Miniature cryogenic diamond-anvil high-pressure cell

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A novel diamond-anvil cell is described which measures 19-mm diameter by 28 mm long to fit the Oxford Instruments CF 1204 flow cryostat, while providing full axial and tilt adjustment of the diamonds. The drive mechanism provides for pressure changes while the cell is in the cryostat. The small dimensions are achieved because the drive force is not transmitted through the tilt mechanism, and by the Bowden cable technique used for the drive mechanism. This cell makes high pressure a readily portable technique that can be brought to many already existing experimental apparatuses.

INTRODUCTION

Diamond-anvil high-pressure cells (DAC) are a recent and very successful application of the Bridgman technique of generating high pressures. A weak material, in a gap of large aspect ratio (length to thickness) between hard anvils, will sustain pressures far in excess of its own strength.[1] Thus, in a typical DAC, a steel gasket (shear strength ~ 8 kbar) of thickness $\sim 100\,\mu$m is squeezed between diamond anvil surfaces ~ 700-μm diameter to achieve pressures up to a few hundred kbar in a sample hole provided in the center of the gasket. For a review of DAC work, see Ref. 2.

The sample space in a DAC is, of necessity, very small. To achieve a pressure on the back surface of the diamond sustainable by other materials (Be:Cu, tool steel, or tungsten carbide), the diamond anvils typically have a culet ~ 500 to 700-μm diameter for a back surface ~ 3 to 4-mm diameter, and this allows the sample hole in the gasket to be $\sim 200\,\mu$m and the sample itself to be $\sim 100\,\mu$m square (to allow for shrinkage of the hole on initial compression). Nevertheless, DACs are usually large, typically several cm in the smallest outside dimension. For cryogenic use, this requires a large purpose-built cryostat. Another problem is that, for pressures of some 100 kbar, a force of up to 1 t (10 kN) has to be applied to the DAC, and the transmission of such forces down a cryostat or their generation at the cold end has been achieved only with bulky drive mechanisms or in clamp cells where the pressure changes require removal of the cell from the cryostat.

Here, we report a novel design of DAC and of a drive mechanism in which both these problems have been overcome, so that the cell can be used in a standard flow cryostat (Oxford Instruments CF1204) or even immersed in a helium storage Dewar, with the pressure changes made while the cell is in the cryostat.

I. DESIGN AND CONSTRUCTION
A. The miniature diamond anvil cell

The cell is shown in Fig. 1. It has a piston–cylinder support for the upper diamond, while the lower diamond is mounted on the anvil plate which gives X–Y adjustment through three screws. The anvil plate is supported by the bottom plate which has tilt alignment with respect to the body by means of three standoff set screws and three secur-

ing cap screws. The force is transmitted to the piston through the top plate.

Two prototypes have been built. EN24 tool steel was used for one; only the anvil plate and piston are hardened. This cell was built by Diacell Products, and, using standard Drukker diamonds with 750-μm culets, has been taken to 110 kbar without problems (see Sec. II). The other cell, built in our own workshops from Cu:Be hardened throughout to 37 Rockwell C, is being further developed for electrical measurements. It has been taken to 50 kbar with 1-mm culets at a force of 8 kN. These pressures are determined by the work we are doing in the cells (optical studies of semiconductors). With smaller diamond culets, much higher pressures should be achievable.

The principle reason for the large dimensions of conventional cells is the necessity of careful alignment of the diamond culets, both in tilt and radially. Some small clamp cells
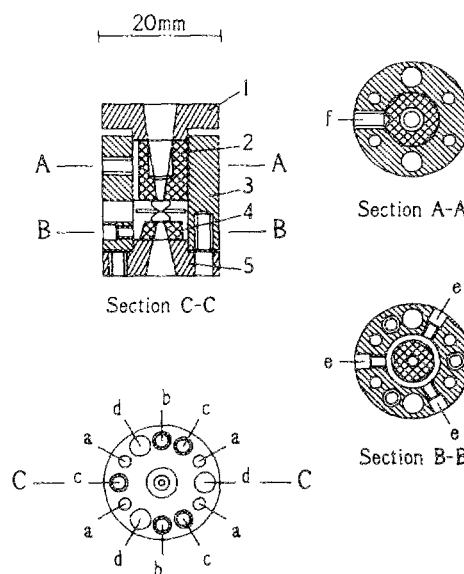

