Materials Science & Engineering A 675 (2016) 110–119

Contents lists available at ScienceDirect

Materials Science & Engineering A

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

Initial texture effects on the thermal stability and grain growth behavior of nanocrystalline Ni thin films

Josh Kacher [a,*], Khalid Hattar [b], Ian M. Robertson [c]

[a] Georgia Institute of Technology, Atlanta, GA, United States
[b] Sandia National Laboratories, Albuquerque, NM, United States
[c] University of Wisconsin, Madison, United States

ARTICLE INFO

Article history:
Received 27 May 2016
Received in revised form
9 August 2016
Accepted 11 August 2016
Available online 12 August 2016

Keywords:
Abnormal grain growth
Nanocrystalline
In situ transmission electron microscopy
Transmission Kikuchi diffraction
EBSD

ABSTRACT

Nanocrystalline Ni thin films with different as-deposited textures were synthesized by varying the substrate temperature during pulsed-laser deposition. The influence of initial texture variations on the thermal stability and annealing behavior of the thin films was studied via *in situ* transmission electron microscopy annealing and transmission Kikuchi diffraction analysis. It was found that, as the substrate temperature during deposition was increased, the initial microstructure varied from a random distribution of grains to one almost entirely composed of (001) – oriented grains, with the volume fraction of (001) – oriented grains increasing with increasing substrate temperature. The (001) – oriented grains increased thermal stability and constrained grain growth during annealing, resulting in a bimodal grain size distribution in the annealed films. Despite different surface orientations after annealing, the misorientation distribution functions for the films were similar, suggesting that the boundary state is a primary factor in dictating equilibrium.

1. Introduction

With their promise of superior properties such as improved mechanical strength [1–3] and fatigue resistance [4,5], nanocrystalline materials continue to be an area of active research. However, due to the high volume-fraction of grain boundaries in comparison to coarse-grained materials, these materials tend to be unstable even at low homologous temperatures, resulting in significant grain coarsening over relatively short time periods [6–15]. Efforts to understand this growth, including strategies to stabilize nanocrystalline structures or promote the development of favorable microstructures, promise to provide greater control in the development and application of nanocrystalline materials [16–18]. For example, experimental work has shown that inducing a bimodal grain size distribution [19] or generating a high density of nanotwins [20,21] can increase the ductility of a sample while still maintaining the strength associated with nanocrystalline materials.

Extensive work using tools such as *in situ* and *post mortem* transmission electron microscopy (TEM) characterization [7,8,10,11,22], transmission Kikuchi diffraction (TKD) analysis [23], and x-ray characterization [6] has shown that grain growth in nanocrystalline materials proceeds in an abnormal manner, with individual grains growing at rates many times higher than grains in the surrounding nanocrystalline matrix. These studies have also demonstrated that grain growth in thin films stagnates at smaller sizes than what is seen in bulk materials. The reasons for the stabilization of the microstructure, that is, the stagnation of grain growth, is still being debated [24], though possible factors include: solute particle pinning [25,26], surface and texture effects including grain boundary grooving [27–32], relative grain boundary mobilities [33], and vacancy buildup [34]. Previous studies have suggested that grain boundary characteristics, specifically grain boundary complexions, strongly influence the stability and abnormal grain growth behavior of the nanocrystalline materials [35–37].

Ongoing research efforts seek a more fundamental understanding of the factors influencing low-temperature grain growth that will enable controlled growth behavior and/or increased thermal stability. Many of these efforts have focused on chemical means to stabilize the matrix, specifically through the addition of solute elements or finely dispersed particles that can have a Zener pinning effect on grain boundary growth [16,38–41]. One area that has received comparatively little attention is the manipulation of initial texture conditions to influence later growth behavior. Computational results have suggested that seeding a grain structure with a strong orientation component can directly influence grain growth kinetics and delay equilibrium [42–46]. Experimentally, little has been done to investigate how variations in the

* Corresponding author.
E-mail address: josh.kacher@mse.gatech.edu (J. Kacher).

http://dx.doi.org/10.1016/j.msea.2016.08.045

initial texture conditions can influence later growth behavior.

This paper reports experimental results on the influence of initial texture conditions on the thermal stability and annealing behavior of nanocrystalline thin films. It provides experimental confirmation of computational results related to grain boundaries and thermal stability and offers additional insight into the influence of surface energetics on grain growth behavior. The results demonstrate the potential of influencing initial texture in thin films as a tool to increase thermal stability and dictate grain growth behavior in nanocrystalline materials.

2. Experimental methods

Nanocrystalline Ni samples were made using pulsed-laser deposition at Sandia National Laboratories following a procedure described previously [9,22,47]. The samples were deposited on a base of (001) oriented rock salt with a beam energy of 500 mJ to a final thickness of approximately 70 nm. The growth chamber achieved a base pressure of approximately 5×10^{-7} Torr. A 99.97% pure Ni target was used during the deposition. The Ni was deposited at a nominal laser pulse frequency of 35 Hz until the desired thickness was achieved. The substrate was maintained at three different temperatures, −150 °C, room temperature, and 200 °C, during deposition. In the remainder of this paper, these samples are referred to as −150 °C, RT, and 200 °C films, respectively.

