Synthesis of Stretchable Gold Films with Nanocracks: Stretched up to 120% Strain while Maintaining Conductivity

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Abstract. With the great deformability of stretch, compression, bend and twisting, while preserving electrical property, metal films on elastomeric substrates have many applications for serving as bioelectrical interfaces. However, at present, most polymer-supported thin metal films reported rupture at small elongations (<10%). In this work, highly stretchable thin gold films were fabricated on PDMS substrates by a novel micro-processing technology. The as deposited films can be stretched by a maximum 120% strain while maintaining their electrical conductivity. Electrical characteristics of the gold films under single-cycle and multi-cycle stretch deformations are investigated in this work. SEM images imply that the gold films are under the structure of nanocracks. The mechanisms of the stretchability of the gold films can be explained by the nanocracks, which uniformly distribute with random orientation in the films.

1. Introduction

Deformable electronics are being developed for many applications, including artificial blood vessel[1], electronic skins[2], electronic textiles[3], paperlike displays[4], and flexible solar cells[5]. Curved structures of conventional rigid materials have been researched and developed intensively during the past decade due to their electrical properties and simple processing technology[6]. However, freestanding metal thin films rupture when stretched beyond 1%-2% [7]. Moreover, most of biological tissues are soft, elastic and curved. Due to the large mismatch in mechanical properties, the implanted electronic devices may irritate and damage the surrounding tissue and cause immune reaction, scarring and hemorrhage[8]. Currently, stretchable electronic devices have received extensive attention due to their great deformability and better Young’s modulus matching with biological tissues. Metal films on elastomeric substrates can be stretched repeatedly by tens of percent over hundreds of cycles without fatigue and remain electrically conducting[9-11]. The devices are usually made from gold, semiconductors, graphene and nano-wires on stretchable substrates. However, most polymer-supported thin metal films rupture at small elongations ( ε <10%), although elongations as high as 20% have been reported in few cases[12-13].

In this paper, we fabricated highly stretchable thin gold films with nanocracks on PDMS using magnetron sputtering. The films can maintain conducting while being stretched at large strains as about 120%. The electrical characteristics of the highly stretchable gold film are investigated, in both single-cycle and multi-cycle deformation. In addition, the surface morphologies of the highly stretchable gold films on PDMS are also investigated.

2. Experiments and Results
2.1 Fabrication process of highly stretchable thin gold films on elastomeric substrates

Fabrication process of highly stretchable thin gold films on elastomeric substrates is described with the schematic illustration showed in Figure 1. A silicon wafer used as a rigid backing is coated with a monolayer of 1H, 1H, 2H, 2H perfluoro-octyl-trichlorosilane (48931-10G, Sigma Aldrich) to facilitate the removal of silicone membranes at the end of fabrication. The silicone prepolymer and the cross linker are mixed in a 10:1 ratio by weight (PDMS; Sylgard 184, Dow Corning), mixing and defoaming in a conditioning mixer (AR-100, Thinky Mixer, Japan), then spun on the silicon wafer at 600 rpm, 60 s for substrate. The PDMS is cured at 80°C for 3 h. This preparation produces a 110-μm-thick PDMS membrane for substrate. After plasma treatment on the PDMS substrate, a 3-nm-thick titanium adhesion layer followed by a 50-nm-thick gold film is deposited through a pre-patterned, stainless steel mask by a direct-current (dc) magnetron sputtering system (JS4S-75G, JSWN, China). After peeled off from the silicon wafer, the electrode sample is accomplished. For the sample, the mask pattern is designed similar to a dumbbell, two pads at the ends are squares (1.5 mm×1.5 mm), the wire in between is 0.5 mm in width and 8 mm in length.

Traditional electrodes are made usually by lithography process. The photoresist is spin on the substrate, in the step of exposure and development, due to the light show characteristics, part of the photoresist dissolves, the other part continues to remain in the substrate serving as the mask. After metal deposition, the photoresist residue is dissolved by stripping liquid. In this paper, the traditional process is improved, we choose a pre-patterned stainless steel instead of photoresist serving as the mask. The stainless steel mask can be used repeatedly. The improved process saves time, cost, and improves the efficiency.

