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(Received 24 September 2007; accepted 2 November 2007; published online 28 November 2007)

When a freestanding plastically deformable metal film is stretched, it ruptures by strain localization, and the elongation is less than a few percent. When the film is deposited on a polymer substrate, however, strain localization may be retarded by the substrate. This paper reports Cu films deposited on Kapton substrates and stretched up to the rupture of the substrates (at an elongation between 50% and 60%). When Cr adhesion layers are introduced between Cu and Kapton, few microcracks in Cu may be found, and the measured electrical resistance agrees with a theoretical prediction. Micrographs show that the strain localization and debonding coevolve. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2817234]

Deformable electronics are being developed for many applications, including paperlike displays,[1] electronic skins,[2] electronic textiles,[3] and flexible solar cells.[4] Such a product usually contains inorganic films deposited on a polymer substrate. While the polymer substrate may survive a large elongation, the inorganic films may not. This paper focuses on the deformability of metal films on polymer substrates.

When a freestanding film of a plastically deformable metal is subjected to a tensile load, the film ruptures by strain localization, by forming a neck within a narrow region, of a width comparable to the thickness of the film. The strain is large within the neck, but is small elsewhere in the film. Because the film has an extraordinarily large length-to-thickness ratio, the net elongation of the film upon rupture is small, typically less than a few percent.[5–9]

For a metal film well bonded to a polymer substrate, finite element simulations have shown that the substrate can delocalize strain, so that the metal film can elongate indefinitely, only limited by the rupture of the polymer substrate.[10,11] Experimentally, however, most polymer-supported thin metal films rupture at small elongations ($<10\%$),[12–19] although elongations as high as 20% have been reported in few cases.[20–22] This discrepancy between experiment and theory may be caused by the effects of a very small grain size and inadequate interfacial adhesion.[11,18] This paper demonstrates that metal films on polymer substrates can indeed be stretched beyond a strain of 50%, and is limited by the rupture of the polymer substrate.

We will also show that the electrical resistance of the metal film increases with elongation in a way predicted on the assumptions that the film retains the rectangular shape during deformation, and that the resistivity ρ of the film remains unchanged. Let R be the resistance of the metal film, which is stretched to length L and cross-sectional area A. Let R_0, L_0, and A_0 be the corresponding initial values. When the deformation is large, the volumetric strain of the metal is much smaller than the tensile strain. Following a common practice, we assume that the volume is constraint, namely, $AL=A_0L_0$. The resistance of the strained film is $R=\rho L/A$, and that of the unstrained film is $R_0=\rho L_0/A_0$. Consequently, the ratio of the resistance of the strained film to the resistance of the unstrained film is

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

This expression is well known in various forms. We will show that this prediction is followed by some of the metal films up to a large elongation.

The polymer substrates were 12.7 μm thick polyimide foils (Kapton 50HN® by DuPont), which were ultrasonically cleaned with methanol and acetone. The substrates were then placed inside a direct-current (dc) magnetron sputter-deposition system with a base pressure of 1×10^{-7} Torr, and were sputter cleaned for 5 min using an Ar plasma at a radio-frequency power of 24 W and a pressure of 2×10^{-2} Torr. Cu films of 1 μm thick were deposited through a rectangular shadow mask (5×50 mm^2 opening) onto the substrates 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 deposition rate was approximately 0.39 nm/s. The films were annealed inside the sputter chamber at 200 °C for 30 min immediately after the deposition and without breaking vacuum. Additional annealing did not further increase the grain size. For one set of specimens, no interlayer was used between the film and the substrate. For the other set, a 10 nm thick Cr interlayer was sputter deposited immediately prior to the Cu deposition.

Tensile test specimens with a width of 5 mm were cut from the coated substrates with a razor blade. The specimens were kept under vacuum for 24–48 h before they were deformed in uniaxial tension using an Instron 3342 uniaxial tensile tester. All tests were performed at room temperature with a gauge length $L_0=30$ mm and at a constant strain rate of 3.3×10^{-4} s^{-1}. During tensile testing, the electrical resistance of the films was measured using a Keithley 2000 multimeter. The surface morphology and cross sections of the deformed and undeformed Cu films were characterized using a LEO scanning electron microscope (SEM) and a FEI dual-beam focused ion beam/scanning electron microscope (FIB/SEM) after the specimens were fully unloaded.

