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Large actuation and high dielectric strength in metallized dielectric elastomer actuators

Sze Hsien Low, Li Lynn Shiau, and Gih-Keong Lau

School of Mechanical and Aerospace Engineering, Nanyang Technological University, Singapore 639798

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Metal films are seldom used as compliant electrodes for dielectric elastomer actuator (DEA) because they tend to restrain deformation of soft dielectrics. This work showed that silver film electrodes formed by electroless deposition (ELD) are indeed stretchable, and the DEA using ELD silver electrodes is able to generate an actuation up to 50% areal strain. Such ELD silver electrodes can self-heal, remain conductive at up to 33% uni-axial strain, and do not stiffen the dielectric layer as much as the sputtered silver electrodes. This metallized DEA can sustain a high breakdown field up to 350 MV/m, which is good for generating a large actuation force. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4709480>]

Metal films have been used as electrodes in plastic capacitors as they can be made very thin, in the nanometer range, and they can adhere well to the plastic dielectric layer. In addition, they are able to self clear by isolating localized defects or electrical shorting from the rest of the active capacitor.^{1–3} These properties make metal films desirable for use as electrodes in dielectric elastomer actuators (DEAs). However, they have been deemed unsuitable for DEAs as they may restrict large deformation of soft dielectric. DEAs with un-patterned sputtered gold electrodes have only allowed small actuated strains of less than 6%.^{4–6} In order to be used as compliant electrodes for DEAs, metal films need to have a small stiffness comparable to that of the soft dielectric layer and they have to remain conductive even when highly strained.⁷

In a bid to make metal films stretchable enough for use in DEAs, several groups have endeavoured to pattern the metal electrodes and/or the dielectric layer. For example, gold has been sputtered in zigzag⁸ and spiral-like^{5,9} patterns, while silver has been deposited on corrugated dielectric layers by means of the e-beam evaporation method.¹⁰ Apart from the sputtering and evaporation deposition methods, other physical or chemical deposition methods have also been used to form stretchable metallic electrodes. These include electrodes formed by metal ion implantation,^{11,12} salt-reduced platinum,¹³ and a silver nanoparticle/polymer gel composite.¹⁴ However, most DEAs using metallic thin-film electrodes produce relatively small actuated area strains. Reported actuated area strains include that of electrodes made of un-patterned evaporated gold (<6%),^{6,9} patterned sputtered gold (<2%),⁵ and silver deposited on a corrugated dielectric elastomer (<7%).¹⁵ Dielectric elastomer actuators with metal ion implanted electrodes, which have metal nanoparticles embedded in soft dielectric, can bulge out-of-plane with an equivalent area strain of up to 40%.¹¹ However, its self-healing properties have not been reported. In addition, it is expensive to batch produce the ion implanted electrodes due to the equipment required.⁷

This letter shows that un-patterned thin metal film electrodes, formed by the electro-less deposition of silver (ELD silver), allow relatively large actuated area strains of up to

50%. The use of ELD silver electrodes also enables high breakdown field strengths of up to 350 MV/m. Such electrodes are able to retain electrical conductivity when mechanically stretched by up to 33% uniaxially and does not stiffen the dielectric layer as much as sputtered silver does. Thin metal films formed by ELD silver are not only more stretchable than that formed by sputtered silver; the fabrication process is also simpler and less costly, as it does not require expensive equipment. The uses of ELD silver are quite versatile, in that on a large, commercial scale, it has been commonly used to form large areas of reflective thin silver films for the fabrication of mirrors. While on a small scale, ELD silver can be used for forming micro-patterns by means of micro-contact printing.¹⁶ Use of ELD silver as electrodes in DEAs has recently been reported by our group in the form of a uni-morph configuration.¹⁷

The ELD silver film electrodes were formed by using a commercially available three-part silvering solution set (HE-300, Peacock Laboratories, Inc.), comprising a silver diamine complex, a sodium hydroxide activator, and a reducing agent. All solutions were diluted in a 1:40 ratio before use. In order to form a thin silver film, the substrate was first washed with deionised water. Following that, a sensitizing solution (No. 93 Sensitizing Solution, Peacock Laboratories, Inc.) was used to wet the surface that is to be plated. After a minute, the sensitizing solution was flushed using deionised water. Next, the three silvering solution parts were mixed together in equal amounts and then dripped onto the wetted area in order to deposit a thin silver film. After the chemicals have been left on for a minute, they were washed off with deionised water and then the newly formed silver film electrodes were blown dry.

