A pressure calibration method for a portable wide-access "panoramic" cell*

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A simple and convenient pressure calibration method is developed for a newly designed portable wide-access 'panoramic' cell. This cell is adapted to angle-dispersive-mode high-pressure *in situ* neutron diffraction of reactor neutron sources. This pressure calibration method has established a relationship between the cell pressure and the anvil displacement (gasket compression) based on the fixed-point calibration technique. By employing TiZr gasket with a thickness of 3 mm and WC anvil with a culet of 4 mm diameter, the average anvil displacements are 1.31 mm and 2.22 mm for Bi phase transitions (2.55 GPa and 7.7 GPa), and 1.85 mm for Ba phase transitions (5.5 GPa), respectively. In this pressure range, the pressure increases quickly with decreasing gasket thickness, and undergoes a linear increase with the anvil displacement. By extrapolating the calibration curve, the cell pressure will achieve 10 GPa when the anvil displacement is around 2.5 mm.

Keywords: pressure calibration, neutron diffraction, phase transition

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1. Introduction

Neutrons are complementary to X-rays because they can specially probe nuclei, magnetic structure, are sensitive to light atoms, and have high penetrating power. Thus, *in situ* neutron diffraction experiments under high pressure can play a key role in obtaining structural information on various materials. A toroid-type high-pressure apparatus with a so-called 'Paris-Edinburgh (PE) press'[1] is a standard high-pressure device in this domain. It can generate pressures of up to 10 GPa on sample volumes of ~ 100 mm^3, and pressures up to 30 GPa on sample volumes of ~ 30 mm^3.[2,3] This device is adapted to time-of-flight (TOF) techniques at a fixed scattering angle, and achieves enormous success in high-pressure structural studies about the pulsed neutron sources, such as the neutron facilities ISIS in the United Kingdom or LANSCE in the United States. In contrast to the energy-dispersive mode in pulse neutron facilities used by TOF spectroscopy, diffraction patterns are usually collected with the angle-dispersive mode in continuous neutron sources, such as reactor based facilities. However, there is some inconveniences to making progress with this technique. First, the opening window for diffraction is limited by the geometry of high pressure devices, and thus it is difficult to cover the wide angle range needed for the angular dispersion method. Secondly, the strong contamination of the pattern is made by the signal from the anvils because of the need to ensure large sample volumes. More recently, to combine high pressures and large sample volumes, Xu[4,5] and his colleagues have developed a panoramic type moissanite (SiC) anvil cell (MAC) that can readily generate pressure to 50 GPa, which is significantly higher than that generated by conventional Paris–Edinburgh cells. However, its sample volume is much smaller than that of a PE cell, although the sample volumes are many orders larger in magnitude than those available in diamond anvil cells (DAC).[6] MAC is driven by hand force, the same as DAC, thereby it is convenient to bring them to the X-ray/neutron beamlines. Analogously, we have designed a portable panoramic type opposed anvil cell with a wide opening window, large sample volume, and mild higher pressure, which is adapted to be employed at reactor neutron sources for angle-dispersive mode diffraction.[7]

In general, the cell pressure of high-pressure devices can be determined by measuring the EOS of pressure markers, such as Pt, Ag, Au, NaCl, MgO, etc., via *in situ* X-ray/neutron diffraction techniques.[8] Another conventional pressure calibration is to measure the fluorescence spectra of ruby grains while applying high pressure.[9,10] This method is adapted to DAC or MAC, which profits from the fact that the fluorescence spectra can pass through the diamond or moissanite anvils. On the other hand, the pressure calibration for a large volume press generally uses the fixed-point calibration technique. This method establishes the relationship of the cell pressure and applied load (or hydraulic oil pressure) prior to the high pressure experiment by measuring the phase transition of some standard materials, such as bismuth, thallium, cesium, barium, etc. Thereby, the accurate and repeatable determination of an applied load is necessary for this method. Generally, it is difficult to measure the applied load directly for 'hand-force' type cells due to the discontinuous force applied, such as our designed

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panoramic type opposed anvil cell where pressure is generated by turning screws. In this paper, we develop a simple and convenient pressure calibration based on the fixed-point calibration technique by monitoring the anvil displacement (gasket compression) of our portable wide-access 'panoramic' cell.

