Cryogenic Loading of Argon Pressure Medium in Diamond Anvil High Pressure Cells with in-situ Pressure Determination

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A versatile system for cryogenic loading of argon pressure medium into the sample space of a diamond anvil cell has been developed. The system has been designed such that, with suitable adaptors, a wide range of diamond anvil cell designs can be pressurized. The pressure in the cell can be monitored during pressurization using the ruby fluorescence method via optical fiber access into the loading chamber. This enables precise and accurate setting of the loading pressure in the cell.

I. INTRODUCTION

The use of high pressure methods to investigate material properties is increasing and a wide range of methods can now be employed at high pressure including optical measurements (such as Raman spectroscopy), neutron and x-ray measurements, ac-susceptibility, Mossbauer spectroscopy, heat capacity measurements, etc. To reach the highest pressures, cells utilizing opposing anvils, such as diamond, must be used. A wide range of diamond anvil cell (DAC) designs are available, but they all have the same basic principle of operation as shown in figure 1.

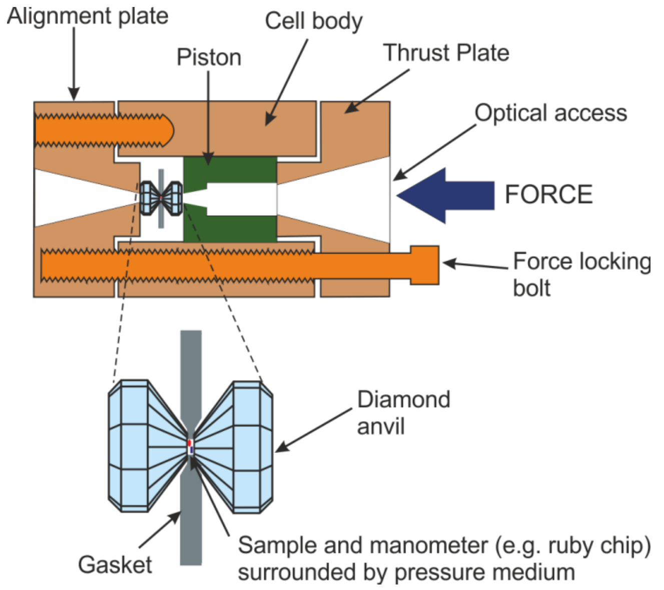

FIG. 1. (Color online) Basic principle of operation of a diamond anvil cell. The sample, manometer and pressure transmitting medium are contained in a small hole (<0.5 mm) in a metal gasket held between two diamond anvils. When force is applied to the cell the anvils are brought together so causing the gasket to plastically flow sealing the contents of the gasket hole between the diamonds and increasing the pressure on the sample.

A typical diamond anvil cell consists of two diamond anvils. One is fixed and the other is free to move with the application of an applied external force. The anvils are aligned such that they are centered and parallel to each other. The pressure is applied to the sample by making use of a gasketed sample chamber between the two anvils. The gasket typically consists of a thin piece of metal with a small sample hole drilled in the center. Inside the sample hole is placed the sample, manometer and pressure transmitting medium. To pressurize the cell, force is applied to the cell such that one diamond anvil is advanced towards the other. The diamonds then press on the gasket causing the sample hole to contract inwards. This causes pressure to build up in the pressure transmitting medium which pressurizes the sample. If the correct pressure transmitting medium is used it is possible to obtain hydrostatic conditions in the cell.

A number of materials have been used as pressure transmitting media in pressure cells[1]. Noble gases make particularly good pressure media as they are typically inert to the sample. If glues or epoxies are used in the sample chamber, such as for wire contacts to samples or for gasket insulation, noble gases are typically inert to those too. They also have good hydrostaticity. Argon, for example, has good hydrostaticity to at least ~10 GPa[1]. Being a gas at ambient temperature and pressure however, means that specialist apparatus is required to load argon into the high pressure chamber of DACs.

