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Highly stretchable printed strain sensors using multi-walled carbon nanotube/silicone rubber composites

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ABSTRACT

The developing fields of wearable electronics and soft robotics have created a strong demand for flexible and stretchable strain sensors. This work presents a printing technique and conductive multiwalled carbon nanotube/silicone polymer nanocomposites applied to achieve embedded piezoresistive large strain sensors. A printing process that allows the sensors to be produced inside a silicone substrate has compelling advantages: fabricated devices are well insulated and protected against wear. These process features simplify fabrication of complex patterns. Printed sensors can withstand stretching of up to 300%, with maximum hysteresis of 11% for 300% maximum strain, and gauge factors between 1.0 and 1.5 depending on the selected composition. The devices developed in this work have potential applications in the measurement of human body motion and position control of soft actuators.

1. Introduction

Common rigid electrical components are frequently ill suited for integration into wearable systems and soft robots. In these applications a fully flexible structure and large strain range are important design goals [1]. Additionally, highly compliant soft devices have infinite degrees of freedom leading to complex, distributed displacements [2,3]. Therefore a pattern of several sensors distributed around a part to measure multi axis deformation is desirable. The nanocomposite printing technique applied in this work has advantages to automate the fabrication of miniaturised strain sensors embedded in stretchable polymer structures.

In traditional, rigid systems the small deflections of parts can be measured using devices such as metal and semiconductor strain gauges. While these devices can precisely sense small deformations, limitations such as small maximum strain (of the order of 5% for metallic strain gauges [4,5]), stiffness, and challenges integrating metals and semiconductors with compliant parts make these conventional devices unsuitable for systems fabricated from soft polymers. As a result, the development of flexible large strain sensors has been an active area of research, with two standout transduction methods: capacitive sensing using conductive layers on either side of a stretchable dielectric [6,7], and piezoresistive sensing using a deformable conductive material [8–10].

Common capacitive large strain sensors consist of two conductive electrodes separated by a polymer dielectric. In this approach, the stretching and thinning of a polymer dielectric layer causes an increase in capacitance [5]. With an appropriate external sensing circuit [11], capacitive large strain sensors can achieve acceptable resolution and hysteresis. However, the requirement for thin, uniform dielectric and electrode layers necessitates specialised production techniques. The required external capacitance measurement circuit, which with current technology cannot be implemented in flexible materials, increases size and cost.

In contrast to capacitive devices, piezoresistive large strain sensors can be fabricated in a single layer, and external circuitry is significantly simplified. Piezresistive devices may therefore be preferred for interoperation with other printed components. Conductive films and embedded liquids are two significant techniques for implementing piezoresistive strain sensors on stretchable polymer substrates. In film based sensors, a thin layer of conductive material such carbon nanotubes [12], graphene [13], or conducting polymer [14] dispersed in a solvent or inert polymer is coated on the substrate.

One disadvantage of sensors of this type is that adhesion issues between the conductive layer and the substrate may cause cracking or delamination [15], frequently limiting the maximum strain of sensors of this type. By using embedded conductive liquids [16]

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or grease, the cracking and lifetime challenges of coated film sensors can be avoided, resulting in achieving excellent stretchability. However, the encapsulated liquid inside the stretchable structure does not provide mechanical strength and may flow within the part.

The printed carbon nanotube/silicone rubber piezoresistive sensors developed in this work are intended to have several advantages in comparison to other sensor designs. The sensors developed in this work are embedded within the stretchable substrate and protected against wear. When judged against the carbon nanotube mesh sensors recently demonstrated by Guo et al. [17], we show linear response at substantially higher maximum strains. By using a cured and highly stretchable matrix material for the conductive composite in place of a conducting liquid, the liquid absorption issues encountered in liquid-filled sensors by Yildiz et al. [18] can be avoided.

Finally, deposition by printing, which allows almost arbitrary geometry to be created [19], is attractive for future integrated fabrication with sensors, electrical interconnection, and other ancillary components. In comparison to the work of Muth et al. [19], where conductive carbon grease was printed inside the substrate and remains fluid, we deposit carbon nanotube/silicone rubber composite that, on curing, forms a solid part of the finished sensor, well-bonded to the surrounding material.

After printing, the sensors presented here were characterised by stretching measurements. In other research, negative temperature-resistance coefficients of carbon nanotube/polymer composites have been noted [20,21], so in order to understand the conduction mechanism and allow temperature compensation in practical use, the temperature dependence of conductivity was measured.