2. Experimental details

2.1. Geometry of the cell

The newly developed cell designed for angle-dispersive neutron diffraction is a wide-access 'panoramic' cell. Two opposed anvils are central and aligned in a piston cylinder, as illustrated in Fig. 1. Eight screws and a lot of disc springs are used to lock-up pressure. The load is applied by turning screws, which disengage the setting from the bulky hydraulic ram and loading frames, and it is convenient to bring them to the neutron beamlines. The entire setting has a mass of 10 kg and a size of Φ 85 mm $\times$ 100 mm, which ensures that it is portable and lightweight. Two symmetrical windows, each with 135° equatorial and 68° azimuthal opening angles, are cut open at the equatorial position of the outer cylinder body. PCD (polycrystal diamond) or SiC anvils are employed to generate higher pressure, while WC (Tungsten carbide) is used to generate a slightly lower pressure range.

This cell is mainly intended for direct observation of the sample under pressure, as well as neutron diffraction. The key features of this new cell design are: to enlarge the diffraction windows to satisfy the angle-dispersive mode in the continuous neutron sources; to lock-up pressure on two opposed anvils with piston-cylinder alignment by turning screws; and, to adapt the PCD anvil to ensure both large sample volumes and high pressure.

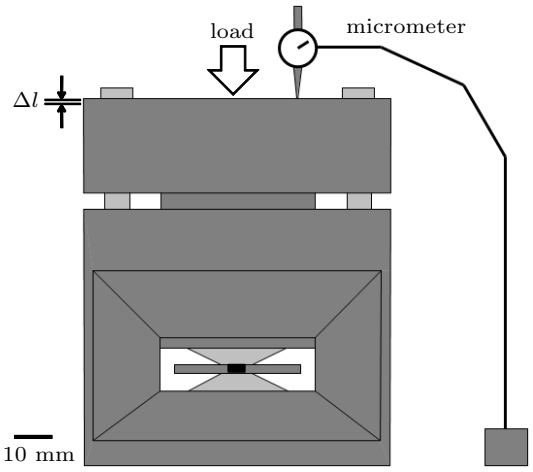


Fig. 1. Schematic drawing of the portable wide-access 'panoramic' cell. The load is applied by turning screws. A micrometer was employed to determine the anvil displacement (gasket compression) by measuring the spacing between the rear tables of cell.

2.2. New calibration method of cell pressure

The fixed-point calibration technique was originally developed by Bridgman.[11] Some materials undergoing phase transitions can be expressed by abrupt changes in electrical resistance at a certain pressure.[12] When these transitions are observed to occur, the corresponding force on the anvils is monitored and a calibration plot of cell pressure versus press load is established.

Since in our case the force is applied by screws, the direct measurement of the press load is rather laborious. After the load is applied step by step, the gasket thickness decreases from an original value of 3 mm to $\sim$ 0.5 mm. However, the measurement of anvil displacement (gasket compression) is advisable.[13] The linear relationship of applied load and squeezed displacement of gasket was proposed by Dunstan.[14] Thus, we propose a calibration method by establishing a relationship between press load and gasket compression. This calibration method has a simple procedure before performing high-pressure experiments, which can ensure the efficiency of the cell, select an optimal value of relationship between the size of anvil culets and thickness of gasket, and establish a simple empirical rule for estimating the press load by measuring the gasket compression.

Taking into account that the sub-millimeter scale of gasket compression is enough for the use of a micrometer (the measurement accuracy is 0.001 mm),[15] a micrometer is used to determine the anvil displacements in this study, which measures the spacing between the rear (supporting) tables of the cell, as shown in the Fig. 1.

2.3. Cell assembly

In this experiment, the cell pressures were calibrated by using the resistance measurement method with Bi and Ba. This method is followed by a careful assembling. The WC anvil, which has a cone-shaped flat culet of Φ 4 mm, is employed in this study. The anvil is cut with 10° slope and is pushed into the steel reinforcement ring. Baked (700 °C, 30 min) natural pyrophyllite is used as gasket, measuring initially 3 mm in thickness. A null-scattering Ti/Zr alloy (67.7 mol% Ti, 32.2 mol% Zr) is located around the pyrophyllite as an additional support. We used copper foil (0.01 mm thickness) as electrodes for the electrical connection and used electrical tape as an insulator to separate the copper foil from the Ti/Zr alloy. The details for the connection of Bi and Ba wires, and copper foils are shown in Fig. 2. During the experiments, we simultaneously recorded the anvil displacement and the resistances of Bi and Ba, while applying the load step by step.

