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Laser-scribed Graphene Oxide Electrodes for Soft Electroactive Devices

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Abstract

Inherently soft electroactive devices will enable a range of novel applications in biomedical, wearable, and robotics technology. In this area, dielectric elastomers (DEs) are particularly promising electromechanical transducers. One major challenge for DEs and other soft devices is the fabrication and precise patterning of thin stretchable conductors. Conventional methods such as pad printing and spray deposition require pre-fabricated molds or stencils, and inkjet printing relies on organic solvents to tune the rheology of inks. In this work, laser-scribed graphene oxide (LSGO) is proposed to fabricate patterned, stretchable electrodes for DE actuators (DEAs). The method uses a low-cost laser engraver that can produce arbitrary patterns with 0.1 mm resolution. The process is green (free of organic solvents) and does not require molds or stencils. DEAs with regular and complex LSGO electrodes were fabricated and characterized. In a direct comparison with DEAs made using conductive carbon grease as electrode material, LSGO DEAs show higher maximum actuation strain. Areal strain of 8.8% was observed in a planar LSGO DEA with patterned electrodes.

Main Text

There is a growing need for biomedical, wearable, and robotic devices that are able to deform with movement and conform to complex shapes such as the human body.^[1–3] To enable such devices, actuators, sensors and other electroactive components need to be developed that are inherently soft and compliant. One particularly promising technology in this field are dielectric elastomers (DEs). DEs are soft electromechanical transducers that can serve as actuators as well as sensors and energy harvesting devices.^[4] They are lightweight, scalable

and have potentially high energy- and power density.^[5] One major challenge for soft and stretchable electronics in general and for DEs in particular, is the fabrication of soft conductors to serve as electrodes. For DEs, graphite or carbon black as conductive fillers in soft composites are most commonly used.^[6] Other materials such as graphene, carbon nanotubes and silver nanowires show great promise for use in conductive elastomer composites^[7,8] but are generally more expensive or more challenging to process, especially on a larger scale. In addition, depositing thin films of these materials with defined shapes and high accuracy remains a challenge. Methods including pad printing^[9] or spray deposition^[10] have been used to apply electrode materials but these rely on pre-fabricated molds or masks. Inkjet printing^[11,12] allows deposition of very thin layers and arbitrary, high-detail patterns but usually requires the use of organic solvents to form inks with suitable rheological properties. To overcome these limitations, we propose laser-scribed graphene oxide as a new method to produce compliant electrodes for application in dielectric elastomer actuators (DEAs) and potentially other soft, electroactive devices.

Graphene and its derivative graphene oxide (GO) have been of great interest for use in electronics for over a decade. GO is particularly interesting because, unlike graphene, it can be produced economically and at a large scale using current methods,^[13] is easily processable in water with no need for organic solvents and it can be converted selectively from non-conductive GO to reduced graphene oxide (rGO), which is electrically conductive. This reduction can be done by various methods, including direct laser scribing of GO films.^[14] Such laser-scribed graphene oxide (LSGO) can be produced under a range of different conditions e.g., using femtosecond lasers,^[15] pulsed CO₂ lasers,^[16] or continuous diode

lasers^[17] and has even been demonstrated using a commercial LightScribe® DVD drive.^[18] Laser scribing is an easy method to produce high-detail, conductive patterns on GO films and LSGO has already been used to fabricate a range of different devices such as supercapacitors,^[18] various sensors,^[19–22] electronic components such as transistors^[23] or memory^[24] and even loudspeakers^[25] and LEDs.^[26] Electrodes for ionic polymer actuators have also been fabricated.^[27] However, with the exception of a simple resistive strain sensor,^[26] LSGO has, to the best of our knowledge, not been used in conjunction with soft substrates to produce truly elastic devices. In this work, we demonstrate that laser scribing of GO can be used to produce compliant, patterned electrodes for DEAs.

