Fabrication of stretchable composites with anisotropic electrical conductivity for compliant pressure transducers

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***Abstract*—We present a simple fabrication approach for anisotropically conductive stretchable composites, towards novel flexible pressure transducers. Flexible electronic systems have gained great interest in recent years, and within this space, anisotropic conducting materials have been explored for enhanced sensing performance. However, current methods for producing these materials are complex or are limited to small fabrication areas. Our method uses film applicator coating to render commercially available conductive RTVs anisotropically conductive. A ratio of in-plane surface resistance to through-thickness resistance of 10^{10} was achieved using our method. Furthermore, we show that when a normal pressure is applied to such films, the in-plane resistance can be reduced by seven orders of magnitude for an applied pressure of 10 kPa. Hence these materials show great promise for the development of novel, robust pressure transducers.**

***Index Terms*—Flexible electronics, stretchable electronics, anisotropic electrical conductivity, flexible pressure sensors.**

I. Introduction

Flexible electronics devices have gained great interest in recent years due to their potential for robust and conformable electronic systems, such as electronic textiles, skin-mounted health monitoring systems and wearable robotics. Recent trends have seen the development of compliant conductors with anisotropic properties for improved performance or to enable novel functionalities [1], [2]. However, fabrication of these anisotropic materials can be complex and limited in scalability in terms of fabrication size.

Here we present a simple method for the production of stretchable conductors with anisotropic conductivity using film applicator coating, a process well suited to large-area fabrication and roll-to-roll manufacturing. We demonstrate control over the level of anisotropy simply by adjusting the separation distance between the film applicator and coating substrate g. Moreover, we show that for films cast using with low values of g, the in-plane resistance can be drastically reduced by the application of a normal pressure. Finally, we utilize this effect to build a flexible pressure switch exhibiting large changes in resistance to pressures above 5 kPa. The results demonstrate the potential of these anisotropically conductive composites for the development of highly responsive and flexible pressure transducers for autonomous soft robotic systems, or wearable pressure responsive devices.

II. Fabrication Procedure

We use the commercially available, conductive, room temperature vulcanize (RTV) silicone SS-25M (Silicone Solutions) as the conductive composite. The composite has nickel coated graphite particles as the conductive filler with average particle diameter of 25 μm (Fig. 1e). The conductive filler loading is high, between 35-70 % of the total mixture weight. We use an automatic film applicator (Elcometer, 4340), with a height adjustable applicator bar, to cast the conductive composite into thin films. The fabrication procedure is as follows: A PET substrate is first coated with a thin layer of poly(acrylic acid) (PAA) aqueous solution (Sigma Aldrich), which acts as a water-soluble sacrificial layer. The PAA solution is diluted with isopropyl alcohol (IPA) prior to coating to reduce drying time (solution composition is 5% PAA: 9.3% water: 85.7% IPA by weight). The PAA coating is left to dry in ambient conditions for 2 minutes, forming a layer approximately 1 μm thick. The silicone RTV is then cast on top of the PAA layer to the desired thickness at a speed of 10 mm/s and subsequently cured in an oven at 70°C for 24 hours (Fig. 1a and Fig. 1b). The film is then released by submerging in warm water and left to dry for a further 24 hours prior to performing resistance measurements.

III. Characterization of Resistance

We characterize the in-plane (x and y directions as defined in Fig. 2a) and through-thickness (z direction) resistances of the cast composite films as a function of film applicator separation g, which preliminary experiments showed to be the predominant variable affecting anisotropy in conductivity. Composite films cast using a separation g = 100 μm, 200 μm, 500 μm and 1 mm (here after referred to as g_{100}, g_{200}, g_{500} and g_{1000}, respectively) are characterized. The cured film thicknesses, h, are 50 μm, 95 μm 270 μm and 370 μm, respectively. The experimental setup for the resistance measurements is schematically shown in Fig. 2a. Two rectangular Neodymium magnets (K&J Magnetics, dimensions: 9.5 mm by 4.8 mm by 1.6 mm) are used to form the electrical contacts to the composite film, which is magnetic due to the nickel present in the filler particles. A precision digital multimeter (Agilent 34401), operating in four-probe measurement mode (to eliminate contact resistances) is used to the measure resistance. For in-plane measurements, a non-conductive spacer is placed

