A Low-Temperature Glass Cryostat for X-ray Diffraction Work

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A simple cryostat made of glass for X-ray diffraction at low temperatures is described. The windows for the X-rays are made from Mylar films fixed to slots cut in the base of a glass helium reservoir. This glass cryostat weighs only 6 kg and specimens can be easily handled in the cryostat. A specimen chamber has also been designed so that in-situ pulse annealing can be easily carried out. Some examples of measurements with this cryostat are described.

Introduction

Many types of metal cryostats for X-ray diffraction studies have been reported [see reference 1 of Peterson & Simmons (1965)]. There are, however, only a few reports of a glass cryostat [for example see Lytle (1964)], although it has the advantages of simplicity, light weight and ease of specimen handling. The difficulties involved in making a glass cryostat are in design of a specimen chamber and X-ray windows. In the glass cryostat described in the present paper, these were solved by a simple design and in particular by use of a technique for fixing Mylar windows on the specimen chamber without using Kovar seal. Furthermore, the present cryostat is designed to permit in-situ pulse annealings. This cryostat was successfully used for a recovery study of the lattice parameter of graphite irradiated by neutrons at 5 K and also for a study of the thermal expansion of molybdenum in the temperature range 4.2 to 300 K.

Cryostat

A schematic drawing of the glass cryostat is shown in Fig. 1. It consists of four coaxial glass tubes, with liquid helium in the innermost tube, a vacuum between the first and second tubes, liquid nitrogen between the second and third tubes, and a vacuum between the third and fourth tubes. The windows for the X-rays are made from 0.003 in (0.075 mm) thick Mylar films (made by E. I. du Pont de Nemours and Co.), adhered to slots which are cut in the lower part (specimen chamber) of the glass tube of the helium reservoir. Epibond 121 (made by Furane Plastics, Inc., U.S.A.) is used as the adhesive on the slots. No Kovar metal seal is used. The outer X-ray windows at room temperature are also covered with Mylar films. The base of the glass cryostat sits on a metal flange with an O-ring. The inside wall of the cryostat is silvered, leaving slits to observe liquid helium levels. The slits are also used for examining the specimen during handling in liquid.
specimen chamber are found to withstand repeated thermal cycling between 4.2 K and room temperature. X-ray measurements at 4.2 K are carried out with the specimen immersed in liquid helium. In order to vary the temperature of the specimen, the top of the inner member (4) is sealed by lowering the central tube (5), liquid helium in the specimen chamber is boiled off through the central tube (5) and the temperature of the specimen is controlled by the heater wound around the copper block (3). The temperature is measured by Au–0.03% Co/copper, or copper/constantan thermocouples, embedded in the copper block (3). A reference junction of the thermocouples is immersed in liquid helium as shown in Fig. 1. The temperature can be easily maintained within 0.05 °C at around 40 K and 0.02 °C above 50 K.

Transfer of specimens in liquid helium

To study recovery after neutron irradiation at liquid-helium temperature, the irradiated specimens must be transferred from a liquid-helium Dewar into the X-ray measurement cryostat without any intermediate warm-up. A simple method of transferring the specimens in liquid helium has been developed (Maeta, Kato & Okuda, 1975) and applied to X-ray measurements as described below. A small vacuum-insulated metal

![Fig. 3. Schematic drawing of the method of transferring the specimens to the X-ray glass cryostat.](image)

helium. The helium reservoir has a standard tapered ground neck [ground joint (1) in Fig. 1], on which another ground taper of an inner member (4) sits and this joint separates the upper liquid helium reservoir from the bottom specimen chamber. A small amount of vacuum grease (or N-Apiezon grease) applied to the joint makes the seal complete. The inner member (4) has a glass pipe (2), the bottom end of which is connected to a copper block (3) by the Mylar film, using Epibond 121. A specimen holder is positioned in this copper block (3), which has an electrical heater element and thermocouples (see Fig. 2). A central tube (5) is inserted into the upper end of the inner member (4). A radiation shield made of copper plate is connected to the bottom of the liquid nitrogen reservoir. The whole cryostat weighs only about 6 kg, so that it can be mounted on an ordinary goniometer of a commercial horizontal diffractometer.

