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# A Cross-Sectional Transmission Electron Microscopy Study of Iron recovered from a Laser-Heated Diamond Anvil Cell

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**Abstract.** This paper discusses the use of a focused ion beam for the preparation of cross-sectional transmission electron microscopy specimens from small samples, typically 150 micron in diameter, that have been recovered from a laser-heated diamond anvil cell. We present preliminary observations of iron melted with helium as the pressure-transmitting medium to pressures near 4 GPa. The project is designed to investigate entrapment mechanisms of the inert gases in metals and silicates at high pressure and temperatures.

## 1. Introduction

The inert gases form a unique trace-elemental and isotopic system that yields information on the formation and evolution of the solid Earth, planets and their atmospheres [1]. But a completely self-consistent evolution of the terrestrial inert gas inventory has not yet been fully developed, leading to much debate on the location of trapped gases and differences between atmospheric and deep sources. High-pressure experiments have shown that the inert gases display unusual physical properties under the conditions of planetary interiors [2] but little is known at the atomic scale of the nature of inert gas entrapment in this environment. To investigate the relationship between the inert gases and solids at high pressure and temperature, transmission electron microscopy (TEM) has been employed to study cross-sectional foils recovered from the laser-heated diamond anvil cell (LHDAC).

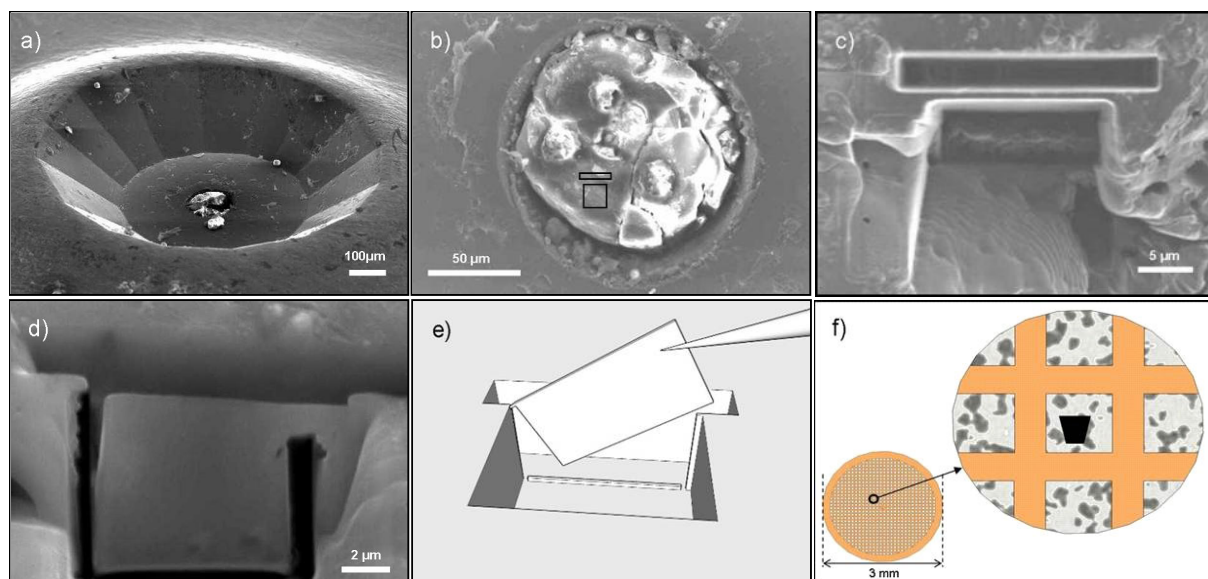
To recreate the extreme conditions present in the interior of a planet, a LHDAC was used, which can in principle reach pressures and temperatures equal to those at the centre of the Earth (350GPa, 6000K). The DAC presses two diamonds together around a chamber of diameter 50–150 $\mu$ m which is drilled within a pre-indented gasket. An infra-red, Nd-YAG laser heats the sample. The experimental method is described in more detail elsewhere [3].