FIG. 1. The miniature cryogenic diamond anvil cell. The principal components marked on the vertical section C–C are (1) top plate, (2) piston, (3) body, (4) anvil plate, and (5) bottom plate. The holes marked in the bottom projection are for (a) the tension wires, (b) the clamp bolts, (c) the standoff screws, and (d) the securing screws. The holes marked in the transverse sections are for (e) the X–Y adjustment and anvil plate securing screws and (f) the piston guide screw.

have been reported in which this constraint has been avoided by working at relatively low pressures ($\lesssim 50$ kbar) without provision for alignment.[3] However, where tilt alignment mechanisms have been used, they have always been built sufficiently sturdy to take the full load applied to the diamonds without loss of alignment accuracy. Where the load is applied through the adjustment screws, relatively large screws are required. On the other hand, where the load is carried by hemispherical or opposed cylindrical rockers, the mechanical complexity including setting screws again necessitates large dimensions. We have solved this problem by not transmitting the load through the tilt adjustment mechanism (Fig. 1). In our cell, the load is applied to the top and bottom plates, which bear directly on the diamonds (through the piston and axial adjustment plate, respectively). The body of the cell is aligned with respect to the bottom plate for tilt, but the load is not transmitted through it.

In a reasonably well-made DAC, the piston–cylinder clearance is typically 10–20 μm. This clearance, for a piston length of 10 mm, implies a tilt, when load is applied, of 10^{-3} rad, or 2–3 fringes across the culet. It should be emphasized that this amount of tilt is both inescapable, short of making the piston an interference fit in the cylinder, and also hard to exceed since the transverse or symmetry-breaking forces are also $\sim 10^{-3}$ times the applied load. This consideration dictates our choice of un undersquare piston, 10 mm long in a bore of 9 mm, to give little more tilt than many conventional designs with oversquare pistons perhaps 15 mm long by 20 mm diameter.

It is appropriate to consider two common misconceptions that have perhaps unduly influenced DAC designs in the past. Both are concerned with the problem of maintaining the alignment of the diamonds under load; that is, with the stiffness of the DAC. The first misconception is that larger components imply a stiffer assembly. This is not so, for moments also increase with size. Thus, a beam will carry the same load, for the same angular deflection, if its length and width are scaled together. In consequence, any DAC design could be scaled down without effect on stiffness-limited angular misalignments of the diamonds. In particular, our cell does not suffer from its small body dimensions because lengths have been reduced in proportion to thicknesses. The second misconception is that hard or strong metals are stiffer than weak or soft metals. In general, this is not so. The Young's and bulk moduli of a given metal are almost independent of its state of hardness (and not very dependent on which metal it is). This is because the moduli depend on the interatomic force laws, while the hardness, tensile strength, etc., depend on dislocations and how they are pinned. Similarly, in a macroscopic component, the ultimate strength depends on the weakest cross section but the stiffness generally does not. Thus, although the body of our DAC is full of holes, it remains stiff, and apart from considerations of piston–cylinder wear could be made of brass instead of Cu:Be or tool steel without disadvantage, provided the diamonds were still supported on hardened material.

B. Drive mechanism

While the cell is provided with two clamp bolts, these are primarily for convenience in loading the cell. It is desirable to be able to change the pressure while the cell is in the cryostat, and so to transmit the load ($\lesssim 1$ t) down the cryostat or to generate it at the cold end. Previous cells have used either push rods of large cross sections ($\sim$ cm^2) or complicated and bulky screw, wedge, or lever mechanisms to achieve this. We transmit the full force down the cryostat in a relatively lightweight structure by using the principle of the Bowden cable, in which the tension member (the wire) supports, continuously along its length, the compression member (the tube) against crippling (Fig. 2). In this way, the components can be made as thin as their ultimate strength allows, without consideration of length-to-diameter ratios, and the assembly can be curved if necessary (Fig. 2). The tube and wire assembly is driven from outside the cryostat by a hydraulic cylinder (PowRlock Miniram 5100A).

