Acta Materialia 120 (2016) 1–13

Contents lists available at ScienceDirect

Acta Materialia

journal homepage: www.elsevier.com/locate/actamat

Full length article

Plasticity-induced restructuring of a nanocrystalline grain boundary network

Jason F. Panzarino [a], Zhiliang Pan [a], Timothy J. Rupert [a, b, *]

[a] Department of Mechanical and Aerospace Engineering, University of California, Irvine, CA 92697, USA
[b] Department of Chemical Engineering and Materials Science, University of California, Irvine, CA 92697, USA

ARTICLE INFO

Article history:
Received 1 April 2016
Received in revised form
16 July 2016
Accepted 14 August 2016

Keywords:
Nanocrystalline material
Cyclic micro-plasticity
Grain boundaries
Grain boundary engineering
Molecular dynamics

ABSTRACT

The grain boundary-mediated mechanisms that control plastic deformation of nanocrystalline metals should cause evolution of the grain boundary network, since they directly alter misorientation relationships between crystals. Unfortunately, current experimental techniques are unable to track such evolution, due to limits on both spatial and temporal resolution. In this work, molecular dynamics simulations are used to study grain boundary restructuring in nanocrystalline Al during both monotonic tension and cyclic loading. This task is enabled by the creation of new analysis tools for atomistic datasets that allow for a complete characterization and tracking of microstructural descriptors of the grain boundary network. Quantitative measurements of grain boundary character distribution, triple junction type, grain boundary plane normal, and other interfacial network characteristics are extracted and analyzed. The results presented here show that nanocrystalline plasticity leads to an increase in special boundary fraction and disruption of two-dimensional boundary connectivity, with the most dramatic evolution occurring in the smallest grain sizes.

1. Introduction

Nanocrystalline metals permanently deform through novel physical mechanisms [1,2] that can be attributed to an increase in the grain boundary volume fraction. As average grain size (d) is reduced below ~10 nm, a crossover from intragranular dislocation-based plasticity to grain boundary-mediated plasticity occurs, with the mechanisms of grain boundary sliding [3,4], grain rotation [5–8], and grain boundary migration [9] beginning to dominate. Because the grains themselves participate, these mechanisms are capable of altering not only grain shape but also interfacial structure during plasticity. For example, a rotating grain will change the misorientation relationship across its interfaces, potentially evolving grain boundary configurations and boundary energies [10,11].

Atomistic models, most often molecular dynamics (MD) simulations, have played a crucial role in detailing nanocrystalline deformation physics and understanding how they alter microstructure. For example, Upmanyu et al. [12] used an atomistic model of an embedded nano-grain to show how simultaneous grain rotation and grain boundary migration reduces the overall interfacial energy of the system. Rotation rates and boundary migration velocities were highly dependent on the lattice misorientation and boundary energy, with larger misorientations and higher energies leading to relative increases in the rate of each mechanism. Similar work has also been used to uncover collective plastic rearrangement in polycrystalline systems. Hasnaoui et al. [13] used MD simulations of polycrystalline Ni ($d = 5$ nm) subjected to constant tensile stress and observed an alignment of interfacial shear planes through collective grain rotation caused by intergranular slip. Rupert [14] also observed collective rotation of neighboring grains in simulated Ni ($d = 3$ nm), reporting the development of shear localization pathways which extended across nanowire samples. A more quantitative approach was recently used by Panzarino et al. [15] to study relative contributions of each mechanism during mechanical cycling of polycrystalline Al ($d = 5$ nm). These authors noted an overall reduction in grain boundary energy, which manifested as several occurrences of rotation-induced twinning as well as coalescence of low-angle grain boundaries within the microstructure. As a whole, prior work suggests that long-range evolution of the grain boundary network is likely during plasticity for extremely fine grain sizes.

* Corresponding author. Department of Mechanical and Aerospace Engineering, University of California, Irvine, CA 92697, USA.
E-mail address: trupert@uci.edu (T.J. Rupert).

http://dx.doi.org/10.1016/j.actamat.2016.08.040
1359-6454/

Evolution of the grain boundary network would open the door for grain boundary engineering of nanocrystalline materials, where the fraction of boundaries with "special" properties can be altered with thermomechanical processing [16]. One approach for identifying special boundaries is to characterize interfaces using the coincident site lattice (CSL) model [17], which assigns a value (Σ) to each interface corresponding to the inverse of the maximum theoretical number of lattice sites which are shared between neighboring crystals, deeming those with $\Sigma \leq 29$ as special. A recent example of grain boundary engineering extended to nanocrystalline metals was presented by Bober et al. [18], who utilized transmission Kikuchi diffraction (TKD) inside of a scanning electron microscope to conclude that nanocrystalline Ni thin films ($d = 23$ nm) subjected to thermomechanical cycling treatments will increase their $\Sigma 3$ boundary content as well as continually refine existing $\Sigma 3$ boundaries to a more perfect misorientation. This study confirmed the potential for altering grain boundaries through nanocrystalline deformation physics, but these authors focused on statistical boundary metrics rather than micro-mechanisms of evolution due to the inherent lack of temporal resolution provided by ex situ characterization. Kobler et al. [19] began to access time-resolved measurements by using automated crystal orientation mapping in the transmission electron microscope (ACOM-TEM) in combination with in situ straining. These authors found evidence of deformation-induced grain growth, grain rotation, and twinning/detwinning in nanocrystalline Pd films with $d = 37$ nm. However, because of the two-dimensional nature of data gathered by both TKD and ACOM-TEM, the orientation maps and boundary character distributions provided by such experimental techniques cannot confidently identify the grain boundary plane orientations (CSL only requires lattice misorientation between grains). In order to fully describe a grain boundary, the boundary surface normal (two parameters) in addition to the lattice misorientation relationship between grains (three parameters) must be determined [20].

