffraction data. File # is the sequence number of the images taken as a function of time. The blue dots are the measured points, and the red curve is a fitted sinusoidal curve.

we summed the data from the ten detectors giving us the equivalent of a 230 s dataset. As we are most confident of the (112) peak, we use it here. We calculate the strain as $(d_{ob} - d_{av})/d_{av}$, where d_{ob} is the observed lattice spacing and d_{av} is the average of the lattice spacing over one cycle time of the sine wave. Figure 3 illustrates the observed lattice strain of the (112) peak in blue and the fitted curve in red over four cycles of oscillation at the target P-T conditions.

In these experiments, the sample pressure and temperature environment is generated in the multi-anvil DDIA apparatus. Due to the new feature required in this type of bulk modulus measurement, we automated the oil pressure control with software. The software designates a number of variables in order to implement sinusoidal pressure waves. These variables include amplitude, period, and motor speed. Other features include automatic x-ray diffraction pattern collection, shutter control, x-ray radiograph image collection, data writing, and log file writing. The software enables the different types of data recording synchronized with the sinusoidal function. Thus, even if the image volume data and x-ray diffraction data are collected at different time, their relative phase is well known.

The bulk modulus is calculated as the oscillation pressure change divided by the oscillation induced volume strain of the sample or $K_{ol}(3\ \varepsilon_{diff})/(\varepsilon_{ax} + 2\varepsilon_{rad})$, where K_{ol} is the bulk modulus of the reference (olivine), ε_{diff} is the olivine volume strain determined by diffraction, ε_{ax} is the axial strain of the entire sample determined from the images, and ε_{rad} is the radial strain from the images. This dataset yields a bulk modulus of 127 GPa. This agrees well with our expectation for this situation. We expect an uncertainty of about 10% due to the high resolution that is needed for both the images and the diffraction. Experiments at half of the amplitude of the driving pressure were considerably less well constrained.

Accuracy can be improved with larger amplitude pressure variations. Our pressurization rate was limited by the press motor and the gear box currently used for pumping the hydraulic fluid. The x-ray patterns can be improved by longer exposures (lower frequencies), but the recrystallization will still be a problem. A change in the cell design that removes rhenium from the x-ray path will help the diffraction pattern.

In summary, we report here an experimental method to measure the bulk modulus in a multi-anvil high pressure device at a low frequency. This method allows us to isolate the role of volume-changing processes like phase transition on the elastic properties from processes that soften shear moduli only. This method targets issues relevant to the deep Earth, allowing us to address questions such as whether the transition zone in the mantle should have low seismic velocity regions because of phase transitions.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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