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Deformation behavior in bulk nanocrystalline-ultrafine aluminum: in situ evidence of plastic strain recovery

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The plastic deformation behavior of bulk nanocrystalline-ultrafine Al was investigated under in situ compressive loading using high-energy synchrotron X-ray diffraction. After one loading–unloading cycle, to 2% strain, we find reversible peak broadening within the nanocrystalline grain volume and tensile residual stress (80 MPa) within the ultrafine grain volume. Upon unloading, we detect recovery of 12% of the plastic strain, and this recovery increases up to 28% at even higher applied deformations to 4%.

Keywords: Bulk samples; Nanocrystalline microstructure; Plastic deformation; Strain recovery

In the last decade significant efforts have been undertaken to determine whether nanocrystalline (nc) metals (crystallite size <100 nm) deform by dislocation movement as observed in coarse-grained metals, or by alternative mechanisms often predicted by atomistic simulations (including deformation twinning, grain-boundary sliding or rotation, and diffusion-related mechanisms) [1–4]. Recently, important observations on thin films of electrodeposited nc-Al and nc-Au [5] have shown that plastic deformation in these films is partially recoverable. These macroscopic observations follow the work of Budrovic et al. [6] in which a thin film of nc-Ni was analyzed during deformation using X-ray diffraction (XRD). They found a complete recovery of the peak broadening after loading–unloading tests, while no comparable recovery was observed for conventional coarse-grained face-centered cubic (fcc) metals.

In general, when a material is deformed the Bragg reflections detected with XRD can be affected in two ways. First, the peaks will shift in position in the presence of homogeneous (mean) strains. Second, the breadth and the shape of the peaks can change for two reasons: (i) the size of diffracting elements (e.g. the grain size) changes; and (ii) changes in the inhomogeneous strain (microstrain). Possible sources of inhomogeneous strain include lattice dislocations, grain-boundary defects and intracrystalline gradients such as high-dislocation-density walls surrounding low-dislocation-density cell interiors [7,8].

The aim of this work was to study the recovery of plastic deformation in bulk fine grained-samples using in situ high-energy synchrotron X-ray diffraction [6], along with complementary ex situ deformation studies. The experiments were performed on bimodal nanocrystalline-ultrafine (nc-UF) Al produced by consolidation of nanostructured powders (average crystallite size 38 nm), by means of spark plasma sintering (SPS) at 620 °C. This technique results in fully dense samples retaining a fine microstructure as a consequence of the short sintering time [9].

In this paper, the Al 111, 200 and 220 reflections are evaluated during in situ compressive deformation to 2% strain. The full width at half maximum (FWHM) and peak shifts of diffraction peaks were analyzed during both loading and unloading. Ex situ compression tests at 2%, 3% and 4% strain and a precise dimensional control of the sample for 30 min at room temperature (after unloading) were performed. These studies provide insight into the relationship between microstructure and plastic deformation behavior in fine (nc-UF) grained metals using bulk samples, which have to date been limited to thin films [6].

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Transmission electron microscopy (TEM) was carried out using a Philips CM12 microscope operating at 120 kV. Extensive post-mortem TEM observations after synchrotron experiment were performed in order to support and clarify the behavior observed during in situ XRD compression tests.

TEM observations reveal a bimodal microstructure (Fig. 1a) of the bulk-deformed nc-UF Al. This fact is critical when plastic deformation occurs. In coarse grains the dislocations are generated by intragranular sources, while for nc-grains atomistic simulations suggest, particularly for fcc metals with high stacking fault energy (e.g. Al), that grain boundaries act as preferential dislocation nucleation sites. Simulations of nc-Al microstructures show nucleation of complete 1/2 [1 1 0] dislocation from the grain boundaries and triple junctions. These dislocation loops terminate at the grain boundaries, which is also distinct from coarse grain behavior [10,11]. Figure 1b–d shows selected individual grains with different sizes after unloading from 2% compressive strain. Dislocations are clearly visible if the grain size is above 100–150 nm; while below this limit alternative deformation mechanisms, such as twinning, are observed. This is consistent with recent experimental observations on nc-Al and nc-Cu, where evidence of partial dislocations emission from grain boundaries was presented [12,13]. These partial dislocations interact to form deformation twins and stacking faults.

For XRD experiments a parallelepiped ($2.5 \times 2.5 \times 5$ mm^3) of bulk nc-UF Al was studied. The samples were deformed in situ using a screw-driven load frame while being interrogated with high-energy X-rays (monochromatic wavelength of 0.15359 Å and beam size of 60×60 μm) at sector 1-ID at the Advanced Photon Source (Argonne, USA). Transmission diffraction patterns where recorded from the central region of the sample using a MAR 345 image plate detector (2300×2300 pixels), mounted perpendicular to the incident beam behind the sample. By using area detector and high-energy X-rays (and associated small θ), scattering from the load and transverse directions was collected simultaneously, over a q-range covering the Al (1 1 1), (2 0 0) and (2 2 0) reflections. The sample was loaded in 100 MPa steps to 400 MPa, and then unloaded, with diffraction data collected for 30 s at every 100 MPa interval.