Samples of DEAs were prepared by first cutting 3M VHB F9473PC films into 28 mm × 28 mm strips and subsequently manually stretching and securing each strip in place to an acrylic frame with an outer border of 70 mm × 70 mm frame and a cutout of 50 mm × 50 mm. The F9473PC films have an initial thickness of 250 μm. They are then pre-stretched at bi-axial strains of 150% × 150%. The above-mentioned silvering process was then used to form a pair of 10 mm diameter silver film electrodes on either side of the

pre-stretched VHB film. Thin conductive trails were also applied, such that they extended from each circular electrode from opposite sides. Following that, thin aluminium foil strips were then attached to the trails of both electrodes, which were in turn connected to a high voltage source (610E, Trek Inc.) by crocodile clips. A digital camera (Canon EOS 550D) fitted with a macro lens (Tamron AF 90 mm f/2.8 Di SP A/M 1:1 Macro Lens) was used to take photographs or videos of the electrode as it was subjected to discrete values of voltage. The photographs were then analyzed using MATLAB and the change in area calculated. The voltage and current across the DEA, with respect to time, was logged by means of a data acquisition card (PCI 6052E, National Instruments).

Figure 1(a) shows a DEA sample, using ELD silver electrodes, achieving a large actuated area strain of 50% at a driving voltage of 9 kV. Upon activation, an areal expansion of the ELD silver electrodes was observed to be accompanied by reduction in opacity. Other electrode materials, such as 10 μm thick tap-on graphite powder and 150 nm thick sputtered silver, were also tested in the configuration of the pre-stretched membrane DEAs. Figure 1(b) shows that actuated area strain of DEAs increases with the driving voltage. DEAs with graphite electrodes produces the largest maximum actuated areal strains (close to 90%), followed by DEAs with ELD silver electrodes (close to 50%) and then DEAs with sputtered silver electrodes (less than 10%). ELD silvered DEAs produces approximately 50% less maximum actuation strain than the graphite-electroded DEAs because metal electrodes are stiffer than graphite ones. Unlike the

parabolic trend of graphite-electroded DEAs, the actuated strain for ELD silvered DEAs increased at a decreasing rate as driving voltage increased and it plateaus at driving voltages above 7.5 kV. This behavior is attributed to the stiffening effect of metal electrodes with increasing strain. Despite producing less actuation than the graphite ones, ELD silver electroded DEAs exhibit desirable characteristics of having a higher breakdown voltage and a higher breakdown field strength. As electrostatic force is proportional to the square of the electric field, this means that ELD silvered DEAs have the potential for generating high pressures, albeit at moderate actuated strains.

A major contributing factor to high breakdown fields is the ELD silver film electrode's ability to self-heal. Such a property not only allows the circumvention of a premature failure of the actuator, but also allows it to be driven at even higher voltages. When electrical breakdown happens at a defect site in the dielectric layer, arc discharge accompanied the breakdown vaporises the metal electrode in the neighborhood of the defect, thereby rendering that region non-conductive.¹ Figure 2(a) shows the time histories of current and voltage as the driving voltage was increased in steps of 500 V. The first self-healing event occurred at 5000 V, and that was characterized by a spike in the current, together with a corresponding drop and recovery of the voltage. In contrast, this phenomenon was not observed in DEAs using graphite electrodes. It was also observed that at higher electric fields, leakage current increased steadily. Apart from metal film electrodes, single-walled carbon nanotube electrodes have also demonstrated self-healing, which is also

FIG. 1. Electro-mechanical activation of bi-axially pre-stretched membrane DEAs: (a) photographs showing the top view of an ELD silver electrode for the DEA before and during activation. A 50% area strain is obtained during the activation at 9 kV. (b) A graph showing the actuated area strain as a function of driving voltage for the DEAs using various electrode materials. Five samples of DEAs using ELD silver electrodes were tested, and error bars indicate the standard deviation.

FIG. 2. Self-healing demonstrated in a DEA with ELD silver electrodes: (a) time histories of voltage and current show that the first self-healing occurs at 5 kV, as indicated by a recovery following a plunge in voltage together with a corresponding spike in current. (b) A self-heal site wherein passivation of the puncture site circumvents a terminal electrical-shortening.

known as self-clearing.¹⁸ A self-heal site is shown in Figure 2(b), wherein the passivation of the surrounding regions of the puncture enabled a terminal short-circuit to be averted, thereby allowing even higher driving voltages to be applied.

