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Size-Dependent Rupture Strain of Elastically Stretchable Metal Conductors

O. Graudejus^{a,*}, Z. Jia^b, T. Li^{b,*}, and S. Wagner^c

^aDepartment of Chemistry and Biochemistry, and Center for Adaptive Neural Systems, Arizona State University, Tempe, Arizona 85287

^bDepartment of Mechanical Engineering, and Maryland NanoCenter, University of Maryland, College Park, Maryland 20742

^cDepartment of Electrical Engineering and Princeton Institute for the Science and Technology of Materials, Princeton University, Princeton, New Jersey 08544

Abstract

Experiments show that the rupture strain of gold conductors on elastomers decreases as the conductors are made long and narrow. Rupture is caused by the irreversible coalescence of microcracks into one long crack. A mechanics model identifies a critical crack length ℓ_{cr} above which the long crack propagates across the entire conductor width. ℓ_{cr} depends on the fracture toughness of the gold film and the width of the conductor. The model provides guidance for the design of highly stretchable conductors.

Keywords

thin films; rupture strain; stretchable conductors; flexible electronics

Elastically stretchable conductors are explored for biomedical applications [1–3], dielectric elastomer actuators [4], and stretchable electronics [5,6]. A class of highly stretchable conductors consists of gold films on substrates of the elastomer polydimethylsiloxane (PDMS). Depending on fabrication conditions, these gold films can adopt three morphologies: microcracked, buckled, and smooth [7]. We are particularly interested in the microcracked films, because they can be elastically stretched to, and relax from, large strains while remaining electrically conducting [8]. For example, 40 μm wide and 5 mm long microcracked gold conductors on 300 μm thick PDMS can be stretched reversibly to about 50% uni-axial strain [9]. However, at 90% uni-axial strain electrical conductance is lost because at one or a few locations microcracks coalesce to one long crack that severs the entire conductor. The parameters that determine the formation of such long cracks have not been understood. Here we show experimentally that the rupture strain ϵ_r increases with the width and decreases with the length of the conductor, and present a mechanics model that explains this size dependence of the rupture strain.

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*Authors to whom correspondence should be addressed: Fabrication and material properties, oliver.graudejus@asu.edu, Mechanics model, lit@umd.edu.

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75 nm thick microcracked gold films on 2 to 3 nm of chromium adhesion layer are deposited at $<65^{\circ}\text{C}$ substrate temperature on 300 μm thick PDMS. The as-deposited film contains 0.5 to 2 μm long randomly distributed microcracks, which delineate a stretchable gold network. The gold film is patterned into $L = 5$ and 20 mm long and $W = 6, 25, 45,$ and 95 μm wide conductors by lithography and wet etching [7,9] (Figure 1a). The distribution of microcracks within these conductors is homogenous without appreciable edge effects because the microcracks in the gold film are produced during deposition. Figure 1 shows scanning electron micrographs (SEMs) of a microcracked gold film before stretching (Figure 1b), an optical micrograph of a 95 μm wide conductor during stretching at 10% strain (Figure 1c), and an SEM after stretching to a maximum strain of $\sim 90\%$ (Figure 1d). The SEM after stretching clearly shows the coalescence of the microcracks perpendicular to the stretching direction. The SEM and optical micrograph images show that the microcracks coalesce homogeneously throughout the conductor, and perpendicular to the stretching direction. Patterned conductors are stretched in steps of 0.1% every 3s up to a designated maximum strain, and then relaxed at the same rate, while the electrical resistance R is recorded. After 3 to 5 cycles, the designated maximum strain is increased by 10%, and the gold conductor is stretched and relaxed again. This procedure is repeated until the conductor fails. Figure 2 shows the plots of resistance R vs. applied strain ϵ for $L = 5$ and 20 mm long and $W = 95, 45,$ and 25 μm , respectively. The indicated rupture strain ϵ_r is the strain at which the resistance increases sharply. Figure 3 summarizes the rupture strain ϵ_r of gold conductors as a function of their width W and length L . The PDMS that carries 95 μm wide conductors often tears before the conductors fail. ϵ_r is seen to decrease with decreasing W and increasing L . As ϵ_r decreases, the conductors become more sensitive to sample handling. For example, conductors with $W = 6 \mu\text{m}$ have such a small ϵ_r that they either fracture while the PDMS is peeled off its temporary glass slide carrier ($L = 20$ mm), or rupture at $\epsilon < 2\%$ ($L = 5$ mm). Adrega et al. also observed lower yields for conductors with $W = 10$ or 20 μm than for $W > 30 \mu\text{m}$ [10]. The macroscopic resistivity of our microcracked conductors is 7 to 12 times higher than the bulk resistivity of gold. The increase is caused by additional electron scattering in the thin film, by the surface roughness of the PDMS, and by the microcracks themselves. The resistivity of gold film controls deposited in the same run on a glass slide (no microcracks, low surface roughness) is 2 to 3 times that of bulk gold.

