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Miniature Diamond Ultrahigh Pressure Cell Adapted for In-reactor Irradiation Studies*

GERALD L. KULCINSKI,† CHARLES W. MAYNARD, AND ROBERT C. WALSH
Nuclear Engineering Department, University of Wisconsin, Madison, Wisconsin 53706

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A special Bridgman opposed diamond anvil apparatus was constructed to generate useful static pressure to 50 kbar at ambient temperatures. It was designed to function in the environment of a nuclear reactor and to occupy 65 cc. A method of bombarding materials under pressure with fission fragments is described.

INTRODUCTION

EXPERIMENTALLY, the main problem with the use of nuclear radiation in conjunction with ultrahigh pressure (>30 kbar) is the sheer bulk of many of the current devices. Normally, the experimental space available for irradiation studies is a 5–7.5 cm diam tube. A quick review of the literature[1] reveals that pressure equipment may vary in over-all size from a few cubic centimeters to 5 m³ and weigh on the order of 1 to 20 000 kg. The geometry of the most popular and functional devices (i.e., cubic,[2] tetrahedral,[3] and 'belt' configuration[4]) prohibits them from being placed near the core of a nuclear reactor without seriously disrupting the neutron flux of the system. Fast fluxes that could be obtained by placing this equipment in a collimated beam outside the reactor would be so small as to make the integrated dose insignificant. Furthermore, materials of construction for high pressure equipment are not usually picked for their activation properties. It is not uncommon to find large amounts of Cr, W, Mn, and Co in the cells. Working around such equipment after appreciable doses of neutrons would be extremely hazardous.

Bridgman anvil[5] and piston-cylinder devices[6] conform more readily to the required geometry, but have some serious drawbacks. The piston or anvil is usually driven by a highly compressed gas or organic liquid. Since the pressure generating equipment is quite bulky, it would be desirable to place it outside the reactor. This in turn would require 3 to 5 m of high pressure tubing to connect the piston or anvil to the generating equipment. An organic liquid would then be subjected to large doses of ionizing radiation which would tend to make it more viscous. It may even cause the fluid to solidify under the combined action of pressure and irradiation.[7] Furthermore, most reactor safety engineers become concerned about the energy stored in a high pressure (10 000 kg/cm²) line which might be placed next to a delicate arrangement of fuel elements. From an energy release standpoint, liquids would be favored over gases.

DESIGN

It seems evident that the desirable characteristics of a pressure cell designed for in-reactor irradiation studies are

(1) small volume (small weight),
(2) materials with low neutron activation cross sections,
(3) self-contained, nonhydraulic pressure generation, and
(4) small energy reservoir.

Such a cell has been designed and is shown in Fig. 1. This cell is constructed on the Bridgman opposed anvil principle and consists of two spring loaded gem cut diamonds acting both as pressure anvils and observation windows. In some respects it incorporates features from

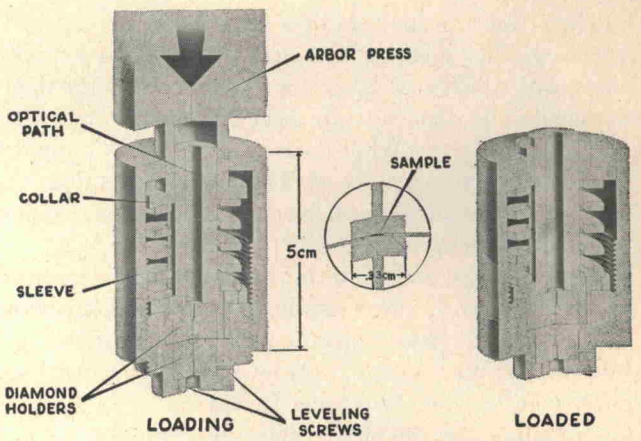


FIG. 1. A schematic of the diamond pressure cell (left) in the process of increasing the pressure and (right) loaded position ready for insertion into a nuclear reactor.

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† Now with Battelle-Northwest, Richland, Washington 99352.