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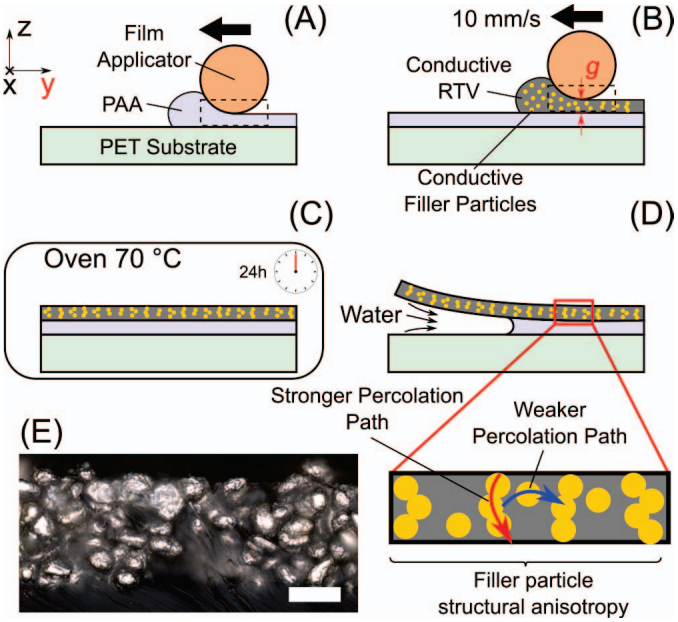

Fig. 1. Schematic of fabrication procedure. (A) Casting of PAA water-soluble sacrificial layer on to the PET substrate using an automatic film applicator. (B) Casting of the conductive RTV on top of the dried PAA layer. (C) Curing of the cast RTV film in an oven at 70 °C for 24 hours. (D) Dissolving of the PAA layer to release the RTV film and schematic hypothesis for film resistance anisotropy. (E) Optical microscope image of a cross-section of an example cast composite ($g = 150$ µm, scale bar is 40 µm).

between the two magnets to prevent electrical contact, and also to define the in-plane separation distance, s (see Fig. 2a). For each test, s is set approximately equal to the composite film thickness h (i.e. the separation of the electrical contact is normalized by the film thickness for each sample). For through-thickness measurements, the magnets sandwich the composite film. Measurements where taken on four locations on each sample and averaged.

The results of the resistance characterization are shown in Fig. 2a, showing that the in-plane resistances, in the x and y dimensions, decrease with increasing g, whereas the through-thickness resistance increases monotonically. The results also show a substantial difference in in-plane resistances across the various samples. With the g_{100} sample, the x and y resistances were larger than the measurement range of the multimeter used, which was 2 GΩ (the resistance for this sample was set to 2 GΩ, representing a conservative estimate of the true resistance) where as the resistance was less than 30 Ω and 50 Ω for the g_{500} sample for the x and y resistances, respectively.

Most notably, the degree of conductivity anisotropy between in-plane and thickness dimensions is extremely high, as highlighted in Fig. 2b, which plots the anisotropy ratio a - the ratio of the mean of the average in-plane resistances to the average through-thickness resistance. The figure shows that an anisotropy ratio of over 10^{10} is achieved with the g_{100} sample.

We hypothesize that the dominant mechanism by which the composite becomes anisotropically conductive is shear-stress-induced structural anisotropy of the conductive filler particles, similar to methods used in the fabrication of aligned nano structures [3]. This leads to a reduced percolation path in the in-plane dimensions, while compacting the fillers in the thickness dimension and thus ensuring percolation (Fig. 1d). There are likely several factors which influence this process, including composite viscosity, casting speed, substrate surface energy and filler type. Increased anisotropy is likely to be achieved as the applicator separation approaches the filler particle diameter, but at the cost of film homogeneity and mechanical integrity. Unlike previously reported methods for the fabrication of anisotropically conductive materials, the method presented here is simple and well suited to fabrication in large areas. Moreover, because these materials are elastomer based, they can be deformed and stretched readily.