For reasons of ready commercial availability, we used 2-mm stainless-steel wire in $\frac{3}{16}$ in. 16SWG (2.7-mm i.d., 4.7-mm o.d.) stainless-steel tube. A closer fit would be preferable to reduce any tendency of the assembly to bend under load; however, these dimensions have proved acceptable (see Sec. II). To avoid bulky fixings at the cell, the wires run down the cryostat, around the cell, and back up the cryostat. (We thank Dr. K. P. Homewood for this idea). Saddles are provided at the bottom of the cell to avoid sharp bends in the wire; these may be semicircular or relatively flat according to the space available. The securing of the wires at the hydraulic ram is dictated by the need to avoid, on the one hand, any increase in diameter (so that the wires can be disassembled from the tubes and the cell) and, on the other hand, any reduction in core diameter such as screw threading (so that
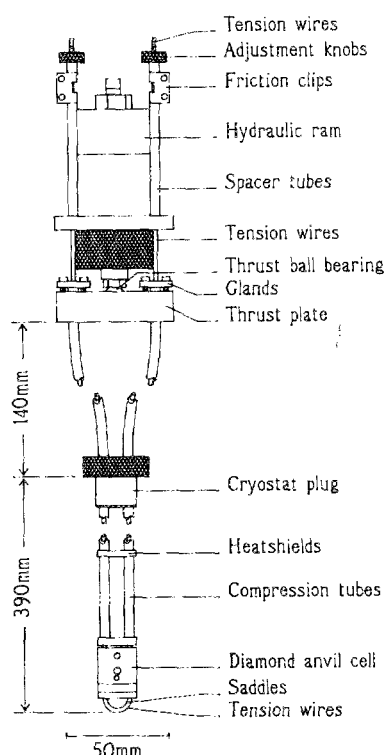
FIG. 2. The drive mechanism.

Tension wires
Adjustment knobs
Friction clips
Hydraulic ram
Spacer tubes
Tension wires
Thrust ball bearing
Glands
Thrust plate

140mm

390mm

Cryostat plug
Heatshields
Compression tubes
Diamond anvil cell
Saddles
Tension wires

50mm

the full strength of the wire may be used). For these reasons, a friction clip is used (Fig. 2). Above the friction clips, the wires are threaded so that the positions of the clips can be controlled before tightening them down. In use, the adjustment knobs are run back 1–2 mm so that any slippage of a clip under load is easy to detect.

The collective cross section of the wires is 4π mm^2, giving a total force of ~ 10 kN for a UTS ~ 1 GPa; the friction clips sustain the maximum force generated by the ram (~ 8 kN) without failure or slippage. A steel ball is interposed between the ram and the thrust plate to ensure that the loading of the four wires is symmetrical. At high loads, the wires also stretch elastically 2–3 mm, and this also ensures equal loading of the four wires.

Glands are provided where the wires enter the thrust plate, and the tubes are also soldered into the plate to provide a seal. This is necessary to prevent air from being drawn into the cryostat.

II. OPERATION

It might be thought that the small size of the DAC would make alignment and loading the cell a more delicate operation. This is not so, because the diamonds remain the conventional size. It is only necessary to note that loading of the sample and gasket are carried out on the upper (piston) diamond, since the bore size and the interdependence of the axial and tilt adjustments make the lower diamond relatively inaccessible. For use with alcohol (4:1 methanol:ethanol), our loading sequence is as follows. Once the cell is correctly aligned, a 0.5-mm stainless-steel gasket is indented to ~ 100-μm thickness, and the 200 to 300-μm-diam sample hole is drilled in the center of the indentation. The sample and the calibration ruby, both cut from material thinned to $\sim 30\,\mu$m, are placed in the center of the upper (piston) diamond and the gasket is lowered over the diamond. Gasket guide pins have been installed in the Cu–Be cell to facilitate this operation. Viewing the sample through the gasket hole as the gasket is lowered enables the position of the sample on the diamond to be checked. The gasket may be secured using Plasticine pillars in the steel cell without guide pins. It is finally settled into position by assembling the piston into the cell and applying finger pressure. The piston is then removed and a drop of alcohol mixture is placed over the indentation with a hypodermic syringe. After checking visually for the absence of bubbles in the sample space, the body of the cell is brought down over the piston. With finger pressure applied to the piston, the cell can now be inverted so that the top plate and clamp bolts can be installed. The clamp bolts are tightened until some yield is felt; at this stage the pressure is in the range of 5–25 kbar. The drive mechanism can now be assembled around the cell and, if desired, the clamp bolts can be slackened or withdrawn completely. The cell is now ready for insertion in the cryostat.

The flow cryostat operates in any orientation. We place it on its side to provide a horizontal beam axis through the end window. Because optical access in the cryostat is limited to the end window, laser light cannot be seen coming through the gasket hole and out of the top end of the cell.