2.2 Electrical properties of highly stretchable thin gold films on elastomeric substrates

A typical electrode sample designed for this study is showed in Figure 2(a). The electrode is designed similar to a dumbbell, two pads at the ends are squares. The electrical resistances are tested between pad #1 and pad #2. A customized stretcher device designed for stretching the electrode at a certain amount of strain is also showed in Figure 2(a). Figure 2(a) shows the states of the electrode before and under stretching. From Figure 2(b), we can see that the critical uniaxial strain or the maximum uniaxial strain while the gold film maintains conduction, characterized as stretchability of the gold film, can reach as high as about 120%. The strain where the resistance is O.R. is defined as electrical rupture strain, at which the sample is still mechanically robust. Multiple samples are tested, and the critical strain value of 120% is highly repeatable. Although the resistance increases along with stretching, the changes in resistance are reversible, and the resistance versus the applied strain shows a hysteresis curve. From Figure 2(b), we can also conclude that the resistance after being fully relaxed is slightly higher than that before stretching. The electrical characteristics of the highly stretchable thin gold film during multi-cycle deformation are also investigated. From Figure 2(c), we can see that the...
resistance of the electrode at a certain strain becomes lower and lower from one cycle to the subsequent cycles with the maximum applied strain of 110%. A continuous, cyclic deformation is applied for testing, and the shift in resistance tends to converge to a stable phase in the end.

Figure 2. (a) Optical images of the 50 nm thick gold films on PDMS. The red double arrow line shows the direction of the stretch. (b) Electrical resistance of the 50 nm thick gold films on PDMS while they are stretched at different applied uniaxial strains (n=5), where O.R. represents that the resistance is out of measuring range, indicating a fracture of the film. (c) Electrical resistance of gold conductors (n=5) under cyclic applied strain when the maximum applied strain is 110%. The curved arrows present the stretching and releasing process.

2.3 The principle of stretchability of highly stretchable thin gold films on elastomeric substrates
Scanning electron microscope (SEM, NOVA NanoSEM 450, FEI) images of highly stretchable thin gold film on PDMS are showed in Figure 3. Figure 3(a) shows the SEM image of the film at the resting state. Initial nanocracks can be found in the film uniformly distributed with random orientation, and the mean length of nanocracks is 90 ± 8 nm (n =100). While the film is being stretched for the first time, some of the initial nanocracks propagate and coalesce into large microscale cracks to ease local stress, which can be seen in Figure 3(b). It is believed that due to so many initial nanocracks, gold films can have such a high stretchability. Figure 3(c) shows the SEM image of the film after stretching. We can see that the microscale cracks close when the films relax, and some turnup flakes appear in the films. Which can be concluded that, the resistance of the electrode after being fully relaxed is slightly higher than that before stretching because of the contact resistance between the two edges of cracks when the cracks close.

Figure 3. SEM images of the 50 nm thick gold films on PDMS before, under, and after stretching. The white double arrow line in (b) shows the direction for the uniaxial stretch.

3. Conclusion
In this work, an improved processing technology was adopted to fabricate highly stretchable nanocrack gold films based on magnetron sputtering and a stainless steel mask, which improved the processing efficiency greatly. The gold films can be stretched as much as 120% uniaxial strain and maintain their
electrical conductivity under stretching. The highly stretchable films can be bent, stretched or twisted while keeping conductive, which create a promising field in biomedical applications for its better modulus match with biological tissues and robust mechanical and electrical properties. They allow for construction of electronic integrated circuits spread over on complex and dynamic curved surfaces, creating a view for the development of stretchable electronic devices. Based on the highly stretchable gold films, we will develop stretchable microelectrode arrays, which will be designed and fabricated using commercially available tape as a deformable substrate.

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