Fortunately, since MD simulations can provide the exact positions of atoms during the entire course of deformation, all of the required information is available to extract and observe the intricacies of grain boundary network restructuring as it is occurring, on a femtosecond timescale. Currently, such data mining is still a daunting task, even with the advent of several tools and metrics capable of quantifying nanoscale microstructural features. Several researchers have constructed analysis methods which can identify grains [21–23], interface atom types [23], and even extract entire dislocation networks including boundary dislocations [24], but there are currently no tools that can extract and track grain boundary character from raw atomistic simulation data. In order to truly understand how grain boundary mediated plasticity can evolve grain boundary networks, new algorithms must be developed which are capable of segmenting interfacial regions and then extracting features like grain boundary surface normals, grain orientation/disorientation relationships, and triple junction structure, while simultaneously tracking these features as they evolve throughout a simulation. Such analysis tools would provide fully characterized five parameter grain boundary data with spatial and temporal resolution that is currently inaccessible with experimental techniques.

In order to understand how plasticity at the nanoscale evolves grain boundary networks, we use MD simulations to subject nanocrystalline Al to both monotonic tension as well as mechanical cycling. Two grain sizes, 5 nm and 10 nm, were examined so that differences in grain boundary network evolution associated with different plasticity mechanisms can be observed. In addition, the effect of temperature on boundary evolution during plasticity was also studied. In order to provide a truly quantitative analysis of the grain boundary network, we have developed several new techniques for characterizing and tracking grain boundary features and incorporated these tools into a recently developed analysis algorithm for atomistic datasets [21]. The tool developed here is able to identify all grain boundaries, triple junctions, and vertex points while also providing interface character and network connectivity information using methods familiar to the experimental community. We find that special boundary fraction dramatically increases with deformation for $d = 5$ nm, with elevated temperature and repetitive loading giving the largest changes. This evolution mainly manifests as an increase in the fraction of $\Sigma 3$ and $\Sigma 11$ boundaries, which we show is related to restructuring to find low-energy boundary configurations. This evolution is contrasted with boundary network evolution during thermal annealing, to highlight differences that are characteristic of mechanically-driven structural reorganization.

2. Simulation methods

Samples with average grain sizes of 5 nm and 10 nm were constructed using the Voronoi method with random Euler angles assigned to each grain nucleation site [4,25]. In addition, a minimum separation distance between grain nucleation sites was employed which allows for more equiaxed grains and a tight grain size distribution to more efficiently analyze the response of each specific grain size. For each sample, the same set of Euler angles was used to ensure that the starting texture and grain boundary structure would be identical despite the difference in grain size. The Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [26] was used and all atomic interactions were described by an embedded atom method (EAM) potential for Al developed by Mishin et al. [27]. This many-body potential was developed using a combination of *ab initio* and experimental data with the intention of simulating internal defects and plasticity in Aluminum and was found to accurately describe point defects, planar faults, grain boundaries, and experimental values of stacking fault energy. A 2 fs time step was used for time integration during the simulation with periodic boundary conditions applied in all directions. Any overlapping atoms which were separated by less than 2 atomic radii were removed and the final structures were then relaxed using a conjugate gradient minimization with an energy tolerance of 10^{-6} eV and a force tolerance of 10^{-6} eV/Å. The resulting structures were fully dense and free of stored dislocations, containing 48 grains. The $d = 5$ nm sample contained 180,982 atoms with a cubic simulation cell side length of 146.5 Å and the $d = 10$ nm sample contained 1,480,503 atoms with a side length of 294.4 Å. The samples were then annealed at 600 K for 100 ps to remove excess grain boundary dislocations and free volume [28–30]. This step ensures that any observed structural evolution is not merely a byproduct of unstable high-energy interfaces caused by the construction technique used to create the samples. After equilibration, samples were cooled at a rate of 30 K/ps until the desired testing temperature was reached. Although these Voronoi samples are the main focus of this work, we also compare with nanocrystalline atomistic models created using two other construction techniques that give slightly different starting microstructures. Further details of this additional analysis are provided in Section 4.3.

For mechanical testing, monotonic tension to 10% true strain and mechanical cycling were performed. Cycling was achieved through tension load-unload cycles. Specimens that had been loaded past the yield point to 5% true strain were first unloaded to 3% strain and then pulled back to 5% strain, with each unloading-loading pair representing a single cycle. This process of mechanical cycling was repeated for a total of 10 cycles. Both sets of mechanical testing simulations were run at temperatures of 300 K,