***In-Situ* Nanoscale Deformation**

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In situ TEM Straining of Nanograined Al Films Strengthened with Al_2O_3 Nanoparticles

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ABSTRACT

Growing interest in nanomaterials has raised many questions regarding the operating mechanisms active during the deformation and failure of nanoscale materials. To address this, a simple, effective *in situ* TEM straining technique was developed that provides direct detailed observations of the active deformation mechanisms at a length scale relevant to most nanomaterials. The capabilities of this new straining structure are highlighted with initial results in pulsed laser deposited (PLD) Al-Al_2O_3 thin films of uniform thickness. The Al-Al_2O_3 system was chosen for investigation, as the grain size can be tailored via deposition and annealing conditions and the active mechanisms in the binary system can be compared to previous studies in PLD Ni and evaporated Al films. PLD Al-Al_2O_3 free-standing films of various oxide concentrations and different thermal histories were produced and characterized in terms of average grain and particle sizes. Preliminary *in situ* TEM straining experiments show intergranular failure for films with 5 vol% Al_2O_3. Further work is in progress to explore and understand the active deformation and failure mechanisms, as well as the dependence of mechanisms on processing routes.

INTRODUCTION

There has been significant interest in nanograined metals and their deformation and failure mechanisms for several decades. Over this time, the processing and mechanical testing of these nanograined structures have improved significantly. Many interesting properties have been reported and attributed to a variety of mechanisms [1, 2]. For example, Wang et al. has reported that with the proper control of rolling at liquid nitrogen temperature, followed by low temperature annealing, a metal that has both high strength and large ductility can be formed. This combination of properties is not typical of either nanograined or coarse grained metals [3]. In order to understand the active mechanisms that permit this unique combination of properties, detailed *in situ* TEM experiments must be done to investigate the effect of microstructural variables within nanograined films.

The development of *in situ* TEM techniques for straining of free-standing thin films and simultaneous observations of the active deformation and failure mechanisms has progressed significantly over the last decade. A variety of *in situ* TEM straining structures used to apply mechanical load to nanostructured metals has been developed. Each of these structures provide benefits and limitations that should be considered when developing an *in situ* experiment to elucidate certain mechanisms [4, 5]. This study will emphasize a custom-made straining structure that was specifically developed for this study based on previous straining structures in the literature [6, 7].

Previous studies using a similar pulsed-laser deposition (PLD) technique coupled with *in situ* TEM straining structures revealed interesting results in PLD Ni thin films of various microstructures [8]. In post-mortem analysis, the straining of nanograined, ultra-fine grained, and bimodal PLD Ni films of nominally 80 nm thickness demonstrated three different, corresponding fracture surfaces: The nanograined films showed a smooth fracture surface with little plasticity, the ultra-fine grained films showed a jagged fracture surface with localized plasticity, and the films with a bimodal grain size distribution showed a mixture of the two. In the latter case, the fracture surface was smooth when it progressed through nanograined regions, but showed necking, dislocation pile-ups, and twinning when the fracture path encountered large grains. Additional observations during *in situ* straining showed further details of the active mechanisms during deformation [8], and thus validated the use of simple TEM straining structures for understanding failure mechanisms.

In a separate study, ultra-fine grained Al sputter deposited and electron-beam evaporated thin films were found to be columnar in nature with deformation and failure dominated by grain boundary grooving. In these films, which were deposited on Si and made free-standing by a series of microfabrication steps, the surface was observed to have significant surface roughness. *In situ* TEM straining experiments of the ultra-fine grained Al films resulted in intergranular failure with limited to no plasticity throughout the film. All of the observed plasticity occurred directly ahead of the crack tip. Large grains in these films acted to hinder crack progression and showed limited dislocation activity and twinning. The intergranular failure was inhibited by increasing the film thickness, which decreased the percentage of stress-riser to film thickness ratio [8]. The insight gained from *in situ* TEM straining experiments of high purity nanograined PLD Ni and ultra-fine grained Al samples provides two experimental parameters to compare with this investigation into nanograined Al-Al_2O_3 films in which greater complexity in the active mechanisms is expected.

EXPERIMENT

Nanograined Al films strengthened with Al_2O_3 precipitates were prepared using PLD. Using two alternating sources, mixed films of Al and Al_2O_3 were deposited onto polished NaCl substrates with final compositions of 1, 5, and 10 vol% Al_2O_3. For each volume percent, three deposition routes were used varying the number of deposition cycles (5, 10, or 20). In total 9 films were deposited, each with a final thickness of 100 nm. Based on past experiments, the films are assumed to show some columnar nature and be predominantly [111] in texture with some surface roughness. To coarsen the Al_2O_3 particles that were precipitated during deposition, and to additionally grow the grain structure to facilitate *in situ* TEM observations, the films were annealed in a vacuum furnace. Anneals took place for 1 hour at 300, 400 and 500 °C at 1 x 10-5 Torr.

