BL02B2
Powder Diffraction

1. Introduction

BL02B2 is a bending-magnet beamline designed specifically for high-resolution powder X-ray diffraction studies of crystalline powder materials. These diffraction studies help elucidate the relationship between crystal structure and physical properties through phase identification, precise structural analysis, and *in situ* observations under different external conditions. The beamline offers monochromatic X-rays with an energy range of 12–37 keV ($\Delta E/E$ is approximately 2×10^{-4}). Since the upgrade of the powder diffractometer in FY2024, powder diffraction patterns have been captured using six one-dimensional (1D) microstrip MYTHEN detectors [1]. There are two types of experiment conducted: (i) high-throughput powder diffraction experiments utilizing a sample changer, and (ii) *in situ*/time-resolved powder diffraction experiments under various conditions. The high-throughput experiments can automatically process up to 50 capillary samples within a temperature range of 30 to 1100 K.

For *in situ* powder diffraction experiments under various conditions, additional equipment needs to be installed in the powder diffractometer. A furnace and a cryostat are available for conducting experiments at high temperatures (up to 1473 K) and low temperatures (down to 5 K), respectively. A remote gas handling system is used for controlling gas and vapor pressures within a capillary [2]. Additionally, users can conduct *in situ* powder diffraction experiments using specialized equipment they bring, such as electric field generators for ceramics, charging/discharging cells for batteries, and light irradiation systems. A two-dimensional (2D) flat panel detector (FPD), XRD3025, has been added to enhance *in situ* powder diffraction measurements using high-energy X-rays, enabling the evaluation of crystal grain size. In recent years, we have developed a high-temperature heating stage capable of reaching 1673 K (in FY2022) and a precise pressure-control cell capable of pressurizing up to 400 MPa (in FY2023), thereby further improving the sample environment for powder diffraction experiments.

Although improvements to the sample environment have enabled an increasingly diverse range of measurements, the detectors most essential to powder diffraction experiments have been undergoing progressive aging. The six 1D MYTHEN detectors on the beamline have been in continuous operation for 10 years since their installation and have consequently deteriorated due to an increase in the number of defective strips and malfunctioning power supplies. Although image plate (IP) detectors have also been used, the IP readers have shown unstable performance and required the frequent replacement of parts. However, since the introduction of FPD around FY2019, the frequency of use of IP detectors has significantly declined. On the other hand, there has been a growing demand for measurements in the high-angle region ($2\theta > 100°$) to access the high-Q region, along with increasing needs for low-angle measurements ($2\theta \sim 1°$), particularly for molecular crystals with large molecular structures. To address this situation, we have recently decided to replace our six sets of MYTHEN 6k detectors in FY2024 with eight sets of MYTHEN2 8k detectors.

2. Powder diffractometer equipped with MYTHEN2 8k detectors

To replace the detectors, the existing MYTHEN 6k and IP detectors were removed from the 2θ arm, and an arc-shaped mounting unit for the MYTHEN2 8k detectors was installed. Figure 1 shows a photograph of a powder diffractometer equipped with MYTHEN2 8k detectors. The sample-to-detector distance is 477.46 mm, identical to that in the previous diffractometer equipped with MYTHEN 6k detectors. The detectors are positioned at 10° intervals along the 2θ direction, with each module covering approximately 7.7°. Furthermore, the size of each strip is 50 μm, allowing measurements with an angular resolution of 0.006° in 2θ. To accommodate measurements in high-angle regions, the positions of the XYZ motorized stage of the beam stop and the sample observation illumination system were modified, eliminating the interference with the 2θ arm and detectors.