This makes optical alignment very much harder. The most satisfactory solution in a photoluminescence or similar experiment would be to have optics outside the cryostat that would focus the laser beam, collect the emitted light, and in addition, provide microscopic viewing of the focal plane.[4] Not having such a system, we find that a ruby chip secured in the tapered bore of the bottom diamond pad (anvil plate) provides a very useful reference. This chip is about 200–300 μm across, big enough to see easily with the naked eye. It is then easy to align the laser and detection optics onto it, after which a known translation brings the sample or cell ruby close to accurate alignment. This reference ruby also provides a 0-kbar reference throughout the experiment; although a neon lamp is used for calibration, this reference is useful since the ruby emission shifts with temperature.

We have not yet tried to load the cell with argon in place of alcohol. It is not, however, anticipated that the design of this cell will present any special problems in argon loading. Using alcohol, the cell can be warmed to room temperature for pressure changes in the cryostat without loss of optical alignment (the signal is never lost completely, although it is usually necessary to shift the focusing lens slightly to recover the maximum). If uniaxial stress is not a problem, the pressure can be changed cold. We find that the shifts in position of the cell with respect to the optics are about 50 μm over the full temperature and pressure ranges.

In Fig. 3, a plot is shown of the cell pressure against the hydraulic drive pressure during a typical experiment. The clamp bolts alone gave 21 kbar; after the hydraulic drive took over the load, the pressure–pressure relationship was nearly linear, at about 0.1 kbar/psi. Note that the cell pressure changes by up to 10 kbar as the cell is cooled to 80 K; the reason for this is not understood. In this run, the clamp bolts were not slackened, resulting in a remarkably slow pressure
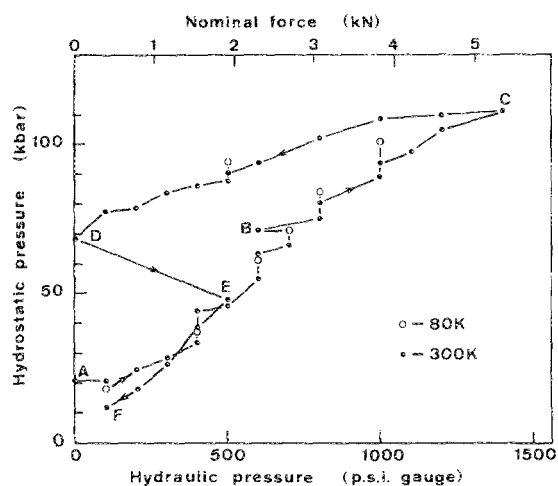
FIG. 3. The pressure in the cell is plotted against hydraulic drive pressure and nominal force for a typical run. 700-μm diamond culets were used, with a stainless-steel gasket preindented to a 100- and a 250-μm sample hole. A: The initial pressure with clamp bolts only was 21 kbar. B: Overnight cooling of the laboratory led to some loss of hydraulic pressure. C–D: The clamp bolts retained the pressure despite removal of the hydraulic load. D–E: 500 psi was applied and the clamp bolts removed. E–F: Note the close matching of the loading and unloading pressures.

decrease on removal of the load. However, on reapplying load and removing the clamp bolts, the descending pressure–pressure curve then follows the ascending curve fairly closely.

III. FURTHER DEVELOPMENT

In this section we consider developments of the present cell and further possible reduction in size of the DAC. If necessary, the diameter could easily be reduced to 10–12 mm. The piston need not be larger than 4–5 mm, and if the alignment screws were made 10-12 BA (say, 1.5–1 mm), the body wall thickness could be reduced to ~2–3 mm without undue loss of stiffness.

However, it would be much more useful to reduce the length rather than the diameter of the cell, so that it could be mounted across the cryostat diameter. Here, we are restricted by the length of a pair of diamonds (3–4 mm) and the length of the piston (to limit tilt error, any reduction in length below ~10 mm would require a concomitant increase in the accuracy of the piston–cylinder fit). Add the support and drive plates, and it is hard to see how a cell complete with alignment and drive mechanisms could be made sufficiently shorter than the 28 mm of the cell presented here, so as to fit, for example, across the bore of a 2-cm cryostat. We conclude that further useful miniaturization of the DAC would require a radical departure from present technology.

Development of the present cell lies in the direction of improved convenience of operation, in particular, argon loading to allow pressure changes at low temperature, and modifications to allow the demounting of the cell from the drive mechanism without total disassembly of the latter. Modifications to facilitate electrical feedthroughs are also in hand. These will be reported at a later time.

ACKNOWLEDGMENTS

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