High ductility of a metal film adherent on a polymer substrate

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In recent development of deformable electronics, it has been noticed that thin metal films often rupture at small tensile strains. Here we report experiments with Cu films deposited on polymeric substrates and show that the rupture strains of the metal films are sensitive to their adhesion to the substrates. Well-bonded Cu films can sustain strains up to 10% without appreciable cracks and up to 30% with discontinuous microcracks. By contrast, poorly bonded Cu films form channel cracks at strains about 2%. The cracks form by a mixture of strain localization and intergranular fracture. The films rupture at large strains when the localization is retarded by the adherent substrates. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2108110]

Deformable electronics on polymer substrates are being developed for many applications, including paperlike displays,[1,2] sensitive skins,[3] and electronic textiles.[4] One design of such a system consists of a polymer substrate on which small functional islands are fabricated with stiff materials and interconnected by thin metal lines. When the polymer substrate is stretched, the stiff islands experience small strains, but the metal interconnects must deform with the substrate. It has been often reported that thin metal films, freestanding or polymer supported, rupture at small strains ($>2\%$).[5–12] On the other hand, it has also been reported that some polymer-supported metal films can sustain strains up to 20%.[13–15] The cause for the substantial difference has been uncertain. This paper demonstrates that metal films well bonded to the substrates can sustain much larger strains than films poorly bonded to the substrates.

Subject to a tensile strain, a freestanding metal thin film can deform plastically. When the tensile strain is large enough to break native oxides or other passivation layers on the surface of the metal film, dislocations in the metal film can readily escape. On further straining, the film does not harden as much as its bulk counterpart. Consequently, once the film thins preferentially at a local spot by forming a neck, further deformation is localized in the neck, leading to rupture [Fig. 1(a)]. By volume conservation, local thinning results in a local elongation of length comparable to the film thickness. The local elongation contributes little to the overall rupture strain, because the film has a small thickness-to-length ratio.

For a metal film deposited on a polymer substrate, a recent finite element simulation has shown that the substrate can suppress strain localization.[16] While the local elongation is accommodated for the freestanding metal film as the ruptured halves move apart, it cannot be so accommodated for a metal film bonded to a substrate [Fig. 1(b)]. This geometric constraint due to the substrate retards strain localization, so that the metal film deforms uniformly to a large strain. The above arguments are phrased in terms of necking, but similar arguments apply to localization by shear band formation.

Under certain conditions, the substrate is ineffective in suppressing strain localization. For example, if the metal film debonds from the substrate, the film becomes freestanding and is free to form a neck [Fig. 1(c)]. Alternatively, if the substrate is too compliant, its constraint is insufficient to prevent strain localization in the film. According to our calculations,[17,18] a substrate of Young's modulus in the megapascal range (e.g., an elastomer) conforms to the localized deformation of the film and the rupture strain of the film is comparable to that of a freestanding film, but a substrate of Young's modulus in the gigapascal range (e.g., a polyimide) should suppress strain localization in the metal film.

We now report on experiments designed to demonstrate the role of film/substrate adhesion on the ductility of metal thin films. Two sets of Cu films were deposited on polyimide substrates with good and poor adhesion. A 125-μm-thick polyimide foil (Kapton® by DuPont) was cut into dogbone-shaped substrates with a rectangular gauge section of 4 mm in width and 24 mm in length. Cu films of 100 and 170 nm thickness were deposited onto the polyimide substrates using a direct-current (dc) magnetron sputter-deposition system with a base pressure of 2×10^{-8} Torr, a dc power of 200 W, and a working gas (Ar) pressure of 5×10^{-3} Torr. For one set of specimens, the Kapton substrates were sputter cleaned using an Ar plasma at a radio-frequency (rf) power of 24 W and a pressure of 2×10^{-2} Torr. A 15-nm-thick Ti sticking

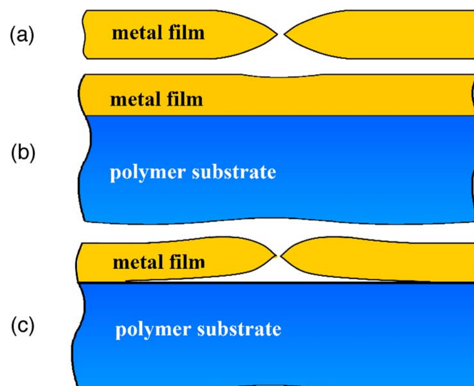

FIG. 1. (Color online) The rupture of a metal film is caused by strain localization. Local thinning leads to local elongation. (a) A freestanding metal film accommodates the local elongation as the ruptured halves move apart. (b) When the film is well bonded to a substrate, the local elongation in the film may be suppressed by the substrate. (c) Debond of the metal film assists rupture.