2. Material preparation and printing method

In this work, multiwalled carbon nanotubes (MWCNT) was selected as the conductive phase due to high conductivity of composites [22] and acceptable material cost. The use of silicon polymer for the substrate and inert phase allows for excellent stretchability. Appropriate material mixtures for printing were prepared by mixing multiwalled carbon nanotubes (MWCNT) with uncured silicone and other additives. This mixture was then printed inside a partially cured silicone substrate to produce a sensor.

2.1. Preparation of conductive nanocomposite mixtures

The conductivity, maximum stretch, stiffness, and resistance-strain relationship of the cured conductive material depends on the size and concentration of MWCNT, silicone, and additives such as solvents or thinners. In this work, the commercially available addition curing silicone Ecoflex® 00-30 [23] (with Shore hardness 00-30) was used in combination with multi-walled carbon nanotubes purchased from Shenzhen Nanotech Port Co. Ltd. (with diameter in the range 10–30 nm and length > 2 μm).

To prepare the conducting material for the printing, nanoparticles were added to uncured silicone and additives in the desired mass ratio, prior to centrifugal mixing to achieve a uniform dispersion (using the mixing equipment Kurabo Mazerustar KK-50S). In preparation for printing, uncured MWCNT/silicone mixture was then loaded into syringes under vacuum to avoid air bubbles. Results for varying mass ratios of MWCNT and silicone are shown in Table 1. In addition to conductivity (which can be adjusted over several orders of magnitude [24]), the mixing ratio of MWCNT affects the viscosity of the mixture. At low mixing ratios, the mixtures are not conducting, while when the mixing ratio is too high, the material is too viscous to print.

Table 1
Effect of MWCNT/silicone mixing ratio.

Ratio MWCNT:silicone	Conductivity	Printability
1:10	Yes	No
1:12	Yes	No
1:15	Yes	Yes
1:17	Yes	Yes
1:20	>40 MΩ	Yes

2.2. Sub-surface printing within uncured polymer

Surface wear and layer delamination are common failure modes of existing stretchable conductive materials. Sub-surface printing of curable conductive nanocomposite inside partially cured insulating matrix is a convenient method to achieve conductive traces which are protected from surface wear, well bonded to the surrounding material, and (unlike conductive liquids or greases) contribute to structural strength. As shown in Fig. 1, in an arrangement similar to a fused deposition modelling 3D printer, a syringe driver is used to extrude prepared uncured conductive composite into a partially cured silicone substrate through a 0.6 mm diameter nozzle. By controlling the X–Y motion of the substrate, arbitrary patterns can be created.

An example of a highly stretchable strain sensor printed inside a silicon rubber substrate is shown in Fig. 2. A cross section of the embedded trace is shown in Fig. 3. Prior to printing, the mixed Ecoflex® 00-30 substrate was left to stand for 20 min. At this point the substrate has partially cured, and is sufficiently viscous that the conductive MWCNT/silicone rubber composite remains in place after deposition, but sufficiently fluid that the material surface reflows to leave no mark. It is important that the composite is printed sufficiently deep inside the substrate so that the surface of the substrate does not crack when stretched. The speed at which the printing head moves through the substrate must be limited to

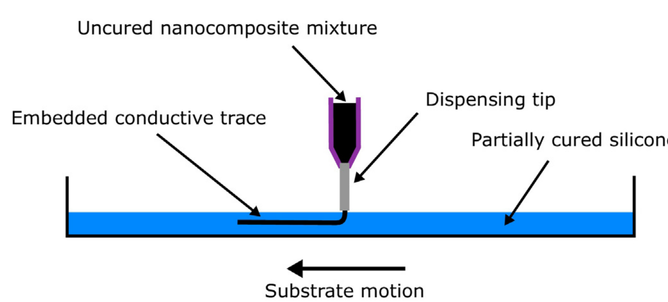

Fig. 1. Printing of embedded conductive traces in partially cured silicone.

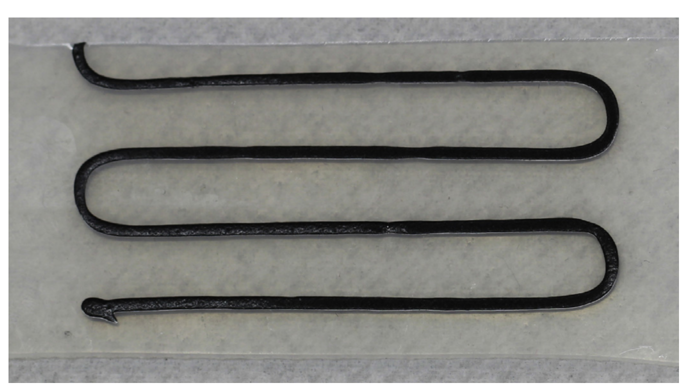

Fig. 2. Example printed conductive pattern embedded in silicone substrate.