The LSGO method consists fundamentally of two simple steps: drop-casting of aqueous GO dispersion on an elastic substrate to produce a thin GO film, and laser scribing of the GO film to produce conductive LSGO patterns. The patterned electrode is produced in-situ rather than transferred to the elastic substrate after patterning, as is the case in previously reported laser-assisted patterning techniques for DEA electrodes.^[28] This simplifies the process and avoids potential defects due to misalignment or inclusions. The LSGO method presented here uses a low-cost (~£100), commercial laser engraver for laser scribing and requires no expensive equipment for film casting or post-processing. As such, our approach provides a uniquely simple, low-cost and green (free of organic solvents) route to produce compliant electrodes with arbitrary, complex patterns.

To fabricate a DEA with LSGO electrodes, an aqueous GO dispersion is drop cast onto a pre-stretched elastic membrane that serves as the dielectric for the DEA. After drying, the drop

casting process is repeated on the other side of the membrane. A small desktop laser engraver with a 250-mW diode laser was then used to reduce the GO film on one side of the membrane in the desired electrode pattern. Afterwards, the membrane is flipped over, and the reverse side is laser scribed. Unreduced GO that has not been laser scribed remains hydrophilic and can be removed by washing with water-soaked cotton wool (see SI for details), leaving only the more hydrophobic and conductive LSGO behind. A schematic of the fabrication process is depicted in **Figure 1a**. Fabrication of multi-layer LSGO DEAs has not been attempted. In principle, this could be achieved by stacking several LSGO DEAs as described above. Electrodes would only have to be applied on one side. Depending on the elastomer, an adhesive would be required to bond layers and, as mentioned above, the additional laminating step introduces the risk of misalignment and inclusions. Alternatively, consecutive dielectric layers could be deposited on top of an existing layer with LSGO electrodes (e.g., by spin coating with liquid silicone rubber) and then casting and laser scribing another LSGO layer. However, process time would be greatly increased because each GO layer needs time to dry and, using this method, several layers cannot dry in parallel.

The laser-scribing process is mainly controlled by two parameters: power and speed of the laser. Both these parameters influence the irradiation energy per area and thus the maximum temperature and heating rates experienced by the GO film. These must be carefully adjusted to obtain good results. Figure 1b shows the effect of laser power on the resulting LSGO films. The major challenge in producing LSGO DEAs arises from heat damage to the elastomer membrane caused during laser scribing. The third column in Figure 1a depicts the silicone elastomer membrane after laser scribing at varying laser power, showing damage at high

intensity (LSGO was removed by washing with ethanol; see SI for details). Thermal reduction of graphene oxide occurs at around 200 °C.^[29] The acrylic elastomer VHB 4905, which is commonly used as a dielectric membrane in DEAs, starts to degrade at temperatures above 149 °C.^[30] It cannot withstand the temperatures necessary for GO reduction and is therefore unsuitable for the laser scribing process. Instead, ELASTOSIL® silicone films, which have a specified operating range between -45 °C and 200 °C,^[31] were used. Maximum local temperatures reached during laser scribing are likely higher than that, but since most silicones show gradual degradation to temperatures over 300 °C, we expect minimal effects from short exposures to local heating above 200 °C. It was found that laser power and speed could be optimized to effectively reduce GO films without visible damage to the substrate (see Figure 1b). All LSGO electrodes presented in this work were laser scribed at 60% laser power at a scan speed of 1000 mm min⁻¹.

Unreduced GO forms very stiff, nonconductive films. These stiff films can restrict the deformation of the elastic membrane and will crack even under very small strain (see Figure S1). Therefore, depending on the electrode geometry, washing of the laser-scribed films to remove excess graphene oxide is important to allow the elastomer to deform freely.

Laser scribing converts GO to LSGO which has a 30% reduced oxygen content on average, as determined by energy-dispersive X-ray spectroscopy (Figure S3). LSGO has a sponge-like structure of loosely stacked graphene sheets.^[21] This makes it softer than GO and prevents crack propagation (see Figure S1). When stretched, LSGO separates evenly into small flakes that behave similar to a film made of graphite particles. The conductive flakes have sufficient

overlap to form a connected mesh and maintain overall conductivity under moderate strain. The LSGO film adheres well to the silicone substrate and is quite resistant to rubbing. Especially when the film has been washed and any loose debris removed from the surface, the material will not visibly rub off at a light touch. Rubbing with gentle pressure will leave a stain reminiscent of graphite from a pencil. Complete removal of the film is difficult even with firm rubbing, unless a solvent is used. In this regard, LSGO makes for much easier handling than CG. Under high strain, LSGO will show large cracks but, unlike unreduced GO, does not delaminate from the silicone substrate (see Figure S1).

