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The effect of film thickness on the failure strain of polymer-supported metal films

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Abstract

We perform uniaxial tensile tests on polyimide-supported copper films with a strong (1 1 1) fiber texture and with thicknesses varying from 50 nm to 1 μm. Films with thicknesses below 200 nm fail by intergranular fracture at elongations of only a few percent. Thicker films rupture by ductile transgranular fracture and local debonding from the substrate. The failure strain for transgranular fracture exhibits a maximum for film thicknesses around 500 nm. The transgranular failure mechanism is elucidated by performing finite element simulations that incorporate a cohesive zone along the film/substrate interface. As the film thickness increases from 200 to 500 nm, a decrease in the yield stress of the film makes it more difficult for the film to debond from the substrate, thus increasing the failure strain. As the thickness increases beyond 500 nm, however, the fraction of (1 0 0) grains in the (1 1 1)-textured films increases. On deformation, necking and debonding initiate at the (1 0 0) grains, leading to a reduction in the failure strain of the films.

Keywords: Metal films; Failure strain; Thickness; Yield strength; Texture

1. Introduction

Flexible electronic devices integrate diverse inorganic materials such as semiconductors, metals and ceramics on a polymer substrate. Benefits such as low weight, ruggedness, low costs, large area and ease of integration promise to open doors to new applications in areas where conventional electronics have not been effective. Applications of flexible electronics include paper-like electronic displays [1], conformable electronic textiles [2], rollable solar cells [3] and flexible RFID tags [4]. Flexible electronic devices are usually made as organic/inorganic hybrids. One architecture uses metallic films that are patterned into interconnects to link isolated functional ceramic islands supported by a deformable polymer substrate [5–8]. When such a structure is stretched or bent, the polymer substrate carries most of the deformation while the ceramic islands experience only small strains. As a result, the whole system can sustain large applied deformations without rupturing the electronic components. In this architecture, the metallic interconnects between the islands have to deform along with the underlying substrate. Failure of these interconnects results in a loss of electrical connectivity between the islands. In this paper, we investigate the fundamental mechanisms that limit the strain of metal films attached to polymer substrates.

Freestanding metal films stretched in tension are often reported to rupture at small strains. When stretched, a freestanding film of a ductile metal ruptures by forming a neck within a narrow region. Although the strain within the neck is large, the strain elsewhere in the film is small. Because thin-film samples often have very large length-to-thickness ratios, the net elongation of the freestanding film upon rupture is small, typically less than a few percent [9–13].

For a metal film that is well bonded to a polymer substrate, finite element simulations have shown that the polymer substrate can suppress necking in the metal film, so that the film can elongate indefinitely, limited only by rupture of the polymer substrate [14,15]. Experimentally, however,

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most polymer-supported thin metal films rupture at small elongations (<10%) [16–23], even though elongations as high as 20% have been reported in a few cases [24–26]. Our recent experiments have achieved elongations over 50% of polyimide-bonded Cu films without detectable fracture [27]. We have demonstrated that good adhesion between film and substrate is critical to achieving large failure strains, an observation that is consistent with both theoretical predictions [15] and a previous experimental investigation [22].

Strong adhesion is not the only condition required to achieve large failure strains. It is also important to ensure that deformation occurs as uniformly as possible. Another set of experiments showed that sputtered Cu films rupture at much smaller strains in their as-deposited state as compared to annealed films [28]. Without thermal treatment, the microstructure of as-deposited Cu films is unstable during deformation and room-temperature grain growth is observed under mechanical loading. Eventually, the films fail by ductile necking as a result of concurrent grain growth, strain localization at large grains and film debonding from the substrate.

In this paper, we present a comprehensive experimental study of the effect of film thickness on the failure strain of annealed Cu coatings supported by polyimide substrates. The yield strength and crystallographic texture of the film are found to have a significant impact on the failure strain. These effects are further elucidated by modelling ductile fracture of the coatings using the finite element method. The paper is organized as follows. Section 2 describes the experimental setup and procedures. Experimental results including resistance–elongation measurements and sample micrographs are provided in Section 3. The finite element simulations are described in Section 4. Final conclusions are given in Section 5.