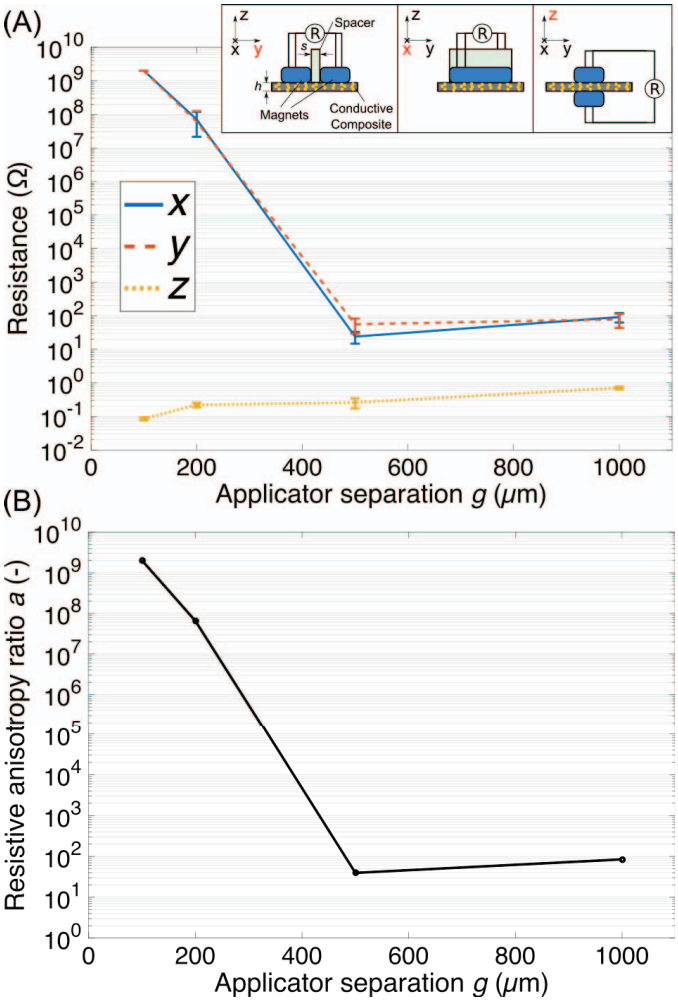

Fig. 2. Characterization of resistance of fabricated composite samples. (A) In-plane (x and y) and through-thickness (z) resistances as a function applicator separation g (average of four measurements, error bars are ± one standard deviation). Inset, schematic depiction of the experimental setup. (B) Ratio of the mean of the average in-plane resistances to through-thickness resistance, a, against applicator separation g.

IV. Flexible Pressure Switch

Applying normal pressure to the composite films cast with low values of g were found to drastically reduce their in-

plane resistance. The proposed mechanism for this effect is a combination of: a) the bunching together of filler particles in the in-plane dimensions, as a result of material incompressibility and shear stresses generated in the film, b) electron tunneling across the small gaps between filler particles, which vary with applied pressure, as proposed by [4]. We utilize this mechanism to construct a flexible pressure switch. The switch is composed of a g_{100} composite sample placed on top of a circular, 5 µm thin, interdigitated copper electrode adhered to a PET flexible substrate. The interdigitated electrode is approximately 4 mm in diameter, with 240 µm line widths and 150 µm wide spaces (Fig. 3a).

The response of the switch to normal pressure is characterized using the experimental setup depicted in Fig. 3a. A stiff, non-conducting rod approximately 8 mm in diameter is attached to an Instron tensile testing machine (model 5544A) via a pneumatic gripper. A compressive force is applied by the Instron via the stiff rod onto the surface of the pressure switch. Resistance is measured using the precision digital multimeter described in the previous section, operating in four-probe measurement mode. Electrical connections are made to the electrodes via magnets (the iron surface beneath the switch keeps the magnets in place).

