Slotted carbide anvils: improved X-ray access for synchrotron-based multi-anvil experiments.

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Abstract

We present a new technique to allow enhanced X-ray access to synchrotron-based multi-anvil experiments with tungsten carbide anvils. Slots of 2 mm width and 1 mm depth are cut into the four anvils which are in the beam path along their <110> diagonals and these slots are filled with baked pyrophyllite to produce a region in the anvils which is X-ray transparent. We have tested this arrangement using a standard 10/5 cell assembly and the X-ray window remains open, allowing imaging of a 2.5 by 2 mm section of the cell to at least 20 GPa. There is no evidence that the slots reduce the life of the anvils and the recovered slotted anvils have been used in subsequent experiments.

Introduction

Pressures from a few, to several tens of, gigapascals are important for a wide range of studies in the Earth, physical and material sciences and are readily achieved in large-volume multi-anvil press devices. Synchrotron-based multi-anvil techniques have become routine for many studies, such as viscosity measurements of inviscid and stiff materials [1,2], phase relations[3], structure of melts[4], P-V-T equations of state[5,6,7], and thermal conductivity[8,9], where physical properties must be determined *in situ*. Diffraction-based studies gain optical access to the sample by passing X-rays through the gasket region between the tungsten carbide anvils which compress the sample, however this limits the diffraction vector to a small proportion of the Debye ring and many imaging applications require larger X-ray windows than is available through the gasket region. Currently, studies which require collection of full Debye rings or images of large volumes of the high-pressure environment require costly X-ray transparent anvils made of cBN or sintered diamond. This cost, along with the high-pressure manufacturing process required to make these anvils, limits the size of cubic boron nitride (cBN) and diamond anvils to 14mm, compared to 26 or 32 mm edge length tungsten carbide anvils often used in multi-anvil experiments. In addition to their high cost, ultra-hard diamond and cBN anvils have a relatively low tensile strength, limiting their use to multi-anvil press designs which maintain a true cubic geometry during compression, such as the DIA-type. The small size of the anvils additionally limits the maximum applied load to ~700 tons, requiring that very small pressure cells (typically 7 or 8 mm octahedra compressed by anvils with 2 mm truncations) are used to generate pressures of 20 GPa or above. This compares badly with off-line experiments where tungsten carbide anvils are used and 10 mm octahedra with 4 mm truncations can generate pressures of 25 GPa, allowing samples up to 10 times the volume of synchrotron experiments (with very large presses, samples of ~10 mm^3 or more can be compressed to the $MgSiO_3$ perovskite stability field, 23.5 GPa[10]). It would therefore be useful if X-ray transparent windows could be inserted into standard tungsten carbide anvils; here we describe, and present initial results from, such a system.

X-ray Transparent Window

In order to perform radiographic observations of samples which are larger than the width of the gasket, the part of the tungsten carbide anvil which is in the X-ray beam path must be replaced by an X-ray transparent material. Ideally, 4 anvils are thus treated since the X-rays pass between two anvils upstream of the sample and a second pair of anvils downstream of the sample. We have ground slots, 1 mm deep and 2 mm wide, along the <110> direction in one face each of four anvils and filled this slot with X-ray transparent material (Fig. 1). The window material consisted of baked pyrophyllite (heated to 1000 °C for 2 hours and slowly cooled) which was cemented into the anvil slots using alumina-based cement ('Feuerfestkitt' from Haldenwanger GmbH). In addition to being inexpensive and easy to machine, baked pyrophyllite has the added advantage that it has a similar X-ray absorption coefficient to the soft pyrophyllite gasket material commonly used in multi-anvil experiments. This reduces the complexity of radiographic images collected using X-ray windows. A similar technique has been used in DIA-type multi-anvil experiments for imaging at lower pressures (up to ~5 GPa, ref. 7) but here we demonstrate that the technique can work to pressures of at least 20 GPa in 6/8 geometries.

High-Pressure Test.

High-pressure synchrotron experiments were performed on a 1700 ton DIA-type multi-anvil press installed on beamline W2 at DORIS, Hasylab. We used monochromatic 100 keV X-rays for the imaging experiment reported here. Tungsten carbide anvils from Hawedia with 32 mm edge-length and 5 mm truncations compressed 10 mm edge-length chromium-doped MgO octahedra. The four anvils with X-ray windows were placed such that they would provide a 2 mm by 2 mm opening even if the anvils were to come into contact with each other. The four anvils not in the X-ray beam were normal WC anvils without any slots cut into them. The sample consisted of a cold-pressed pellet of ZnS, 0.8 mm in diameter and 2 mm long filling a cylinder of polycrystalline corundum with an outer diameter of 1.6 mm. The insulator-metal transition in ZnS at 15.6 GPa was used for pressure calibration. Aluminium rods with 1.6 mm diameter provided electrical contacts between the sample and two of the normal anvils. Thin foils of platinum placed between the aluminium rods and ZnS sample acted as markers of the length of the sample for the radiographic observations (Fig. 2). Radiographic images were collected by placing a florescent YAG crystal in the X-ray beam downstream from the high-pressure cell. A Leica DFC350FX microscope camera combined with long-focal length lens was used to capture the fluorescence image on the YAG crystal. Experiments were performed by slow compression at room temperature (taking about 3 hours to reach the maximum press load of 400 bars), during which the resistance of the ZnS was constantly monitored and radiographic images collected once per minute. Once the insulator-metal transition had been observed the sample was slowly decompressed (over ~ 9 hours) and the anvils containing the X-ray windows inspected for damage.

