

## Milliseconds Quick XAFS System using Quasimonochromatic Undulator Radiation

Time-resolved X-ray absorption fine structure (XAFS) technique is a powerful tool for investigating local structure and electronic state of materials in both crystalline and non-crystalline state during chemical or physical reaction process. Recently, dilute or thin film samples in transient reaction states have been highly interested in many research fields, such as catalysts, functional devices, environmental materials and so on. In this report, we describe the newly developed milliseconds time-resolved XAFS measurement system adapted for actual dilute or film samples in transient reaction states [1].

Two major techniques are used in time-resolved XAFS measurements for transient reactions: energy dispersive XAFS (DXAFS) and quick XAFS (QXAFS). In DXAFS measurement, polychromatic X-rays are focused on the sample by a bent-crystal, which have different energies depending on the diffraction angle. It is capable of measuring XAFS spectra with a one-shot exposure of several-tens microsecond using a position sensitive detector. DXAFS can only be used in transmission mode, which is adapted mostly for concentrated samples. QXAFS is carried out by quick scanning of a monochromator to obtain XAFS spectra within a short time, of which time resolution is seconds in most beamlines. QXAFS has a big advantage in that it can be applied to the fluorescence mode, which enables the measurements of dilute or thin film samples.

To achieve faster QXAFS, a higher X-ray flux on

the sample and faster scanning monochromator are needed. We used helical undulator radiation in **BL40XU** [2,3]. This undulator generates on-axis fundamental radiation in the energy range from 8 - 17 keV. The total power was reduced to 7 - 15 W by eliminating off-axis higher harmonics radiation in the front end without significant loss of the fundamental radiation, which greatly mitigated the heat load on the X-ray optics. Low heat load radiation allowed the cooling devices to be removed and the monochromator to be downsized.

We newly developed a compact Si(111) channel cut monochromator (size: 27 (l) × 13 (w) × 14 (t) mm). The monochromator crystal was mounted on a galvano scanner stage (**Fig. 1**) which repeatedly oscillate a Bragg angle of crystal with up to 100 Hz in the EXAFS range. The height of the exit beam from the channel cut monochromator changed during an XAFS measurement but was reduced to about 40 - 5 μm at 8 - 17 keV due to a small gap of 3 mm between the reflecting surfaces of the crystal. Detectors were the ionization chambers for the transmission mode and a photomultiplier with a plastic scintillator for the fluorescence X-ray detection. The monochromator, detectors and a sample were arranged on an experimental stage placed in an experimental hutch (**Fig. 2**). The energy at each data points was calculated from the angle of the galvano scanner, and was corrected with simultaneously measured XAFS spectrum of a standard sample using the beam

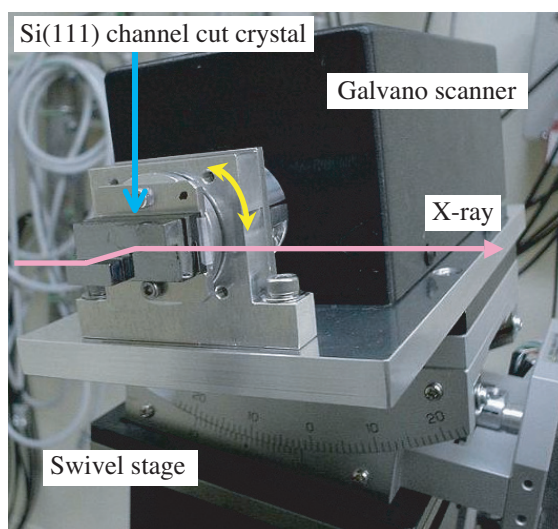


Fig. 1. Si(111) channel-cut monochromator mounted on galvano scanner stage.

