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Abstract

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The bending movement of the three interphalangeal joints of the finger were recorded simultaneously. The sensing elements (i.e. soft strain sensors) showed an outstanding linearity between the change of resistance and the strain during the finger bending, with a gauge factor of ~ 1 , repeatability and stability ($>10,000$ cycles), fast response and low creep, with negligible mechanical and electrical hysteresis. These sensors have a significant potential to establish prosthetic devices with built-in sensing elements, paving the way towards prosthetic limbs compatible with natural control.

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Large area and ultra-thin compliant strain sensors for prosthetic devices

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Abstract

There is an increasing need to establish skin-like ultra-thin sensors which can directly be deposited (e.g. sprayed) on a soft active structure representing the limbs (e.g. fingers) of prosthetic devices such as a prosthetic hand. Such sensors are essential to control the actuation behaviour of the soft structure, and to obtain information about temperature of an object which the finger is touching. This study employed an inexpensive and scalable technique to place elastomeric sensors directly onto a monolithic structure representing the finger of a soft robotic prosthetic hand. The chemical affinity between the solvent used, and the soft elastomers (thermoplastic urethane, TPU, and styrene-butadiene-styrene, SBS) allowed the blending of the TPU and the SBS at the finger interface, creating a highly compliant piezoresistive sensor, that was directly deposited in the desired place.

The bending movement of the three interphalangeal joints of the finger were recorded simultaneously. The sensing elements (i.e. soft strain sensors) showed an outstanding linearity between the change of resistance and the strain during the finger bending, with a gauge factor of ~1, repeatability and stability (> 10,000 cycles), fast response and low creep, with negligible mechanical and electrical hysteresis. These sensors have a significant potential to establish prosthetic devices with built-in sensing elements, paving the way towards prosthetic limbs compatible with natural control.

1. Introduction

Wearable devices and stretchable sensors have recently received significant attention due to the nature of their interaction with the human body, their easy integration, and their long-term monitoring capabilities with minimal maintenance [1]. Flexibility and stretchability – the

ability to conform to and cover active and arbitrarily shaped objects needs to be studied in order to design wearable devices such as sensors which can be embedded into clothes or attached directly to skin for real-time monitoring of human activities such as monitoring personal health and in therapeutics, or human movement [1, 2].

The measurement of the strain involved in human motion often requires sensors with high stretchability. Conventional strain gauges have a limit of 5 % strain in order to show a linear strain response [3]. Polymeric materials, especially elastomeric ones, are known for their outstanding stretchability with minimal mechanical hysteresis and this makes them suitable for high strain applications. The use of one dimension (1D) conductive materials such as carbon nanotubes or graphene is a popular solution to produce conductive nanocomposites with outstanding chemical, mechanical and electrical properties [1, 4, 5]. The use of conductive nanofillers with high aspect ratio allows the construction of conductive nanocomposites with a lower percolation threshold, which improves the mechanical performance without compromising the electrical behaviour [6]. Two-dimensional networks of 1D materials have outstanding mechanical and electrical performance because they can slide or deform while maintaining a percolating pathway [7]. These nanocomposites can find applications as skin-mountable [1, 8], wearable [9], temperature [10], strain, [2] pressure sensors for soft robotic applications.

Intrinsically stretchable sensing devices are based on the use of a soft elastomeric matrix and conductive 1D nanofillers. The polymeric matrix gives mechanical stretchability, while the conductive fillers add the electronic properties to the nanocomposite, creating a large strain sensing device. Polydimethylsiloxane (PDMS) and Ecoflex are the commonly used elastomers to build large strain gauge sensors. Usually, these sensors are printed or painted on the surface of the elastomeric matrix and then encapsulated with another layer of the elastomer [1]. The use of a sandwich system with soft materials requires numerous fabrication steps to fabricate the sensors which are difficult to replicate, especially at large scale. The sensors also need to be assembled onto the surface of the target object, compromising the intimate contact between the sensing material and the target object due to the incompatibility between the compliance of the sensor and target object, reducing the sensor's accuracy, repeatability, and operation life, especially under cyclic loading.

