New Large-volume High P-T *In Situ* X-ray Diffraction System at BL04B1

Earth's interior is under extremely high pressure and high temperature conditions. Therefore, knowledge of the physical and chemical properties of Earth's materials at high pressures and temperatures are indispensable for studying the structure, dynamics and evolution of Earth. *In situ* X-ray diffraction is a useful and powerful method for studying crystal structures, phase relations, equations of state, viscoelastic properties of minerals and melts at high *P-T* conditions. The KAWAI-type apparatus, together with a diamond anvil cell, is one of the most widely used high-pressure apparatuses, particularly in geosciences. The reason is that high pressures up to 30 GPa can be generated in relatively large volumes, rendering it possible to conduct sophisticated experiments with a complex sample assembly in fairly homogeneous high P-T conditions. In SPring-8, a KAWAI-type apparatus for *in situ* X-ray observation, named SPEED-1500, was installed at the bending magnet beamline **BL04B1** in 1997 [1], and a number of important experimental results have been obtained.

Despite the great success of SPEED-1500, further developments of high P-T *in situ* X-ray observation in a KAWAI-type apparatus are necessary to extend the pressure and temperature ranges. SPEED-1500 was originally designed for experiments using tungsten carbide (WC) anvils, which limit the generated pressures to *ca*. 30 GPa. In order to extend the pressure range, the use of sintered diamond (SD) is necessary. Because SD is very brittle, however, we need much more precise dimensions of the guide block system and control of press load than those of SPEED-1500. At high temperatures, grain growth of the sample materials is very rapid in a Kawai-type apparatus. As a result, we often miss many lines in diffraction patterns taken by SPEED-1500 at high temperatures. In such cases, we had very low precision in determining pressures and unit cell volumes of samples, and sometimes failed to identify the phases present. Hence, we need an oscillation system to obtain high-quality diffraction patterns against grain growth. Mainly because of these two reasons, we have designed and installed the second KAWAI-type high P-T apparatus, SPEED-Mk II, at the same beamline BL04B1 (Fig. 1) [2].

Fig. 1. Photograph of the press part of SPEED-Mk II.

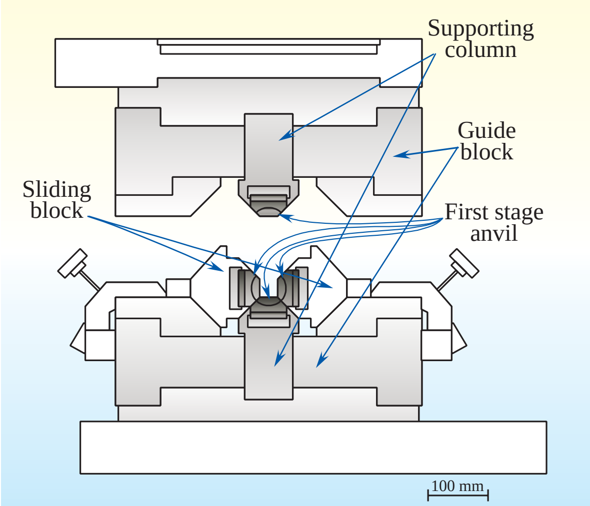

Fig. 2. Schematic drawing of the high-pressure vessel. The columns that support the upper and lower first-stage anvils (supporting columns) are separated from the main parts of the guide blocks so that the change in the relative dimension of the cubic compression space with press load can be minimized through adjustment.

The basic design of SPEED-Mk II has followed that of SPEED-1500. Namely, it is equipped with a DIA-type guide block system, a hydraulic system for maximum press load of 15 MN, a heating system, and a horizontal goniometer with a Ge-SSD for energy-dispersive X-ray diffraction.

Six first-stage anvils on the DIA-type guide block system compress a cubic space for a KAWAI-type assembly composed of eight second-stage cubic anvils, each with a truncated corner. One of the serious problems for pressure generation in such a system is that the compression space deforms with increasing press load, which may cause blow-out or breaking of SD anvils. In fact, the horizontal dimension of the compression space increases relatively to the vertical one with increasing press load at a rate of 13 μm/MN in SPEED-1500. In order to fix this problem, the columns supporting the top and bottom first-stage anvils are separated from the main part of the guide blocks so that the strength supporting these anvils can be adjusted while examining the relative deformation of the compression space (Fig. 2). By repeating adjustments, the rate of the relative deformation has been suppressed to 4 μm/MN.