The small size of the samples taken from the DAC, makes traditional sample preparation techniques such as tripod polishing or ultramicrotomy difficult and very destructive [4]; however, the focused ion beam (FIB) system, commonly coupled with an SEM would seem to lend itself perfectly to this task. This tool has been widely used across the sciences, from creating thin films of biological samples [5] to cross-sectioning micro-diamonds [6] and can produce numerous foils from a very small area with minimum destruction of the original sample. The ion beam can be controlled to mill specific shapes, while the attached SEM enables very precise milling of specific areas. The literature contains numerous methods for preparing samples for TEM using the FIB system, all very similar but with

slight modifications to suit different substances [6-9] The following method has been tailored to suit the particularly challenging samples used in the current project.

## 2. Experimental Methods

Samples of iron surrounded with helium to act as the pressure medium were pressed to 4GPa, and then heated to 2500K, using the LHDAC at Oxford. The form of the sample after removal from the LHDAC is shown in Figure 1a: the sample is at the centre of a depression created within the stainless-steel gasket by the diamonds. Since the gaskets are too large for the sample holder of the FIB, the centre portion ( $\text{\O} \sim 7\text{mm}$ ) was punched out and mounted on a  $45^\circ$  angled stub using a conductive carbon adhesive disc and the area from which the sample was to be taken was then located, Figure 1b.



**Figure 1.** Various steps of the FIB specimen procedure; (a) the depression created by the diamond anvil cell, with the sample visible in the centre of the gasket; (b) a suitable area is chosen for cross-sectioning; (c) two trenches are milled, one either side of the region of interest; (d) the sample is tilted and the sides and underside are cut; (e) a glass rod is used to remove the specimen from the sample; (f) the final specimen ( $\sim 10 \times 0.1 \times 10 \mu\text{m}$ ) is then placed on a carbon film on a copper TEM grid.

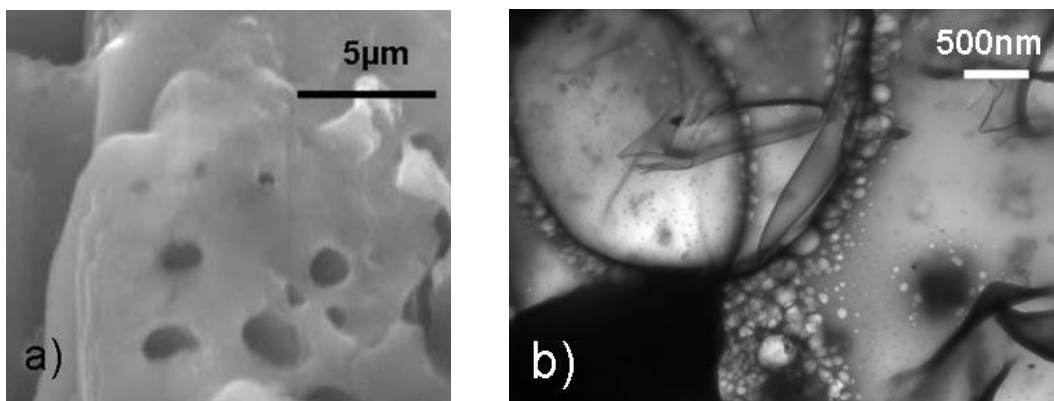
Once the area of interest had been chosen, a large beam current ( $\sim 3\text{nA}$ ) was used to mill two trenches one either side of the feature; a  $10 \times 10 \times 10 \mu\text{m}$  and a  $15 \times 2 \times 10 \mu\text{m}$  trench with approximately a  $3 \mu\text{m}$  gap (the sample area) between the two (Figure 1c). Owing to the conditions in the DAC the surfaces are highly irregular; this can cause the so-called ‘curtain’ effect when milling, where inhomogeneities on the surface and within the sample locally affect the milling rate, this is especially pronounced with larger beam currents. The next stage was to begin to thin the sample area from the  $3 \mu\text{m}$  down to a suitable thickness for the TEM work ( $\sim 100\text{nm}$ ). A succession of smaller beams was used; firstly a  $1\text{nA}$  thinning down to  $1.5 \mu\text{m}$ , followed by  $250\text{pA}$  to  $300\text{--}400\text{nm}$ . At this point the stage was tilted by  $45\text{--}60^\circ$  so that the sample could be cut partially free (Figure 1d). This prevents the sample from warping or breaking. A final thinning step was then performed at  $115\text{pA}$  reducing the foil to  $\sim 100\text{nm}$ , followed by a final step at  $50\text{pA}$  to give a final ‘polish’ in order to remove surface roughness. The final cut was then made to free the sample.