To create fully compliant strain sensors, not only new materials but also new fabrication approaches are needed to provide additional features to the devices, without compromising the mechanical and the electronic properties. Spray coating is one inexpensive, easy to implement manufacturing technique but it remains unexplored as a method for the

development of stretchable sensors. In preparation of the conductive inks to establish soft strain sensors, the chemical affinity between the solvents and the materials used as a sensitive part and the target object has not been studied. This has resulted in problems such as peel off from the target object, limited repeatability, and low operation cycles.

In this work, we introduce a new type of stretchable electronic nanomaterial consisting of polymers and solvent with favourable chemical affinity, creating high performance mechanically compliant strain sensors deposited on a prosthetic finger made of a soft thermoplastic. Multiwall carbon nanotubes (MWCNT) – SBS ink was prepared and sprayed on the outer surface of the interphalangeal regions of the three dimensional (3D) printed soft robotic finger that was used as the soft target object. The sprayed ink on the soft strain sensor does not require encapsulation or protective layers. Experiments conducted with the soft finger with an ultra-thin strain sensor sprayed onto it show the sensor linearly follows the bending behaviour of the soft finger at different speeds, with a high repeatability (more than 10,000 cycles were applied), with fast response and low creep. The strain sensor was also shown to can recreate the temperature sensation of the skin, by changing its resistance with temperature. Based on the scalable fabrication technique, the strain sensors can be formed into complex shapes that will fit on almost any surface to reliably generate signals indicating strain in the surface they are assembled on.

2. Materials and Methods

2.1. Soft Robotic Finger Design and 3D Printing

The soft robotic finger has been designed as a monolithic structure with three flexural hinges, resembling the human index finger in geometrical dimensions [11]. While the length and other geometric dimensions of the soft finger model are from a typical human index finger, the proximal phalange of the finger has been designed shorter by moving the metacarpophalangeal (MCP) joint location closer to the proximal interphalangeal (PIP) joint to increase the conformability of the soft robotic finger. Channels for tendon-cables were designed to house tendon-cable routing to lower the friction between the tendon-cable and the soft channels and increase the bending moments acting onto the flexure hinges of the finger (Figure 1).

Filaflex, a polyurethane based thermoplastic (TPU) was used as a soft material to 3D print the soft finger with predesigned flexure hinges (Figure 1a). Non-symmetric elliptical flexure

hinges are used in the soft finger and this generates larger displacements under the lowest force input [12]. Geometric relation for the thickness of the non-symmetric elliptical flexure hinge is given as [13]:

$$t_{\text{elliptical}}(y) = \frac{t + 2c \left[1 - \sqrt{1 - \left(1 - \frac{2y}{c}\right)^2} \right]}{2} \quad (1)$$

where t is the thickness (3.0 mm) of the flexure hinge at its thinnest region, c (15.5 mm) and l (16.0 mm), are the depth and length of the hinge groove, respectively, and w (20.0 mm) is the width of the flexure hinge (Figure 1a).

2.2. Sensor Material Preparation and Deposition

Multi-walled carbon nanotubes (MWCNT) were provided by Nanoamor (purity 95%, outer mean diameter = 20 – 40 nm; length = 5 – 15 μm) and commercial Calprene C540, a linear tri-block SBS copolymer was supplied by Dynasol Gestión, S.A. The elastomeric matrix is composed of 40% styrene. To promote good dispersion and disentanglement of the MWCNT bundles, the fillers were placed inside a beaker, 5.5 mL toluene (Sigma Aldrich) was added and kept in an ultrasound bath (*SoniClean*) for 4h. Then, 1 gram of the elastomeric SBS copolymer was added to the solution and stirred at room temperature until complete dissolution. The amount of MWCNT in the solution was selected to provide a concentration of 4 wt% in the SBS matrix. The solution was then transferred to a spray gun chamber to spray onto the soft finger in the desired configuration using a mask including the geometry of the strain sensor, and the solvent evaporation was performed at room temperature (Figure 1b).

2.3. Materials Characterization

The morphology of the sprayed sensors was analyzed by scanning electron microscopy (SEM, JEOL 7500). Samples were fractured after immersion into liquid nitrogen for 10 min and platinum coating was deposited on the surface and cross-section of the samples by sputtering (Dynavac Sputter Coater) with the thickness of ~ 10 nm. Infrared measurements (FTIR) were performed at room temperature in a, IRAffinity-1S (Shimadzu) in ATR mode from 4000 to 650 cm^{-1} . FTIR spectra were collected with 32 scans and a resolution of 2 cm^{-1} , at room temperature.

