Improvements for high-pressure cell experiments using the latest single crystal laboratory systems

Hiroyasu Sato*

1. Introduction

Pressure is one of the important parameters that define the structure and state of materials.

A high-pressure diamond anvil cell (DAC) is commonly used to measure the "*in-situ* analysis" of the structural change against pressure in the X-ray diffraction experiment. In a DAC experiment, it is necessary to enclose the sample in a space less than ~200 µm in diameter in order to apply high pressure. The diameter of the X-ray beam should be smaller than the window of the DAC. There are thus three issues that make this experiment difficult to perform using a laboratory X-ray system. A small X-ray beam requires a large amount of collimation that significantly reduces flux, the window for the DAC absorbs X-rays, and finally the sample size has to be small and this reduces the diffraction intensities. Because of these restrictions, it is often believed that a synchrotron radiation X-ray source is needed to observe the X-ray diffraction in a DAC experiment.

However, recent changes in fundamental technologies related to X-ray sources and X-ray detectors allow us to obtain single crystal structures from crystals as small as a few microns in diameter with a laboratory system. These new laboratory configurations improve the S/N of the DAC experiment to allow elucidation of structural information in the laboratory.

The combination of a micro focus rotating anode X-ray generator (MicroMax-007HF) and a multilayer focusing optic system (VariMax), produces a brilliance that is an order of magnitude better than that of the combination of the conventional rotating anode X-ray generator and graphite monochromator. This high brilliant X-ray source is integrated in the latest single crystal systems (Fig. 1). The DAC experiment can easily be measured with such a system.

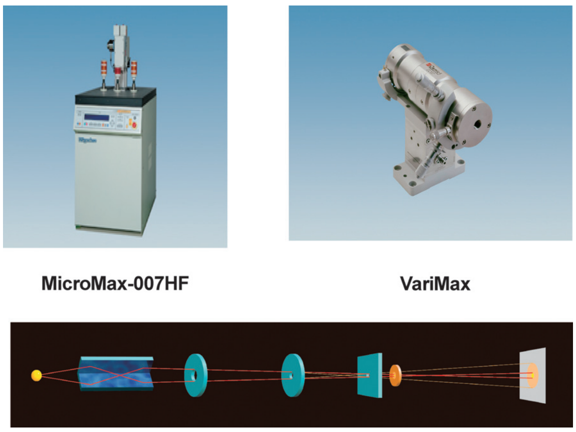

Fig. 1. The MicroMax-007HF (above left) and the VariMax optic system (above right). The geometry of the X-ray focusing and collimation system (lower).

2. Elemental technologies

Rigaku's microfocus X-ray generator improves the collection efficiency of the multilayer optics by concentrating the entire power into a circle (ϕ70 µm) that is smaller than the area viewed by the optics. The comparison between the conventional and the latest X-ray generators are summarized in Table 1.

In the single crystal system, the beam diameter at the sample position is approximately 200 µm when using the VariMax HF. In order to minimize the diffraction from the gasket material built in the high-pressure cell, it is necessary to have the beam much smaller than the opening of the gasket. Therefore we created a new collimator having a 30 µm pinhole aperture. An additional positive effect of using such a fine beam is to reduce the background originated mainly from air scattering. The detector is also an extremely important component for a high performance system. Therefore, both an IP and a CCD system were tested in the following experiments.

3. Experiments and results

3.1. System configuration

The systems integrating the IP and the CCD detectors were shown in Figs. 2 and 3. The DAC is mounted on the conventional goniometer head with an XYZ stage. The collimators ϕ100 µm and ϕ30 µm were used. The sample position is adjusted by using the CCD camera. The CCD camera should be placed at the same height of the DAC window to eliminate the parallax originating from the diamond.

Table 1. Comparison of the performance of X-ray generators.

X-ray generator	ultraX18	MicroMax-007HF
Power [kW]	5.40	1.20
Focal spot size [µm]	300	70
Brightness [kW/mm^2]	6.0	31.2
Relative brightness	1.00	5.20

* Application Laboratories, Rigaku Corporation.