LSGO films fabricated by our method have a sheet resistance of $7.2 \text{ k}\Omega \text{ sq}^{-1}$ ($n=3$, $\sigma=1.2 \text{ k}\Omega \text{ sq}^{-1}$) at 0% strain. When the LSGO film is stretched, resistance increases by about 3 orders of magnitude at 50% strain. Cyclic stretching shows a residual increase in resistance by one order of magnitude after the first cycle but almost no further increase after the second and third cycle (see Figure S2). Even at relatively high resistance, LSGO remains suitable as a conductor for high-voltage, low-current applications.

To characterize the performance of LSGO as electrode material for DEAs, three groups of actuators were fabricated. Group 1: LSGO electrodes, used as-is without washing (6 samples). Group 2: LSGO electrodes, washed to remove unreduced GO (4 samples). Group 3: Carbon grease (CG) electrodes, applied by hand using a mask to ensure identical geometry as the LSGO electrodes (9 samples). CG is used as a baseline to compare LSGO electrodes against. Since some commercial CG contains silicone oil which can degrade the silicone membrane, CG made with castor oil was used (see SI Section S6). Except for the electrode material, all

actuators were prepared in an identical fashion, to eliminate other influencing factors. Each DEA consists of a circular, 100% biaxially pre-strained silicone membrane ($\text{\O}45$ mm) with matching circular electrodes ($\text{\O}40$ mm) on either side. **Figure 2a-c** show one sample of each group, respectively.

In the test setup, a vertically suspended steel rod of mass 31.7 g is resting on the center of the horizontally mounted membrane. Actuation of the DEA causes the membrane to expand in area and the rod to move up and down (Figure S6, Video S1). The difference in the vertical position of the rod between the relaxed and actuated state is defined as the stroke of the actuator and recorded as a performance metric. The test setup is depicted in Figure 2d.

The initial stretching that breaks and softens the LSGO film requires a higher stress than the DEA can exert through actuation and must be done by application of an external force. We term this process “training”, which, in addition to loosening the LSGO film, also softens the elastomer membrane through the Mullins effect.^[32] All samples were first tested untrained and then a second time after training. Training is performed using the test setup described above, by applying force to the steel rod to stretch each DEA to 16 mm stroke for approximately one second. The process is repeated three times per sample.

Figure 2e shows actuation stroke over voltage for each group of actuators. Untrained LSGO samples showed very small stroke and are not shown (see SI for full results). Overall, unwashed LSGO DEAs showed the highest stroke in our experiment of 1.94 mm at 10 kV. Washed LSGO produced a stroke of 1.40 mm at 10 kV and 18% lower stroke than unwashed LSGO on average. This can be explained by two complementary effects. Firstly, washing

might cause loose flakes of LSGO to be removed from the surface. These flakes help to maintain interconnection between domains of LSGO as the membrane is stretched. Without these flakes, some areas of the electrode may become isolated and do not actuate, leading to reduced performance. Secondly, the remaining GO in the unwashed DEAs restricts deformation in the passive part of the membrane (i.e., the area around the edges that is not covered by the reduced GO electrode). Because the membrane is pre-stretched, the passive area will contract when the DEA is actuated. This in-plane stretch absorbs part of the actuation strain and reduces the amplitude of out-of-plane motion. The restricting effect of the stiff GO film therefore creates a slight advantage in this actuator configuration.