During large-strain actuation of DEAs, the electrodes for DEAs need to remain conductive when highly stretched. If conductivity is lost, parts of the electrode would become isolated from the voltage source, and those regions of the DEA would become inactive, which would in turn limit the actuated strain. Uni-axial tensile tests were conducted in order to determine the limit as to which the ELD silver electrode can be stretched before losing its conductivity. Here, an electrode with a resistance higher than $1\text{ G}\Omega$ was deemed to be non-conductive.

Samples of electrodes used in the uni-axial tensile tests were prepared by first cutting 3M VHB 4905 films into strips of $25.4\text{ mm} \times 76.2\text{ mm}$ and then silvered on one side. Following that, the centre section spanning 25.4 mm was left exposed, and two copper electrical leads were placed on each end of the exposed area. Silver grease was applied at the interface in order to improve contact between the ELD silver film and the copper electrical leads. Following that, two pairs of acrylic pieces were used to clamp the ends of the VHB film, such that only the middle section, spanning $25.4\text{ mm} \times 25.4\text{ mm}$, was free to deform. These samples were then strained uni-axially, at a rate of 3 mm/min , by a mechanical testing system (Instron 5565), where the force and extension were logged. For resistance logging, a digital multimeter (34410A, Agilent Technologies) was connected to the two electrical leads using the four-probe resistance measurement method. In this way, the change in resistance across the electrode, with respect to an increase in strain, could be determined.

As shown in Figure 3(a), the resistance across the ELD silver electrode started low at $1\text{ }\Omega$ but lost its conductivity at 33% uni-axial strain. The sputtered silver film electrode also had a low starting resistance of $1\text{ }\Omega$, but it lost its conductivity at a lower uni-axial strain of 21%. In contrast, the graphite electrode had a much higher starting resistance of $94\text{ k}\Omega$ but managed to retain its conductivity till more than 300% uni-axial strain. Among these electrode materials, graphite was able to sustain the most strain despite having a higher initial resistance, which is acceptable in DEAs as higher resistances do not affect its functionality.

As shown in Figure 3(b), micro-cracks are formed in an ELD silver electrode, which was subjected to increasing uni-axial strains. The introduction of micro-cracks, with increasing strain, caused grains in the ELD silver film to get smaller. However, the grains remained connected up to 33% strain. Consequently, the metal film electrode is able to remain electrically conductive when stretched. In addition, the presence of these micro-cracks may help in lowering the stiffness of the electrode in the in-plane direction and, consequently, allow a larger area expansion of the DEA during actuation.

Figure 3(c) shows the change in resistance of an ELD silver electrode over 15 cycles of loading and unloading, wherein uniaxial strain varied between 0% and 20% strain. The resistance increases with the loading strain, and it decreases with the unloading strain. This sample has an ini-

FIG. 3. (a) Change in resistance of an ELD silver electrode, a graphite electrode, and a sputtered silver electrode under uni-axial strain, up to the loss of conductivity; (b) micrographs showing the formation and then widening of micro-cracks in an ELD silver film under uni-axial strains at 0%, 10%, and 20%; and (c) change in resistance of an ELD silver electrode under cyclic loading and unloading of uni-axial strains between 0% and 20%.

tial resistance of $100\text{ }\Omega$ at 0% strain before loading, and it has a peak resistance of $362\text{ }\Omega$ when first loaded to 20% strain. In the subsequent cycles of loading and unloading, the peak resistance at 20% does not increase but decreases below the first peak resistance. This suggests the micro-crack does not propagate further during cyclic loading. In addition, a higher resistance at 0% in the subsequent cycles indicate that the metallic film have plastically lengthened during loading and buckled during unloading. ELD silver's ability to maintain its conductivity through cyclic loading is also confirmed by cyclic electro-mechanical actuation of ELD silvered DEAs.

Figure 4 shows the stress-strain curves obtained from VHB 4905 strips coated with the different electrode materials. The strips have an initial thickness of $500\text{ }\mu\text{m}$ before being stretched. Stress in the samples increases with uniaxial strain. Young's modulus for the samples can be obtained by least-square fitting each stress-strain curve using the Mooney-Rivlin model.¹⁹ Young's Modulus of a VHB 4905 strip coated with graphite, ELD silver, and sputtered silver was 1.03, 1.20, and 1.58 times that of an uncoated VHB 4905 strip, respectively. It is apparent that sputtered silver stiffens the VHB film the most, followed by ELD silver and then graphite. The high stiffening effect and a small strain limit, at which electrical conductivity could be maintained, explain why DEAs with sputtered metal electrodes could not obtain as large an actuated strain as that obtained by DEAs with ELD silver.