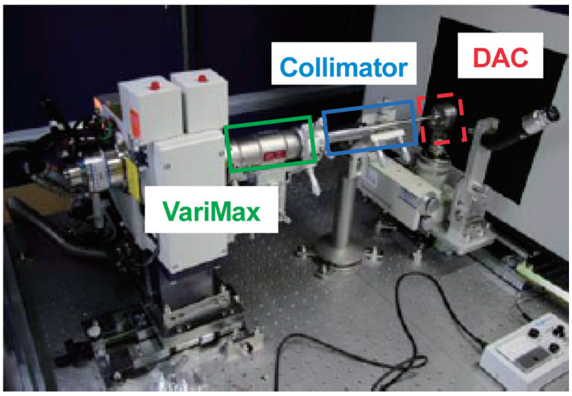

Fig. 2. The R-AXIS VII with the DAC. This particular system is equipped with the VariMax Mo optic system.

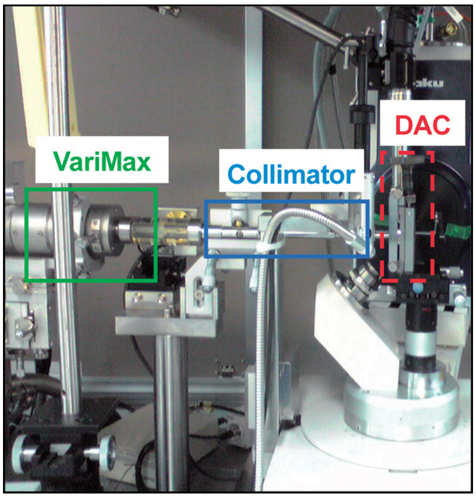

Fig. 3. The VariMax and Saturn CCD with a DAC.

3.2. Experimental Conditions

Sample and measurement conditions are shown in Table 2.

The direction of the incident X-ray beam relative to the DAC was determined so that the scattering from the gasket is minimized. The Mo radiation was chosen for the penetrating power and the lower reflection angles to extend the resolution limit.

3.3. Effect of collimator size

In the diffraction profile when using a 100 µm collimator, a diffraction line from the gasket is observed because the beam diameter is comparable with the size of the sample chamber that is determined by the opening of the Re gasket (the blue line in Fig. 4). On the other hand, the peak originating from the gasket disappeared when using the ϕ30 µm collimator (the magenta line in Fig. 4).

3.4. Comparison of detector properties

Figure 5 shows diffraction images and the X-ray diffraction profiles measured with the same exposure time. With this exposure time, the effect of the high

Table 2. Conditions of sample and diffraction measurement.

Sample conditions	
Sample	MgO+Au powder
Sample size	30 µm
Gasket	Rhenium
Sample chamber	150 mm
Measurement conditions	
CCD system	
Exposure time	1000 s
Collimator size	ϕ100 µm, ϕ30 µm
Camera distance	100.0 mm
Detector 2θ	−10°
IP system	
Exposure time	1000 s
Collimator size	ϕ30 µm
Camera distance	127.4 mm
Detector 2θ	0°

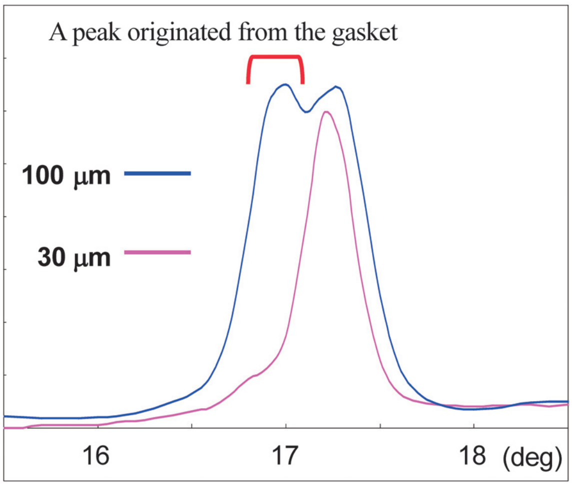

Fig. 4. The XRD pattern of the powder sample (MgO+Au mixture).

sensitivity of the CCD is prominent. However, when a longer exposure time is used, the IP system produces similar results to the CCD system.

