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Statistical analysis of the size- and rate-dependence of yield and plastic flow in nanocrystalline copper pillars

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ABSTRACT

The effects of specimen size and strain rate on the plastic deformation response of sub-μm-sized nanocrystalline Cu pillars were examined through a series of micro-compression experiments, with particular emphasis on the stochastic nature of the measured responses. A large number of micropillars two different diameters, both with an average grain size of 6 nm, were prepared by employing the single batch process of e-beam lithography and electroplating and tested. By recourse to statistical analysis, it was recognized the yield strength and flow stress increase with pillar size and strain rate. Further, the rate sensitivity in smaller pillars was more pronounced, implying synergetic interactions between the deformed volume and the strain rate imposed. The coupling influence of size and rate on yield was analyzed by estimating the parameters in a statistical distribution having Weibull-like formula, revealing that the enhanced role of free surface in smaller pillar may make it easy to trigger yielding. The size-dependence of rate-sensitive plastic flow was also statistically examined in detail and discussed in terms of strain-rate sensitivity, activation volume, and the combined roles of free surfaces and grain boundaries.

1. Introduction

The strain-rate sensitivity (SRS) of plastic deformation in metals and alloys is an extensively researched topic, as it is essential not only for better understanding of thermally activated processes, but also for developing improved manufacturing processes such as metal forming, high-speed machining, and other dynamic processes. This is accomplished by examining the mechanical properties of the material under investigation over a wide range of strain rates, $\dot{\varepsilon}$, and SRS is expressed in terms of the parameter m which is given by $\left(\frac{\partial \ln \sigma}{\partial \ln \dot{\varepsilon}}\right)_{\varepsilon,T}$ where σ is the flow stress. The published literature suggests that m is both the intrinsic length scales such as the grain size as well as extrinsic parameters such as the experimentally variable size of the specimen. For example, an increase in m with decreasing average grain size, d, has long been observed in face-centered cubic (fcc) metals [1–6]. This trend extends even to nanocrystalline (nc) metals (with d < 100 nm) of which m values are now known as ~0.01–0.03 [2–7]. This enhancement was attributed the increased role of grain boundaries (GBs) in the plastic deformation with decreasing d [3,4]. Likewise, the dependence of m on the sample diameter, D, – higher m for a smaller D–is attributed to the enhanced contribution of free surface [8–10]. Then, it is reasonable to expect that considerable enhancement in m could occur when micro-/nano-pillars having nano-sized grains are tested. This aspect remains unexplored hitherto. Further, only limited efforts have been made for investigating the possible synergetic effects of intrinsic and extrinsic size effects on the rate dependence of deformation (i.e., for the pillars having both D < 1 μm and d < 100 nm). A previous work by Zhang et al. [9,10] reported relatively high m (~0.18) of poly-crystal Cu pillars with D~500 nm. However, their pillars had relatively larger d (110 and 180 nm). Recent works by Mohanty et al. [11] and Wehrs

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et al. [12] explored the m of nc Ni pillars with $d < 30$ nm. However, these studies do not complete the picture as only size pillars (D larger than 1.5 μm) was utilized in both the studies [11,12]. We [13] have also reported the m of nc Ni pillars (with d~12 nm), but still the used pillars had single D of 1 μm. Therefore, better understanding of the synergetic effects of both intrinsic and extrinsic sizes on the rate-dependent deformation becomes the first motive of this study.

The second motive of this study, which is perhaps relatively more important, is related to the stochastic nature of the mechanical responses measured on small-volume sample, which is imparted by the smaller number of grains in combination with the finite number of dislocation sources at the very small scale (for example, see recent review [14]). Such inevitable stochastic behavior in the analysis of the size effects on the rate-dependent deformation is, hitherto, one of the issues remaining unsolved in the literature on the micro-compression experiments of small-sized pillars. To examine this, statistical analysis of the large data is essential. However, in most of the studies, which are concerned with plasticity, only limited number of the pillars were probed, e.g., only three pillars were tested for each condition in Refs. [11,12]. In prior studies, the tested pillars were usually prepared by focused ion beam (FIB) milling, which requires long time and hence is costly. Therefore, it is economically-unviable to conduct statistically significant number of micro-compression experiments on FIB-prepared pillars. For this reason, among a variety of nano-mechanical tests, nanoindentation test has been the most popularly used for statistical analysis of the strength fluctuations (using hardness and pop-in stress data [14–22]) thanks to its merits such as simple testing procedure and easy sample preparation.

Keeping the above factors in mind, we explored the stochastic nature of size effects on the rate-sensitive deformation of sub-μm-sized nc Cu pillars (having both $d < 10$ nm and $D \leq 1$ μm) through a series of micro-compression experiments under three different $\dot{\varepsilon}$. More than ~380 pillars (with d~7 nm, and D~550 and ~1000 nm) were prepared by a single batch process of e-beam lithography and electroplating. This fabrication technique offers several advantages: First is the high throughput with the possibility of simultaneous manufacturing hundreds of pillars. Second, the produced pillars are free from any surface damage that is typically attributed to FIB milling process [23]. Last, but not the least, strong sample uniformity across each substrate can be obtained through this fabrication method. From the statistical analysis of the results, the coupled influences of both size and rate on the yielding and plastic flow of the sub-μm-sized nc pillars were discussed in terms of statistical parameters, strain-rate sensitivity, activation volume, and combined roles of free surfaces and GBs.

