Stretchability of thin metal films on elastomer substrates

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Many flexible electronic surfaces comprise inorganic films on organic substrates. Mechanical failure of such integrated structures of stiff and compliant materials poses a significant challenge. This letter studies the stretchability of metal films on elastomer substrates. Our experiment shows that, when stretched, elastomer-supported metal films rupture at strains larger than those reported for freestanding films. We use a finite element code to simulate the rupture process of metal films. A freestanding metal film ruptures by forming a single neck. By contrast, a metal film on an elastomer substrate may develop an array of necks before rupture. While the pre-rupture necks do not change the electrical conductance appreciably, they elongate the metal film, leading to a large overall rupture strain. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1806275]

Flexible electronics are being developed for diverse applications, including paper-like displays,[1,2] electronic textiles,[3] and sensitive skins.[4] Many such macroelectronic surfaces comprise thin films of stiff materials (e.g., metals, transparent conductors, and amorphous silicon) deposited on substrates of compliant materials (e.g., polyimide and silicone). When stretched or bent, the compliant substrate deforms, but the stiff films may crack or debond. Mechanical failure poses a significant challenge in the development of flexible and stretchable electronics. This letter studies the deformability of metal films on polymer substrates.

It has been often reported that gold, aluminum and copper films, freestanding or polymer-supported, rupture at strains of 1%–2%.[5–9] These values are smaller than the rupture strains of the corresponding bulk metals. Two alternative reasons are suggested to account for this observation. First, a thin polycrystalline metal film (e.g., 10–100 nm thick) may be inherently brittle. The grains may be too small to mediate dislocation motion, and the grain boundaries may be embrittled by impurities or be ineffective for diffusional creep. Second, the metal film may be inherently ductile, but deformation is localized. The film does not harden appreciably beyond a modest strain. For a freestanding metal film, once an incipient neck grows, further deformation of the film essentially takes place only in this region. The single neck leads to rupture. Because the film has a small thickness-to-length ratio, the elongation at the single neck contributes little to the overall rupture strain.

In a recent paper,[10] we used a finite element simulation to show that a polyimide substrate can suppress strain localization in a ductile metal film. By volume conservation, necking causes a local elongation. If the metal film is bonded to the polyimide substrate, the substrate suppresses large local elongation in the film, so that the metal film may deform uniformly to a large strain. If the metal film debonds from the substrate, however, the film becomes freestanding and ruptures at a smaller strain. Kang[11] reported that some of his aluminum films on polyimide substrate survived a tensile

strain of 20%. More recent experiments have shown that well-bonded copper films on a polyimide substrate can be stretched to strains exceeding 10% without rupture, but poorly bonded copper films rupture at smaller strains.[12]

While polyimide (Young's modulus ~5 GPa) may be a suitable substrate for paper-like displays, more compliant elastomer substrates (Young's modulus 1–200 MPa) are needed for applications such as sensor skins and retinal implants. This letter studies whether such elastomer substrates can still suppress strain localization, assuming that the metal film is inherently ductile. Recall that Young's modulus of a metal is 10^3 to 10^5 times that of an elastomer.

Figure 1 shows an experimental observation. On a 1-mm-thick silicone substrate (Young's modulus ~1 MPa), we deposited a 5-nm-thick chromium layer, and a 50-nm-thick gold film. The chromium layer improved the adhesion between gold and silicone. The film was patterned as a 25-mm-long and 1- or 2-mm-wide stripe. The deposition process generated a compressive stress in the plane of the film: the film wrinkled, but still bonded to the substrate. The sample was stretched using a homemade tester.[13,14] At a strain of ~1%, the wrinkles disappeared. The electrical re-
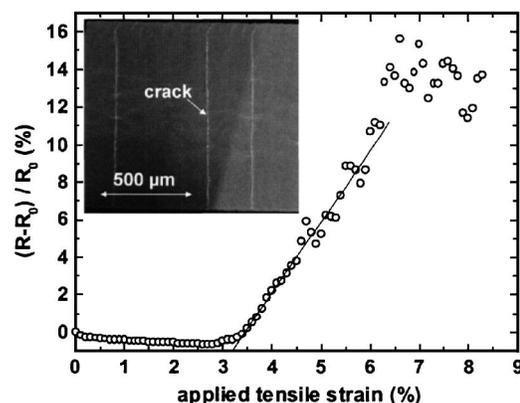

FIG. 1. A 50-nm-thick gold stripe on a silicone elastomer substrate is stretched, while the changing electrical resistance R of the gold stripe is recorded. R_0 is the reference value measured before stretching. The plan-view scanning electron micrograph is taken after the tensile load is released. The cracks are perpendicular to the stretch direction.