Figure 1: a) Geometric parameters of a non-symmetric elliptical flexure hinge and a 3D printed monolithic soft robotic finger (3D Printing Systems, UP Plus 2) using Filaflex filament, b) Spray-painted schematic process of the soft finger, and c) soft robotic prosthetic finger with joints (flexure hinges) labelled. Finger and interphalangeal sensors correspond to the final prosthetic device.

2.4. Electrical Conductivity Measurement

Electrical resistance of the elastomeric composites was calculated from the slope of I–V data measured with an automated Keithley 487 picoammeter/voltage source. I–V data points were collected between Au contacts, deposited on both sides of the samples with a Smart Coater sputter coater (JEOL). Samples resistance (R) was measured with rectangular contacts of 5 mm x 1 mm, on each side of the U-shaped sensor. The sheet resistance of the samples (R_s) was calculated applying equation 2:

$$R = R_s \frac{L}{w} \quad (2)$$

where w is the distance between the two metallic contacts and L is the length of the metallic contacts.

2.5. Actuation of The Soft Robotic Finger and Acquiring its Displacements

The tendon-cable actuation method was employed to flex the soft finger where a single tendon-cable is passed through the designated channels within the finger. This actuation method was chosen not only because of its simplicity but also to construct an adaptive robotic finger which is underactuated and fully compliant with its flexure hinges. Although an underactuated robotic finger is unable to make various finger movements, it is a simple and effective method for conformal grasping which wraps the finger around the object grasped. The finger was flexed by the tendon-cable that was attached in the other end to a servo motor. A pulley with a diameter of 82 mm was designed and fabricated from 3 mm acrylic sheets using laser cutting, and attached onto the servo motor to pull the tendon-cable at different rates, from 2.1 up to 16.7 mm/s. The motion of the soft finger was recorded with a high speed digital camera (Nikon D5100) from a relaxed state to a fully flexed state and vice-

versa. Red markers were used at the tip and joints of the finger to aid image analyses and obtain bending displacements.

2.6. Strain Measurement on the Flexure Hinge Surface

Strain rates on the flexure hinges where the elastomeric strain sensors were printed were obtained experimentally, using image processing. The strain rates of the flexure hinges are also validated using finite element (FE) models of the hinges. Images of the 3D printed soft finger were taken with a digital camera placed vertical to the motion plane of the finger. Two images were taken at initial and final bending configurations of the finger to determine the change in the effective length of the elastomeric strain sensors. Six discrete lines overlaying on the surface of the flexure hinge of the finger are used to form the curve of the finger surface. Initial and final effective lengths of the flexure hinges were measured by summing up these discrete line lengths with an image pixel to mm conversion of 2.833. The experimental strain was compared and validated by the numerical results from the finite element model simulations of the flexure hinges.

2.7. Electro-Mechanical Performance

Electro-mechanical measurements were performed by measuring the electrical resistance of the sample, through silver electrodes painted on the edges of the sensors, and the electrical resistance was obtained (Figure 1).

The change in the electrical resistance dR/R_0 due to the mechanical deformation (dl/l_0) are quantitatively determined by the Gauge Factor (GF), defined as:

$$GF = \frac{dR/R_0}{dl/l_0} \quad (3)$$

GF can be also written as:

$$GF = \frac{dR/R}{\varepsilon_l} = \frac{d\rho/\rho}{\varepsilon_l} + 1 + 2\nu \quad (4)$$

where $dl/l = \varepsilon_l$ and ν is the Poisson's ratio. Per equation 4, the GF has a contribution from the intrinsic piezoresistive effect $\left(\frac{d\rho/\rho}{\varepsilon_l}\right)$ and another from a geometrical effect $(1 + 2\nu)$. The movement of the finger bending was performed at different strain rates (between 2.1 and 16.7

mm.min⁻¹) and equation 3 was used to calculate the GF, using the variation of the resistance during the actuation-strain measurements.

