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ScienceDirect

Acta Materialia 92 (2015) 255–264

Acta MATERIALIA

www.elsevier.com/locate/actamat

An indentation-based method to determine constituent strengths within nanolayered composites

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Received 1 January 2015; revised 2 April 2015; accepted 3 April 2015

Available online 22 April 2015

Abstract—This work presents a new method to determine the flow strengths of the constituents in nanolayered composites, by coupling finite element simulations of nanoindentation with experimental hardness and micropillar compression data. This enhances the capability of separate nanoindentation and micropillar compression tests, which provide only bulk values of flow strength. This expanded capability is critical to understanding how interfaces mediate dislocation nucleation and propagation in each phase. The new method is validated using in-situ diffraction studies of deforming Cu/Ni nanolayered composites with [0 0 1] interfaces. Here, 20 nm thick Ni layers are shown to have ~3 times the flow strength of neighboring 20 nm Cu layers. These flow strengths are comparable to those for pure electrodeposited nanocrystalline ($d = 20$ nm) Cu and Ni, respectively. A Tabor factor of 2.7—often assumed in the literature—may be inappropriate if the ratio of constituent flow strengths is large (>1.5:1).

Keywords: Nanoindentation; Nanocomposite; Multilayer thin films; Finite element simulation; Micropillar compression

1. Introduction

This work presents a new method to determine the flow strengths of individual constituents within nanolayered composite materials. The approach involves coupling finite element analysis and small-scale mechanical testing. First, finite element simulations are used to determine the hardness H and compressive flow strength $\sigma_{8\%}$ (*i.e.*, at 8% strain) of nanolayered composites in terms of the constituent properties. This output is inverted so that the constituent flow strengths are known in terms of $\sigma_{8\%}$ and H or equivalently in terms of the Tabor factor, $c = H/\sigma_{8\%}$ and H. If experimental values of c and H are available, then estimates of the constituent flow strengths can be obtained.

It is well known that nanolayered composites [1,2], submicrometer-scale single crystals [3,4], and nanocrystalline metals [2,5,6] exhibit much larger strengths compared to conventional (micro-) grained metals. The near-theoretical strengths of single crystal submicrometer and nanometer-scale samples are associated with the statistical nature of a small number of dislocations and the truncation of sources in such small volumes [3,7]. Here, surfaces can mediate the nucleation, motion, and removal of dislocation content. In nanolayered composites and nanocrystalline (nc) material, however, surfaces are supplanted by interfaces and grain boundaries that serve as dislocation sources and sinks.

Further, nc metals can undergo quantized plastic events whereby a single dislocation loop can depin from a grain boundary [8] and spontaneously impart ~1% average plastic strain within a ~25 nm grain [9]. Continued plastic deformation is contingent on a succession of quantized events that percolate throughout the polycrystal. A consequence is that electrodeposited nc-Ni with a relatively weak texture is found to have a wide distribution of critical strengths [9]. In contrast, nanolayered composites can have single-crystal-like layers and orientation relationships between phases, so that specific interfaces rather than a variety of grain boundaries are involved. The interfaces can inherent dislocation content with specified Burgers vectors and line directions from one or both of the adjoining phases, in contrast to the varied content inherited by grain boundaries in nc metals. Thus, interfaces in nanolayers can incrementally transition from a more to less coherent state during deformation [10] and therefore offer a more controlled system in which to study the effect of area defects on plasticity.

Few experimental attempts have been made to quantify the effect of interfaces on the strength of constituents in nanolayered composites. At present, nanolayered composite strength is characterized primarily at the macro scale using nanoindentation, micropillar compression, and tensile testing. The few instances in which constituent strengths have been measured underscore the important role of confining interfaces. For example, recent work on

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http://dx.doi.org/10.1016/j.actamat.2015.04.002

1359-6462/

Cu-20 nm/Ni-20 nm films suggests that the biaxial yield strength of the Ni layers can decrease if the adjoining Cu layers yield [10]. Apparently, deformation in the Cu layers deposits dislocations at interfaces, which can then serve as sources of plasticity in the Ni layers. Furthermore, the Cu-20 nm layers withstand in-plane stresses in excess of 700 MPa when confined by Ni [10]. Yet, Cu-30 nm layers withstand only 400 MPa when confined by Nb-30 nm layers [11]. The geometry (film-on-substrate for Cu/Ni vs. free-standing for Cu/Nb) and method of loading differ for these cases. Yet, the results underscore that interfacial density and structure (fcc/fcc vs. fcc/bcc) control constituent strengths. An efficient method to acquire constituent strength data is needed.

