Advanced aging study in Pd
In-situ 3D characterization of He bubble and displacement damage in dense and nanoporous thin films

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Abstract

This initial work attempted to determine the feasibility of using advanced in-situ, electron tomography, and precession electron diffraction techniques to determine the structural evolution that occurs during advanced aging of Pd films with nanometer resolution. To date, significant progress has been made in studying the cavity structures in sputtered, evaporated, and pulsed-laser deposited Pd films that result from both the deposition parameters, as well as from He ion implantation. In addition, preliminary work has been done to determine the feasibility of performing precession electron diffraction (PED) and electron tomography in these type of systems. Significant future work is needed to determine the proper conditions such that relevant advanced aging protocols can be developed.

Introduction

Understanding the structural evolution that occurs in various Pd microstructures as a function thermal environment, He gas evolution, and radiation induced displacement damage is of important in predicting materials aging in complex or extreme enviroments. This approach will utilize in-situ transmission electron microscopy (TEM) and electron tomography to provide real time nanoscale observation of the various Pd structures response to heat, He implantation, and combinations thereof that are complimented by sequential tomographic tilt series. The goal of this work was to determine the utility of advanced in-situ TEM techniques to determine the structural evolution that occurs in Pd films as a function of temperature and He implantation.

Approach

All of the Pd films utilized in this study were directly sputter deposition, electron-beam evaporation, or pulsed-laser deposited onto optically polished NaCl using to thickness up to 100 nm. The substrates were then dissolved in deionized water and the placed on Cu TEM grids. These in-situ TEM He implantation experiments were performed at the In-situ Ion Irradiation TEM facility at Sandia National Laboratories [1]. The He ion were implanted into a TEM sample mounted in a JEOL 2100 TEM operated at 200 kV using a 10 kV Colutron accelerator that are both part of the I^3TEM facility. The in-situ TEM annealing was done utilizing a Phillips CM30 TEM operated at 300 kV. All micrographs were processed using standard image characterization techniques. The precession electron diffraction technique was captured using the ASTAR/Nanomegas system and the electron tomography was collected by manual tilt series and reconstructed using the eTomo software set. Both of which are installed on the JEOL 2100.

Results and Impacts

This study was able to show that adequate TEM samples could be prepared for in-situ TEM studies of the microstructural evolution of Pd films via sputter deposition, electron-beam evaporation, and pulsed-laser deposition. In addition, this study demonstrated and combination of in-situ TEM He implantation and annealing studies can be utilized to observe the formation and evolution of gas bubbles in the Pd thin films despite the potential surface effects. Finally, the benefits of electron tomography and precession electron diffraction for the detailed characterization of these materials were demonstrated.