Argon is a gas at ambient temperature and pressure and as such must be condensed to a liquid before it can be loaded into the sample space. There are two main ways that this can be achieved. The first is the cryogenic loading technique which liquefies the gaseous argon by cooling the gas to low temperature. Examples of loaders utilizing this method include those given in references 2-5. The second method is the high pressure gas loading technique in which the gaseous argon is compressed into a liquid phase by a gas compressor which densifies the gas until it forms a liquid such as those shown in references 6-11. Both methods have their own advantages and disadvantages but the cryogenic loading technique is relatively simple and inexpensive compared to the high pressure gas loading technique and does not have the safety issues associated with handling high pressure gases. The high pressure loading technique requires the use of high pressure gas seals which can make future adaptation to accommodate different designs of pressure cell difficult.

This paper describes a versatile, easy to use apparatus for cryogenic loading of argon into the sample space of diamond anvil cells. The design permits different designs of cell to be loaded with the same apparatus.

Most designs of loading apparatus are only useable by one design of pressure cell due to the geometrical restrictions imposed by the force application mechanism, which is typically by use of some aligned turn-screw mechanism to the tightening bolts of the cell which hold the piston in position in the loaded cell. The design presented here makes use of a hydraulic ram to apply the force to the cell via a stainless steel adaptor connected to the thrust or force plate of the pressure cell. By using different adaptors, the same apparatus can be used to pressurize numerous designs of cell. Utilizing a hydraulic ram with pressure gauge means we know exactly the initial loading force applied to the cell. This capability is typically absent from loaders employing turn-screw force application mechanisms.

Importantly the new argon loader allows for in-situ pressure determination during the loading process by the ruby fluorescence technique. Being able to measure the pressure during the loading process is particularly useful as it enables the desired starting cell pressure to be accurately set. It also means that the maximum loading force of the anvils will not be exceeded and so prevents accidental breakage of the diamond anvils. Typically for simple culet shapes the maximum loading force that can be applied to diamonds before they break has been found to be around 10 kN, and is independent of culet diameter[12]. The use of beveled culets in precision engineered cells allows forces in excess of 40 kN to be applied to the anvils resulting in pressures in the Mbar regime to be reached at the sample[12].

The design permits safe and efficient, in terms of both time and cost, liquefaction of argon gas from a typical laboratory argon cylinder into the loading chamber. The system has proved reliable and easy to use with a success rate of over 90% loading of argon into the sample space of diamond anvil cells.

II. APPARATUS

The argon loader uses argon as a pressure transmitting medium in diamond anvil high pressure cells. In overview the system works by a) cryogenically condensing liquid argon around the pressure cell and in the sample chamber, b) applying the loading force using a hydraulic ram and in so doing trapping liquid argon in the sample chamber, c) locking the force on the cell so that the cell can be removed from the system, d) measure the pressure in the cell during the entire procedure using the ruby fluorescence technique via an optical fiber.

There are two main components to the apparatus. The first is the argon loading ram system comprising the liquid argon condensation chamber and the force application hydraulic ram and associated gauge. The second is the ruby fluorescence pressure measurement system and optical fibers. Figure 2 shows the entire system and figures 3 and 4 the argon loading system in more detail.

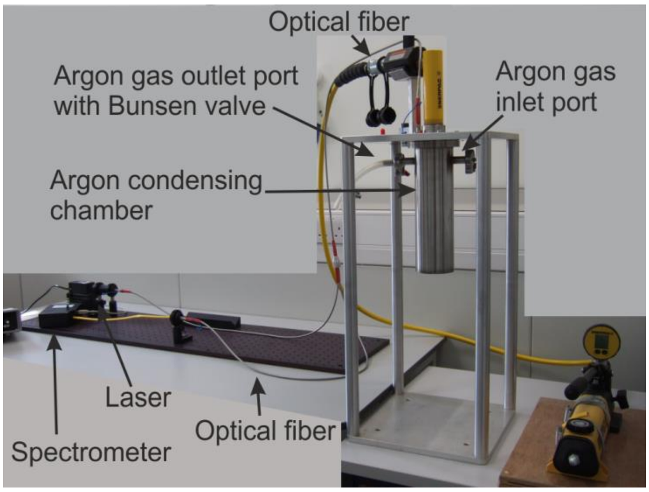

FIG. 2. (Color online) Argon loading apparatus with ruby fluorescence system.