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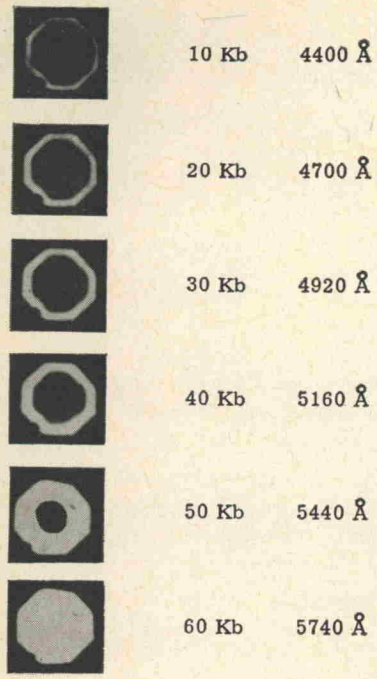
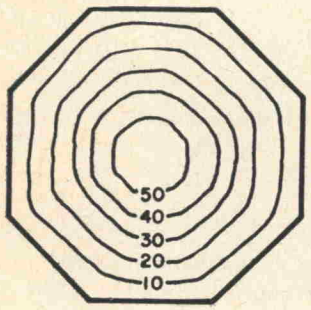


FIG. 2. (Top) Typical calibration pictures for high pressure cell. Black areas represent region of sample above pressure designated at the side of photo. (×13) (Bottom) Typical pressure distribution for a well aligned cell.



cells designed by Lawson and Tang,[8] Jamieson,[9] and Lippincott *et al.*[10]

The pressure is applied in the following manner. A hand operated arbor press acts through the collar to compress the spring which applies a compressive force to the upper diamond. The diamonds are held in stainless steel seats with plaster of paris. While the required pressure is applied, the sleeve is screwed down until it engages the spring. The arbor press is then released, leaving the threads to support the compressed spring.

Conditions (3) and (4) of the design criteria were satisfactorily met by the use of a spring. Negligible energy tie-up and internal pressure generation made the unit self-contained with no external leads or tubing. However, one must sacrifice sample volume in order to use a spring loaded mechanism. Typically, the sample size was 2×10^{-3} cm^2 by 5×10^{-4} cm and weighed on the order of 5 to 10 μg. It was usually in the shape of the small diamond face, namely an octagon wafer (Fig. 3).

The cells are generally 7 cm long and 3.8 cm in diameter, although this is not the minimum, size. The bulk of the cell is made of 6000 series aluminum, and the diamond seats and leveling screws are made of 303 stainless steel. The spring is made of spring steel. Typically, the total weight of the cell is 170 g, consisting of 100 g Al, 60 g Fe, 5 g Cr, 2 g Ni, 2 g Mg, and 1 to 2 g Cu. No provisions are made for heating or cooling the sample volume under pressure, and the temperature of the sample depends on gamma ray heating and reactor ambient temperature. This is usually less than 60°C. Temperatures of −50 to 250°C can be obtained with suitable temperature control systems incorporated in the design. The samples, usually in powder form, are placed between the diamond faces and the pressure is slowly applied. To minimize alignment problems and to take advantage of the massive support principle, the two diamond culet faces are purposely ground to have different surface areas. There are eight leveling screws on the bottom diamond seat. Four of them, spaced at 90° intervals, act perpendicular to the sample surface and four more at 90° intervals act parallel to the sample surface.

It has been observed that proper alignment of the diamonds before each run is extremely important in the extension of their useful lifetimes. Before the sample is introduced, the cell is assembled and the diamond surfaces are placed lightly against one another. White light is directed through both diamond surfaces perpendicular to the interface between the two. A microscope is focused on the interface and white light interference patterns are centered with the alignment screws. The pattern is gradually eliminated until one or two fringes can be seen. This means that the two diamond surfaces are essentially parallel and that any sample placed between them has a uniform thickness and symmetrical pressure distribution.

In order to calibrate the cells, one must have a feeling for the nature of the pressure or stress generated. In the early use of such diamond cells, the pressure was calculated by simply determining the force applied and then dividing by the area on which this force acted. Three things are wrong with this. First, frictional effects on the piston walls are neglected; second, the assumption of a linear spring constant might be in error, particularly when highly compressed; and third, and most important, a uniform pressure over the entire face of the diamond is assumed. Observations[11] have verified that nonuniform pressures are imposed, since phase changes observed optically always seemed to start in the center and move progressively outwards as the pressure is increased. In some cases, as much as 112 kbar of pressure difference has been observed between the center of the diamond and the edge.