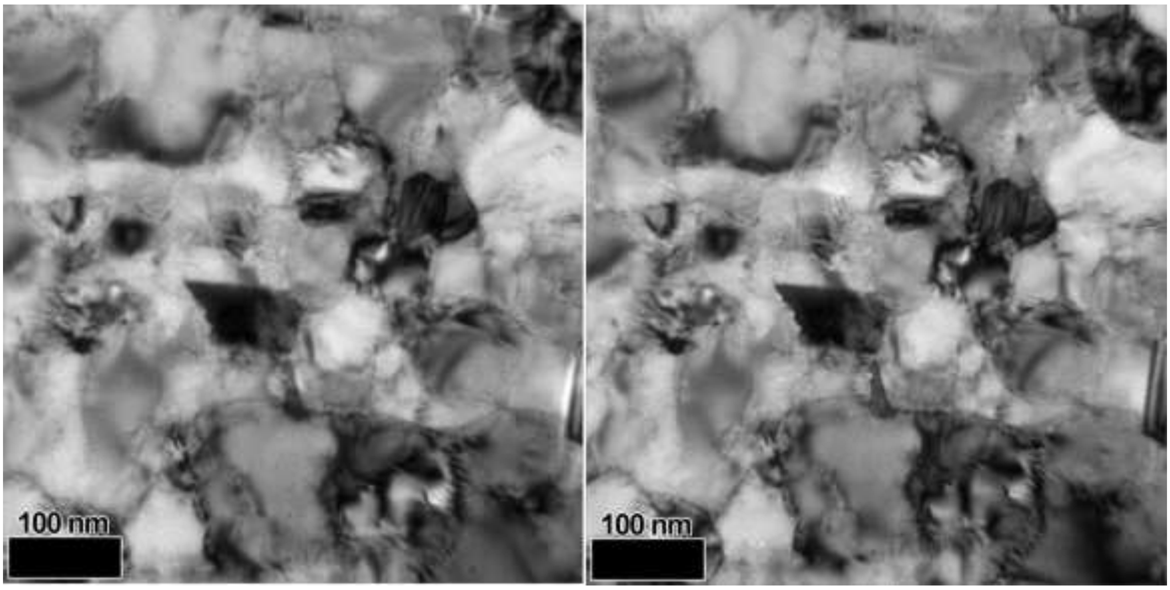

Figure 1. Sputter deposition alone produced significant porosity in the Pd films. a) +1000 nm defocus results in the voids showing up as black. b) -1000 nm defocus results in the voids showing us as white.

The three deposition techniques resulted in very different structures of the thin Pd films. This study will not focus on the variation in grain size, texture, grain boundary character although information on all of these were collected during this study through the use of precession electron diffraction. An important difference between the films was the porosity observed in the sputter deposited films that were not present in the evaporated or pulsed-laser deposited films. The presence of the nanoscale pores can be observed in the through focus images seen in Figure 1. Cavity formation in the annealed sputtered films without He implantation suggests incorporation of gas from the sputter deposition process. This dissolved noble gas likely affects the behavior of He in the film, and may have implications for any component made using this technique. Previous work has shown that the nanoporous structure can be tailored extensively in Pd by controlling the deposition parameters [2]. He implantation was performed on the as-deposited sputtered samples in-situ in the TEM to observe microstructural changes in real time. During this real time observation, no new pores were clearly observed to form. However, after 1 month of aging the sample in a standard lab environment, the same sample was found to contain a high density of small cavities. Unfortunately, the size and distribution of the pores present in the sputter deposited films was on the order of those produced from He implantation making it difficult to discern the source of each cavity structure. As such, the remainder of the study focused on the full dense films produced by evaporation and pulsed-laser deposition.

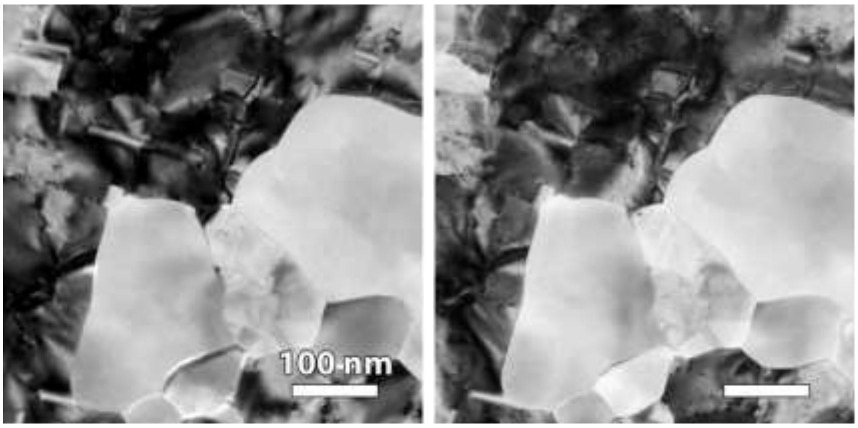

Figure 2. Pulsed-laser deposited Pd films a) as deposited and b) after implantation with 10 keV He.