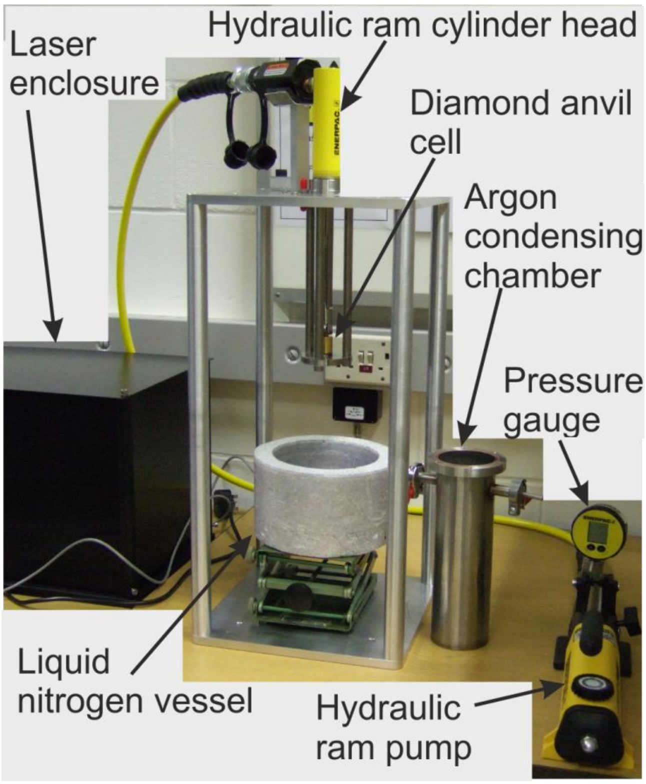

FIG. 3. (Color online) Argon loading hydraulic ram system with condensing chamber.

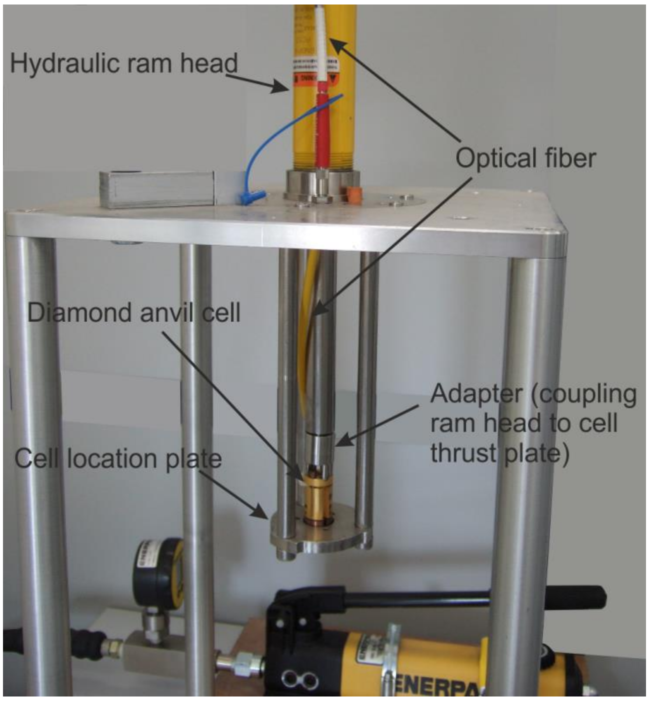

FIG. 4. (Color online) Argon loading system showing close up of cell mounting. The adaptor and location plate can easily be changed to accommodate a wide range of cell designs.

