BL13XU
X-ray Diffraction and Scattering I

1. Introduction

BL13XU is a beamline designed for the structural analyses of various materials at the atomic scale using X-ray diffraction techniques. Originally, this beamline targeted mainly the study of surface and interface structures [1]. Recently, increasing demands for advanced techniques, such as *in situ*, operando, and automatic measurements have been made by many users in various fields. To meet these requests, we integrated and rearranged the various diffraction systems in this beamline in FY2022. At the same time, we upgraded the optical components such as the monochromator and mirrors, for use with high-energy X-rays up to 72 keV.

After the upgrade, BL13XU has four experimental hutches (EHs): the multi-axis diffractometer at EH1, the diffraction measurement multipurpose frame at EH2, the high-resolution powder diffractometer at EH3, and the nanodiffraction system at EH4. With the high performance and versatility of these instruments, we offer users X-ray diffraction experiments using various types of samples under a wide range of conditions.

Below, we report the upgrades made in FY2024 on the multi-axis diffractometer (EH1) and high-resolution powder diffractometer (EH3).

2. Multi-axis diffractometer (EH1)

In recent years, there has been a growing demand for real-time and highly accurate X-ray diffraction techniques to observe structural changes under operating conditions in materials and devices during product development. In particular, there is growing interest in *in situ* observation techniques using X-ray diffraction. However, when applied to thick materials and complex layered structures, diffraction and scattering occur from the entire X-ray beam path. This makes it difficult to pinpoint the diffraction center and extract specific, target information.

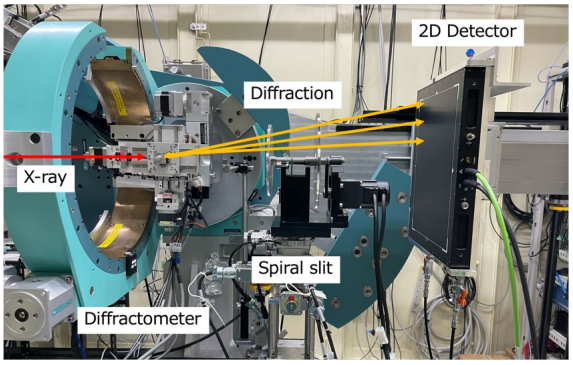

Fig. 1. *In situ* XRD observation system using spiral slits.

To address this issue, we developed an *in situ* observation system at BL13XU EH1 using a multi-purpose 6-axis X-ray diffractometer (Huber), high-energy X-rays, and a spiral slit. The spiral slit, which consists of two rotating steel plates with spiral-shaped grooves, selectively collects diffraction signals from spatially limited regions without requiring angular scanning. Suzuki *et al.* have already applied this technique to evaluate residual stress in steel materials [2]. We developed a new configuration, based on their spiral slit design, that is better suited for the *in situ* observation of a wider range of materials and devices [3]. The constructed system supports high-energy X-rays in the range of 40–70 keV, enabling the structural evaluation of dense and thick materials. To define a gauge volume of approximately 0.1 mm and to

cover a measurable 2θ scattering angle range from 2 to 20°, a 1-mm-thick tantalum plate was fabricated with 0.1-mm-wide slits. The working distance from the slit surface is 150 mm, which enhances its adaptability to diverse sample environments. We used a VAREX XRD3025 detector (detection area 300 mm × 250 mm, spatial resolution 100 μm/pixel) to cover the entire slit area.

The constructed system is shown in Fig. 1. Since the diffractometer and the 2-D detector can be controlled in tandem, the system can be used for various experimental techniques by placing the sample at the center of the diffractometer. In the future, we plan to characterize the system's spatial and temporal resolutions, and apply it to a wide range of *in situ* observation experiments, including those on structural materials, energy devices, and chemical reaction fields.

2. High−resolution powder diffractometer (EH3)

A high-resolution powder diffractometer [4] was installed at EH3 in FY2022, enabling high-throughput measurements as well as *in situ* and operando experiments. The instrument accommodates a large sample space (600 mm × 600 mm) and a load capacity of approximately 500 kg. Its 2θ arm is equipped with six 2-D CdTe detectors (LAMBDA 750k), offering high detection efficiency for high-energy X-rays. The detectors allow rapid data acquisition at energies exceeding 60 keV. The system also allows data collection beyond a Q-range of 20 Å^{-1}, making it suitable for pair distribution function (PDF) analysis. Currently, it is operational for X-ray energies between 16 and 72 keV and is used under a variety of sample environments. Standard configurations include N_2 gas low- and high-temperature blowers, enabling experiments over a temperature range of 90 to 1100 K. Combined with an automatic sample changer, up to 100 samples can be measured in sequence, including in variable-temperature experiments. In FY2023, additional sample-environment devices were commissioned, including a gas-handling system operating from 1 Pa to 130 kPa [5], a closed-cycle cryostat for powder diffraction covering temperatures from 5 to 400 K, and an electric furnace (HTK1200N, Anton Paar) capable of reaching 1473 K.