In contrast to the sputtered deposited films in which the same stock material was annealed for a study in coarser grained material; cavities were found after annealing, prior to He implantation; The pulsed-laser deposited films did not exhibit bubble formation after either He implantation or annealing, as can be seen in Figure 2. The experiment by pulsed laser deposited films, avoids the gas incorporation of sputtering process. Unlike those shown previously, annealed pulsed-laser deposited films did not have cavities. It is suspected that this is possibly due to either the implant dose being lower than used in the sputter films or the density and type of grain boundaries present in the pulsed-laser deposited films providing larger number of sinks, improved diffusion path, or both. After implantation with He, the same areas were examined daily for 3 weeks to check for microstructural change.

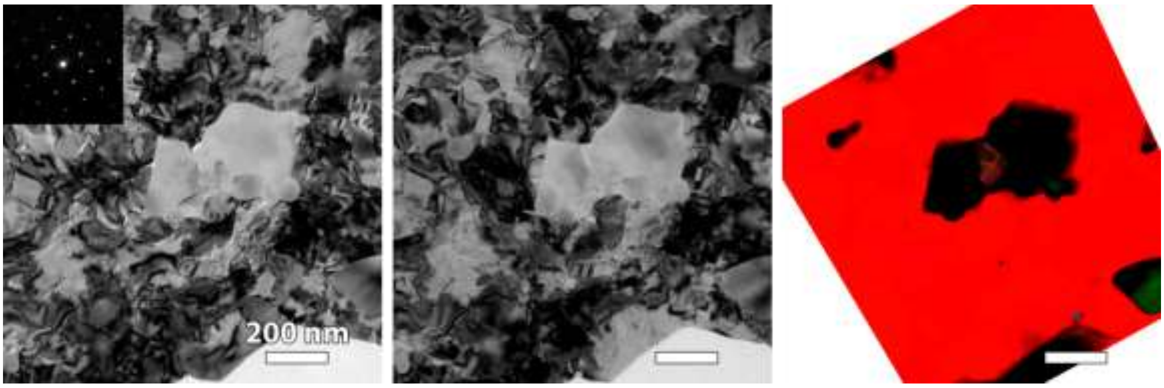

Figure 3. a) Nanostructure of the He implanted film directly after implantation with a SAD insert taken from the large grain present in the center of the image. b) The identical region located in the same sample aged at room temperature and lab environment conditions for 1 month. c) Initial PED investigation revealed through phase mapping significant contamination (regions in black).

Cavities were not observed during any of these observations, suggesting that the dissolved gas content or other unknown factors play a significant role in cavity formation. A newly installed automated orientation mapping capability (similar to EBSD in the SEM) was used to analyze the microstructure on the grain level. This technique called, precession electron diffraction, is a relatively new technique in the electron microscopy field that provides a range of information on phase, microstructural texture, local strain, and grain boundary information [3]. We have previously used it to determine the local phase distribution in a bi-phase PLD Ni sample [4]. In this study, precession electron diffraction was used to try to elucidate factors that might be playing a role in the lack of bubble formation that was observed. During this study, orientation mapping revealed phases that could not be matched to Pd, suggesting contamination of some kind, and necessitating new material to study.

