International Journal of Fatigue 103 (2017) 444–455

Contents lists available at ScienceDirect

International Journal of Fatigue

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

Thickness effects on fatigue crack propagation in submicrometer-thick freestanding copper films

Toshiyuki Kondo *, Hiroyuki Hirakata, Kohji Minoshima

Department of Mechanical Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan

ARTICLE INFO

Article history:
Received 23 March 2017
Received in revised form 12 June 2017
Accepted 22 June 2017
Available online 24 June 2017

Keywords:
Thin films
Fatigue
Crack propagation
Thickness effects

ABSTRACT

To investigate the thickness effects on fatigue crack propagation properties and mechanisms of as-deposited submicrometer-thick freestanding metallic films, fatigue crack propagation experiments in freestanding copper films with a thickness B of approximately 100 nm were conducted at a stress ratio R of 0.1, and compared to the films with $B \approx 500$ nm. Electron-beam-evaporated copper films with $B \approx 100$ nm had smaller grains compared to the films with $B \approx 500$ nm. In addition, fine-grained regions with grain sizes of a few tens of nanometers were observed dispersedly. In films with $B \approx 100$ nm, fatigue crack initiated and stably propagated from a notch root by applying a cyclic load. However, the arrest or retardation of fatigue crack propagation often occurred even under constant maximum stress. The fatigue crack propagation rate (da/dN) of films with $B \approx 100$ nm is higher than that of films with $B \approx 500$ nm, except for the crack arrest cases; films with $B \approx 100$ nm showed lower fatigue fracture toughness. Moreover, fatigue crack in films with $B \approx 100$ nm propagated even below the fatigue threshold of films with $B \approx 500$ nm. Intermittent field emission scanning electron microscope (FESEM) observations of fatigue crack propagation behavior and fracture surface observations as well as microstructural analyses, using electron backscatter diffraction (EBSD), revealed that the fatigue cracks propagated by intergranular cracking and transgranular cracking with intrusion/extrusion formation. Compared to the films with $B \approx 500$ nm, the fatigue damage along the crack path became narrower and the frequency of intrusion/extrusion formation decreased. EBSD analysis also revealed that fatigue crack stably propagated in the coarse-grained region, whereas the arrest or retardation of the fatigue crack propagation occurred when the fatigue crack tip reached the fine-grained region, with grain sizes of a few tens of nanometers, indicating that the fine-grained regions have higher resistance to fatigue damage formation than do the coarse-grained regions.

1. Introduction

Submicrometer-to-nanometer-scale metallic materials show size effects on plasticity: The yield strength of metallic materials increases as the sample dimensions decrease [1–5]. These size effects can be explained by dislocation starvation mechanisms [1,3,4] or source truncation mechanisms [5–7]. The submicrometer-to-nanometer-scale metallic materials have characteristic structure such as large surface-to-volume ratio. In such materials, mobile dislocations easily annihilate at a free surface due to image force, which accelerates the elimination of dislocations from the materials. Moreover, available dislocation sources such as Frank–Read sources are limited. In this condition, higher shear stress is needed for the nucleation and multiplication of dislocations as the sample dimensions decrease. In addition, when metallic materials are prepared by a physical vapor deposition (e.g. electron beam evaporation, sputtering), grain size is on the order of the sample thickness. Therefore, grain boundary strengthening also contributes to thickness effects on yield stress of as-deposited metallic films, known as the Hall–Petch relationship.

Some researchers reported size effects on the mechanisms or strength of fatigue damage formation in submicrometer-to-nanometer-scale metallic materials [8–12]. Zhang et al. [9] conducted fatigue experiments on micrometer-to-submicrometer-thick copper (Cu) films fixed on a deformable substrate. They reported that extrusions on free surface and intrusions or void on the film/substrate interface were formed in micrometer-thick films. In submicrometer-thick films, however, the intrusion/extrusion formation was suppressed and interface-mediated damages, such as cracks, grooves and voids, along grain boundaries or twin boundaries, were formed. Moreover, the fatigue life of Cu films fixed on

---

* Corresponding author.
E-mail address: kondo@mech.eng.osaka-u.ac.jp (T. Kondo).

http://dx.doi.org/10.1016/j.ijfatigue.2017.06.029
0142-1123/© 2017 Elsevier Ltd. All rights reserved.

a deformable substrate increased as thickness and/or grain size of films decreased [10]. Fang et al. [12] conducted resonance vibration fatigue experiments on freestanding single crystal gold cantilever specimens with various cantilever thickness and width of ~0.4–4 μm. They reported that intrusions/extrusions were formed at all specimen sizes by fatigue loading, whereas the width of the intrusion/extrusion decreased as the specimen size decreased. Moreover, analysis by the elastic finite element method (FEM) revealed that resolved shear stress, needed for intrusion/extrusion formation, increased as the specimen size decreased. These experimental results indicate that the strength needed for fatigue initiation, such as intrusion/extrusion formation, increases as the size of materials decreases.

In contrast, a few studies have reported the size effects on the properties or mechanisms of fatigue crack propagation in submicrometer-thick films [13–16]. Meirom et al. [15] conducted fatigue crack propagation experiments in approximately 500-nm- and 1-μm-thick freestanding nanocrystalline platinum (Pt) films with high thickness-to-grain size ratio, and reported that the fatigue crack propagation properties of thicker films were similar to bulk microcrystalline Pt. In the case of thinner, 500-nm-thick films, the fatigue crack propagation rate was accelerated and fatigue fracture toughness was decreased. In both films, grain coarsening around the crack path and ahead of the crack tip by cyclic deformation and intergranular fatigue crack propagation mainly occurred in most of the stable crack propagation regime.