**Experimental procedures**

*Helium consumption and temperature control*

Under a steady-state condition, the rate of loss of liquid helium is about 80 cm$^3$ h$^{-1}$ (1.1 litres of liquid helium in the reservoir can maintain the specimen at 4.2 K for about 12 h). The vacuum spaces in the cryostat are evacuated by a vacuum pump with a liquid-nitrogen trap. The Mylar films of the X-ray windows on the

![Fig. 4. A view of the cryostat mounted on the goniometer set on a type SG-6 diffractometer.](image)
Dewar (a bucket Dewar) is suspended in liquid helium in a separate Dewar containing the irradiated specimens. A specimen is placed in this bucket Dewar in liquid helium and the bucket Dewar is lifted into a transferring box (shown at the top of Fig. 3). The bottom shutter (Fig. 3) is closed and the box is moved to the top of the X-ray cryostat, the central tube (5) having been removed beforehand. The bottom shutter of the box is then opened and the bucket is lowered into the cryostat. After the specimen has been taken out of the bucket and set in the measuring position, the cryostat cover with the central tube (5) is positioned as in Fig. 1. The whole procedure can be accomplished in a short time and with a low consumption of liquid helium.

**X-ray measurements**

Fig. 4 shows the cryostat set on a goniometer mounted on a type SG-6 diffractometer (made by Rigaku Denki Co., Ltd, Japan). Fine adjustment of the specimen position is achieved by moving the goniometer arcs and X-Y cross slides. X-ray measurements were carried out with a monochromated beam (a silicon single crystal was used) and a scintillation counter. The lattice parameter is measured by rotating the cryostat counterclockwise with the goniometer. Liquid helium around the specimen is not found to affect the diffraction profile within experimental error (see Fig. 5).

Two typical examples of the results of lattice-parameter measurements are given below.

**Recovery after neutron irradiation**

The change in the c-axis spacing in pyrolytic graphite has been measured after neutron irradiation at 5 K and subsequent isochronal pulse annealings in the temperature range 5 ~ 900 K. Specimens were well crystallized, monochromator-grade pyrolytic graphite. Fast-neutron irradiation was performed at 5 K in LHTL (Liquid Helium Temperature Loop) of the JRR-3 reactor at the Japan Atomic Energy Research Institute. All measurements of the c-axis spacing were carried out at 4.2 K using the monochromated Cu Kα₁ beam and the 00.8 reflexion. The diffraction peak was at 134.6° in 2θ. The c-axis spacing of a dummy specimen of graphite was examined after several pulse annealings at various temperatures. The accuracy in the present measurements was checked with this dummy specimen, and that of the relative lattice-spacing change at liquid-helium temperature was shown to be better than δ(Δc/c) = ± 1 x 10⁻⁵. The total irradiation-induced change in the lattice spacing (Δc₀/c) was determined by comparing the Bragg peak positions before and after irradiation.

Fig. 6 shows the results of isochronal pulse annealings on the specimens irradiated with fast neutrons at doses of 1.2 x 10¹⁷ and 3.1 x 10¹⁷ neutrons cm⁻² (Δc₀/c = 4 x 10⁻⁴ and 1.0 x 10⁻³), respectively. The pulse annealing time was 6 min. A detailed account of the results has been described elsewhere (Maeta, Iwata & Okuda, 1975a, b).

**Thermal expansion measurements**

The thermal expansion of the lattice parameter of a molybdenum single crystal was measured in the temperature range 4.2 ~ 300 K. The crystals were purchased from MRC (a nominal purity of 99.99%). The measurements were carried out with monochromated Co Kα₁ radiation, and the 222 diffraction...
peak occurred at $2\theta = 161.5^\circ$. Typical examples of the reflexion profiles at 40 and 60 K are shown in Fig. 5. The $Kz_2$ component of Cu $K\alpha$ reflexion is not present because a fine-focus source and the monochromated beam were used at a high-angle reflexion from the specimen. Fig. 7 gives the lattice parameters vs. measuring temperatures. The value of the lattice parameter at 273.2 K has been adjusted to fit the 3.14673 Å value of Straumanis & Shodhan (1968). For comparison, results of other workers are also shown. The present results are in excellent agreement with the values obtained by optical interferometry (Nix & MacNair, 1942).

**Other applications**

The glass cryostat can be applied to the study of optical spectrometry, and with the method of transferring specimens in liquid helium it can also be used for the study of rare-gas crystals, since in-situ crystal growth becomes unnecessary.

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**References**