Both unwashed and washed LSGO DEAs (trained) show larger stroke than untrained CG DEAs. This can be explained by softening of the elastomer membrane due to the Mullins effect. After training, the average stroke of CG DEAs increased by 81%. Trained CG DEAs show the highest strain of all tested groups for any given voltage. At 9 kV they produce an average stroke of 1.64 mm. At the same voltage, unwashed LSGO shows 1.31 mm and washed LSGO shows 1.00 mm stroke, respectively. However, all trained CG DEAs broke down above 9 kV while both groups of LSGO DEAs withstood 10 kV, which is the maximum voltage of the high-voltage driver used in the experiments. Figure 2f shows the cumulative percentage of failed samples due to dielectric breakdown over applied voltage for each group of samples. While none of the untrained CG DEAs failed up to 10 kV, the trained CG DEA all failed above 9 kV. Three of the LSGO DEAs also failed at 8.5 kV and 10 kV respectively, but unlike trained CG DEAs, neither of the two LSGO groups showed systematic failure of all samples. This indicates that training can cause damage to the membrane, leading to premature

failure in CG DEAs. Interestingly, LSGO DEAs did not show the same reduction in breakdown strength after training. The mechanism for this phenomenon is yet unknown, but it is an interesting observation that warrants further investigation. Diffusion of oil from CG into the dielectric membrane is not a likely explanation since a castor oil-based CG was used specifically to avoid this sort of interaction (see SI Section S6). Castor oil is not known to degrade ELASTOSIL® silicone rubber.^[33] Due to the systematic premature failure of CG DEAs, unwashed LSGO DEAs showed the highest actuation stroke overall.

To demonstrate the patterning ability of the laser scribing process, a 3-segment DEA was manufactured where the circular membrane surface is covered with three segments of LSGO electrodes of equal size (shown in **Figure 3a**). Actuating a segment results in areal expansion of that segment as well as lateral displacement of the center of the membrane as passive segments contract (Video S2). Figure 3c shows the average areal strain in the actuated segment (one segment active at any one time) for the given actuation voltage. At 10 kV, a strain of 8.8% is achieved. This is small compared to some previously reported DEAs with areal strain near 150%,^[34] but lies in the range that a contractile stack actuator might experience.^[35]

The laser scribing method can produce electrode patterns with sub-millimeter details. The minimum resolution is dictated by the laser engraver hardware used and here limited to 0.1 mm (see SI Sections S11 and S12); even higher resolution could be achieved with a different engraver. To showcase high-detail patterning, a DEA with electrodes in the shape of the University of Bristol logo was laser scribed. This is shown in Figure 3b (after washing). The

logo expands when actuated, with Figure 3c showing the areal strain in the electrode when increasing the voltage from 5 kV to 10 kV. Because the shape of the logo is not well suited for DEA actuation, the observed strain is smaller than in the 3-part DEA but is clearly visible by eye (Video S3). Effective actuation in DEAs requires interaction between active (actuated) and passive areas in the membrane. With interlaced active and passive areas covering most of the membrane surface in case of the logo, little effective strain in any particular direction is achieved. A reduction of strain was observed over three actuation cycles, with the areal strain measured in each cycle shown in Figure 3c. During actuation, electrical sparks were visible across some parts of the membrane at voltages above 6 kV as the membrane was charged. This did not lead to membrane failure but indicates that some areas of the electrode became disconnected as the membrane deformed. This is much more likely in an intricately patterned electrode, as many small areas are only connected by narrow pathways, which may break under strain. When the actuation voltage was sufficiently high, small gaps within the LSGO electrode were bridged by sparks. This leads to degradation of the electrode, which eventually results in isolation of disjointed areas and a decrease in the active electrode area. As can be seen in Figure 3c, this gradual degradation counteracts the increasing voltage, leading to an overall decrease in actuation strain that is retained in subsequent cycles. For practical applications, it may be necessary to ensure that connecting traces of LSGO have a specific minimum width to avoid sparking and degradation. For DEAs with circular electrodes (no narrow connecting pathways), preliminary fatigue tests do not show significant degradation in actuator stroke, capacitance, or resistance over 120 actuation cycles (see SI Section S10). Only washed LSGO DEAs show a very slight reduction in stroke over the course of the test,

although no increase in electrode resistance is evident. Future work will include further investigation of the long-term stability of LSGO. The preliminary results show great promise and warrant further investigation of LSGO as an electrode material for DEAs and soft electronic devices.