After the specimen had been almost cut free (Figure 1d), the entire sample was removed from the SEM/FIB and placed under an optical microscope ( $\times 20$  magnification), where using a

common ex-situ lift-out method [9], a glass rod that had been pulled to a point was used to lift out the specimen of typical dimensions  $10 \times 10 \mu\text{m}$  (Figure 1e). A slight modification was needed to the normal lift-out procedure as the sample resides at the bottom of a depression, which reduces the manoeuvrability of the glass rod. By having the sample mounted at  $45^\circ$ , the needle's movement is uninhibited. The specimen was then placed on a TEM grid with 300mesh coated in carbon film (Figure 1f), ready for microscopy.

The samples were prepared on a JEOL JSM-5910 scanning electron microscope fitted with an Orsay Physics FIB system, with both electron and ion beams operating at 30keV. The lift-out was performed on a Prior optical microscope fitted with a capillary action micro-manipulator. The transmission electron microscopy was performed on a JEOL 200CX operating at an accelerating voltage of 100kV. Images were recorded on photographic negatives and subsequently scanned (as positive images at 1200dpi) into Adobe Photoshop CS where the images were optimised for printing by alteration of the contrast and brightness.

### 3. Results & Discussion



**Figure 2.** a) A SEM micrograph illustrating the large craters at the surface. b) Bright-field TEM image of small cavities surrounding a large lenticular cavity. A small degree of objective lens underfocus (approximately 1000nm) has been used to enhance the contrast from the small cavities

Figure 2a is a SEM image recorded prior to the FIB sample preparation, showing a collection of large craters evident on the surface indicating that at least these large surface cavities do not result from the FIB processing. Figure 2b shows a large density of small cavities clustered at the edge of a much larger cavity of the order of a few micrometres. As the foil is only approximately 100nm thick, the large cavity must have a lenticular form. We tentatively identify the cavities as helium inclusions resulting from turbulent mixing at the interface during the pressing and heating within the DAC. The images are qualitatively similar to emulsions formed as a result of agitation at the interface between two immiscible fluids. In the present case, this morphology has been preserved in the system as movement of the focused laser spot — diameter  $\sim 20 \mu\text{m}$  — that has resulted in a quenching of the area. If our interpretation is correct then we may have identified a new mechanism for the incorporation of inert gases into molten solids at high pressure, however, the presence of helium in the cavities will have to be confirmed by additional measurements.

### 4. Conclusions/Further Work

The macroscopic mechanical trapping of rare gases may have had some role in the early, highly energetic stages of the Earth's formation. The next step that will likely determine how rare gases are sequestered on the atomic scale (leading to ppm and ppb concentrations) will require searching for much smaller features or clusters of rare gases formed in metals under pressure. Future work will therefore include studies into other inert gases and their interaction with silicates and metals, as well as further investigation into the Fe/He system.

The FIB system has already shown itself to be an excellent sample preparation method for TEM, and lends itself particularly well to DAC samples. A foil can be prepared within 12 hours with the major limiting factor being the lift-out procedure with a current success rate of 1 in 4. Two further means of improvement are to:

- (i) coat the rough surface of the sample with a uniform tungsten layer using the FIB. This reduces the curtain effect and also provides a barrier against gallium implantation into the surface, during the milling process;
- (ii) reduce the accelerating energy of the ions to 5–10keV for the final polishing stage to lower the surface roughness and the damage caused by the gallium ions within the sample.

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