This work shows that the ELD silver electrodes are compliant enough for use in DEAs, contrary to common

FIG. 4. Stress-strain curves for VHB 4905 samples with coating of three electrode materials under uniaxial stretching. The calculated Young's modulus for each sample was obtained by using a least-square fitting of measured stress and strain to the Mooney-Rivlin model.

conception that metal films are too stiff for DEAs. The present work shows that DEAs using ELD silver achieve a large area strain up to 50%, higher than the values reported so far for DEAs using metal electrodes. This large actuation strain is made possible because the ELD silver can retain its conductivity even when stretched and it does not stiffen the DEA much. Furthermore, ELD silver electrodes can self heal, and consequently, DEAs using such electrodes can reach high breakdown voltages and breakdown fields, which enable high actuated pressure. In addition, ELD silver can be made very thin and can adhere well to most dielectric layers. This makes them highly suitable for use in stack or roll configurations.

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- ¹J. Tortai, N. Bonifaci, A. Denat, and C. Trassy, *J. Appl. Phys.* **97**, 053304 (2005).
- ²M. Rabuffi and G. Picci, *IEEE Trans. Plasma Sci.* **30**, 1939–1942 (2002).
- ³C. Reed and S. Cichanowski, *IEEE Trans. Dielectr. Electr. Insul.* **1**, 904–922 (1994).
- ⁴R. Pelrine, R. Kornbluh, J. Joseph, R. Heydt, Q. Pei, and S. Chiba, *Mater. Sci. Eng., C* **11**, 89–100 (2000).
- ⁵A. Pimpin, Y. Suzuki, and N. Kasagi, in *17th IEEE International Conference on Micro Electro Mechanical Systems (MEMS)* (IEEE, 2004), pp. 478–481.
- ⁶N. Galler, H. Dittbacher, B. Steinberger, A. Hohenau, M. Dansachmüller, F. Camacho-Gonzales, S. Bauer, J. Krenn, A. Leitner, and F. Aussenegg, *Appl. Phys. B* **85**, 7–10 (2006).
- ⁷B. Balakrishnan and E. Smela, *Proc. SPIE* **7642**, 76420K (2010).
- ⁸R. Kornbluh, R. Pelrine, J. Joseph, R. Heydt, Q. Pei, and S. Chiba, *Proc. SPIE* **3669**, 149 (1999).
- ⁹A. Pimpin, Y. Suzuki, and N. Kasagi, *J. Microelectromech. Syst.* **16**, 753–764 (2007).
- ¹⁰M. Benslimane, P. Gravesen, and P. Sommer-Larsen, *Proc. SPIE* **4695**, 150 (2002).
- ¹¹S. Rosset, P. Dubois, M. Niklaus, and H. Shea, in *Transducers 2009 International Conference on Solid-State Sensors, Actuators and Microsystems* (IEEE, 2009), pp. 2401–2404.
- ¹²S. Rosset, M. Niklaus, P. Dubois, and H. Shea, *Adv. Funct. Mater.* **19**, 470–478 (2009).
- ¹³M. Urdaneta, R. Delille, and E. Smela, *Adv. Mater.* **19**, 2629–2633 (2007).
- ¹⁴D. Hyun, M. Park, C. Park, B. Kim, Y. Xia, J. Hur, J. Kim, J. Park, and U. Jeong, *Adv. Mater.* **23**, 2946–2950 (2011).
- ¹⁵M. Benslimane, H. Kiil, and M. Tryson, *Polym. Int.* **59**, 415–421 (2010).
- ¹⁶Y. Xia, N. Venkateswaran, D. Qin, J. Tien, and G. Whitesides, *Langmuir* **14**, 363–371 (1998).
- ¹⁷G. K. Lau, S. C. K. Goh, and L. L. Shiau, *Sens. Actuators, A* **169**, 234–241 (2011).
- ¹⁸W. Yuan, L. Hu, Z. Yu, T. Lam, J. Biggs, S. Ha, D. Xi, B. Chen, M. Sene-sky, G. Grüner *et al.*, *Adv. Mater.* **20**, 621–625 (2008).
- ¹⁹M. Mooney, *J. Appl. Phys.* **11**, 582–592 (1940).