In conclusion, we have shown that LSGO is a viable method to produce complex, compliant electrodes for DEAs. In a direct comparison with CG DEAs using the same dielectric membrane, LSGO DEAs showed higher actuation stroke since they did not appear to suffer from the same reduction in breakdown strength that was observed in CG DEAs after training. The laser scribing method is simple, green, low-cost and allows for intricate patterning of electrodes on a sub-millimeter scale. In addition, LSGO DEAs are also easier to handle during preparation and testing than (unencapsulated) CG DEAs since the electrodes do not rub off as easily. We expect that this method will ensure broad uptake of such LSGO electrodes in a range of applications.

Experimental Section

Materials: GO dispersion in water (4 mg mL^{-1}) was purchased from Graphenea Inc. (USA) and diluted with DI water. ELASTOSIL® silicone film (Wacker Chemie AG, Germany) was purchased from Silex Ltd. L-ascorbic acid was purchased from Sigma-Aldrich Company Ltd. (UK) and used as received.

Membrane Preparation: To fabricate dielectric membranes for DEAs, ELASTOSIL® silicone membranes were pre-stretched biaxially to 2 times their original diameter (i.e. 300% areal strain) and attached to circular frames using Sil-Poxy® silicone adhesive (Smooth-On, Inc., USA). Frames are laser-cut from 1.5 mm thick PMMA and one frame is attached on either side of the membrane to maintain the pre-strain and serve as a mold for drop casting.

GO Preparation and Drop Casting: L-ascorbic acid (0.2 mg mL^{-1}) was added to GO dispersion (1 mg mL^{-1}). It serves as a mild reducing agent^[36] and was found to improve the reduction of GO during laser scribing. The dispersion is sonicated in an ultrasonic bath for 10 minutes and then immediately drop cast onto the pre-stretched silicone membranes (0.1 mL cm^{-2}). The films are dried at ambient conditions.

Actuator fabrication: GO films are cast on both sides of a pre-stretched silicone membrane. Laser scribing of GO films is performed with a 250 mW, 405 nm diode laser (see SI Section S11) at 1000 mm s^{-1} and 60% relative laser power.

Washing: After laser scribing, the unreduced GO can be washed off by gentle rubbing with a water-soaked piece of cotton wool. The hydrophilic GO will easily rub off when wet, while LSGO stays in place.

Electrical contacts: For easy connection, adhesive copper tape is attached to the frame to serve as contacts. Carbon grease is used to establish an electrical connection between the LSGO electrode and the copper tape on the inside of the frame.

See SI for detailed explanations and experimental conditions for the LSGO DEA fabrication process.

Characterization: Microscope images were taken on an Olympus BX50 polarized light microscope with no polarizing filters used, fitted with an Olympus Digital Camera. For actuation testing, two Ultravolt 5HVA24-BP1 HV amplifiers (Ultravolt, Inc., USA) were used to generate a maximum of 10 kV differential voltage to drive the DEAs. Stroke length was recorded using a Keyence LK-G152 laser displacement sensor (Keyence, Japan). To determine areal strain, DEAs were filmed using a Lumix G7 digital camera (Panasonic, Japan) mounted at a fixed distance above the membrane. Strain is determined by pixel counts of LSGO in segmented images using MATLAB (MathWorks, Inc., USA).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author. All relevant research data is available from the University of Bristol Research Data Repository: data.bris, at <https://doi.org/10.5523/bris.35xripkf6hemi21msqq6i662g6>.