To prepare free-standing thin films for straining, the following preparation steps were followed, see Figure 1. First, sacrificial straining structures were prepared by cutting thin sheets of 301 stainless steel into 2.4 x 11 mm rectangles, drilling holes at each end, and using a diamond saw to cut a small slit from edge-to-center at the middle of each rectangle, Figure 1a. Next, the above described films of PLD Al-Al_2O_3 deposited onto NaCl substrates were cleaved into rectangular blocks, approximately 1.5 x 5 mm in cross section. Each NaCl block with PLD film attached was glued using M-bond, film side down, to a straining structure, Figure 1b. After the M-bond had cured at room temperature, the NaCl substrate was dissolved in a bath of deionized (DI) water, Figure 1c. A series of steps were then taken to remove the dissolved NaCl from the DI water by pipetting out most of the fluid, refilling with DI water, and repeating. In the final steps, ethanol was added to the solution, instead of DI water, to reduce the surface tension of the fluid and prevent rupture of the free-standing thin film upon drying. After these steps, the film was removed from the solution with tweezers and held in air to dry prior to straining in the TEM. An actual straining structure with an 80 nm-thick PLD film arrowed is shown in Figure 1d.

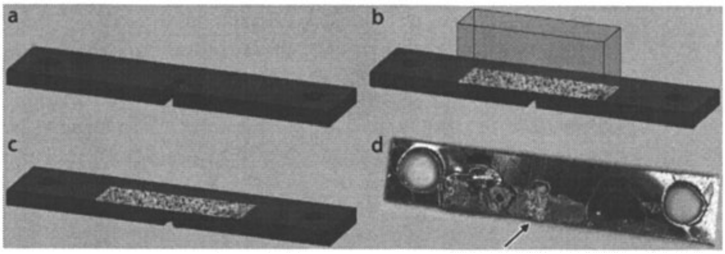

Figure 1. a) Sacrificial straining structure; b) NaCl with deposited film attached to straining structure; c) straining structure with the NaCl removed, leaving the film adhered to the structure; and d) an example of an actual structure with an 80 nm-thick PLD Ni film attached to a straining structure.

DISCUSSION

The as-deposited microstructures for Al-Al_2O_3 films were characterized using plan-view bright-field TEM over a range of vol.% Al_2O_3 and number of deposition cycles. The films were all nanograined, but did not all contain identifiable particles. No particles were seen in the case of the 1 vol.% films, and the particles are sparse in the 5 and 10 vol.% films.

In order to develop microstructures with observable Al_2O_3 particles and larger grain sizes to facilitate TEM imaging and analysis, a series of annealing experiments were performed for each film: 300, 400, and 500 °C for 1 hour. An example comparison of a 1 vol.% Al-Al_2O_3 film deposited using 10 deposition cycles is shown in Figure 2 for (a) an as-deposited and (b) a post-annealing microstructure. Here it is clear that the annealing experiment both increased the number of observable precipitated Al_2O_3 particles and the average grain size of the film. The average grain size in the as-depostied film, Figure 2a, was 31 nm with no distinguishable Al_2O_3 particles. After annealing, Figure 2b, the average grain size was found to be 110 nm and the average particle size 9 nm. Over the range of the 18 post-annealing films analyzed, the average particle size ranged from 3 to 12 nm and the average grain size from 30 to 110 nm.

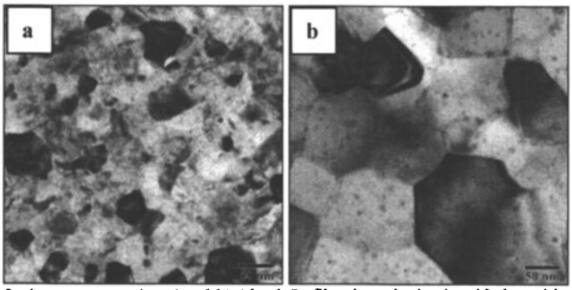

Figure 2. Examples of microstructures in a 1 vol.% Al-Al_2O_3 film deposited using 10 deposition cycles. (a) As deposited microstructure, and (b) microstructure after annealing 1 hour at 500 °C showing increase in grain size and development of Al_2O_3 particles.

Shown in Figure 3 is an example matrix of microstructures showing the relationship between vol.% Al_2O_3 and annealing temperature. All films were deposited using 5 layer pairs. This figure shows a few key features: In films with lower vol.% Al_2O_3, the final grain sizes were larger (compare, for example, g, h & i). This is likely due to fewer precipitates present, and thus fewer obstacles pinning the movement of grain boundaries during annealing. Additionally, anneals at higher temperatures led to larger grain sizes, as well (compare, for example, a, d, & g). This is due to increased mobility of grain sizes, as temperature is increased, resulting in faster grain growth rates. Microvoids are observed to form at grain boundaries and triple points in 1 vol% films annealed at 500 °C for one hour. This is associated with the rapid growth at 0.83 of the melting temperature and no Zener drag effect and will serve, as the limit for thermal processing of these films. Finally, in all 9 films, Al_2O_3 precipitates were observed after annealing. These observations for films deposited with 5 deposition cycles are consistent with films deposited using 10 and 20 deposition cycles.