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It has been discovered that TlBr has a pressure sensitive absorption edge which progresses toward the red end of the spectrum as pressure is increased.[12] At 1 atm the edge occurs at 4170 Å, i.e., TlBr is transparent to wavelengths greater than 4170 Å and opaque to wavelengths less than 4170 Å. As pressure is increased, it is necessary to use longer wavelengths to observe transparency. However, in our cell, when monochromatic light from a model 90 Perkin-Elmer monochromator was shone through the diamonds and TlBr under pressure, the entire specimen did not become transparent at once. Rather, when going from violet to red, the edges of the sample became transparent first and the circle of opacity decreased as wavelength increased. This indicated that we have a much higher pressure in the center of the sample than at the outside.

Realizing that different parts of our samples were subjected to different pressures, depending on their distance from the center, it was necessary for us to know the pressure distributions. This was easily done by noting the position of the opaque circle as a function of the wavelength of light (see Fig. 2). The position of the circles was noted photographically with 3000 ASA Polaroid film, and then the pressure contours were placed on an outline of the sample. By integrating the area representing pressures above the reference pressure, we could accurately describe the state of our sample. Hence, it was possible to cover a range of pressures, from the peak pressure to the shear strength of the material in a single calibration. Typically, the highest useful pressure (greater than 10% of the sample above this pressure) was 50 kbar and most often 80 to 90% of the sample was above 10 kbar. It was found that the pressure distributions were constant for periods of 7 to 10 days (no spring relaxation).

Neutron activation problems are reduced by the choice of cell materials. However, even with this design, there are maximum limits to thermal flux exposures. The two isotopes which are troublesome are ^{51}Cr and ^{64}Cu. Since ^{64}Cu has a 12.9 h half-life, its activity is insignificant after 4 to 5 days. ^{51}Cr, with a 27.8 day half-life, is a nuisance in that the cell acquires about 30 mR/h (at contact) of ^{51}Cr gamma activity for every 10^{16} nvt thermal flux. This limits useful thermal fluxes to 10^{18} nvt, and, depending on the cadmium ratio, fast fluxes to about $1–5\times10^{17}$ nvt, for hand unloading without the benefit of hot cells.

After proper decay periods (~4 to 5 days), the cells are unloaded by releasing the load on the threads through a slight compression of the spring. The sleeve is then unscrewed and pressure is released with an arbor press. With the sleeve, collar, and spring removed, the remaining two parts are carefully pulled apart. The sample, depending on its nature, usually remains bound by friction to one of the diamond faces. If it is a brittle material, it is removed under

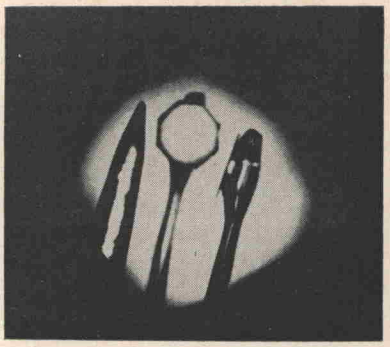


FIG. 3. Sample mounted on 0.3 mm diam glass fiber (center) as compared to pinpoint (left). Edgewise view (right) of sample that is 0.01 mm thick. (×13)

a binocular microscope with a de Foubrune micromanipulator, which has a ratio of movements of 1:2500. A fine hook, 75 μ in diameter, is used in this operation.

In a current set of experiments, the phase stability of materials was studied.[13] Therefore, after the sample was removed, it was stuck onto a fine glass fiber with nail polish (see Fig. 3) and placed in a Debye-Sherrer powder x-ray camera. If the sample could not be removed in one piece, the diamond to which it adhered was removed from its plaster of Paris mounting and the sample plus diamond was placed in the camera with the diamond face oriented parallel to the x-ray beam.

By proper techniques, the sample can be bombarded with fission fragments while under pressure. A thin foil of uranium metal is mechanically pressed onto one of the diamonds. The material to be studied is placed on the other diamond and the two are then forced together in the usual manner. The equilibrium thickness of this "sandwich" is about 25 μ evenly distributed between the two materials. Fission fragments born in the uranium are then deposited in the sample material.

It is worth mentioning that after fast neutron bombardment ($1–5\times10^{6}$ nvt) diamonds become dark green in color and absorb all the wavelength below 4600 Å (or about 20 kbar pressure using TlBr calibration). If the diamonds are to be reused, they must be periodically annealed. It was found that heating the diamonds to 700 to 800°C in an inert atmosphere for 1 to 2 h removes a large fraction of the color and leaves the diamonds bright gold in color. Irradiation to 10^{17} nvt does not appear to affect their mechanical properties.

ACKNOWLEDGMENTS

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