Next, to meet a broader range of experimental needs, several measurement methods—double-step, single-step, fine-step, and high-angle modes—were developed and implemented into the measurement program. The detector control system was developed using Python and LabVIEW. In the double-step mode, as in previous implementations, exposures are performed at two fixed 2θ positions to obtain continuous powder diffraction. The resulting data are then merged, enabling measurements across a wide angular range of 2θ = 1° to 83°. The single-step mode is a single-shot measurement method that uses an asymmetric detector arrangement at +2θ and −2θ. While the measurable 2θ range is limited to approximately 40°, this mode is suitable for time-resolved measurements. The fine-step mode involves shifting the detector by increments of 0.003°, allowing for high-resolution measurements in fine angular steps (0.003°). This method is primarily used for highly crystalline materials. The high-angle mode is an optional measurement mode that covers the 2θ range up to 140°, enabling access to a wide Q range. Note that fine-step and high-angle modes require measurement at four positions at 2θ.

Figure 2 shows the powder diffraction patterns collected using the high-angle mode. The measurement sample was NIST 660C LaB_6, sealed in a 0.1 mm capillary. The incident X-ray energy was set to 25 keV, and the exposure time was 300 s. As shown in the figure, the diffraction pattern from the sample can be observed up to 2θ = 140°, with diffraction peaks clearly visible even beyond 2θ = 100°, demonstrating the capability of this mode for high-angle measurements. This configuration enables the measurement of powder diffraction patterns with Q values exceeding 23 Å^{-1}, with the full automation of high-Q measurements using the MYTHEN2 8k detector.

This upgrade enables measurements over a wide angular range, from 2θ = 1° to 140°, and supports the analysis of crystalline materials with Q values of 25 Å^{-1} or higher. This allows not only precise structural analysis using Rietveld refinement but also pair distribution function analysis, particularly for functional materials containing light elements. Furthermore, by utilizing the fine-step mode in combination with diffraction peaks in the low-energy X-ray and high-angle regions, the angular resolution can be improved, facilitating the observation of subtle lattice distortions and other fine structural phenomena.

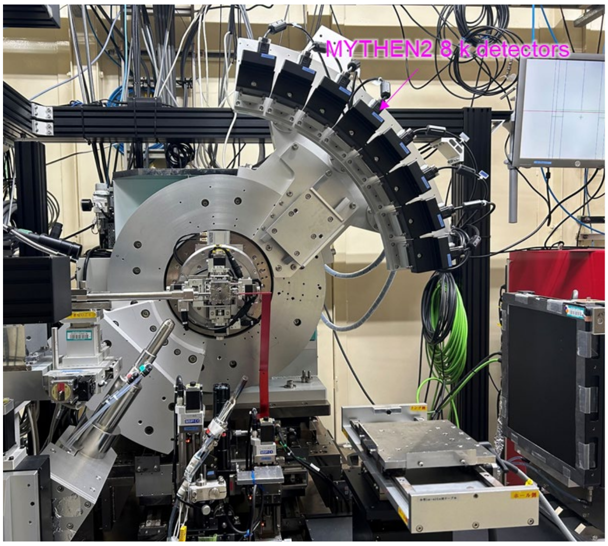

Fig. 1. Photograph of the new powder diffractometer equipped with the MYTHEN2 8k detectors at BL02B2.

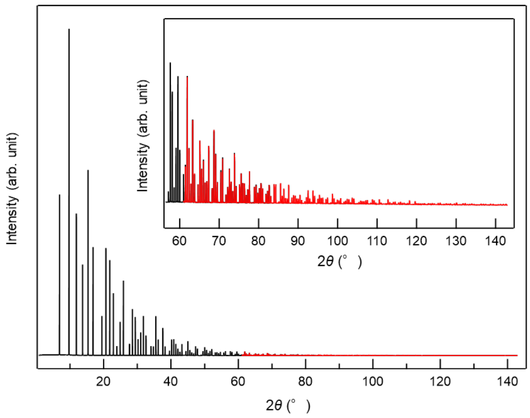

Fig. 2. Powder diffraction pattern of LaB_6. Black lines indicate the data measured in the double-step mode. The red lines show the data measured in the high-angle mode in the extended region.

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