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Abstract

Microtomography combined with hard X-ray imaging microscopy has been employed to observe nanoscopic features in a material, which has, to date, only been done by the transmission electron microscopy (TEM). Here, we show a characteristic microstructure in an aluminum alloy, such as a slant gap between growing precipitates that impinge on each other due to the presence of a solute-depleted zone. Such observation is not possible by conventional projection microtomography even using the highest resolution available. It would appear that the present technique has a unique potential to observe nanoscopic features inside materials that are several orders of magnitude thicker than TEM specimens.

Key words: Hard X-ray, microtomography, X-ray microscopy, Fresnel zone plate, precipitate, aluminum, synchrotron radiation

The technique for measuring the spatial distribution of the X-ray attenuation coefficient is known as computed tomography (CT). As is well known, the CT technique has been employed in industry as well as medical diagnostic imaging. Meanwhile, third-generation synchrotron radiation (SR) facilities, such as the ESRF and SPring-8, have been identified as ideal for CT, providing a high-energy X-ray beam with excellent lateral coherence and monochromaticity^{1,2,3}. CT performed using SR facilities has enabled the reconstruction of volumes with a maximum spatial resolution of about 1 μm and a voxel size down to 0.3 μm ^{4,5,6,7}. Physical limitations caused by the Fresnel diffraction and the diffraction limit of visible light, prevent further improvement in spatial resolution in the ordinary microtomography⁸. X-ray imaging microscopy with an X-ray objective provides an alternative for the observation of microstructural features^{9,10}. An X-ray image is directly magnified with X-ray optics instead of being magnified after conversion into visible light using a scintillator. Soft X-ray microscopy using various reflecting and diffracting X-ray optical components has been developed primarily for the study of biological specimens⁹, prior to the development of the hard X-ray region owing to the availability of suitable X-ray objectives. Thanks to recent advances in the fabrication of high-resolution Fresnel zone plates (FZP), encouraging experiments have been conducted within the hard X-ray region, demonstrating 3D images of a diatom fossil, glass spheres and a meteorite⁸. The FZP is a circular diffraction grating with radially increasing line density. It can be used as a diffractive image-forming lens for monochromatic X-rays. The present study employs a hard X-ray microscopy setup with a FZP for the tomographic observation at SPring-8. The measured spatial

resolution of the FZP used has been reported to be approximately equal to the theoretical Rayleigh resolution¹, i.e., $\delta = 122 \text{ nm}$, given by $\delta = 1.22\Delta r_n$, where Δr_n is the outermost zone width of the FZP. An Al-Ag alloy, which generally exhibits the typical crystallographic alignment of age-hardening precipitate particles, is prepared to demonstrate the feasibility of the present technique. This would be an attempt to investigate the feasibility of applying an X-ray imaging technique to materials science and engineering.

The experiment was performed at the X-ray undulator beamline, BL47XU, of SPring-8 using the set-up shown in Fig.1. A monochromatic X-ray beam having a photon energy of 9.8 keV from a liquid-nitrogen-cooled Si (111) double crystal monochromator was used. A beam diffuser rotating at 600 rpm, which was packed with graphite powder, was used to modify the high coherence that often causes strong speckle-like noise⁸. The specifications of the FZP used are available elsewhere¹¹. The diffraction efficiency of the FZP is expected to be 10-15% at 9.8 keV. Only the first-order diffracted beam was used for imaging. The 0th-order diffraction was cut off using a center beam stop. The image detector consists of a scintillator ($\text{Gd}_2\text{O}_2\text{S:Tb}$ powder), relay lens, and a cooled CCD camera (Pixel size of the camera: $(6.45 \text{ }\mu\text{m})^2$, number of pixel: $1344 \text{ (H)} \times 1024 \text{ (V)}$). The image detector has an effective pixel size of $(4.34 \text{ }\mu\text{m})^2$.

The measured magnification of the X-ray microscope system was 49.3 times, and an isotropic voxel with an 88-nm edge was achieved in the reconstructed slices. The practical field of view was $56.8 \text{ }\mu\text{m} \text{ (H)} \times 35.3 \text{ }\mu\text{m} \text{ (V)}$. An X-ray radiograph of

the test patterns showed that 180 and 240 nm pitches were barely resolved in the vertical and horizontal directions, respectively¹¹. Image slices were reconstructed from the series of projections based on the conventional convolution backprojection algorithm.

An Al-Ag system was chosen so that the large difference in atomic number between Al and Ag would yield strong compositional contrast in X-ray imaging. An Al-10mass%Ag binary alloy was prepared by casting. The cast ingot was first soaked for 7.2 ks at 863 K followed by hot-rolling at 673 K to 92% reduction. The sample was then solution heat-treated for 14.4 ks at 813 K and artificially aged for 10.8 ks at 673 K. Small needle-like specimens were machined manually.

