A new full-field transmission x-ray microscope has spatial resolution as high as 20nm and an in situ imaging capability.

Over the past decade, technology breakthroughs in the field of x-ray optics have enabled the development of advanced imaging nanoprobes at third-generation synchrotrons. X-rays have unique capabilities in terms of resolution, sensitivity, and speed, and by combining these properties with their ability to penetrate matter, these new instruments have played an important role in the recent advent of nano-material-related research. The gap—in terms of spatial resolution—between such x-ray instruments and electron microscopes, however, still needs to be reduced. In addition, it remains a challenge to offer in situ measurement capabilities while simultaneously pushing the spatial resolution limits.

Conceptually, transmission x-ray microscopes (TXMs) are similar to optical visible light microscopes. In these instruments, tunable monochromatic x-rays illuminate the condenser—either an ellipsoidal glass mono-capillary or special type of diffraction grating known as a beam-shaping condenser (BSC)—and a Fresnel zone plate (FZP) is used as the objective lens to magnify the images or radiographs (see Figure 1). TXMs are also full-field imaging instruments, meaning that the snapshot images of absorption contrasts inside samples are acquired with 2D detectors (commonly four megapixel sensors). It is this type of full-field imaging—much faster than raster scan modes of pencil beam nanoprobes—which makes dynamic studies possible.

To take on the challenge of nano-materials science in the fields of energy storage, microelectronics, nano-porous material functions, as well as life, Earth, and environmental sciences, we have developed a new in-house TXM at the Advanced Photon Source (sector 32-ID) of the Argonne National Laboratory. This instrument has replaced an older, first-generation commercial system by providing a superior analytical imaging performance and in situ capabilities. In addition, our TXM supports a large user community. The TXM is also equipped with a rotary stage that enables image acquisitions of samples over 180° and makes it possible to perform x-ray nano-computed tomography (nano-CT). In the nano-CT process, algorithms are used to reconstruct the images of volumes (whose voxels correspond to linear attenuation coefficients).

One of the technical challenges and key design features of our TXM is the ability to perform routine operations with a flexible sample environment for in situ experiments, together with a maximum spatial resolution of 20nm (i.e., a resolving power large user community. The TXM is also equipped with a rotary stage that enables image acquisitions of samples over 180° and makes it possible to perform x-ray nano-computed tomography (nano-CT). In the nano-CT process, algorithms are used to reconstruct the images of volumes (whose voxels correspond to linear attenuation coefficients).

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of 50,000 line pairs per millimeter), as illustrated in Figure 2(a).
To achieve this, it was necessary to develop an ultra-stable design in which the degrees of freedom were reduced to the strict minimum. We thus developed, in-house, a special ultra-stiff vertical stage. In addition, we achieve angular and lateral alignment of the rotary stage with the use of a stage-free procedure. Samples are rotated with high-stiffness, low-runout, air-bearing rotary stages. As a result, the amplitude of the vibrations at the sample position can be as low as 4nm root mean square.

The FZP in our instrument also has primordial stability and is carried with long-travel-range stiff piezo-stages that are mounted upside down on a 60cm-tall granite pyramid. This pyramid overhangs the sample stack to accommodate short working distances. The high-stiffness sample stack has a load capacity of several kilograms and enough clearance around the sample to allow the handling of various environmental cells for in situ experiments. Since we began operations with our TXM, we have successfully led in situ experiments on materials under high pressure (50GPa) within diamond anvil cells, on ‘in operando’ batteries, and on still fuel cells operating at 900°C under a controlled atmosphere.

It is typical for tomogram acquisitions to last for between five and 15 minutes, with 720 or 1500 projections recorded, respectively, in step- or fly-scan mode over 180° (with an exposure time of 250–500ms). Analytical techniques that are currently available for use with our TXM include absorption (see Figure 3), phase contrast, and x-ray absorption near edge structure spectroscopy nano-tomography. We have also integrated a micro-CT module when multi-scale approaches have been required. To ensure high efficiency, high resolution, and the implementation of these various analytical techniques, we have made significant efforts to develop the necessary in-house optomechanical systems. For instance, our development of a large-aperture BSC has improved the instrument efficiency, the homogeneity of the sample illumination, and has enabled Zernike-type phase contrasts—see Figure 2(b) and (c)—for the study of low-atomic-mass elements (e.g., carbon-based materials).

Our high-resolution, 20nm-outermost-zone-width FZP (with a thickness of 0.4μm) has also been produced in-house and can be stacked with a stacking system derived from a scanning nanoprobe instrument. Once stacked in the near-field, the FZP pair behaves like a thicker single lens with an enhanced efficiency. Our TXM also benefits from sophisticated reconstruction algorithms that we implement in TomoPy (an open source collaborative framework developed at the Advanced Photon Source for the analysis of synchrotron tomographic data). With the use of these algorithms we can deliver high-quality reconstructions from noisy or a limited number of projections, and we can thus speed up data acquisitions.

In summary, we have designed and constructed a versatile nano-tomography instrument that has been proven to have outstanding stability and efficiency during operations (even when loaded with in situ cells). The integration of new algorithms with different schemes of acquisition has opened up the possibility of conducting dynamic studies with our TXM instrument. In particular, our development of optics for Zernike phase contrast imaging has enabled new types of application for carbon-based materials. Within the next few months we will be developing an FZP with a 16nm outermost zone width, which should provide a resolving power close to 10nm. In addition, the integration of TomoPy on supercomputers is part of our ongoing work. This will make such resource-hungry algorithms available for routine use.

Figure 2. (a) Absorption radiograph of a Siemens star. This image demonstrates the 20nm spatial resolution of the TXM instrument. (b) Absorption and (c) Zernike phase contrast images of a graphite electrode. (Images are provided courtesy of S. Farhad, University of Akron.)

Figure 3. (a) Reconstructed cross section and (b) 3D view of cerussite (PbCO₃) replacing calcite (CaCO₃) in a single crystal, obtained using the TXM. (Image courtesy of K. Yuan.)

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References