3. Results

3.1. Strain Analysis Obtained from Computer Simulation and Image Processing

Finite element (FE) analysis was performed to characterize the distribution of the strain along the developed prosthetic device. Figure 2a shows the simulation results, and the positions that are submitted to higher strains during the bending process are the interphalangeal ones. Based on the results provided by FE simulation, MWCNT-SBS elastomeric strain sensors were deposited in the positions that were exposed to higher deformations (Figure 2). The experimental value of the strain was calculated based on the images recorded during the bending process. Discrete lines overlying the surface of the sensor were used to generate the curvature of the soft prosthetic device, in the places where the sensors were deposited, and a maximum strain of 10 % was recorded experimentally, and was in accordance to the value estimated through the FE analysis (Figure 2). It was also validated that the flexion (bending) angle of the MCP and PIP interphalangeal joint are approximately 58°, and for the DIP interphalangeal joint the value is 68°.

Figure 2. Soft robotic finger strain measurement: a) Finger interphalangeal region with the strain distribution during the bending process, obtained by FE analysis with a maximum strain of 10 %, and b) experimental method used to record the strain of the interphalangeal regions.

3.2. Chemical Affinity and Morphology of the Printed Strain Gauge Sensors

A key feature of the printed parts (sensor and finger) are the chemical properties of the materials to obtain a strong chemical affinity between the Filaflex used to printed the finger part and elastomeric piezoresistive sensor, and therefore, eliminate the need of an intermediate layer between the moving part (finger) and the sensor material.

The relative affinity of the polymers (TPU and SBS) and the solvent (toluene) used can be estimated through the Hansen solubility parameters (δ) by:

$$\delta^2 = \delta_d^2 + \delta_p^2 + \delta_h^2 \quad (5)$$

According to the Hansen and Skaarup equation, δ includes a dispersive force component (d), a dipole – dipole interaction (p) and hydrogen bonding forces component (h) [14]. Materials with similar levels of Hansen solubility parameter are likely to have a higher miscibility. The difference in the solubility parameter for the polymer-solvent interaction can be estimated from:

$$\Delta\delta_t = \left[(\delta_{P,p} - \delta_{S,p})^2 + (\delta_{P,h} - \delta_{S,h})^2 + (\delta_{P,d} - \delta_{S,d})^2 \right]^{1/2} \quad (6)$$

where, p , d and h indicate the polar, dispersive and hydrogen bonding components, respectively, and the symbols P and S identify the polymer and the solvent, respectively. Equation 6 can be used to predict the affinity between the solvent and the TPU polymer used to print the soft finger and the SBS elastomeric strain sensor. The values obtained for the $\Delta\delta_t$ between the solvent and the TPU was $8.23 \text{ MPa}^{-1/2}$ and between the toluene and the SBS was $2.35 \text{ MPa}^{-1/2}$. This result suggest that the solvent used had a strong affinity with the SBS elastomeric matrix and is a good solvent to dissolve it, but on the other hand, was a weaker solvent for the TPU (table 1).

Table 1: The solubility parameters ($\text{MPa}^{1/2}$) of the polymers, and solvent used for the spray-printing process [14, 15].

Material	δ_D	δ_P	δ_H	δ
Toluene	18.00	1.40	2.00	18.16
SBS	17.29	0.38	0.00	17.29
TPU	18.45	3.66	9.90	21.26

Figure 3a shows the surface morphology of the strain sensors placed on the joints of the finger. Scanning electron microscopy was performed to the spray deposited strain sensors to evaluate the morphology of the 3D printed soft robotic finger (Figure 3a), the microstructure of the MWCNT-SBS nanocomposite, and the interface between the piezoresistive sensor and

the 3d printed finger (Figure 3b). Regular Filaflex printed lines with a width of approximately 230 μm was observed, and the spray printed sensor followed the roughness of the soft finger (Figure 3a). The surface of the elastomeric nanocomposite presented a non-interconnected porosity, with pores smaller than 10 μm that are mainly distributed over the surface, rather than the bulk. We postulate that this porosity is probably due to the fast solvent evaporation process that occurs at the surface of the sensors during spraying and the polymer chains do not have enough time to occupy the free space left behind by the solvent.