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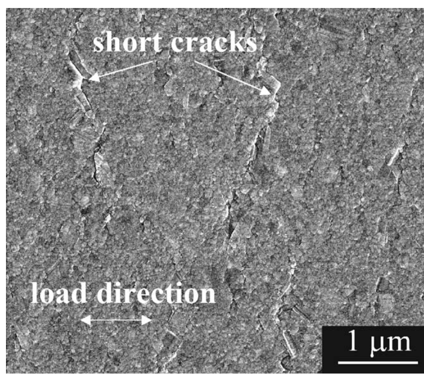

FIG. 2. A 100-nm-thick Cu film is well bonded to a polyimide substrate and is stretched to a strain of 10%. Except for isolated short cracks, the majority of the Cu film is intact.

layer was sputter deposited immediately prior to the Cu deposition to improve the adhesion. For the other set of specimens, no Ar plasma treatment was applied to the Kapton substrates and a 20 nm carbon-release layer was sputtered prior to the Cu deposition to weaken the adhesion. While the critical adhesion level required to suppress strain localization is not known, the two sets of samples clearly represent two extreme cases of adhesion in this material system. As-deposited films were kept under vacuum for approximately 72 h before they were deformed in uniaxial tension using an MTS uniaxial tensile tester. All tests were performed at room temperature with a constant strain rate of 2.5×10^{-4} s^{-1}. The surface morphology and cross section of the tested Cu films were characterized using a FEI Dual-Beam focused ion beam/scanning electron microscope (FIB/SEM) after the specimens were fully unloaded.

For specimens with good adhesion (i.e., with the Ti sticking layer), the Cu films sustained large strains without rupture. Figure 2 shows a micrograph of such a film after having been subjected to a strain of 10%. Only isolated short cracks that formed during loading and closed during unloading were visible. No debonding of the film and substrate was observed in cross sections perpendicular to the cracks.

For specimens with poor adhesion (i.e., with the carbon-release layer), the Cu films started to form channel cracks at tensile strains about 2%. The cracks traversed the entire gauge width of the specimens, perpendicular to the tensile direction. More cracks appeared as the tensile strain was increased (Fig. 3). Cu strips between adjacent cracks easily

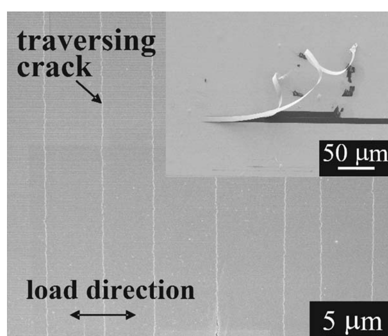

FIG. 3. A 100-nm-thick Cu film is poorly bonded to a polyimide substrate and is stretched to a strain of 6%. Channel cracks start to appear at strains about 2%. The density of the channel cracks increases with the applied strain. The Cu strips between adjacent channel cracks easily peel off, indicating the poor adhesion between the film and substrate.

peeled off (inset of Fig. 3), confirming the poor adhesion between the Cu film and the polyimide substrate. Similar channel cracks have been observed in metal films deposited on polymer substrates by other researchers.[9,10]

The process to form such channel cracks may be understood as follows. When the composite of the film and the substrate is stretched, if the Cu film deforms uniformly, no traction will act on the film/substrate interface, so that the substrate will remain adherent and suppress strain localization in the film. If, however, imperfections in the film initiate strain localization, traction will arise on the film/substrate interface. This traction can cause delamination between the Cu film and the polyimide substrate, provided the adhesion is poor. Once delaminated, the Cu film is locally freestanding and necks easily. The two processes, strain localization and delamination, promote each other and coevolve, leading to the propagation of the channel cracks.

The above picture can also be paraphrased in terms of microscopic processes. The metal film deforms by the motion of dislocations. Consider two extreme cases. In one case, on approaching the film/substrate interface, the dislocations spread out readily into the interface over a region much larger than the film thickness. Consequently, these dislocations do not hinder further dislocation motion in the film, allowing strain localization. Alternatively, the dislocations do not spread out readily into the interface, and therefore hinder further dislocation motion in the film, suppressing strain localization. The extent over which the dislocations spread on the interface is related to the adhesion of the interface.

How did a sample with good adhesion behave under even higher tensile strains? Figure 4 shows a set of micrographs with increasing magnifications, taken from a 170-nm-thick Cu film well bonded to the substrate and stretched to a strain of 30%. The behavior of this film was similar to that of the thinner film: In the Cu film, zigzag cracks formed in the nominal direction perpendicular to the loading direction [Fig. 4(a)]. The zigzag cracks resulted from microcracks initiated at angles about 60° from the loading direction [Fig. 4(b)]. The microcracks formed by a mixture of local thinning and intergranular fracture [Figs. 4(b) and 4(c)].