Results

Figure 2 shows radiographic images of the sample assembly collected at a press load of 20 and 400 bars; based on linear extrapolation of the ZnS transition (at 300 bars end load) these loads correspond to approximately 1 and 21 GPa sample pressure; this pressure efficiency is identical to when normal carbide anvils are used. The gap between the anvils reduces from 1.5 mm at 1 GPa to 0.5 mm at 20 GPa, meaning that if there were no X-ray windows cut into the anvils approximately half of the ZnS sample would have been obscured by the anvils. However, the anvil slots remain open providing a 2 mm by 2.5 mm X-ray window even at 20 GPa. On recovery one slotted anvil and one normal anvil were found to be broken; this is a normal breakage rate for tungsten carbide anvils with 5 mm truncations which have been taken to 20 GPa. The other three slotted anvils showed no signs of damage (Fig. 1), suggesting that the slots do not significantly weaken tungsten carbide anvils. We have subsequently re-used the slotted anvils for further experiments with no adverse effects.

Discussion

The present experiment demonstrates that X-ray windows can be successfully cut into standard tungsten carbide anvils for synchrotron-based multi-anvil experiments. This has several advantages over traditional sintered diamond anvils for imaging large volumes. Firstly, the press load which can be applied to sintered diamond anvils is limited by the small size of the anvils. For a DIA-geometry 14 mm diamond anvils are limited to about 700 tons before the steel wedges used to compress the anvils will plastically deform. This limits the anvil truncation size to 2 mm for 25 GPa pressure generation. The most commonly used sizes of tungsten carbide anvils (26 mm or 32 mm) have 3.4 and 5.2 times larger surface area respectively, allowing concomitantly larger press loads. This in turn allows for larger truncation sizes and hence larger samples for a given target pressure. Second, tungsten carbide conducts electricity, allowing the anvil truncations to be used as electrodes. In the present study this allowed the resistance measurement of the ZnS sample. Third, there is a significant cost saving in replacing sintered diamond with tungsten carbide. Even factoring in the larger size of tungsten carbide anvils they are approximately ten times cheaper than sintered diamond or cBN anvils. In addition the increased toughness of carbide compared to diamond or cBN means that they tend to have a longer experimental life, further increasing the saving.

While the maximum pressure achievable with tungsten carbide is limited to around 30 GPa by the strength of the carbide this pressure range is sufficient for many studies, particularly in the Earth and material sciences. The present study used baked pyrophyllite to fill the anvil slots in order to minimise the absorption contrast between this and the pyrophyllite gaskets. The slot material, however, could be replaced by low atomic mass amorphous materials, such as the boron-epoxy mixtures commonly used for pressure media in DIA-type experiments, in order to reduce the absorption and the structure of the background for applications requiring greater X-ray transmission and for diffraction studies. A further improvement for diffraction studies would be to taper the slots in the region behind the inserts to enable collection of full Debye rings during diffraction experiments using tungsten carbide anvils, as shown in figure 3. At higher pressures than in the present experiment it might be necessary to replace the inserts with a harder material such as sintered diamond or cBN. In such a case, the inserts are best prepared as segments from cylinders such that they provide support to the slots in the carbide: we are currently testing one millimetre deep segments cut from three millimetre diameter and ten millimetre long cylinders. Such cylindrical sintered diamond segments are relatively inexpensive (<$100 each) meaning that they would not add significantly to the cost of a synchrotron-based multi-anvil study.

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Figure Captions

Figure 1.

Photographs of four anvils prepared for the radiographic experiment (a) and recovered from the experiment (b). About one centimetre in length of slots are filled with baked pyrophyllite, corresponding to the final distance to which the gaskets extrude during the experiment. The top two anvils which are in the X-ray beam path are also slotted in a similar manner (not shown here). The scale bar is in millimetres.

Figure 2.

X-radiographic images collected using slotted anvils. (a) at 1 GPa the gasket opening is still ~2 mm so the total X-ray window is 2 by 4 mm. The diagonal dark lines in the centre of the image are the Pt marker foils placed at the ends of the ZnS sample. (b) a sketch of the cell components overlain on the 1 GPa image. (c) at 20 GPa, the gaskets have compressed to about 0.5 mm thickness. If there were no slots the anvils would partially obscure the sample at this stage but the slots maintain a 2 by 2.5 mm X-ray window through the centre of the cell.

Figure 3.

Schematic sketch of anvil with slot and cone cut for diffraction studies. The solid angle of diffraction is limited by the distance of the back of the slot from the diffracting volume. It is therefore desirable for the cone to extend as far across the cube face as possible without it reaching the region of gasket extrusion. In the current experiments the effective slot length could have been reduced from 50 mm to 20 mm.