For a well diffracting sample, the CCD system can produce better images with higher S/N in a relatively short period of time compared to the IP system. However, if the strong diffraction spots from the diamond curettes and weak diffraction lines from the powder sample co-exist, the IP system is advantageous because of its wide dynamic range (Fig. 6).

4. Conclusions

By using the latest single crystal lab system, we were able to observe the diffraction line from the sample of about 30 µm. Additionally, by reducing the size of the collimator to ϕ30 µm, we were able to eliminate the

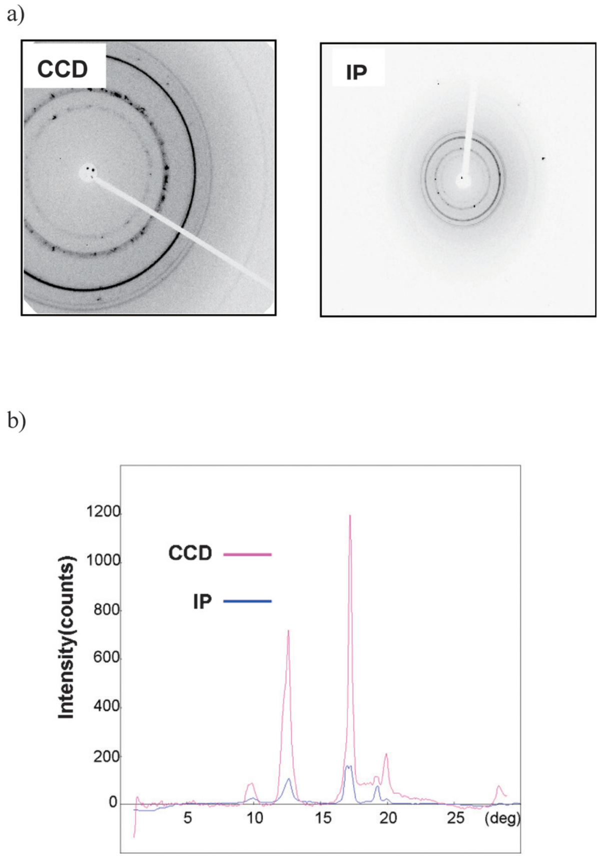

Fig. 5. Comparison of the diffraction images (a), and the X-ray diffraction pattern (b) at the same exposure time.

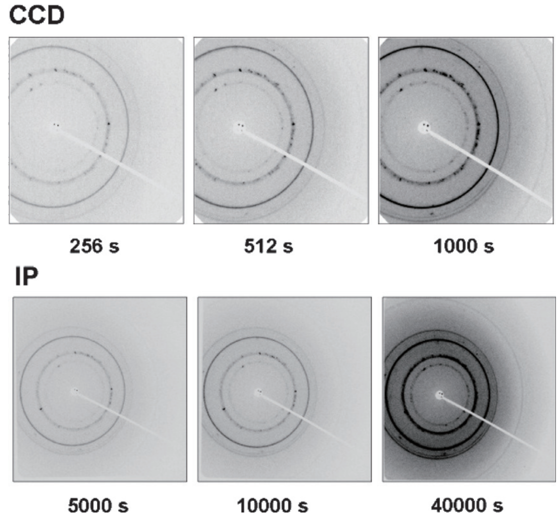

Fig. 6. Diffraction images recorded by the IP and CCD detectors. Long exposure times were employed to elucidate the features of the detectors.

diffraction lines from the gasket.

In this particular experiment, the diffraction intensity resulting from the high sensitivity of the CCD was remarkable. However, similar results can be obtained from the IP system by employing a long exposure time. When both strong and weak diffraction events co-exit, the wide dynamic range of the IP system is better suited. The latest single crystal laboratory system is capable of performing DAC experiments that were believed to be exclusive to synchrotron beam lines.