For the first set of specimens, Cu films were deposited directly on Kapton substrates, with no Cr interlayers. Figure 1 shows the normalized resistance R/R_0 as a function of the normalized length L/L_0. This tensile experiment was terminated when the Kapton substrate ruptured at an elongation of 52%. The dots are experimental data and the curve is the theoretical prediction [Eq. (1)]. Deviation starts at an elongation of approximately 30%. This deviation resulted from

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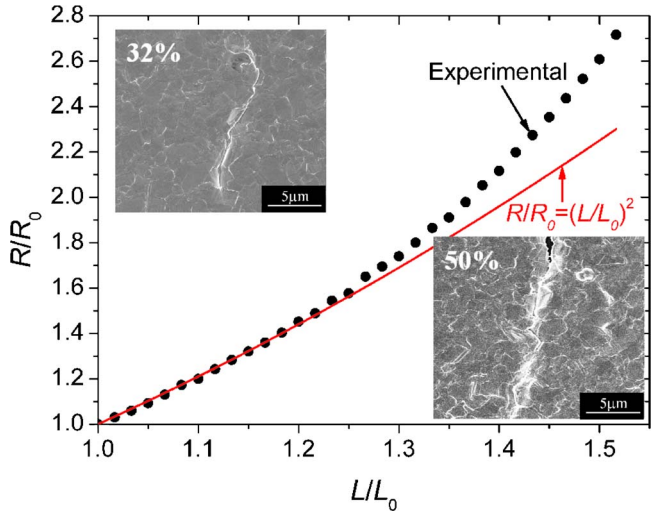

FIG. 1. (Color online) A Cu film was deposited directly on a Kapton. The sample was annealed and then subjected to a tensile load. The electrical resistance R increased with the length L of the specimen, both quantities being normalized by the initial values. The dots are the experimental data and the curve is the theoretical equation $R/R_0=(L/L_0)^2$. Microcracks are shown in SEM micrographs taken postmortem at two elongations, 32% and 50%, with the tensile loading direction being horizontal.

the formation and propagation of cracks in the Cu film, as confirmed by postmortem SEM observation. One inset shows appreciable microcracks at an elongation of 32%, and the other shows larger cracks at an elongation of 50%. The direction of the tensile load is horizontal. Our FIB and SEM images (not included in this paper) indicate that the film is sufficiently flat so that the roughness contributes negligibly to elongation.

We also subjected bare Kapton specimens of the same shape to a tensile load, and found them to rupture at an elongation of about 60%, indicating that the small cracks observed in the coated samples did not trigger the rupture of the entire structure.

For the second set of specimens, 10 nm thick of Cr interlayers were sandwiched between the Cu films and the Kapton substrates. Figure 2 shows the resistance as a function of the length of the film. This experiment was terminated when the Kapton substrate ruptured at an elongation of 56%. In the presence of the Cr layer, there was no deviation between the measured resistance and the theoretical prediction for the entire experimental range. Using SEM, we could not find any cracks in a film strained to an elongation of 40%; the surface morphology is shown in the inset on the left. At an elongation of 50%, only a few isolated short cracks could be identified, as shown in the inset on the right. We have also studied the reversibility of the resistance during unloading. Specimens were strained to 40% and then unloaded at the same rate as they were loaded. The measured resistance-elongation curve during unloading still followed Eq. (1) closely until the total load on the composite was reduced to zero at an elongation of approximately 32%, confirming that no cracks had formed in the Cu film during stretching.

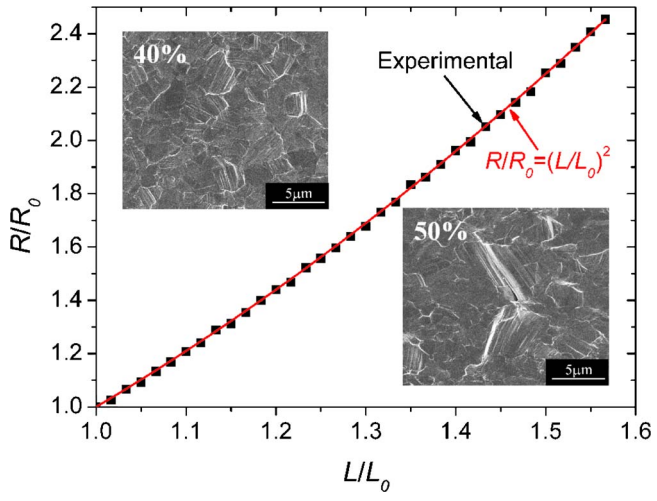

FIG. 2. (Color online) A Cu film was deposited on a Kapton substrate with a Cr interlayer. The sample was annealed and then subjected to a tensile load. Microcracks were observed at an elongation of 50%, but not at an elongation of 40%.

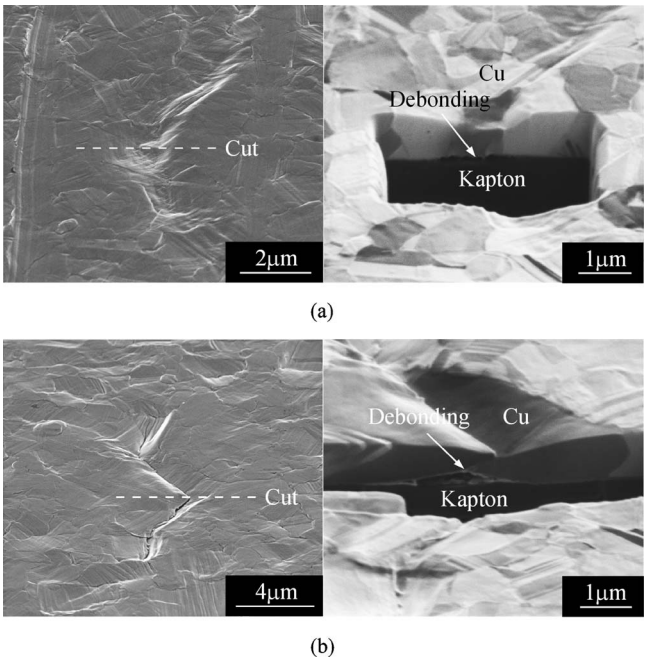

FIG. 3. Images of Cu/Cr/Kapton specimens after elongation of (a) 30% and (b) 50%. Local thinning and debonding coevolved.