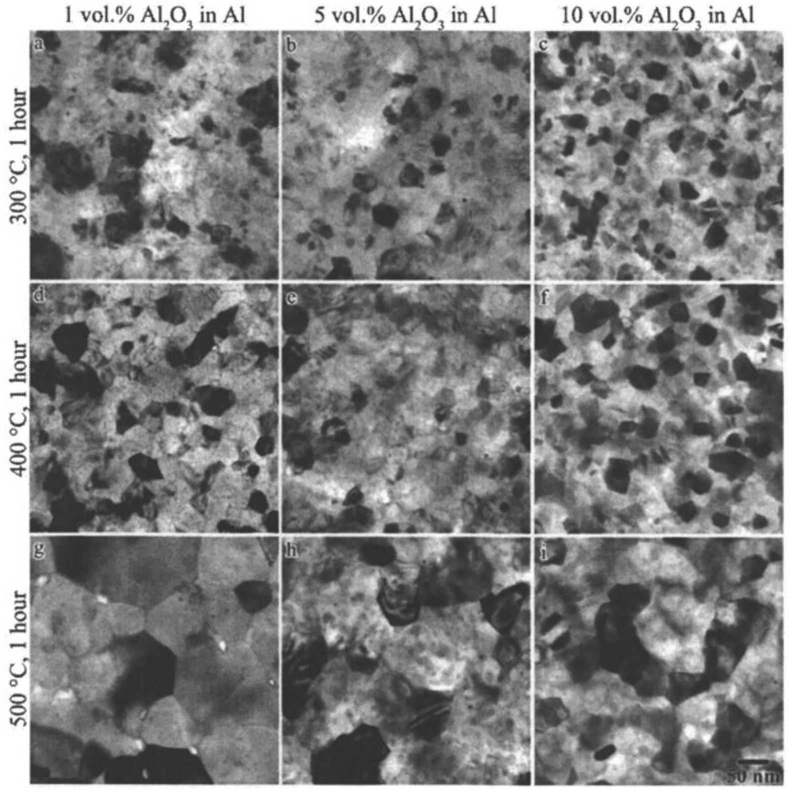

Figure 3. Post-annealing microstructures formed in Al-Al_2O_3 films deposited using 5 layers pairs showing relationship between vol.% Al_2O_3 and annealing temperature.

Following the development of these microstructures and preparation of free-standing straining samples, preliminary *in situ* TEM straining experiments were performed. Figure 4 shows a 1 vol.% Al-Al_2O_3 film, deposited using 10 layer pairs and annealed 1 hour at 400 °C during *in situ* TEM straining. Although the crack path was intergranular, which is associated with brittle fracture, microcracking ahead of the crack tip and dislocation emission during crack opening indicate the presence of toughening mechanisms in the film.

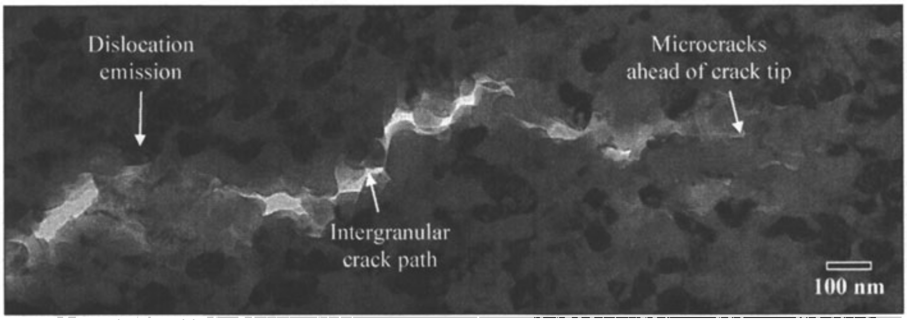

Figure 4. Crack formed in a 1 vol.% Al-Al_2O_3 film (deposited using 10 layer pairs, annealed 1 hour at 400 °C) during *in situ* TEM straining.

Future work in the Al-Al_2O_3 system will include additional annealing experiments and *in situ* TEM straining experiments to understand the link between structure and mechanical properties in nanoparticle-strengthened, nanograined thin films over a range of controlled microstructures. The aim will be to develop stable microstructures that enhance the ductility and toughness of these films and determine the underlying mechanisms that operate in these microstructures resulting in enhanced properties. In addition to the investigation into Al-Al_2O_3, future work will include understanding the effects of particle type (e.g. soft vs. hard, coherent vs. incoherent) and the behavior of more complex systems (e.g. multiple constituents, multiple particles/phases, solute) on the active mechanisms and resulting properties.

CONCLUSIONS

In situ TEM investigation into the deformation and failure mechanisms in nanograined and ultra-fine grained metals was pursued using a custom-built straining structure. Details of an initial investigation into the deformation and failure mechanisms in free-standing PLD Al-Al_2O_3 films were presented. Thermal processing was used to tailor the Al_2O_3 particle size from 3 nm to 12 nm and the Al grain size from 30 nm to 110 nm. The initial results suggest that failure in the 5 vol% Al_2O_3 in Al films is predominantly intergranular with limited signs of plasticity. Further work is underway to understand the effect of various particle sizes and interfaces in nanograined alloys.

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Poster Session: *In-Situ* TEM