To understand the pronounced size-dependence of the rupture strain ϵ_r , we focus on the few long cracks that form when microcracks coalesce under strain, as illustrated in Figure 4a. Such long cracks shield the surrounding microcracks from stress, because the force that drives their propagation is significantly higher than that for the microcracks. Under certain conditions – discussed below – the advance of the long crack of length ℓ becomes unstable, so that it runs across the entire width of the conductor ($\ell \rightarrow W$) and electrical continuity is lost. Therefore the longest crack determines electrical failure. When stretched too far, a conductor fails because the longest crack runs through the conductor width. We observe that such a “deadly” crack develop from a long crack that first develops in the conductor’s interior. It then continues to grow out to the edges, or it merges with cracks that come in from the edges. In Figure 1c, the long crack in the middle is typical of those that become deadly. On the other hand, cracks that grow from the edges tend to terminate in the interior instead of running across the entire width. Such cracks also can be seen in Fig. 1c. Similar observations were found for other samples. In view of this experimental evidence we model the formation of long cracks as illustrated by Fig. 4b.

To quantify this mechanism, we introduce a simplification that is common practice in fracture mechanics [11,12]. We approximate the conductor of Figures 1b and 4a by Figure 4b, as having no microcracks but only the long crack in the middle, and having a stiffness equivalent to that of the microcracked conductor. Crack propagation is driven by the energy release rate G . A crack is stable when G is less than a critical value, the fracture toughness

of the gold film G_c . When $G > G_c$ the crack propagates. Under applied strain ϵ , the energy release rate G of the long crack of Figure 4b is [13]

$$G = \pi E \epsilon^2 \ell \sec(\pi \ell / 2W) / 2, \quad (1)$$

where E is the equivalent stiffness of gold, ℓ the length of the longest crack, and W the width of the conductor. While Eq. (1) gives a good estimate of the driving force for crack propagation when $\ell \ll W$, electrical failure results from the propagation of the long crack across the whole conductor, i.e., $\ell \cong W$. In this case, Eq. (1) tends to overestimate G , given that Eq. (1) assumes no deformation of the two free edges of the conductor while in reality the free edges can deform locally to mitigate the stress concentration near the crack tips when $\ell \cong W$. Therefore, to precisely compute the driving force for crack propagation near conductor failure, we simulate the deformation of the gold film as in Figure 4b under remote elongation using the finite element code ABAQUS. The gold conductor is modeled as a W by L rectangular film with displacement u applied to the two remote vertical edges as in Figure 4b in opposite directions to simulate the relative elongation of $\epsilon = 2u/L$. In the simulations, for a given conductor width W (25 μm , 45 μm and 95 μm respectively), we vary the crack length ℓ to study its effect on the driving force for crack propagation. In the regions near two crack tips, the film is densely meshed into four-node quadrilateral plane stress concentric-circle elements to capture the stress concentration effect. The film portion outside these two regions is less densely meshed. The gold film is taken as a linear elastic material with Young's modulus of 100 GPa and Poisson's ratio of 0.3. The energy release rate G can be calculated directly through contour integral in ABAQUS [14]. Eq. (1) and FEM model are applied to freestanding gold conductors, without substrate, an assumption justified by the huge stiffness ratio between gold and PDMS of $>10^5$. Recent simulations show that, under tension, a patterned thin gold film on a PDMS substrate deforms almost like a freestanding thin gold film with the same pattern [15].