Figure 2 shows a 3D rendered perspective view of the whole specimen (35.3×35.3×61.2 μm), representing the spatial distribution of γ -Ag₂Al precipitates. Cross-sectional scanning electron microscopy reveals the thickness of the precipitates to be in the 200-700 nm range. These precipitates would be coherent forms of equilibrium precipitates as has been reported in the literature¹². The volume fraction, as measured by means of the 3D image analysis of the tomographic volume, is 22.5%, which seems reasonable considering the alloy composition. The regular hexagonal shape of the plate-like precipitates is clearly confirmed in the extracted precipitate image. These precipitates are seen to orient along several crystallographic planes of the matrix aluminum alloy. It is well known that the γ precipitates are hexagonal-shaped within the $(0001)_\gamma // (111)_{\alpha\text{-Al}}$ habit plane, with edges parallel to the close-packed $[11\bar{2}0]_\gamma // [1\bar{1}0]_{\alpha\text{-Al}}$ directions¹². It can be clearly confirmed that the

precipitate plates lie on all four $\{111\}$ planes in the tomographic volumes, as indicated by letters A-D in Fig. 2. The angles between the plates, which can be measured as the angle between their normal vectors, were 58.7° - 78.4° . The reason why these angles differ from the dihedral angle of an octahedron (70.53°) may be mainly the plastic deformation of specimens during sample preparation. Although its extent may also be comparatively small, there may be some effects of image distortion.

As has been reported, the γ plates lengthen very rapidly, rather than thicken, which increases the aspect ratio with increasing ageing time¹². Indeed, plate intersections are frequent, i.e., most of the precipitates are interconnected throughout the volume. Complex growth around the plate intersections is also commonly observed, presumably due to the overlap of strain fields between neighboring plates. Figure 3(a) shows several extracted precipitates where a plate intersection is observed. Plates 1 and 2 lie on the same crystallographic plane of the aluminum matrix, while Plate 3 lies on a different plane. The growth of Plate 3 is seen to be interrupted due to impingement with Plate 1. There exists a gap between Plates 1 and 3, which is clearly confirmed in the virtual tomographic slice of the same region (Fig. 3(b)). Fig. 3(b) also demonstrates the presence of a characteristic aluminum zone surrounding each precipitate that has an X-ray absorption appreciably lower than the remaining aluminum matrix. The low-X-ray-absorption zone appears to be an Ag-depleted zone, which has been reported in the literature¹². The formation of such solute-depleted zones around precipitates can be attributed to the coherent strain around the

precipitates. TEM observation has revealed the width of the Ag-depleted zone to be one or two plate thicknesses wide¹², which seems to be consistent with the present results.

It has been confirmed that the internal precipitates in the present study could hardly be observed by conventional projection microtomography even using the highest resolution set-up¹¹. For 3D observation of microstructure in engineering materials, electron tomography using TEM¹³ may be a potential approach. However, despite the overwhelming advantage of the TEM-CT technique in spatial resolution, the TEM-CT specimen thickness is severely restricted to the thickness of a thin foil, i.e., 0.1-0.5 μm ¹⁴. This precludes the use of TEM-CT for the observation of aspherical and/or coarse precipitates, such as those observed in this study. The focal depth inevitably imposes a constraint on sample size in X-ray imaging microscopy as well. For example, the actual focal depth, Δf , for the present setup is about 193 μm . The Δf value gives the maximum allowable sample diameter, which indicates that samples that are ~2000 times thicker than a thin foil specimen for the TEM observation can be observed by X-ray imaging microtomography. Thus, in-situ observation under external disturbances such as loading may very well be possible by imaging microtomography, as has been performed by projection microtomography^{1,2,6,7,15}. There are several other shortcomings of TEM-CT, such as inevitable diffraction effects and reconstruction with incomplete projection data (a specimen holder typically has a tilt range of around $\pm 70^\circ$), which cause inaccurate morphological assessment, the generation of artifacts, and so on. Overall, it can be concluded that

the present technique is clearly advantageous over other currently available 3D visualization techniques from various viewpoints.

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Figure captions list

- FIG.1 Schematic illustration of the imaging microtomography set-up with a FZP objective.
- FIG. 2 A 3-D rendered perspective view of the whole tomographic volume. The matrix aluminum has been removed to show the internal spatial distribution of θ' age-hardening precipitates, which crystallographically lie on all four equivalent $\{111\}$ planes.
- FIG. 3 Characteristic region in the tomographic volume. (a) 3-D perspective view of several precipitates extracted from the whole volume, representing the existence of a gap between a couple of encountered precipitates. (b) Virtual slice representing the existence of the gap together with a solute-depleted zone (in black) around each precipitate (shown in white). Aluminum matrix is shown in dark grey in the figure.

Fig.1 Schematic illustration of the imaging microtomography set-up with a FZP objective.

Fig.2 A3-D rendered perspective view of the whole tomographic volume. The matrix aluminum has been removed to show the internal spatial distribution of θ' age-hardening precipitates, which crystallographically lie on all four equivalent $\{111\}$ planes.

(a) 3-D perspective view

(b) Virtual slice

Fig.3 Characteristic region in the tomographic volume.

(a) 3-D perspective view of several precipitates extracted from the whole volume, representing the existence of a gap between a couple of encountered precipitates.

(b) Virtual slice representing the existence of the gap together with a solute-depleted zone (in black) around each precipitate (shown in white). Aluminum matrix is shown in dark grey in the figure.