In addition to the dimensions of the compression space, the press load is also controlled precisely in SPEED-Mk II. The press load is controlled with divisions of 20 ~ 30 kN in SPEED-1500, whereas it is controlled with 3 kN divisions in SPEED-Mk II. Furthermore, the hydraulic system is equipped with two inverter pumps, which rotate in proportion to the difference between the set and real loads so as to allow smooth compression and decompression.

Because of these improvements, we have succeeded in extending the pressure range generated by a Kawai-type apparatus. The highest pressure generated by SPEED-1500 is 40 GPa, whereas pressure of 63 GPa has been achieved by SPEED-Mk II (in Feb. 2004, by E. Ito, ISEI, Okayama University).

SPEED-Mk II is placed on a stage system mainly for positioning the samples to the diffraction area. The stage system has one rotation axis around the vertical direction (κ-axis) from $-7°$ to $13°$. This κ-axis is used for controlling the sample oscillation. Figure 3 shows diffraction patterns of MgO at 3 GPa and 2070 K collected with and without operating the oscillation system. Many of the MgO peaks are not observed in the diffraction pattern taken without oscillation (Fig. 3(a)). On the contrary, most of them are observed if the oscillation is operated (Fig. 3(b)). The press stage could be bent during oscillation because the press is very heavy (20 ton). Special attention was paid to designing the stage such that shifts of the sample with oscillation can be suppressed to 50 μm.

Thus, we have improved a Kawai-type apparatus for *in situ* X-ray diffraction to obtain high-quality diffraction patterns at higher temperatures and pressures. We expect that a number of important experimental results will be obtained using this apparatus.

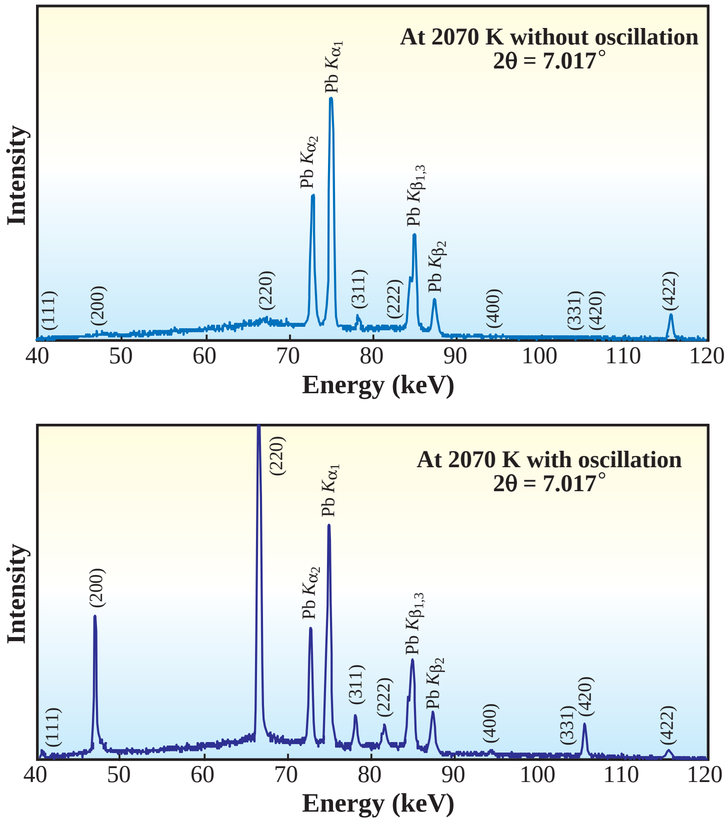

Fig. 3. Diffraction patterns of MgO with and without oscillation at 3 GPa and 2070 K: **(a)** diffraction pattern taken without oscillation, and **(b)** with oscillation. The MgO peaks are labeled by the lattice index. The peaks by Pb fluorescence are labeled as PbKα_1 and so on.

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References

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