To expand the temperature range, a powder diffraction setup with a Linkam TS-1500 heater was installed at BL13XU in FY2024. Similar to the BL02B2 system, it enables heating up to ~1673 K in controlled gas using a flat sapphire cell [6]. Kapton windows reduce diffraction from sapphire/quartz under high-brilliance X-rays. The setup yields ~1 s resolution XRD data suitable for quantitative analysis.

In addition, a high-temperature powder diffraction method employing an infrared heating furnace was developed [7]. This system achieves temperatures up to approximately 1973 K with heating/cooling rates of up to approximately 1000 K/min. Radiation from a near-infrared lamp is focused onto the sample by a gold-coated curved mirror, while the temperature is monitored via a Pt/PtRh thermocouple positioned near the sample and controlled by a dedicated controller.

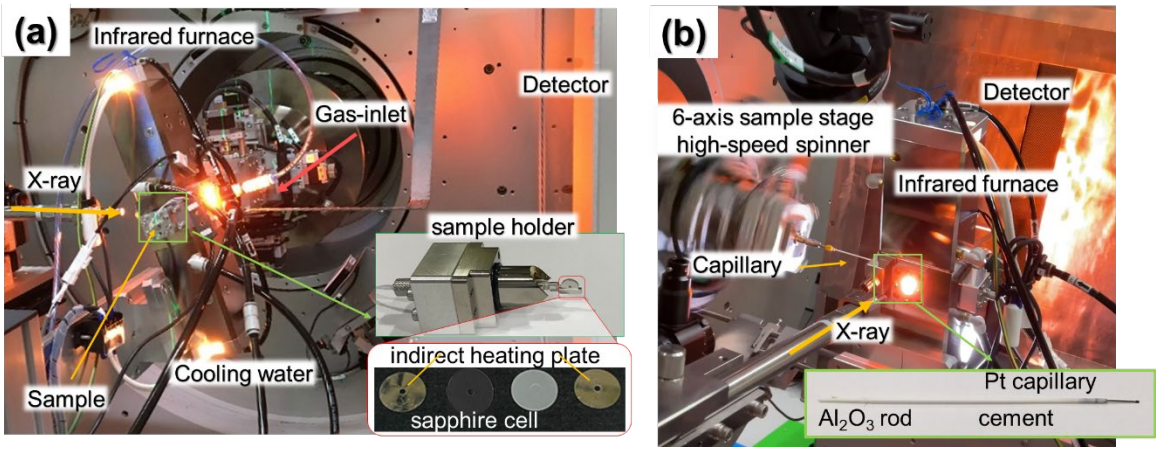

Fig. 2. (a) Infrared heating measurement setup using a flat sapphire sample cell and Pt plates for indirect heating. (b) Infrared heating measurement setup using a capillary sample [7].

The infrared furnace supports two distinct configurations. In the first configuration, a flat sapphire cell is mounted between Pt plates along with a thermocouple (Fig. 2(a)). The Pt plates, which have a 1-mm-diameter hole to allow X-ray transmission, are efficiently heated by infrared radiation, thereby indirectly heating the sample. Gas tightness is maintained, enabling experiments under controlled gas atmospheres. The second configuration, in contrast to the flat cell setup, is designed for capillary samples and includes a dedicated sample port with a 5-mm-diameter opening. Capillaries mounted on the six-axis spinner stage of the diffractometer can be easily aligned and continuously rotated during exposure. For high-temperature experiments up to 1973 K, Pt capillaries are employed. Although Pt has high X-ray absorption, the combination of a thin-walled capillary and high-energy X-rays (60 keV) yields a transmission rate of approximately 60 %. The actual sample temperature is evaluated by monitoring the thermal expansion of the Pt diffraction peaks. These sample-environment systems enable the real-time observation of both material synthesis under controlled atmospheres and structural phase transitions occurring at elevated temperatures. Further development of the powder XRD platform continues to broaden the range of *in situ* and operando experiments that can be conducted.

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