BL04B2
High Energy X-ray Diffraction

1. Introduction

Beamline BL04B2 is used in structural studies of disordered materials by pair distribution function (PDF) analysis. The PDF analysis using high-energy X-ray diffraction is useful for quantitatively determining the local structure of disordered materials with a wide *Q* range. BL04B2 is equipped with two Si crystals as a monochromator, which provides fixed-energy X-rays of 37.7 keV from Si(111), 61.4 keV from Si(220) and 113.1 keV from Si(333) (third-harmonic generation). The energy at 61.4 keV is mainly used in PDF analysis. A dedicated diffractometer for PDF analysis was developed in FY1999 [1] and the diffractometer has been operated for more than twenty years. Recently, we have developed a rapid time-resolved diffraction measurement system, using a large two-dimensional flat-panel detector, which successfully extracted crucial information on the process of crystallizing amorphous solid electrolytes for lithium batteries [2].

In FY2020, we installed a new sample changer combined with a high-temperature furnace and a fully automated alignment system on beamline BL04B2 at SPring-8. The system enables automatic X-ray total scattering measurements of up to 21 samples at various temperatures. Examples of typical measurements of X-ray total scattering and PDF analyses are also discussed to show the validity and usefulness of this system.

2. Improvement of the dedicated sample changer and X-ray total scattering measurement procedure

The new automated sample changer system (manufactured by Rigaku Aihara Seiki, Japan) can load up to 21 samples (Fig. 1). The previous sample changer could only operate with 10 samples at most and was also limited to room-temperature (RT) operation with manual alignment. Capillaries containing the samples of interest are placed in the boron nitride (BN) holders and held by the sample plate. The BN holders accept capillaries with a diameter of 1.5 or 2.0 mm. Before measurements, the sample is inserted into the furnace (HT1500, high-temperature attachment, Rigaku, Japan) from the sample plate and sealed in vacuum. Automated high-energy XRD and X-ray total scattering measurements at various temperatures (from RT to 1200ºC) can also be carried out using the same system.

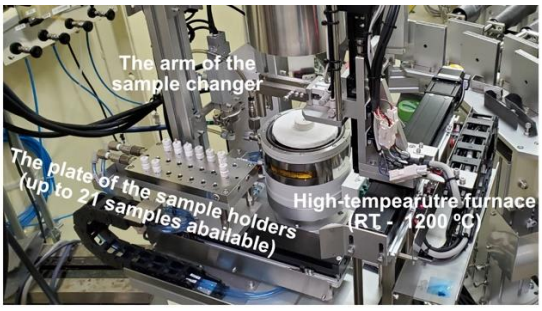

Fig. 1. Experimental setup of the horizontal diffractometer and automated sample changer with furnace in BL04B2.

The fully automated measurement system in BL04B2 is achieved by the combination of the hardware and software developments described above. The detailed operation procedure is as follows. First, the sample capillary in its BN holder is inserted into the furnace. The furnace is evacuated

to suppress any scattering from air around the sample. If heating is required, the temperature is raised until the target temperature is reached. After the temperature is stabilized, the sample is automatically moved to the center of the diffractometer. After correctly aligning the sample, the scattering pattern at low angles (up to around 8°) is obtained using the FPD detector to examine the sample quality and Bragg angle. Further analysis of the X-ray scattering data at around the angle with the highest intensity using the first detector is conducted to avoid saturation. The scattering intensity is tuned by adjusting the receiving slits. Typically, this alignment process takes around 5 min. Finally, measurements of the high-energy XRD and/or X-ray total scattering are conducted.

After measurement under one condition, the program checks the input sheet to determine whether a change of sample is required for the next measurement. If yes, the previous sample is automatically ejected from the furnace and the next sample is inserted by the sample changer. If no, the temperature is adjusted until the new target temperature is reached. By continuing this sequence until reaching the end of the measurement condition input sheet, one can obtain all the data automatically with no further input.

We conducted X-ray total scattering experiments on silicate glass at RT. The X-ray energy used in these measurements was 61.27 keV and data were obtained in the scattering angle range between 0.3° and 48°. The maximum Q (Q_{max}, where $Q=4\pi \sin\theta/\lambda$) observed was 26 Å^{-1}. The obtained data were processed by applying established analytic procedures [3] including absorption, background, polarization, and Compton scattering corrections, and were then normalized to get obtain the Faber–Ziman total structure factor $S(Q)$.

Figure 2 shows the $S(Q)$ of the silicate glass obtained using both automatically and manually collected data. The obtained $S(Q)$ is fairly consistent with the previous work adjusted manually, indicating the validation of this automated system [4]. This result is not surprising when the sample is put at the center of the diffractometer by the fully automated system.

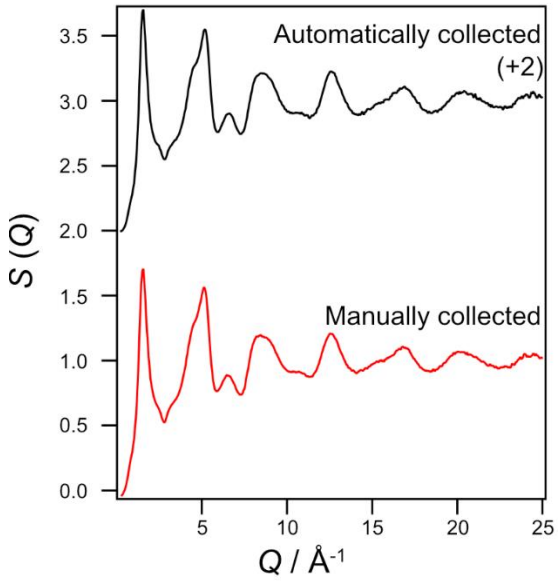

Fig. 2. Total $S(Q)$ for glassy SiO_2 up to 25 Å^{-1}. The manually collected pattern is shown in red, and the automatically collected one in black.

Koji Ohara[*1], Hiroki Yamada[*1], Saori Kawaguchi[*1], Bagautdinov Bagautdin[*1], and Michitaka Takemoto[*2]

[*1] Diffraction and Scattering Division, Center for Synchrotron Radiation Research, JASRI

[*2] Engineering Support Group, Center for Synchrotron Radiation Research, JASRI

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