The orientation of the microcracks appears to agree reasonably well with the prediction of a bifurcation analysis. At low tensile strains, a metal film deforms uniformly. According to the bifurcation analysis, above a critical strain, the uniform deformation becomes unstable against perturbation of small amplitude. The critical strain and the corresponding orientation of the perturbation depend on the type of constitutive law assumed for the metal, as well as on the wavelength of the perturbation.[19–22] For example, for a freestanding metal sheet under uniaxial tension, the lowest critical strain results from perturbation of a wavelength much larger than the thickness of the sheet, and the angle between the incipient neck and the loading direction is 54.7°.[19] For a metal film well bonded to a polymer substrate, however, the lowest critical strain corresponds to perturbation of a wavelength much shorter than the thickness of the film.[16] This short-wave instability is equivalent to the surface instability of a metal sheet. For a metal characterized by a pure power law in uniaxial tension, i.e., $\sigma = K\varepsilon^N$, where σ is the true stress, ε the natural strain, K the prefactor, and N the hardening exponent, the bifurcation analysis predicts that the angle between the orientation of surface instability and

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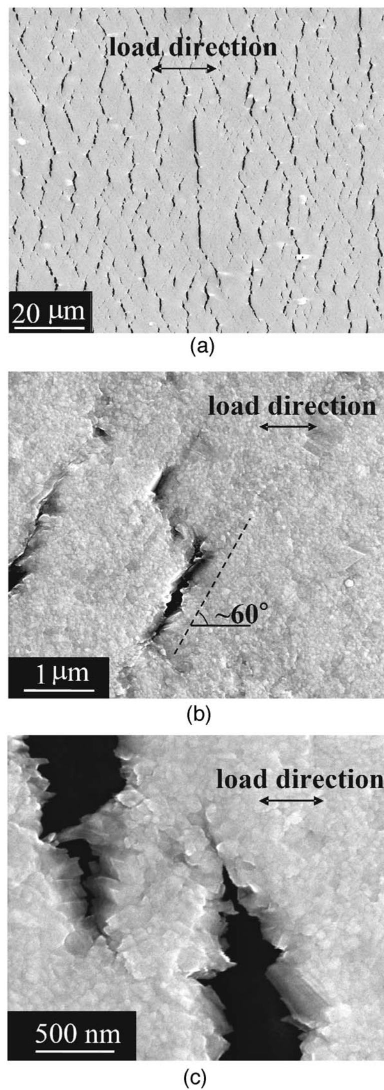

FIG. 4. A 170-nm-thick Cu film is well bonded to a polyimide substrate and is stretched to a strain of 30%. (a) Zigzag cracks appear in the Cu film. (b) Microcracks initiate at angles from the load direction, then coalesce to form the zigzag cracks. (c) Microcracks form by a mixture of local thinning and intergranular fracture.

the loading direction ranges from 43° (for $N \rightarrow 0$) to 61° (for $N=1$).[22]

While the orientation of the microcracks relates reasonably well to the predictions of the bifurcation analysis, the strains for the microcracks to fully develop and coalesce can be much higher than the critical strains predicted by the bifurcation analysis. The adherent substrate significantly elevates the critical strain for the long-wave perturbation.[16] In the other limit, for rupture to occur at short wavelengths, huge strains must occur. Therefore, deformation instability occurs at a tensile strain much higher than that for a freestanding film, at an intermediate wavelength. Upon initiation, the nonuniform deformation localizes strain in the film. Apparently the stress becomes so high that intergranular fracture leads to microcracks.

In summary, we demonstrate that the ductility of a thin metal film depends on its adhesion to the substrate. A well-bonded substrate carries the film to large tensile strains without rupture by delocalizing deformation in the film. A poorly bonded substrate allows the film to form channel cracks at relatively small macroscopic strains by facilitating the coevolution of strain localization and delamination. Furthermore, our experiments show that the ductility of metal films is also limited by intergranular fracture. While we did not systematically investigate the effect of film thickness, no obvious differences were observed between films of different thickness. The geometric constraint of the substrate is effective in retarding strain localization, but is less effective in retarding intergranular fracture, because the latter requires little additional space to proceed. On the other hand, if intergranular fracture does not intervene, the finite element simulation has suggested that a metal film well bonded to a substrate can sustain strains far beyond 30%.[16] It is hoped that new experiments will soon succeed in demonstrating metal films of such extraordinary ductility. While we did not systematically investigate the effect of film thickness, no obvious differences were observed between films of different thickness.

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