Figure 3b shows that the thickness of the strain sensors is less than 500 nm, and the absence of a straight boundary between the finger TPU substrate and the MWCNT-SBS sensor suggests that there is very good compatibility between the two polymers: TPU (finger) and the MWCNT-SBS. Finally, there are no MWCNT clusters on the sensor surface or cross-section, which suggests that the ink preparation along with the spray procedure has resulted in a homogeneous dispersion of the multiwall carbon nanotubes in the SBS matrix.

Figure 3. Spray coated MWCNT-SBS sensors: a) surface and b) cross-section of the sensor. Inset shows a detail of the filler among the polymer matrix.

Recent strategies to develop strain sensors are based on sandwiched structures, where the piezoresistive material is placed in the middle of polydimethylsiloxane (PDMS) or silicon substrates, and the whole sandwich structure is then transferred and attached to the desired device by applying an intermediate layer to fix the sensors to the mechanical part [1]. Sandwich structures require an enormous number of steps to fabricate the sensors, which are very difficult to replicate at a large scale, and some peeling and leaking are unavoidable [1, 4]. Embedding the sensor materials into the soft and flexible materials shows that the performance of the sensor can be compromised due to the damping effect that often occurs due to the differences in the mechanical properties of the different materials used in the entire process (sandwich structure, intermediate fixing material and mechanical part), and they can be damaged even by mild touching [2, 16].

The present strategy, allows that the solvent droplets that reach the surface of the TPU finger are capable to dissolve a thin layer of TPU polymer, that will blend with the sprayed SBS elastomer, creating a covalent bond between the two materials, eliminating the need of an intermediate layer, and consequently the mechanical compliance between the two materials is

high. Furthermore, no peel off, separation or crack or any other mechanical failure problems was observed, even after mild scratching (see supplementary video 1). Moreover, the liquid inks are easily deposited onto the surface of complex shapes like clothes or prosthetic devices to form or deposit the strain sensors with no damage to the nanocomposite MWCNT-SBS thin film, or to the applied surface.

An infrared spectrum represents a fingerprint of the material with absorption bands which corresponds to the frequencies of the vibration modes between the bonds of the atoms present in the material. Attenuated total reflectance – Fourier transformed infrared spectroscopy (FTIR) was performed on the surface of the soft TPU finger and on the sprayed MWCNT-SBS sensor (Figure 4). The main vibration mode characteristics of the butadiene segments within the SBS are at 1449 cm^{-1} (CH_2 scissoring) and 964 cm^{-1} , attributed to the out of the phase deformation trans-1,4 $\text{C}=\text{C}$, $=\text{CH}$ stretching at 3005 cm^{-1} , and at 2844 and 2916 cm^{-1} attributed to the CH stretching. Styrene segments of the SBS elastomer presented absorption bands at 3060 cm^{-1} ($\text{C}-\text{H}$ out of plane deformation) and at 1601 cm^{-1} ($\text{C}-\text{C}$ stretching) [17, 18]. The characteristic absorption band of the TPU matrix can be observed at 3328 cm^{-1} for $\text{N}-\text{H}$, and at 1701 cm^{-1} for the $\text{C}=\text{O}$ stretching. The symmetric and asymmetric of the CH_2 can be observed between $2800 - 3000\text{ cm}^{-1}$. The band that appears at 1595 cm^{-1} , is related to the phenyl ring characteristics of TPU elastomers. The band at 1540 cm^{-1} is due to the $\text{C}-\text{N}$ stretch and $\text{N}-\text{H}$ deformation. The bands that appear at 1162 and 1076 cm^{-1} are related to the $\text{C}-\text{O}-\text{C}$ stretching [19] of the Filaflex polymer.

Figure 4. FTIR spectra obtained for the soft finger substrate and for the MWCNT/SBS printed sensors.

