BL28XU
Advanced Batteries

1. Introduction

BL28XU is managed and operated by Kyoto University for realizing technological innovations in rechargeable batteries. It was renamed “Advanced Batteries” from “RISING2” in FY2021. The RISING2 project had been running from FY2016 to FY2020 as a contract research project of the New Energy and Industrial Technology Development Organization (NEDO) to promote technology development for practical uses of storage batteries. The project exclusively used the beamline for this purpose. In FY2021, the RISING3 project was launched as a successor to RISING2. The project focuses on the two types of post-lithium-ion battery (LIB) system: (1) fluoride batteries, which show great potential in terms of both energy density and safety, and are based on highly original technologies developed in Japan, and (2) zinc-anode batteries, which offer significant safety advantages and cost benefits. From FY2021, the RISING3 project has used most of the beamtime of BL28XU for the research and development of these battery systems.

The main subjects of the current and previous projects that are being conducted in the beamline are as follows: (1) the elucidation of reaction distribution generation factors, (2) the analysis of active material reactions and nonequilibrium behaviors, (3) the elucidation of electrode/electrolyte interface phenomena, (4) the elucidation of the formation mechanism of random materials such as an electrolytic solution and electrolytes at the electrode interface, and (5) the elucidation of thermodynamic or physical instability phenomena inside the storage batteries. Measurement techniques for *in situ* observations of the reaction inside storage batteries via X-ray diffraction (XRD), confocal X-ray diffraction, X-ray absorption spectroscopy (XAS), and hard X-ray photoelectron spectroscopy (HAXPES) are mainly employed for this purpose.

2. Restructuring of control system

The main control system in BL28XU was built on *spec* software, and many features were added to it by tens of thousands of lines of programming codes beamline engineers wrote. However, after a decade of patches and modifications without consistent development policies, the source codes of the system became complicated and difficult to maintain. The customization codes relied too much on the implementation of an old version of *spec* software, and the side effects caused a big problem. Therefore, the beamline engineers started to overhaul the control system a few years ago. The new system is also based on the *spec* software (of a newer version), and most features have been migrated to the new system after its codes were rewritten in a more modern programming manner. In FY2022, the migration will be completed and the old system will be retired.

A considerable amount of the beamtime in BL28XU is spent on the operand XRD measurements of batteries in the charging and discharging processes. These types of measurement generally require high temporal resolutions. For this purpose, a photon-counting two-dimensional (2D) detector, Pilatus3 300K-W CdTe, has been employed, which captures photons in a wide range of diffractometer angles in a single shot with very low background signals. Improvements related to the technique are introduced below.

The images obtained by a 2D detector are usually converted to a one-dimensional diffraction pattern for further analysis. Previously, a user in BL28XU needed to convert them manually after XRD measurements. Recently, a conversion program written in Python has been integrated with the new control system. With this software, an XRD pattern file can be automatically generated immediately after an image is captured. The new system is also integrated with a Python server that controls Albula software (an easy-to-use 2D image viewer) so that a user can use Albula software as a live-view display. These improvements enable a user to check the quality of data on the spot. Operand measurements yield a large number of images and thus these improvements are especially beneficial.

Another improvement is a *spec* code that controls 2D detectors. The controller is implemented as an abstract "macro counter" that covers multiple 2D detectors BL28XU owns, i.e., Pixirad, ORCA series controlled by HiPic software (Hamamatu), and Pilatus (Dectris). Owing to this implementation, a user can transparently use multiple types of 2D detector for image conversion to an XRD pattern, the intensity summation of the region of interest (ROI) on the detector plane, or a live-view display.

3. Improvement of optical alignment for confocal X-ray diffraction

Confocal XRD measurement can extract an XRD pattern of limited sample depth. This technique has been proven to be powerful for the analysis of the inhomogeneous reaction process of a thick electrode [1]. The installation of a 2D detector instead of a nondimensional detector has an advantage in terms of signal-to-noise ratio because stray light which does not go through the two slits between the sample and detectors can be eliminated from the photon count in the ROI.

For this reason, in the confocal XRD setup in BL28XU, Pilatus 300K-W CdTe had been used as the detector for a number of years. A problem with this setup is that the distance of the two slits on the diffractometer arm is small because the detector is large, as shown in Fig. 1(a). When the distance is small, the observation area is widened, which lessens the spatial resolution. To increase the slit-to-slit distance, the beamline staff has recently introduced a compact photon-counting 2D detector, AdvaPIX TPX3 (AdvaCAM). The new setup is shown in Fig. 1(b). A small server program that makes the detector cooperatively work with *spec* software was also developed.

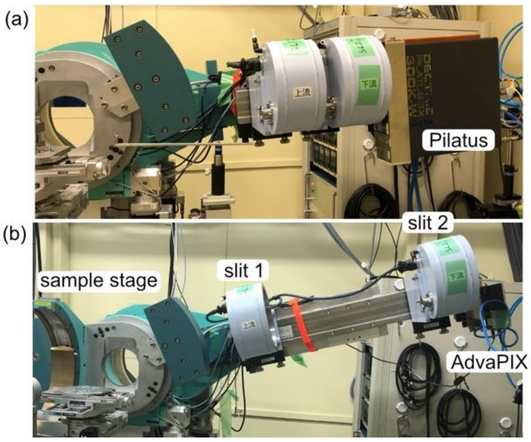

Fig. 1. Layouts of the downstream slits and a detector for the confocal XRD measurements. (a) old and (b) new setups.

To demonstrate the effect of the improvements, a 100-µm-thick silica plate sandwiched by aluminum plates was prepared. The sample and diffractometer angles, θ and 2θ, respectively, were set at an aluminum peak and scanned the sample height. The vertical beam widths of both the incident beam and downstream slits were set at 30 µm. Figure 2 shows the results of the scan in the two layouts. The width of a flat bottom in the new layout is close to 70 µm, which is the expected value when the slit-to-slit distance is sufficiently large.

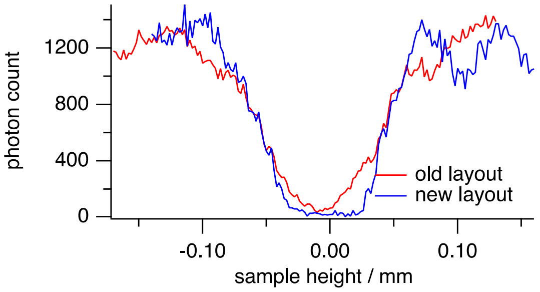

Fig. 2. Intensity of an aluminum peak as a function of sample depth. It was measured by the confocal XRD method on a 100-µm-thick silica plate sandwiched by aluminum plates.

Acknowledgments

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Reference:

[1] Kitada, K. et al. (2016). *J. Power Sources* **301**, 11–17.