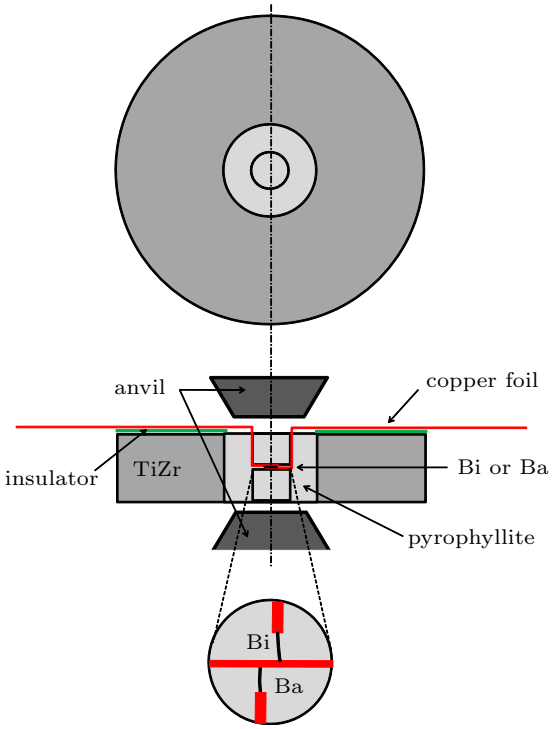


Fig. 2. (color online) Sample assembly for Bi and Ba resistance measurement.

3. Pressure-induced phase transition of ZrW_2O_8

Upon applying pressure at room temperature, the cubic α-ZrW_2O_8 transforms into a quenchable orthorhombic phase λ-ZrW_2O_8. This phase transition takes place at a pressure of 0.2 GPa.[16] As the pressure further increases to 1.5–3.5 GPa, λ-ZrW_2O_8 gradually becomes amorphous, and amorphous ZrW_2O_8 is also irreversible upon decompression.[17]

An additional experiment is conducted with α-ZrW_2O_8 (Alfa, 99.7%, −200 mesh) used as starting material. A 3-mm-thickness Ti/Zr alloy gasket with a Φ 2 mm hole for assembling sample, and the WC anvil with Φ 4 mm culet were used as described in the previous section. Four experiments were performed, with the anvil displacement being 0.5 mm, 0.85 mm, 1.0 mm, and 1.5 mm, respectively. The recovered samples were characterized by X-ray diffraction (Philips X'pert diffractometer).

4. Results and discussion

The electrical resistances of Bi and Ba, and the anvil displacements have been simultaneously measured as the load was applied step by step. We repeated the measurements three times with the same anvil and the same size of gasket. The results are shown in Table 1. Two discontinuities of Bi resistance were observed at 1.314(12) mm and 2.219(8) mm of anvil displacements, which correspond to I–II (2.55 GPa) and III–V (7.7 GPa) phase transitions, respectively, and one abrupt change at 1.850(9) mm for Ba was observed, which corresponds to I–II (5.5 GPa) phase transitions.[18] The changing trends of the resistance of Bi and Ba are consistent with previous reports.[19] Figure 3 is a typical measurement of resistance changes of Bi and Ba as a function of the anvil displacement. The anvil displacement Δl corresponds to the compression of the gasket. Zero of Δl implies the contact position of the anvils against the initial gasket, and a maximum Δl of 3.00 mm for 3.00-mm-thickness gasket was used in this experiment.

Table 1. Measurement results of anvil displacements.