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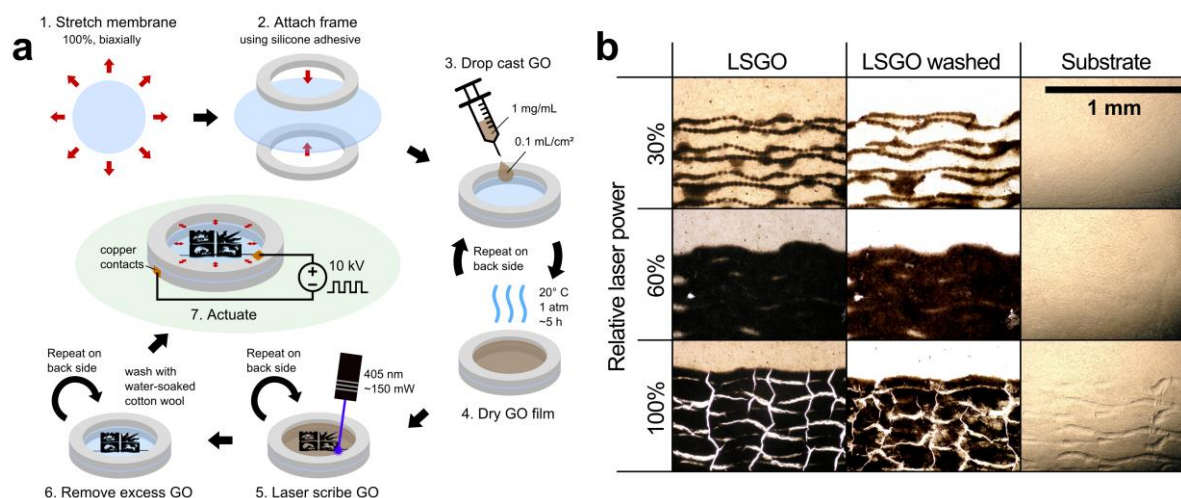


Figure 1. a) Fabrication process of a DEA with laser-scribed GO electrodes. b) Light micrographs of LSGO films laser scribed at 1000 mm min^{-1} with varying laser power. The first column shows LSGO (dark) and some remaining GO (light). The second column shows the same LSGO films after washing off unreduced GO with water. The third column shows the silicone substrates after completely removing GO and LSGO by washing with ethanol (the lighting was adjusted to highlight surface structure). At 100% laser power, damage to the membrane is clearly visible.