The results calculated from Eq. (1) and FEM are plotted in Figure 4c as the normalized energy release rate $G/E\epsilon^2 h$ vs. crack length ℓ for the conductor widths W of the experiments. Note that $G/E\epsilon^2 h = \pi \ell \sec(\pi \ell / 2W) / 2h$ depends only on geometry, not on material properties. The most important result is that the driving force for crack propagation monotonically increases with ℓ for any W . When $\ell \ll W$, the analytical solution from Eq. (1) agrees with the FEM results; however, as ℓ approaches W , Eq. (1) overestimates the crack driving force compared with FEM, but both predict that the crack driving force approaches infinity. The critical crack length ℓ_{cr} demarcates the regime of stable from that of unstable crack propagation. In Figure 4c the intersections with $G_c/E\epsilon^2 h$ of the dashed FEM curves (circles) for the normalized energy release rate $G/E\epsilon^2 h$ determine the critical crack length ℓ_{cr} . If the fracture toughness G_c and the stiffness E of the gold conductor can be determined, Figure 4c can be used to correlate the critical rupture strain ϵ_r and the corresponding critical crack length ℓ_{cr} for a given conductor width. For a given $G_c/E\epsilon^2 h$, ℓ_{cr} increases as W is raised. A crack longer than ℓ_{cr} will propagate unstably and make the conductor fail. In other words, the wider the conductor, the longer ℓ_{cr} and the greater the resistance to failure. Additionally, the driving force for propagation of a crack of length ℓ increases with decreasing W . Both factors, the smaller ℓ_{cr} and the larger driving force for crack propagation, contribute to the decrease in rupture strain with decreasing W . Thus, the mechanics model qualitatively explains the experimentally observed dependence of the rupture strain ϵ_r on width W . The experimentally observed dependence of rupture strain ϵ_r on conductor length L can be understood from the rising probability for the formation of long microcracks with increasing L . Under tensile strain, the probability that microcracks coalesce to a crack of length $> \ell_{cr}$ is higher for a longer conductor. This length dependence of the rupture strain is similar to the often-observed size dependence of the statistical strength of brittle materials [16,17].

In summary, our experiments demonstrate that the rupture strain of a microcracked gold conductor increases with increasing width and decreasing length of the conductor. A fracture mechanics model offers a systematic account of this size dependence. When a microcracked gold conductor is stretched, the microcracks coalesce and form longer cracks. The conductor remains electrically conducting provided that the longest crack is shorter than a critical crack length ℓ_{cr} . These findings highlight the importance of limiting the longest crack from exceeding ℓ_{cr} (e.g., by nanopatterning the silicone substrate [18]) in the design of elastically stretchable metal conductors of high deformability for flexible electronics. Furthermore, the mechanics model sheds light on the correlations between conductor material properties (e.g., fracture toughness), critical defect size, and conductor deformability, and provides guidance for materials selection and morphology control of stretchable metal conductors.

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Figure 1.

(a) Schematic of a gold conductor on PDMS and its cross-sectional view (not to scale). (b) SEM image of the microcrack morphology in the gold conductor before stretching, (c) optical micrograph of a microcracked conductor during stretching (width = $95\ \mu\text{m}$, 10% strain; $R = 230\ \Omega$), and (d) SEM image after stretching to 90% strain.

Figure 2.

Electrical resistance R measured on gold conductors as a function of applied strain. The strain rate is 0.1% every 3s. The rupture strains ϵ_r are marked. (a) Length L / Width $W = 5$ mm / 95 μm , (b) L / $W = 20$ mm / 95 μm , (c) L / $W = 5$ mm / 45 μm , (d) L / $W = 20$ mm / 45 μm , (e) L / $W = 5$ mm / 25 μm , and (f) L / $W = 20$ mm / 25 μm .

Figure 3.

Experimentally determined rupture strain ε_r as a function of width W and length L of gold conductors on PDMS. The $\approx 95 \mu\text{m}$ wide conductors were still electrically conducting at the indicated strains. Mechanical failure due to tearing or slipping out of the clamp prevented the determination of the exact rupture strain.

Figure 4.

(a) When stretched, pre-existing microcracks in a metal conductor coalesce locally to form a long crack. (b) Approximation for FEM simulation. (c) Normalized energy release rate $G/E\epsilon^2h$ as a function of crack length l for conductor widths $W = 25\mu\text{m}$, $45\mu\text{m}$, and $95\mu\text{m}$. Solid lines from Eq. (1), dashed lines from FEM. The fracture toughness of the gold G_c and the applied strain ϵ result in a value for $G_c/E\epsilon^2h$ that is marked by circles on the FEM curves. These identify critical crack lengths l_{cr} . For example, for $G_c/E\epsilon^2h = 750$, $l_{cr} \approx 21\mu\text{m}$, $38\mu\text{m}$, and $80\mu\text{m}$ for $W = 25\mu\text{m}$, $45\mu\text{m}$, and $95\mu\text{m}$, respectively.