	Anvil displacements/mm			Average	Error
	1	2	3		
Bi I–II (2.55 GPa)	1.315	1.326	1.302	1.314	0.012
Ba I–II (5.5 GPa)	1.850	1.842	1.859	1.850	0.009
Bi III–V (7.7 GPa)	2.221	2.210	2.226	2.219	0.008

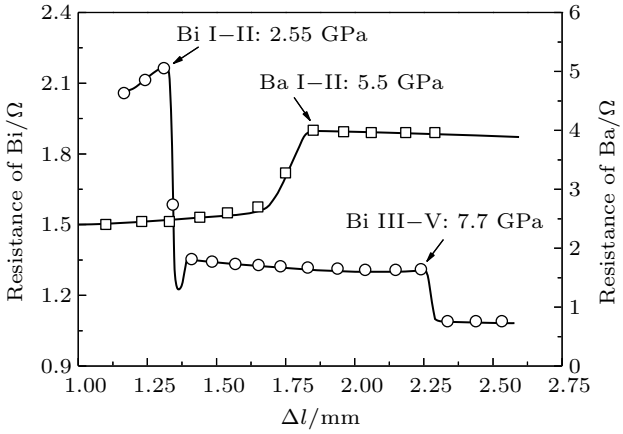


Fig. 3. Resistance changes of Bi (circle) and Ba (square) as a function of the anvil displacement. The lines are used as guides for the eye.

Figure 4 shows the XRD patterns of the recovered ZrW_2O_8 products. Figures 4(a)–4(d) show different applied loads, corresponding to various anvil displacements (Δl) of 0.50 mm, 0.85 mm, 1.00 mm, and 1.50 mm, respectively. As we know, the pressure of α-ZrW_2O_8 transforming to λ-ZrW_2O_8 starts from 0.2 GPa, and then gradually becomes amorphous from 1.5 to 3.5 GPa. Only α-ZrW_2O_8 peaks were observed when Δl is at 0.50 mm, indicating that the cell pressure is below 0.2 GPa at this load (anvil displacement) (Fig. 4(a)). When the anvil displacements are 0.85 and 1.0 mm, intense peaks of λ-ZrW_2O_8 appear, as shown in Figs. 4(b) and 4(c). This indicates that the pressure is between 0.2 GPa and 1.5 GPa at these loads. The percentage of λ-ZrW_2O_8 in Fig. 4(c) obviously increases compared with Fig. 4(b), which should be attributed to the increase of the pressure. When Δl is at 1.5 mm, the peaks of α-ZrW_2O_8 disappear completely and the peaks of λ-ZrW_2O_8 become broad and considerably decrease its intensity, suggesting that the cell pressure reaches up to 3.5 GPa or above.

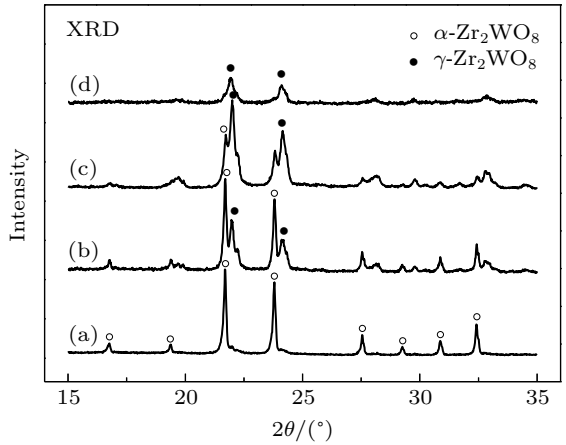


Fig. 4. XRD patterns of recovered ZrW_2O_8 products from different anvil displacements: (a) 0.50 mm, (b) 0.85 mm, (c) 1.00 mm, and (d) 1.50 mm, respectively.

The pressure calibration curves can be obtained based on phase transition of Bi and Ba, as shown in Fig. 5. The result shows that the pressure depends linearly on the anvil displacement, and it has a positive slope ($\sim$ 5.7 GPa/mm). At a lower pressure range (0–1.5 GPa), the pressure increases slowly before the anvil displacement reaches 1.0 mm. Furthermore, the result shows that an anvil displacement is required before any pressure is generated. According to the XRD results of recovered ZrW_2O_8, until the displacement achieves 0.5 mm, the pressure is still below 0.2 GPa. On the basis of an extrapolation of the curve, at an anvil displacement of about 2.5 mm, the pressure can achieve about 10 GPa. However, further squeezing the gasket does not produce a significant increase in the cell pressure.