The samples were prepared for TEM and TKD analysis by scoring the surface in a 2×2 mm grid pattern and submersing the substrates with the films into deionized water. The foils floated to the surface as small squares, from which they were lifted using 3 mm Cu grids. *In situ* TEM annealing and characterization was performed in a Philips CM 300 TEM operated at 300 kV. A Gatan-model heating holder was used to anneal the samples. Temperature ramping occurred in stages, 0–200 °C, 200–350 °C, and 350–450 °C, and the film was allowed to stabilize between stages. Grain growth was recorded digitally using a CCD camera at 3 frames per second.

Orientation mapping of the pre- and post-annealed films was performed in an FEI XL30 scanning electron microscope using TKD (also known as transmission EBSD or tEBSD). TKD, originally developed by Geiss and Keller [48] and since applied to a number of material systems [49–52], relies on the same principles as EBSD to recover local crystal orientation, but collects forward scattered electrons from thin films rather than backscattered electrons. This variation has been shown to improve the spatial resolution to below 10 nm [50]. The TKD setup used in this study is the same as that outlined previously [23]. The beam was kept at an accelerating voltage of 30 kV to maximize spatial resolution. All scans were acquired over a 2×2 μm area with a 5 nm step size. All points indexed with low confidence were filtered from the scans but no data interpolation was used except for when calculating the misorientation distribution functions. The thinness of the foils, and possibly relief of internal stress from the mismatch strain between the salt substrate and the Ni film, led to wave formations, altering the surface normal. To correct for this, the sample orientation was adjusted post-acquisition by aligning the expected (001) with the center of the 001 pole figure (this process is illustrated in Fig. 1). This correction does not affect characterization of the grain boundaries, only the surface normal orientation. Characterization of the films before and after annealing was done on different sample locations due to the difficulty of relocating previous scan sites. Orientation maps, pole figures, and misorientation distribution functions were generated using EDAX/TSL OIM analysis software.

3. Results

3.1. TEM

Thermal stability of the films, the temperature at which no further grain growth was observed, was reached by 350 °C for the film deposited at −150 °C (*i.e.* no further microstructural evolution occurred when the temperature was increased to 450 °C). Thermal stability was seen at 450 °C for the film deposited at RT. This stabilization was reached after approximately 2 min annealing at temperature. The 200 °C film remained stable throughout the annealing process. Bright-field micrographs of the films before and after annealing with their associated diffraction patterns are shown in Figs. 2, 3, and 5. Videos of the annealing process are also provided (Suppl. Videos 1 and 2).

In Fig. 2a and b, it is seen that the initial nanocrystalline structure of the −150 °C film is composed of randomly oriented grains with sizes varying from approximately 5 to 30 nm. The video of the annealing process (Suppl. Video 1) shows that abnormal grain growth initiates from dispersed nucleation sites and quickly grows to consume the surrounding nanocrystalline matrix. After annealing, the nanocrystalline matrix was replaced almost entirely by grains in the larger end of the nanogrdepartments spectrum and ultrafine

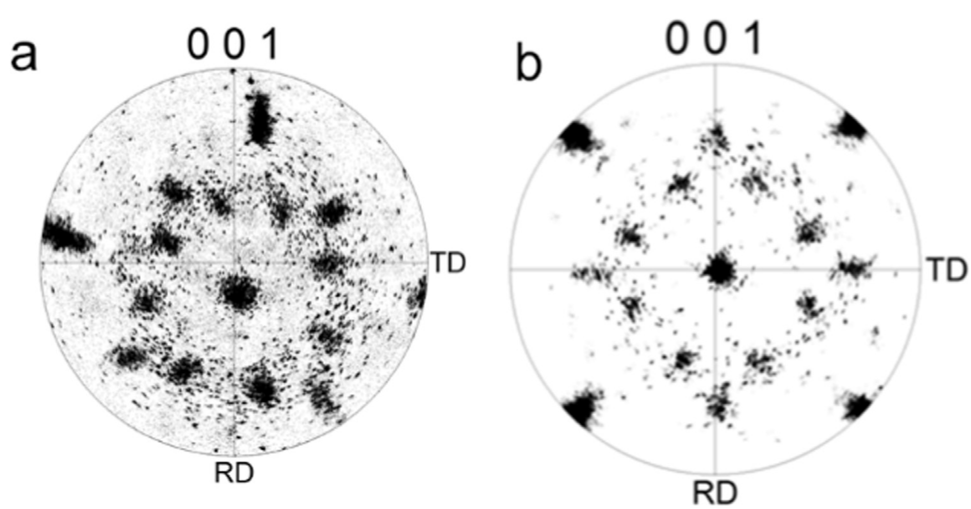

Fig. 1. Pole figure of raw data collected from annealed Ni film (a). Pole figure after low confidence points filtered out and (001) plane aligned (b).