3.3. Electrical and Electromechanical Performance

The temperature-dependent DC electrical resistance was recorded between $20 - 90^\circ\text{C}$ (Figure 5) between two parallel contacts placed on the finger tip. It was observed that the sheet resistance obtained through the I-V data (Figure 5a) decreases with increasing temperature (Figure 5b). Using the data in Figure 5b, the temperature coefficient of resistivity (TCR) was determined to be between $20 - 50^\circ\text{C}$ and $60 - 90^\circ\text{C}$, based on equation 7:

$$TCR = \left(\frac{1}{\rho(T_1)} \right) \left(\frac{\Delta\rho}{\Delta T} \right) \quad (7)$$

where, $\Delta\rho = \rho(T_1) - \rho(T_2)$ and $\Delta T = T_2 - T_1$. The TCR values obtained for the MCNT - SBS nanocomposite printed sensor in the temperature range of 20 - 50 °C and 60 – 90 °C are 0.0097 and 0.021 °C⁻¹, respectively. The values obtained are in the range of the ones reported for other polymer systems like polypyrrole/polyvinylidene fluoride [20] and for polypyrrole layer synthesized by wet chemistry, on the surface of bacterial cellulose fibres [21]. The change in resistance as the temperature increases can be used to track the temperature of objects, and distinguish the cold from warm surfaces and activate an electrical resistance change that can be tracked and measured by a remote system (see supplementary information), thus avoiding the destruction of the prosthetic device and creating a sense of feeling that mimicks the human hand and thermal skin sensations [6, 10], and ultimately improve the adaptability of the device.

Figure 5. Electrical behavior of the MWCNT-SBS sensor printed on the surface of a TPU soft finger: a) Current-Voltage data recorded at different temperatures and b) evolution of the sheet resistivity with temperature.

The printed soft robotic finger with dried and compliant MWCNT-SBS sensors was submitted to a dynamic mechanical loading that mimics the open and closed positions of the human index finger, and the change in the resistance of the three sensors were recorded simultaneously during the bending process (Figure 6). The measurement was performed by bending the finger 15 times in each cycle, followed by a 30 s pause with the finger in the bent position, and another 15 mechanical cycles were applied, as can be seen in Figure 6a. During the finger bending, the change in resistance linearly follows the mechanical pattern applied in all the three sensors (Figure 6a and b). Even after stopping the bending process, and returning later to the mechanical load, the sensor follows the same pattern as observed in the first cycle of experiments, with negligible differences in the recorded change in resistance, between the different measurements, which demonstrates the outstanding performance of the sensors to follow human finger movements, not only the position of the finger but also the individual interphalangeal joints. The electrical hysteresis curve for the different strain sensors is presented in Figure 6c. For the deformations that are characteristics of the bending of the human finger (Figure 2), the strain sensors show almost perfect elastomer behaviour, with a small electromechanical hysteresis loss, and this behaviour follows a similar pattern for the

different sensors, independent of the finger interphalangeal region (Figure 6c). Our results have shown an outstanding performance of the MWCNT-SBS sensors when compared to the ones reported for silver nanowires [4] and carbon nanotubes [2] sandwiched between an elastomeric matrix. The excellent change in resistance and recovery, linearity and negligible hysteresis obtained for the sprayed sensors is probably due to the strong adhesion of the elastomeric SBS to the TPU soft finger matrix (Figure 3 and table 1), reducing buckling and fracture, and promoting a suitable formation of the conductive MWCNT network.

The applied mechanical strain caused an electrical resistance change ($\Delta R/R_0$) that is related to the mechanical strain (ϵ) by the Gauge Factor (GF) (equation 3). The GF was calculated for each interphalangeal joint, and they are 0.93, 0.76, and 0.76 for the metacarpophalangeal joint (MCP), the proximal (PIP) joint, and distal (DIP) interphalangeal joints, respectively, for a finger bending rate of 2.14 mm.min^{-1} . The variation of the gauge factor under different bending rates is presented in Figure 6d. In general, the GF remains similar for the different conditions tested, and the sensors placed in the different interphalangeal joints follow the same pattern. Despite this, the strain sensor placed in the MCP joint presented higher GF values when compared to the other two strain sensors (DIP and PIP). This difference is probably related to the thickness difference that the MWCNT-SBS printed sensors may have. During the spray deposition of the conductive polymeric solution, it is difficult to control with delicate accuracy the amount of material deposited, slight differences in air pressure or deposition rate could be responsible for the decrease of the surface resistivity and consequently contribute for the observed increase of the GF values placed on the MCP joint (Figure 6d).