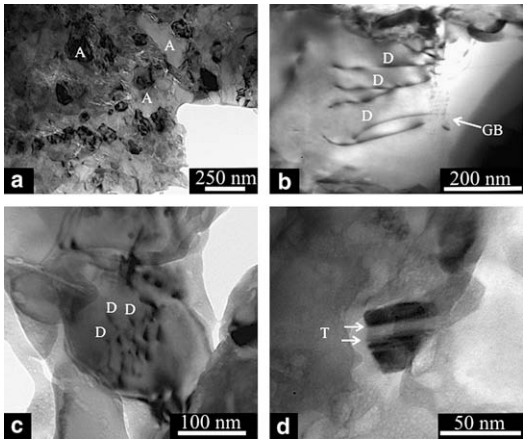

Figure 1. Bright-field (BF) TEM images after 2% strain: (a) BF TEM image shows a bimodal microstructure where some UF grains (200–350 nm) are marked with the letter A. (b) Dislocation lines in a relatively large grain (average size ≈ 380 nm). (c) Still complete, extended dislocations in an equiaxed grain (average size ≈ 150 nm). (d) Twinning and staking faults in a nc-grain (average size ≈ 40 nm).

Diffraction data were converted to TIFF images, using the FIT2D software package [14], and then integrated over five azimuthal sectors to produce 72 one-dimensional lineouts. A reference specimen of ceria (NIST SRM-674a) was used to calibrate the instrumental parameters for the integrations. Resulting lineouts were subsequently entered into the program MAUD (Materials Analysis Using Diffraction) [15], and fit using the Rietveld procedure [16,17].

A triaxial stress model (included in MAUD) with Young's modulus (E) and Poisson's modulus (ν) fixed as constants (71 GPa and 0.33, respectively) was applied to account for the first-order macroscopic strain (averaged over all grains within the macroscopic irradiated volume) induced by macroscopic external stress applied. Using the Popa approach [18], the anisotropic crystallite size (D) and microstrain ($<\varepsilon^2>^{1/2}$) were evaluated. According to the Popa model, $<R_{hkl}>$ can be considered the mean crystallite size in the crystal direction $<hkl>$, so $<R_{hkl}>$ can always be developed in a convergent series of symmetrized spherical harmonics. For fcc-Al, which belongs in the Laue group $m3m$, $<R_{hkl}>$ is expressed as:

$$< R_{kkl} >= R_0 + R_1 K_4^1(x,\phi) + R_2 K_6^1(x,\phi) \quad (1)$$

with:

$$K_4^1(x,\phi) = 0.3046972 P_4^0(x) + 0.3641828 P_4^4(x)\cos 4\phi$$

$$K_6^1(x,\phi) = -0.1410474 P_6^0(x) + 0.527751 P_6^4(x)\cos 4\phi$$

The first term R_0 corresponds to the mean crystallite size over all the **hkl** directions, $x = cos\chi$, P_i^j is the Legendre function and χ and ϕ are the polar and azimuthal angles in a crystallite orthogonal coordinate system.

The microstrain series developed for the cubic system is:

$$< \varepsilon_{hkl}^2 > E_{hkl}^4 = E_1(h^4 + K^4 + l^4) + 2E_2(h^2K^2 + h^2l^2 + K^2l^2). \quad (2)$$

In order to obtain reliable information from in situ deformation experiments on the nc-UF samples studied, it is necessary to consider several aspects, including the bimodal distribution of the crystallite size, the different deformation mechanisms that may be active during compression, the possible inhomogeneous strain and a possible preferred orientation developed during plastic deformation. In microcrystalline fcc metals the inhomogeneous strain developed during deformation can be ascribed to the plastic strain mismatch between the soft cell interior (low-dislocation-density) and hard cell walls (high-dislocation-density) [19]. On the other hand in nc-UF Al, the heterogeneity of the crystallite size may play a key role during plastic strain where UF regions deform plastically and nc regions with high grain-boundary volume fraction are still subjected to elastic accommodation. Particularly inhomogeneous strains might be generated as a consequence of extrinsic grain-boundary dislocations due to the presence of unrelaxed grain boundaries and triple junctions, especially in nanocrystalline structures [20].

Figure 2a shows the Al (2 2 0) peak profile and position at different levels of applied load. At the outset of the test