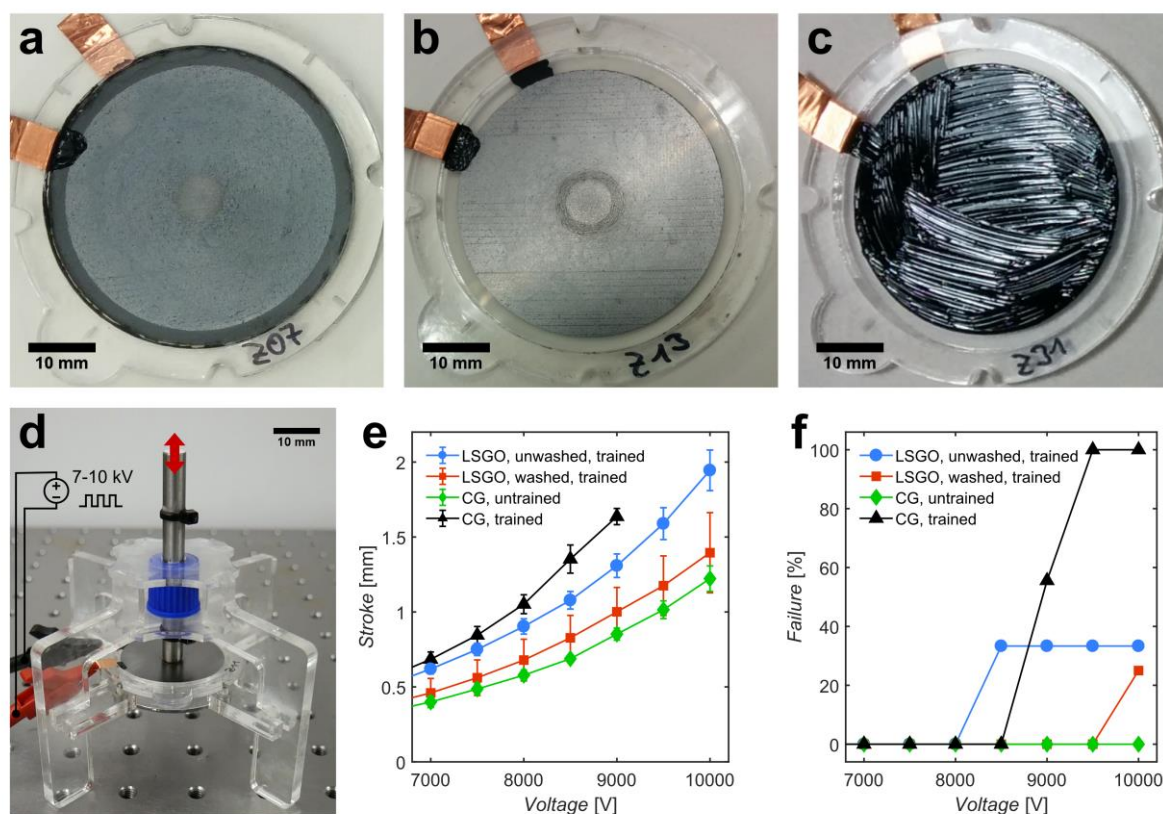


Figure 2. a) Unwashed LSGO DEA. b) Washed LSGO DEA. c) CG DEA. d) Actuation test setup. The vertical steel rod is moved up and down by membrane actuation (see Video S1). e) Stroke of LSGO and CG DEAs for given actuation voltage. Unwashed LSGO DEAs show the largest stroke recorded at 10 kV. f) Percentage of failed samples at given voltage in each group of DEAs. Untrained CG DEAs show systematic failure above 9 kV.

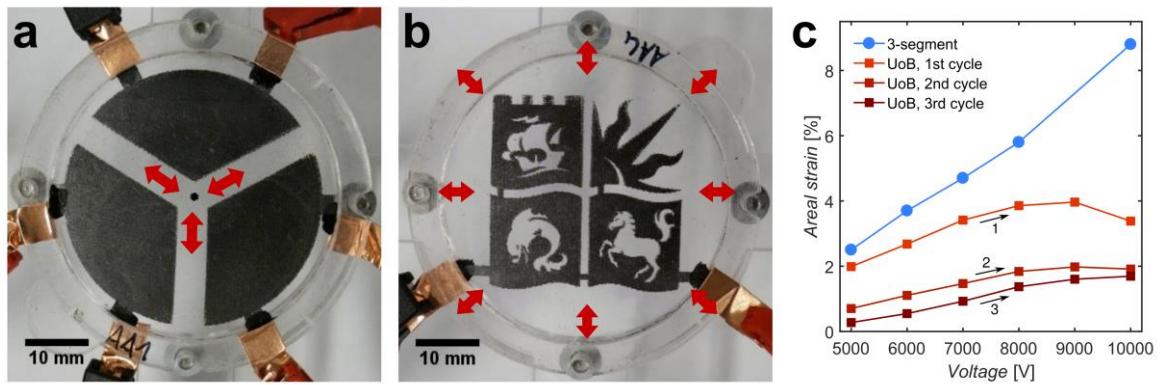


Figure 3. a) LSGO DEA with 3-segment electrode pattern, washed. b) LSGO DEA with University of Bristol (UoB) logo pattern, washed. c) Areal strain over voltage for 3-part and UoB DEAs. Three actuation cycles from 5 kV to 10 kV were recorded for the UoB DEA. Videos S2 and S3 show actuation of LSGO DEAs.

Easy Electrodes!

A major challenge for soft electroactive devices such as dielectric elastomers is the fabrication of thin, precisely patterned, stretchable conductors. In this work, laser-scribed graphene oxide (LSGO) is proposed to fabricate intricate, stretchable electrodes for dielectric elastomer actuators. The method is simple, low-cost and green (free of organic solvents) and produces arbitrary geometries with 0.1 mm resolution.

Keyword: dielectric elastomer actuators, soft electronics, laser scribing, graphene oxide, stretchable conductors

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Laser-scribed Graphene Oxide Electrodes for Soft Electroactive Devices

ToC figure