2. Experimental

The nc Cu pillars examined in this work were fabricated via electron beam lithography and electroplating methods [23] as following. First, the silicon substrates covered with thin Ti (~25 nm) and Au (~25 nm) seed layers were spin coated with a polymethylmethacrylate (PMMA) resist. Subsequently, arrays of circular via-holes with diameters, D, of ~550 and ~1000 nm were patterned in the PMMA film using electron beam lithography. Next, these patterned molds were filled with nc Cu by electroplating by using a commercial grade pure Cu as anode. The solution was made of sulfuric acid, Cu (II) sulfate pentahydrate, thiourea, and ultra-pure water. After electroplating, the remaining PMMA resist was removed with acetone, so as to obtain pillar arrays.

Quasi-static micro-compression tests were performed on the pillars at room temperature (RT) using Nanoindenter XP (formerly MTS; now Keysight Tech., Oak Ridge, TN) with a FIB-milled cylindrical diamond punch having a top diameter of ~8 μm. During the tests, the pillars were loaded with nominal strain rates, $\dot{\varepsilon}$, ranging from 0.0002 to 0.005/s. The morphologies of pillars were imaged before and after the micro-compression tests through scanning electron microscopy (SEM) with Nova NanoSEM 450 (FEI Inc., Hillsboro, OR). Additionally, *in-situ* micro-compression tests were performed on pillars inside a Quanta 250 FEG SEM (FEI Inc., Hillsboro, OR) using a PI 85 picoindenter (Hysitron Inc., Mineapolis, MN). The microstructure of the pillars was examined with the aid of

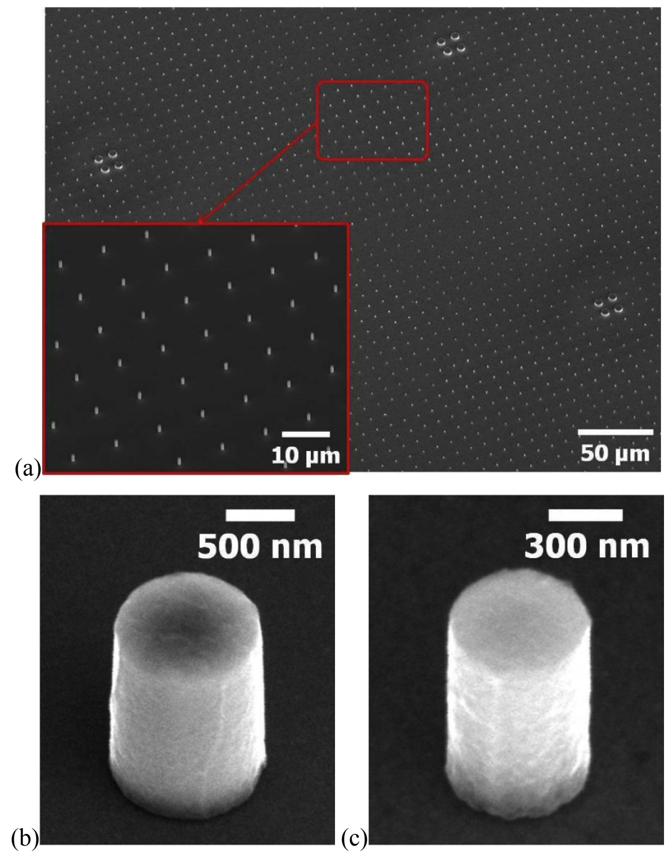

Fig. 1. Representative SEM images of prepared sample geometry; (a) electroplated pillar array; (b) morphology of as-fabricated pillar with D of ~1000 nm and (c) of ~550 nm.

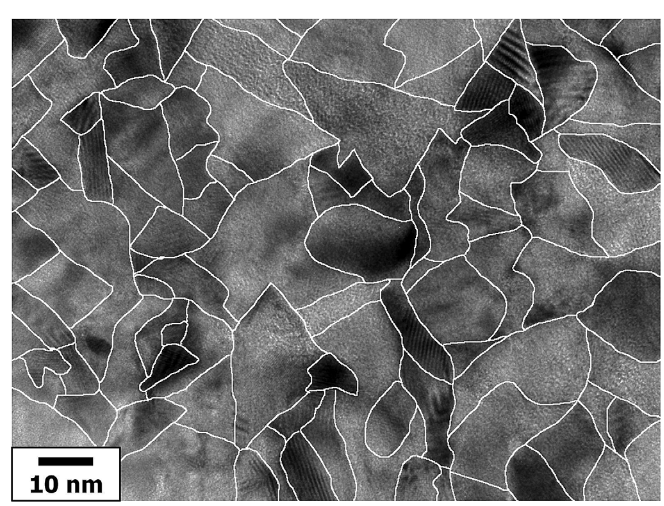

Fig. 2. Typical-high resolution TEM image revealing the nano-sized grains.