2. Experimental detail

The polymer substrates used in this study were 12.7 μm polyimide foils (Kapton 50HN® by DuPont, Circleville, OH). Immediately prior to the deposition of metal films, the substrates were ultrasonically cleaned with acetone and methanol. The substrates were then covered by a shadow mask with seven $5 \times 50\ mm^2$ rectangular windows and placed inside the chamber of a direct-current (DC) magnetron sputter-deposition system with a base pressure better than 1×10^{-7} Torr. After sputter cleaning for 5 min with an Ar plasma at a radio-frequency power of 24 W and a pressure of 2×10^{-2} Torr, a Cu layer was deposited onto the substrates through the windows in the shadow mask. The deposition was performed using a 50.8 mm Cu target at a DC power of 200 W and a working gas (Ar) pressure of 5×10^{-3} Torr. The nominal target–substrate distance was 100 mm and the corresponding deposition rate was $0.39\ nm\ s^{-1}$. The thickness of the film was controlled by varying the deposition time. These deposition conditions are similar to those used in Refs. [27,28]. Immediately after deposition, the Cu films were annealed at 150 °C for 1 h. Annealed specimens were removed from the vacuum chamber after 12 h to allow them to cool down prior to breaking vacuum. Additional annealing under these conditions did not further increase the grain size.

The microstructure of the films was characterized by means of focused ion beam (FIB) imaging in a Zeiss NVision40 Dual-Beam FIB/SEM (Carl Zeiss Inc. Thornwood, NY). Fig. 1a shows typical FIB images for the 50 nm and 1 μm films, illustrating the change in grain size with film thickness. The crystallographic texture of the films was measured using electron backscattered diffraction (EBSD). The EBSD measurements were performed in a Zeiss Supra55VP SEM. Fig. 1b shows orientation maps for the 500 nm and 1 μm films respectively. Grains marked blue are oriented with the (1 1 1) plane parallel to the surface, grains in red[1] are (1 0 0) oriented and grains labelled green possess a (1 0 1) orientation. Both films show a strong (1 1 1) texture, but the 1 μm film contains more (1 0 0) grains than the 500 nm film, as also observed in similar polyimide-supported Cu films [29]. The volume fractions of (1 0 0) grains were estimated from at least five EBSD images for each film thickness and are listed in Table 1. The average grain size was determined by the intercept method with twins counted as separate grains, the same method as used in Ref. [30]. The relation between film thickness and grain size is shown in Fig. 1c. The volume fraction of (1 0 0) grains and grain size are tabulated in Table 1 for further reference.

Rectangular tensile test specimens with a width of 5 mm were cut from the metal-coated substrates using a razor blade. The specimen gauge length was $L_0 = 30$ mm. The specimens were then subject to uniaxial tension using an Instron 3342 tensile tester. All tests were performed at room temperature at a constant strain rate of $3.3 \times 10^{-4}\ s^{-1}$. During tensile testing, the electrical resistance of the films was measured in situ using a Keithley 2000 multimeter in a four-point measurement setup. We used the Zeiss NVision40 Dual-Beam FIB/SEM to examine specimens stretched to different elongations after unloading. At least six samples were tested for each thickness.

3. Results

3.1. In situ resistance measurements and failure strain determination

Fig. 2a shows typical experimental resistance–elongation curves for films with thicknesses ranging from 50 nm to 1 μm, along with the theoretical curve

$$R/R_0 = (L/L_0)^2 \tag{1}$$

which holds as long as there are no resistivity or volume changes during the experiment. We define the failure strain,

[1] For interpretation of color in Figs. 1, 2 and 4–8, the reader is referred to the web version of this article.