The results of the experiments show a substantial decrease in resistance from greater than 2 GΩ to less than 150 Ω for an applied pressure of less than 10 kPa. The change in resistance generated for the applied pressure is significantly greater than that previously reported for other pressure transducers based on anisotropically conductive films [2]. This transduction mechanism is extremely attractive for a variety of applications where large changes in resistance, as a function of applied pressure, are desired. Such devices could be employed in intelligent soft robots, or wearable electronics systems, capable of responding autonomously to discrete changes in the mechanical environment by directly controlling electrical power to an actuator.

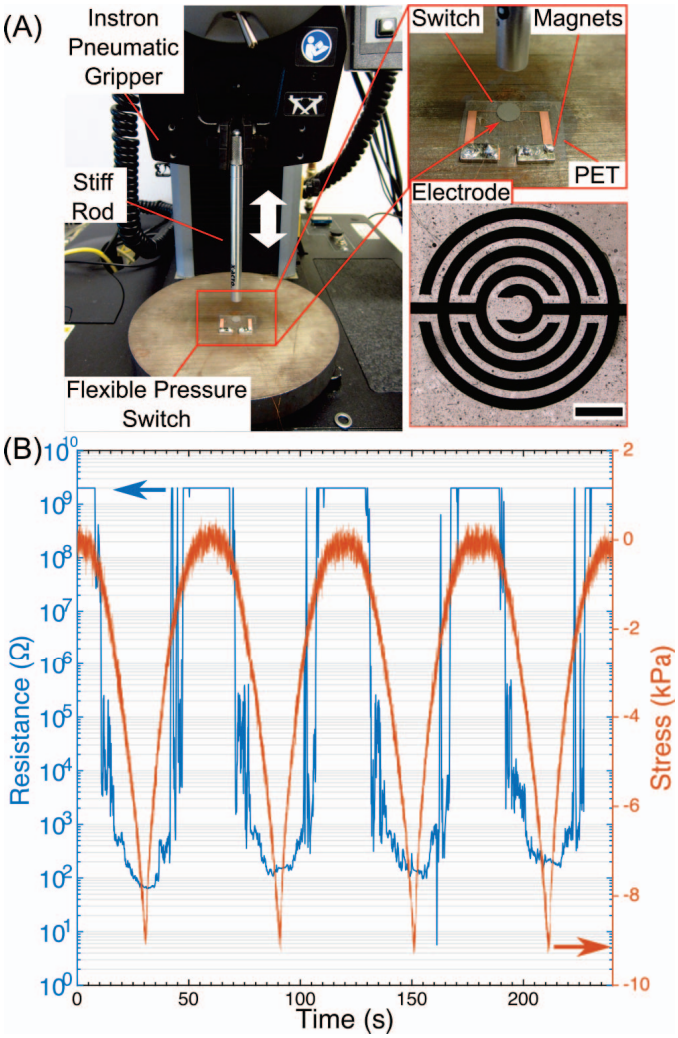

Fig. 3. Response of flexible pressure switch to normal pressure. (A) Experimental setup. Insets show close-up of test sample (top) and circular interdigitated electrode geometry (bottom, scale bar 1 mm). (B) Graph of in-plane resistance (i.e. resistance between the interdigitated electrodes, left y-axis, shown in blue) and normal pressure (right y-axis, shown in red) against time.

V. Conclusions

Here we present a simple approach to the fabrication of stretchable conductors with anisotropic electrical conductivity. Our approach involves the casting of commercially available conductive RTV composites using an automatic film applicator. Films cast at low applicator separation values exhibit extreme resistance anisotropy, as high as 10^{10}, between the in-plane resistance and through-thickness resistance. Furthermore, application of a pressure normal to the surface the films cast at low applicator separator values causes the in-plane resistance to decrease substantially. We utilize this effect to build a flexible pressure switch capable of a 10^7 order magnitude change in resistance (from 2 GΩ to below 150 Ω) in response to an applied pressure of 10 kPa.

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