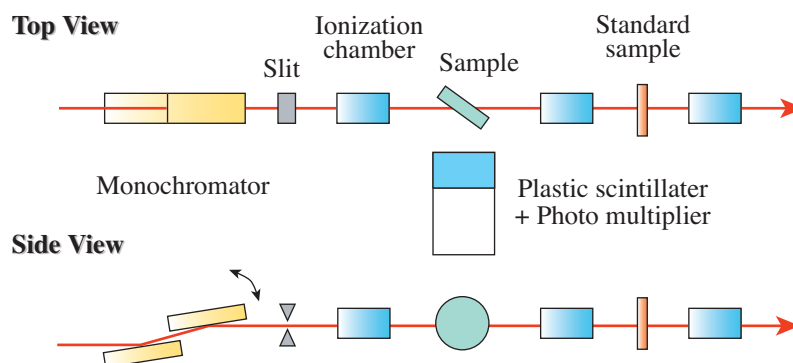


Fig. 2. Layout of monochromator, sample and detectors on experimental stage.

transmitted through the target sample.

Figure 3(a) shows an XAFS spectrum around the Pt- $L_3$  edge (11.6 keV) of Pt foil in the transmission mode together with the incident X-ray intensity. The peak of the fundamental radiation from the undulator can be set to the maximum energy of the target energy region. We set it at 12.5 keV to measure spectra with a high S/N ratio up to  $k = 15 \text{ \AA}^{-1}$ . The

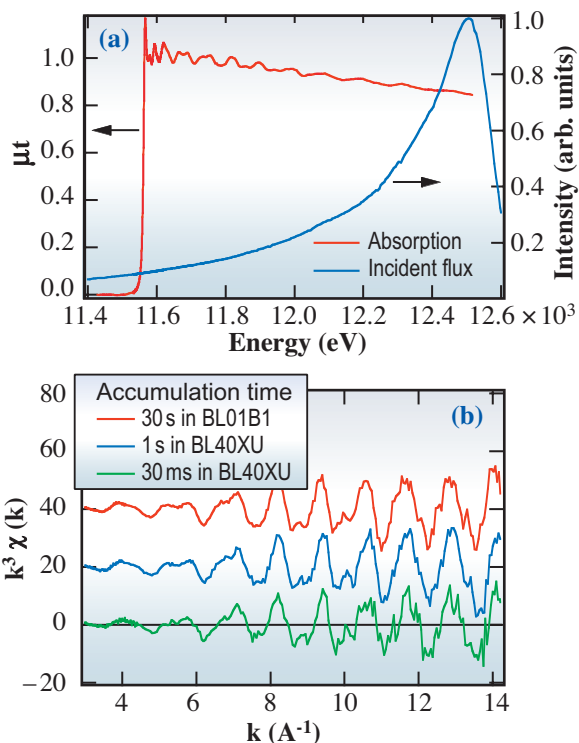


Fig. 3. (a) Pt- $L_3$  edge (11.6 keV) XAFS spectrum of Pt foil in transmission mode together with incident beam intensity, and (b)  $k^3\chi(k)$ -XAFS spectra in fluorescence mode measured in BL40XU and BL01B1.

intensity of the incident beam gradually decreased toward the lower energy region but was still higher than that of the bending magnet source in SPring-8. Such an incident beam profile of the helical undulator is adequate for XAFS measurements because the EXAFS spectrum in the high  $k$  region requires a high incident X-ray flux.

Figure 3(b) shows a  $k^3\chi(k)$ -XAFS spectra of Pt foil in the fluorescence mode measured in BL40XU in comparison with that measured at the bending magnet beamline BL01B1. The data were not corrected for the self-absorption effect. Good quality spectra were obtained in 30 msec. We have adapted so far milliseconds quick XAFS measurement system to dilute samples, such as fuel cell catalysts and automotive exhaust catalysts, with collaborators, and successfully measured spectra with the time resolution of 50 - 100 msec. The results will be published in the near future.

Tomoya Uruga\*, Hajime Tanida and Hiroshi Yamazaki

SPring-8 / JASRI

\*E-mail: urugat@spring8.or.jp

#### References

- [1] T. Uruga, H. Tanida, K. Inoue, H. Yamazaki and T. Irie: *AIP Conf. Proc.* **882** (2007) 914.
- [2] T. Hara *et al.*: *Nucl. Instrum. Meth. A* **467-468** (2001) 165.
- [3] K. Inoue *et al.*: *Nucl. Instrum. Meth. A* **467-468** (2001) 674.