The hydraulic ram and head were supplied by Enerpac and could apply a maximum force of 50,000N. The pressure gauge in series with the hydraulic hose ensures the pressure in the hydraulic system, and hence the force applied to the cell, is known during loading.

A stainless steel adaptor was made to couple the ram head piston to the thrust plate of the DAC. A location plate positions the DAC central to the piston of the ram head (figure 4). By using different adaptors and location plates the apparatus can easily be used to pressurize a wide range of DAC designs.

The pressure in the cell was determined by the ruby fluorescence method. A small chip of ruby was included as a manometer in the sample space and excited by a 3 mW, 532 nm laser via an optical fiber which entered the argon loading chamber and coupled to the cell by a GRIN lens[13]. The pressure dependent fluorescence spectrum was collected via an optical fiber to an Ocean Optics HR4000 spectrometer.

The pressure condensing chamber was made of stainless steel and was attached to the frame by 6 x M6 bolts. An 'O' ring between the frame and the chamber ensured the system was gas tight. A tube inlet port on the chamber allowed argon gas into the chamber from a standard argon gas high pressure cylinder at 300 bar fitted with a gas regulator valve to control the flow of argon into the chamber. The argon cylinder was connected to the condensing chamber by a silicone hose. Another port had a Bunsen over pressure release valve and prevented excessive pressure build up in the argon condensing chamber.

III. EXPERIMENTAL PROCEDURE

The process of using the apparatus to load argon and pressurize a diamond anvil cell is described below. The cell used was a modified Dunston-Shearer DAC[14], although by utilizing different ram head to DAC thrust plate adaptors, other cells can be pressurized and the procedure outlined would be the same. We used a cell with 1.0 mm diamond culets mounted on moissanite backing plates[15] using copper electroplated mounting[16]. A 500 μm hole was drilled[17] in a stainless steel gasket and pre-indented from 500μm to 100μm. The sample hole was loaded with a sample and a small chip of ruby (~2-5μm diameter). The cell was loosely assembled such that the piston anvil was not tight against the gasket so as to allow the argon to flow into the sample space. The assembled DAC was fitted on the argon loader. The optical fiber with GRIN lens coupler[13] was attached to the cell to allow pressure determination using the ruby fluorescence technique during pressurization. The condensing chamber was fitted around the cell and bolted to the support frame. A high pressure argon gas cylinder was attached to the condensing chamber by silicone hose.

To condense the argon around the cell in the condensing chamber the gas regulator on the cylinder was adjusted such that gas at a pressure of a few bars flowed into the chamber. A bucket of liquid nitrogen was then placed around the condensing chamber causing the argon gas to liquefy around the cell and into the gasket hole. Tests showed allowing ~20-30 minutes of gas flow into the chamber produced liquid argon around the cell. If the nitrogen is left too long (>1 hour) around the condensing chamber then argon solidifies around the cell as the melting point of argon at ambient pressure is 84 K which is slightly higher than the nitrogen boiling point.

Before the loading force was applied to the cell the optical system was turned on so that the pressure in the cell could be monitored. The force was applied via the hydraulic ram which forced the piston into the cell causing plastic deformation of the gasket and hence pressurized the sample space of the cell. For the cell described above we typically use an initial loading force of 40 bar in the ram system, which corresponds to ~2500N loading force at the cell and gives ~0.6 GPa pressure in the cell. We have found these loading pressures inside the sample space of the cell to be reproducible to within +/- 10%. The force can be increased if a higher cell pressure is desired. Figure 5 shows the force vs pressure curve for our DAC obtained with the argon loading apparatus. It was obtained by increasing the force on the DAC by the hydraulic ram in increments and measuring the corresponding pressure in the cell using the integrated ruby florescence system. Once the cell was under pressure, the flow of argon gas was stopped and the condensing chamber removed. The cell pressure locking bolts were then tightened to lock on the pressure in the cell to enable it to be removed from the argon loader. During this process the pressure in the cell was constantly monitored by the ruby fluorescence optical system. Figure 6 shows a ruby spectrum produced during pressurization at 77K.