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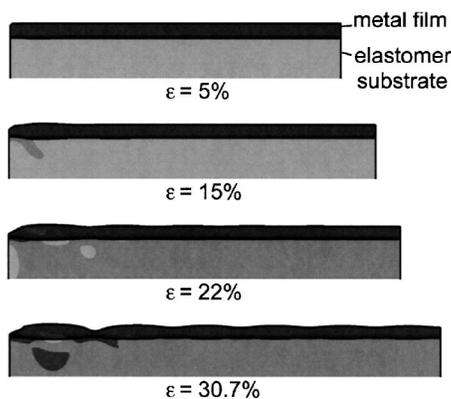[b)]Electronic mail: wagner@princeton.edu

FIG. 2. Deformation sequence of a metal film on a substrate of Young's modulus $E_0 = 150$ MPa. Note the formation of multiple necks in the film. The right halves of the sample and only part of the substrate thickness are shown. An initial imperfection $0.1h$ deep was introduced at the top left corner. Contour colors represent the Mises stress level.
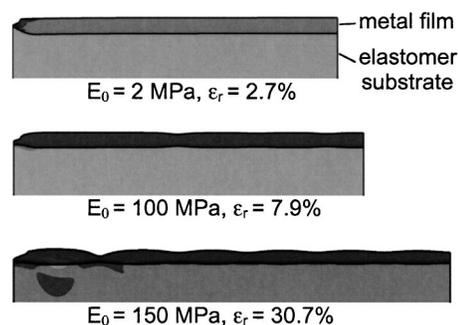

FIG. 3. Finite element simulations of metal films on substrates of various Young's moduli. All films are stretched to rupture. The stiffer the substrate, the larger the rupture strain.

sistance of the gold stripe changed little when the strain was below ~3%, and increased linearly with the strain up to 6%. The sample became electrically open at 8% tensile strain, corresponding to cracks traversing the gold stripe.

We use the finite element code ABAQUS to simulate the deformation process of blanket metal films on elastomer substrates. The metal is modeled as a plastically deformable solid, obeying the J_2 deformation theory.[15] Under a uniaxial stress state, the true stress σ and the natural strain ε obey the power law: $\sigma = K\varepsilon^N$, where $K = 114$ MPa and $N = 0.02$. The elastomer is modeled as a Mooney–Rivlin solid[15] with initial Young's modulus E_0, whose value is varied to study the effect of substrate stiffness. The finite element simulation assumes plane strain conditions. The film thickness is h, and the substrate is a $40h \times 40h$ block. The horizontal displacement is set to be zero along the centerline of the laminate, and set to be u along both sides of the laminate. We report the natural strain $\varepsilon = \ln(1 + u/20h)$. Uniform deformation is a trivial solution to this boundary value problem. An imperfection needs to be introduced to induce nonuniform deformation. At the center of the film surface, we place a V-shaped notch, $0.2h$ wide and $0.1h$ or $0.05h$ deep. Taking advantage of symmetry, we model only the right half of the laminate.

Figure 2 shows the deformation sequence of a metal film on an elastomeric substrate ($E_0 = 150$ MPa). At small strains, the film deforms uniformly; the $0.1h$ deep notch does not cause any significant nonuniformity in the deformation. At large strains, nonuniform deformation becomes pronounced, and develops into multiple necks. The film eventually ruptures near the notch.