The micrographs in Figs. 1 and 2 show that the cracks in the Cu film were transgranular and that they formed predominantly along bands of intense shear within individual grains.

Existing finite element calculations have suggested that plastically deformable metal films on polymer substrates rupture by coevolution of strain localization and debonding.[23,10] This is understood as follows. Without strain localization, there is no traction exerted on the interface to cause debonding. Without debonding, there is no room for large localized strain. Even though cracks cannot be found in Cu/Cr/Kapton specimens deformed up to 40% strain, local thinning can be observed in the specimens after 30% elongation. In Fig. 3, cross-sectional imaging of regions with local thinning by means of a FIB clearly shows the interface debonding that occurs along with the necking. In FIB cross sections at areas without strain localization, however, no deboning was observed. At 50% elongation, sporadic microcracks are observed in the film. As shown in Fig. 3(b), these cracks seem to initiate in regions of intense plastic shear where local thinning can be observed and the debonding width is on the order of the film thickness.

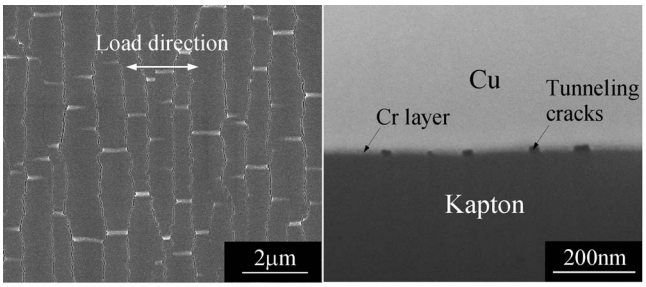

FIG. 4. (a) Channel cracks in Cr layer for bare 10 nm Cr/Kapton specimen (without annealing) after 20% strain. (b) Tunnel cracks in Cr interlayer for Cu/Cr/Kapton specimen after 50% strain.

The failure mechanism discussed above also explains why specimens with better adhesion ruptured at larger strain. It is well known that Cr interlayers enhance adhesion between the metal films and polymer substrates. A poorly bonded substrate provides less of a constraint on the metal film. A slight strain localization as a result of a surface perturbation or film imperfection can easily induce local delamination, and the two processes facilitate each other thereafter, resulting in microcracks and final rupture of the film. A well-bonded substrate, on the other hand, can delocalize deformation in the film even at a large strain because of the strong constraint on the metal layer. Hence the film can be carried to a very large elongation with little debonding or local thinning, preventing the formation of microcracks. In the Cu/Cr/Kapton specimens, local thinning and debonding are already observed at strains around 30% with the first microcracks appearing at 50% strain. It seems reasonable to expect that a stronger interface would retard local thinning and eventual film failure even further.

It should be noted that the failure mechanism described in this paper only occurs for ductile films. Metal films with grain sizes that are too small to allow abundant dislocation activity, materials with weakened grain boundaries, or films of intrinsically brittle materials such as ceramics and intermetallics would all fail at much smaller strains. Indeed, experiments on Kapton specimens coated with just 10 nm Cr show that the brittle Cr layer fails at small strains by channel cracks (Fig. 4(a)). The same is true for the Cr interlayer between the Cu and Kapton. Tunnel cracks in Cr are readily observed at 30% strain. Figure 4(b) shows a SEM cross-section image of tunnel cracks in the Cr layer after 50% strain. Most of the Cr interlayer, however, still provides good adhesion between the Cu film to the polymer substrate. Without debonding, these tunnel cracks cannot provide enough space to accommodate local thinning of the Cu films. It is highly probable, however, that these tunnel cracks may help with the initiation of the debonding, after which local thinning and debonding will coevolve.

In summary, we have fabricated polyimide-supported copper films that deform without rupture up to very large strains. Our experiments show that the films ultimately fail through a ductile mechanism where the coevolution of film debonding from the substrate and of strain localization leads to the formations of microcracks in the copper films. Because debonding plays a critical role in this failure mechanism, adhesion layers that improve the adhesion between film and substrate, retard strain localization, and cracking of the copper film. Though tunnel cracks are observed in the Cr interlayer, they have little effect on the rupture Cu film.

This work was supported by the NSF under Grant No. CMS-0556169 and by the MRSEC at Harvard University. The authors would like to thank David Mooney for use of the Instron tensile tester.

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