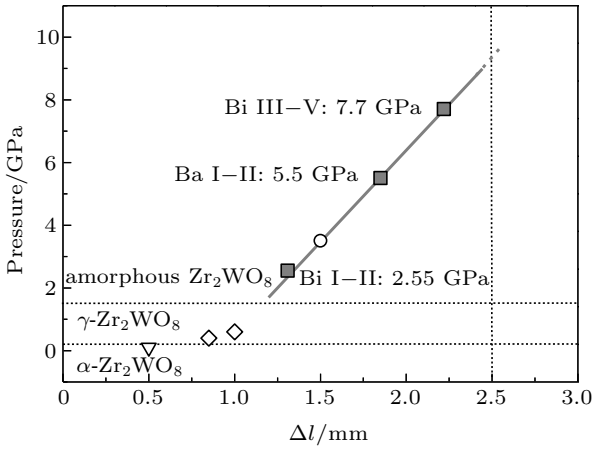


Fig. 5. Pressure calibration curves of cell pressure versus anvil displacement. The solid squares are determined anvil displacements of Bi and Ba by resistance measurement. The opened circle, opened diamond, and opened triangle are amorphous ZrW_2O_8, λ-ZrW_2O_8, and α-ZrW_2O_8, respectively.

According to the operation theory of the gasket in the diamond anvil cell (DAC) by Dunstan,[14], the hydrostatic pressure can be estimated by the following relation:

$$\sigma_{\rm n} = nk + \frac{2k}{\sqrt{3}}\left(\frac{r_0}{t} - \frac{r_{\rm g}}{t}\right),$$

where σ_n is the normal stress on the gasket surface, k is the yield strength of the gasket material, r_0, $r_{\rm g}$, and t are the sample hole radius, anvil culet radius, and gasket thickness, respectively. The hydrostatic pressure is equal to $\sigma_{\rm n}$, if the cell pressure is assumed to be in a hydrostatic condition. Thus, the plateau in pressure generating is most likely related to the gasket aspect ratio, anvil culet diameter, and the yield strength of the gasket. For an anvil displacement below about 1.0 mm, the pressure generating is slow because of some sample leaking that occurs at the beginning of the compression. In order to avoid this, the anvil faces against the gasket should be tightly sealed; namely, prepared preindentation gasket is generally necessary.[14] When the anvil displacement is above 1.0 mm, the pressure increases quickly with the decrease in gasket thickness, and then increases linearly as a function of anvil displacement. In this pressure range, the gasket works in the thick gasket regime, which means that the surface layers of the gasket remain stationary on the anvil face due to the fact that the friction between the gasket and anvil exceeds k (yield strength of the gasket). On the other hand, outside this pressure range, slipping between the gasket and the anvil prevails and the gasket undergoes an outward plastic extrusion and, consequently, the increase in pressure was never observed. In this case, the gasket is in an unstable regime and further squeezing does not induce a pronounced pressure increase.[20] This means that a plateau in pressure generating occurs.

5. Conclusion

In order to adapt to angle-dispersive diffraction in reactor neutron facilities, we designed a panoramic type opposed anvil cell with wide opening window. This portable device can generate and lock the pressure by turning screws, which is convenient way to bring it to neutron beamlines. WC (tungsten carbide), PCD (polycrystal diamond), and SiC can be employed as anvil material depending on the highest pressure needed. Because it is difficult to measure the applied hand force (turning screws by hand) directly, we develop a simple and convenient pressure calibration based on the fixed-point calibration technique. This method can determine the pressure by monitoring the anvil displacement. In this study, we observe the abrupt changes of resistance of Bi and Ba at the anvil displacement of 1.31 mm (Bi I–II), 2.22 mm (Bi III–V), and 1.85 mm (Ba I–II), respectively, by using 3-mm-thickness gasket with a 2mm diameter hole and WC anvil with a culet diameter of 4 mm. According to XRD result of the recovered ZrW_2O_8 samples, the pressure increased slowly with increasing anvil displacement up to around 1.0 mm. When the anvil displacement is above 1.0 mm, the pressure increases quickly with the decrease in gasket thickness, and undergoes a linear increase as a function of the anvil displacement due to the fact that the gasket works in the thick gasket regime, as explained

by Dunstan[14] and Bonetti.[20] On the basis of extrapolation of the curve, with a displacement at about 2.5 mm, the cell pressure can achieve around 10 GPa. However, a plateau in pressure generating will occur because further squeezing does not induce a pronounced pressure increase, although the plateau pressure was not determined in this work.

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