Figure 3 shows ruptured films on substrates of different elastic moduli. We note three types of behavior within the cases simulated. First, when the substrate is very compliant ($E_0 = 2$ MPa), the film forms a single neck, and the substrate locally distorts to follow the neck in the film; the film ruptures at a strain of 2.7%, comparable to that of a freestanding film.[10] As expected, the very compliant substrate does not provide sufficient support to impede strain localization. Second, when the substrate is of intermediate stiffness ($E_0 = 100$ MPa and $E_0 = 150$ MPa), the film forms multiple necks, stretches to a much higher strain, and then ruptures near the notch. Third, when the substrate is sufficiently stiff ($E_0 = 300$ MPa, not shown in Fig. 3), deformation remains

uniform at strains larger than 50%; this behavior is similar to that of a metal film on polyimide.[10]

Figure 4 plots the rupture strain as a function of elastomer stiffness. Initial notch depths of $0.05h$ and $0.1h$ are used in the simulations. As the elastomer stiffness increases, the substrate provides more constraint to the metal film. The plots show that the rupture strain is insensitive to the imperfection size.

Multiple cracks have long been observed experimentally in metal films on polymer substrates and subject to tensile strains.[7,8] One possible scenario is that these cracks form sequentially. Under tension, a single crack forms first, emanating from a site of imperfection. The crack cuts through the metal film, but arrests at the metal/polymer interface. In the presence of the substrate, the crack does not affect the stress elsewhere in the film. Consequently, more cracks can form in the film from other sites of imperfection. In this scenario, the first crack electrically opens the metal film, and the subsequent cracks make no contribution to the stretchability. This scenario is similar to multiple crack development in a brittle film on a substrate.[16] To form cracks sequentially, a metal film must either be inherently brittle, or debond from the substrate.

The ductility of the metal films may allow a different scenario. Under tension, a single incipient neck forms, emanating from a site of imperfection. The ductility of the metal requires the film to thin down and elongate. The elongation is constrained by the substrate. Before the first neck cuts through the film thickness, other incipient necks emerge elsewhere in the film. In this scenario, the pre-rupture necks do
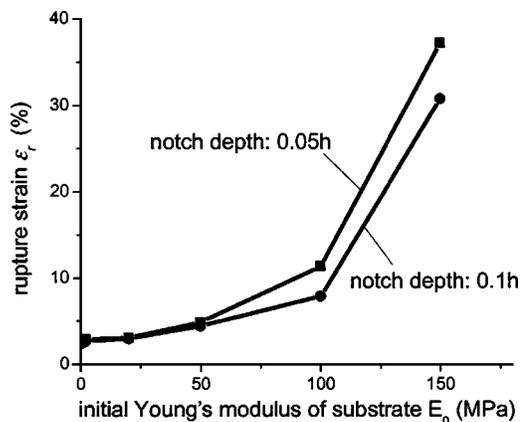

FIG. 4. Rupture strain as the function of Young's modulus of the substrate E_0. Two notch depths are used in the simulations.

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not change the electrical resistance of the metal film appreciably. Each pre-rupture neck contributes an elongation of about one film thickness h. A rough estimation of the metal film rupture strain is $\varepsilon_r \approx h/S$, where S is the average spacing between the pre-rupture necks. This estimate is consistent with the simulations shown in Figs. 2 and 3.

Multiple pre-rupture necks have been observed in other situations, such as during high strain rate extension of bulk metals.[17] However, we are unaware of any experimental observation of multiple pre-rupture necks in thin metal films on polymer substrates. To do so one has to ensure that the metal film is inherently ductile, and that the substrate is of intermediate stiffness. If the substrate is too compliant, the film deforms as if freestanding, and ruptures by a single neck. If the substrate is too stiff, the spacing between the necks is too small, and some other failure mode may intervene before the necks grow appreciably. The stiffness of the substrate may be adjusted by its Young's modulus or its thickness.

The experiments and simulations presented in this letter both indicate that a polymer-supported metal film has a larger rupture strain than a freestanding metal film. However, it is difficult to make a quantitative comparison between the experiments and simulations. Our experiments are for films on 1-mm-thick elastomer substrates. The numerical simulations are for 1-μm-thick substrates. The simulations assume that the metal films obey the J_2 deformation theory, with a hardening exponent $N = 0.02$. To what extent such a model reflects a metal film in experiment is uncertain.

In summary, large rupture strains can be achieved if the metal films are well-bonded to an elastomer substrate of sufficient stiffness. Even better stretchability is possible with recent design concepts such as zigzag interconnects.[9,14] These stretchable interconnects are promising for applications in biologic systems and severe dynamic environments. Further research is needed for such interconnects under cyclic loading.

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