The authors [17] conducted fatigue crack propagation experiments in approximately 500-nm-thick freestanding Cu films with grain sizes of several thousands of nanometers (small thickness-to-grain size ratio films), and reported that intrusions/extrusions, which penetrated the film in the thickness direction, were formed along the Σ3 twin boundaries ahead of the crack tip, and the fatigue crack then propagated preferentially through these intrusions/extrusions at stress ratios R of 0.1 and 0.5 in the region of maximum stress intensity factor $K_{\max} < 4.5\ \mathrm{MPam}^{1/2}$. The fatigue crack propagation rate ($\mathrm{d}a/\mathrm{d}N$), against effective stress intensity factor range (ΔK_{eff}) of the films, was higher than that of bulk Cu, although the resistance to plastic deformation or fatigue damage formation was higher as the sample dimensions decreased [18]. This is because the fatigue crack in the films propagated preferentially through out-of-plane fatigue damage such as intrusions/extrusions. This fatigue fracture mechanism is different to the one in bulk metals: Here, a fatigue crack propagates mainly by in-plane crack tip blunting and re-sharpening. This means that fracture in the thickness direction occurs easily in films, leading to lower resistance to fatigue crack propagation compared to bulk Cu.

As for the size effects in submicrometer-thick films, the amount of accumulated fatigue damage in the thickness direction, for fatigue fracture, would decrease as film thickness decreased. However, thinner films have higher resistance to plastic deformation and fatigue damage formation. Because of these opposite effects, it is not clear whether resistance to fatigue crack propagation increases or decreases as film thickness decreases in submicrometer-thick films.

The purpose of this study is to clarify the thickness effects on fatigue crack propagation properties and mechanisms of as-deposited submicrometer-thick freestanding metallic films. First, fatigue crack propagation experiments were conducted on approximately 100-nm-thick freestanding Cu films. The mechanisms of fatigue crack propagation in the films are discussed on the basis of intermittent field emission scanning electron microscope (FESEM) observations and electron backscatter diffraction (EBSD) analysis. Next, compared to the results obtained in approximately 500-nm-thick freestanding Cu films [17], the thickness effects on the properties and mechanisms of fatigue crack propagation are discussed.

2. Experimental

2.1. Materials and specimens

The tested materials were Cu films with a thickness (B) of approximately 100 nm. The films were deposited by the electron beam evaporation method. The purity of the Cu evaporant was 99.999%. Film thickness was measured by using a stylus surface profiler (Tencor, Alpha-Step 500, resolution: 0.1 nm). The average film thickness and standard deviation are listed in Table 1. The specimen code represents average film thickness. Note that the specimens “B118-1” to “B118-3” were prepared from the same batch. The average film thickness was in the range of 93–121 nm.

The microstructure of the Cu films was evaluated by an EBSD analysis system (AMETEK Inc., DigiView) attached to an FESEM (JEOL Ltd., JSM-7001F). Fig. 1(a) shows the crystal orientation map of the surface of the Cu films with $B \approx 100$ nm. The distance between adjacent EBSD analysis points was set to 50 nm. For comparison, the crystal orientation map of Cu films with $B \approx 500$ nm, deposited under the same conditions of this study, is also displayed. Black lines indicate grain boundaries, and gray regions indicate regions where the crystal orientation could not be determined due to the low confidence index (≤ 0.2). Twin boundaries were observed within a grain on both films, and most of the twin boundaries were Σ3 coherent. Fig. 1(b) shows the logarithmic-normal probability distributions of the grain sizes of films with $B \approx 100$ nm and 500 nm. The analysis results of gray regions were not included in Fig. 1(b). Grain sizes considering and not considering twin boundaries as grain boundaries are shown. Because of the limitation of spatial resolutions of the EBSD analysis, the smallest grain size evaluated was 0.07 μm. It is clear that the grain size became smaller as film thickness decreased. The median values of grain sizes of films with $B \approx 100$ nm, considering and not considering twin boundaries as grain boundaries, were 0.12 μm and 0.15 μm, respectively.

To clarify the details of the microstructures in the gray regions in EBSD analysis, transmission electron microscope (TEM, Hitachi, Ltd., H-800) observation was conducted as shown in Fig. 1(c). Fine grains with diameters of a few tens of nanometers were observed within the gray region. The diffraction pattern obtained from a dashed-circular region in the TEM image also indicates that fine crystalline grains exist within the gray region. The percentage of the gray region (fine-grained region) area in the film was ~21%.

Fig. 2 shows the shape and dimensions of the specimens. A sacrificial layer wet etching method [17,19] was employed to fabricate freestanding film specimens. A single-edge notch (SEN) was introduced to the specimens (SEN specimens), or an asymmetrical center notch (ACN) (Fig. 2(b)) was introduced to the specimens (ACN specimens). The ACN specimens had a sharp notch at one end, and a blunt notch at the other end. The fatigue crack starts only from the sharp notch. The initial notch lengths (68–123 μm) were much larger than the grain size (Fig. 1(b), maximum grain size is ~1.4 μm). The radius of curvature of the notch root was smaller than 100 nm, except the blunt notch in ACN specimens. *In situ* optical microscope observations of fatigue crack propagation were conducted on SEN specimens, and intermittent FESEM observations of fatigue crack propagation were conducted on ACN specimens. Details of the experimental procedures of intermittent FESEM observations are described later.

Table 2 summarizes the mechanical properties of Cu films with $B \approx 100$ nm and 500 nm, previously reported by the authors [17,20]. The yield stress was defined as the stress deviating 2% from the linear approximation line of the elastic region. 0.2% proof stress of films with $B \approx 100$ nm was not obtained because no significant plastic deformation occurred before fracture. The yield stress of films with $B \approx 100$ nm was higher than that of films with