The literature documents the use of micropillar compression [12–14], in-plane tension [15–17], nanoindentation [1,18,19], and in-plane X-ray diffraction [10] to determine the mechanical properties of nanolayered composites. These can furnish the uniaxial compressive/tensile flow strength σ_y, bulk hardness H, and lattice strains within each phase, respectively. If uniaxial data are not available, a common practice is to estimate $\sigma_y = H/c$, where c is the Tabor factor. This approach has several potential complications. Although $c = 2.7$–3.0 for conventional metals, it is smaller for high strength materials and approaches $\sim$1.7 when $E/\sigma_y = 10$ [20]. If there is also an in-plane tensile prestress on the film, then H and therefore c can decrease further, particularly for high strength materials [21]. Nanolayered composites can embody both conditions—high flow strength ($E/\sigma_y \leqslant 50$ [12]) *and* large thickness-averaged in-plane prestress ($\sim$0.5 GPa, based on substrate curvature measurements [22–24]). In particular, Cu-20 nm/Ni-20 nm nanolayered composites—a focus of this study—showed in-plane prestress $\geqslant$0.6 GPa, based on X-ray diffraction studies [10]. A final complication is that c can depend on whether comparable strain rates are used for nanoindentation and micropillar compression tests. Nanolayered composites can have large strain rate sensitivities [25], yet comparable strain rates have *not* been used when calculating c in Al/TiN [26], Al/SiC [27], Cu/Nb [12], and other systems.

The proposed new method to determine constituent flow strengths exploits the sensitivity of the Tabor factor c to mismatches in constituent flow properties in nanolayered composites—a finding first reported by Tan and Shen [28]. Section 2 to follow presents finite element results for the indentation hardness H of layered composites. A *forward* process is identified whereby H and $\sigma_{8\%}$ are predicted in terms of the constituent properties (elastic modulus, flow strength, and in-plane prestress for each of the two constituents) of the composite. The key observation is that H depends not only on the composite flow strength $\sigma_{8\%}$ but also the ratio of the constituent flow strengths. Section 3 presents experimental measurements of $\sigma_{8\%}$, E, H, and in-plane prestress for a Cu-20 nm/Ni-20 nm layered composite. Section 4 introduces a *reverse* process whereby the results of Section 2 are inverted so that the flow strengths of the constituent layers can be determined from bulk properties (H, $\sigma_{8\%}$), bulk in-plane stress, and constituent elastic constants. Section 5 applies the reverse process to the Cu-20 nm/Ni-20 nm system and the predicted constituent flow strengths are compared to values from in-plane X-ray diffraction data on the same system. The versatility of the method is then assessed and the strengths of 20 nm Cu and Ni layers are compared to those for nanocrystalline ($d = 20$ nm) Cu and Ni. The strength of 20 nm grains and 20 nm thick layers is found to be similar, suggesting that grain boundaries and [0 0 1] interfaces have a similar strengthening effect for Cu and Ni.

2. The forward process: finite element (FE) simulations of the Tabor factor

2.1. Finite element simulations

The 3D indentation of nanolayered composites (Fig. 1(a)) was modeled using a 2D axisymmetric geometry (Fig. 1(b)) that was discretized into a finite element mesh (Fig. 1(c)) using ABAQUS 6.11-1 [29]. To reduce computational time, the Berkovich indenter was modeled as a sharp, rigid cone with a half-included tip angle of 70.3°. This provides the same depth-to-area ratio as a Berkovich tip indenter ($A = 24.5h_c^2$) and produces load–displacement curves within 5% of full 3D FE models for the cases reported here [30]. The displacement boundary conditions $u_z(z = 0) = 0$ on the composite base and $u_r(r = 0) = 0$ on the left edge were applied (Fig. 1(b)).

The features in Fig. 1 are the result of studies to optimize computational efficiency and retain numerical accuracy. A composite radius and height of 5 μm (Fig. 1(a)) was deemed sufficient since increases to 25 μm did not change the load–displacement curve or hardness. At depths $>\sim$2 μm (Fig. 1(a)), the composite was modeled by a homogeneous medium with average constituent properties given in Eq. (4). The 300 nm × 300 nm region closest to the indenter (Fig. 1(c)) used 5 nm square, axisymmetric, four-node bilinear, displacement-temperature coupled elements (CAX4T in ABAQUS 6.11-1 [29]). Increasingly larger elements were used in the rest of the composite.

Load and contact area were recorded at each time step during the displacement-controlled simulations. The projected contact area *under load* was calculated from the contact radius—the radial distance from the left edge to the furthest node in contact with the indenter (Fig. 1(b)). Hardness was computed by dividing the load P on the indenter by the actual projected contact area (A_{actual}) measured under load.

$$H = \frac{P}{A_{\text{actual}}} \tag{1}$$

A friction coefficient $\mu = 0.1$ typical of polished metallic surfaces and diamond was used [31,32] and a non-penetrating "hard" contact was assumed in the normal direction. A lower value ($\mu = 0$) increases the pile-up and decreases H by 5–10%. This decrease is nearly independent of the constituent strength ratio for strengths typical of nanolayered composites.

2.2. Effect of constituent and average uniaxial properties

In the forward process, the Tabor factor c was determined over a range of input constituent properties. This was applied to Cu/Ni nanolayered composites with Young's moduli and Poisson's ratios $E_{\text{Ni}} = 200$ GPa, $\nu_{\text{Ni}} = 0.31$, $E_{\text{Cu}} = 128$ GPa, and $\nu_{\text{Cu}} = 0.34$ equal to bulk values. Elastic anisotropy was not observed to affect hardness significantly (<3% change across all tested strengths), consistent with the literature [33]. The average elastic properties are defined by the isostress rule-of-mixtures for a 50/50 volume fraction