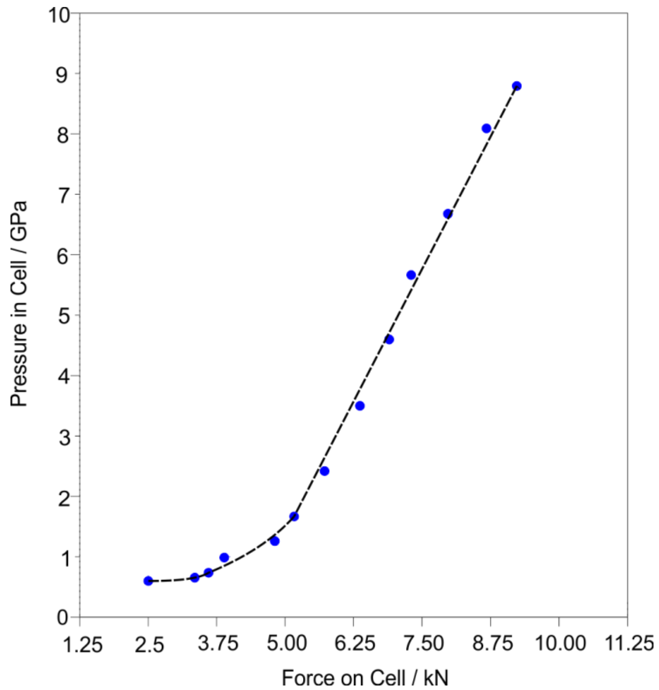

FIG. 5. (Color online) Force vs pressure curve for DAC loaded with argon pressure medium. The diamonds have 1.0 mm culets. (The dotted line is a guide to the eye).

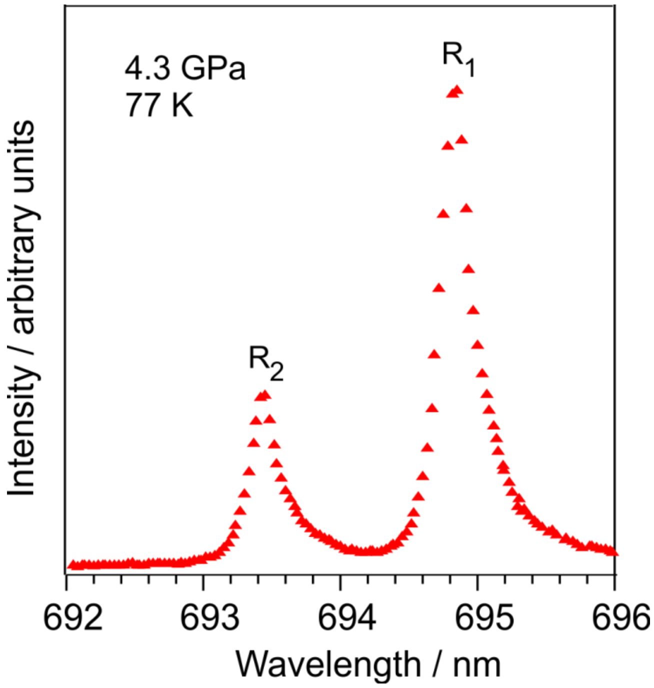

FIG. 6. Ruby Florescence spectra showing increase in pressure of sample space within a diamond anvil cell loaded with argon pressure transmitting medium.

IV. SUMMARY

We have developed a versatile system for cryogenically loading argon as the pressure transmitting medium in diamond anvil high pressure cells. Utilizing a hydraulic ram system means that with suitable adaptors a wide range of DAC designs can be accommodated. The inclusion of a fiber optic system allows for in-situ pressure determination of the DAC during the entire pressurization procedure. This results in sensitive control of the whole process resulting in a success rate of greater than 90% of argon loading into DACs.

ACKNOWLEDGEMENTS

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