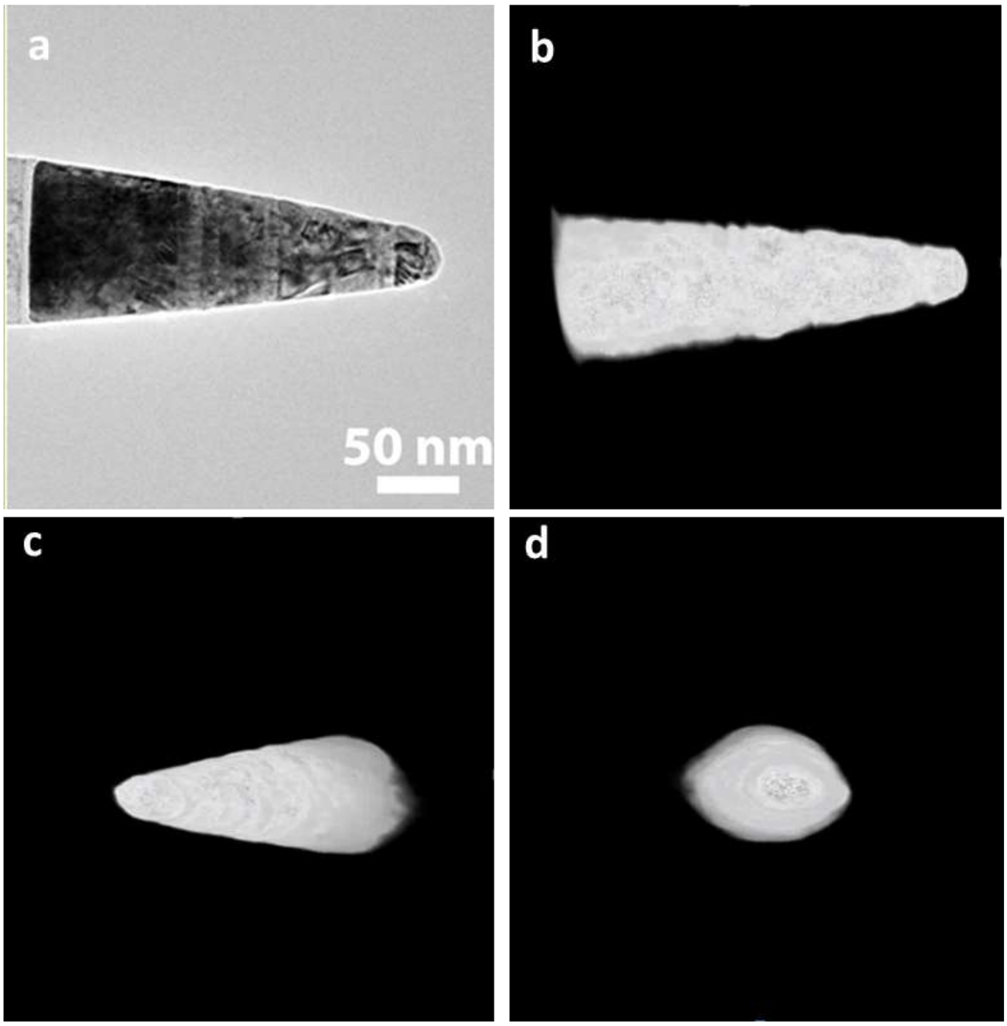

Figure 4. a) Bright Field image of a metal pillar produced by in-situ FIB sample preparation. b-d) Still frames taken at different tilt angle. (b) is taken at a different θ tilt. (c and d) are taken at two different ϕ tilts.

In a similar way to how precession electron diffraction, provides insight into phase, texture, and grain boundary information, electron tomography provides insight into the 3D structure of the sample. Electron tomography is a computational and image intensive electron microscopy technique that provides the ability to reconstruct 3D nanoscale images from a 2D electron micrograph [5]. In Figure 4, an image set taken from a 3D reconstruction model shows the surface and internal structure of a metal nanopillar. It is the hope that continued work in this area will permit full 3D reconstruction of the cavity distribution. Work by other groups have shown the ability to characterize the 3D structure of both deformation and radiation damage in metal sample [6,7]. No work known to the author has looked at internal void distribution using electron tomography. If continued, this will be the focus of follow-on research.

Conclusions and Future Work

Understanding the aging of key material systems is important, but is often time constrained. Accelerated aging of complete components can often overlook critical microstructural phase changes or microstructural evolution. In this work, we have shown In-situ TEM annealing and ion implantation is a potentially feasible method to investigate the role of accelerated on the nanostructure of a material, but will be highly system and process dependent. In addition, both precession electron diffraction and electron tomography can provide significantly more information about the microstructure composition and 3D structure than standard TEM alone. Significant further work is needed to determine the proper in-situ TEM accelerated aging conditions of interest to limited lifetime components of interest to Sandia.

Summary of Findings and Capabilities Related to Aging

No findings relevant to specific component/material aging or capabilities were obtained this year.

References

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