The GF found in this work is similar to the ones reported by Yamada et al. [2] for a carbon nanotube sandwiched strain sensor stretched linearly under a tensile load, and lower than the metallic ones, that have a GF of approximately 2.0, but they are limited to 5% strain [3].

It was reported that the MWCNT-SBS elastomeric nanocomposites with 4 wt% conductive filler had a GF of approximately 2 when submitted to a four point bending deformation [22]. In the present work, the change in resistivity was recorded following the bending movement of the soft TPU finger, rather than stretching it uniaxially or four-point bending the strain sensor, mimicking the real movement of the human fingers. This more realistic approach could explain the low GF values found for the spray printed sensors. Negative GF values (-4.9) were found by Lu et al. [8] for the carbon black doped poly(dimethylsiloxane) (PDMS) and this phenomenon was attributed to the Poisson's effect, where lateral contraction occurs

in conjunction with longitudinal elongation. MWCNT-SBS sprayed sensors considered in this study have less than 500 nm thickness, which is small when compared to the length and width of the printed conductive path. Furthermore, according to equation 4, the Poisson's effect could lead to a decrease of the overall GF value, as previously observed for graphene oxide [8] and MWCNT [23] strain sensors.

Figure 6. Simultaneous measurement of the change in resistance in the different phalanges, when the soft robotic finger was flexed: a) a series of 15 bending cycles with 30 s interval between each cycle, b) electrical behavior of the sensor placed in the proximal phalange, and the corresponding mechanical actuation, c) Strain sensors hysteresis obtained at a bending rate of 8.6 mm.s^{-1} , and d) Evolution of the GF for the three strain sensors recorded at different bending rates of the soft finger matrix.

Long term operation of the strain sensors was investigated by subjecting the strain sensors to more than 10,000 bending cycles. Even after more than 10,000 bending cycles, the strain sensors placed on the different interphalangeal joints showed a negligible change in their electrical response, as shown in Figure 7, showing negligibly small fatigue behaviour (Figure 7b and c).

In the present work, the soft sensors are directly placed on the interphalangeal joints and due to the chemical affinity between the polymers and the solvent, the deformation generated by the finger during the bending process is directly applied to the sensor without constraints. This approach showed that the strain sensors can follow the human finger bending process from lower to higher bending speeds, and accurately monitor its motion. Previous studies and devices developed showed a linear response for uniaxial deformation, rather than bending, and they were unable to linearly follow the bending process of fingers or other human parts such the knees or elbows [1, 2, 5].

The deposition of a MWCNT-SBS thin film onto the surface of the soft finger provides a minimum interfacial area between the strain sensor and the supporting matrix, thereby minimizing fatigue, friction and damping issues between the substrate and the strain sensor, and ultimately takes full advantage of the electrical properties of the conductive filler. **Figure 7.** Resistance change during the bending of the soft TPU finger during more than 10,000 cycles, recorded simultaneously for the three phalanges at a bending rate of 8.6 mm.s^{-1} .

4. Conclusions

The proposed method to fabricate stretchable strain sensors takes the chemical properties and affinity of the raw materials into consideration. The materials used in the development of the sensor are chemically compatible with each other and this maximizes the mechanical compliance without compromising the mechanical and electromechanical properties of the sensing elements. Spray coating of MWCNT-SBS elastomeric polymer can easily be scalable, and the sensors can be fabricated in complex shapes directly on their desired place, without the need of extra intermediate or protective layers. The proposed fabrication route can create human-friendly devices with realistic functions and abilities such as the detection and monitoring of the complex bending behaviour of a soft robotic prosthetic finger. The strain sensors demonstrated an outstanding electromechanical performance, with negligible hysteresis, excellent repeatability and stability. This research has proposed a method to process innovative stretchable devices, with complex shapes that can act as part of the human body and can find a wide range of applications in recreation, soft robotics, prosthetic devices or electronic skin.

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Figure 1: a) Geometric parameters of a non-symmetric elliptical flexure hinge and a 3D printed monolithic soft robotic finger (3D Printing Systems, UP Plus 2) using Filaflex filament, b) Spray-painted schematic process of the soft finger, and c) soft robotic prosthetic finger with joints (flexure hinges) labelled. Finger and interphalangeal sensors correspond to the final prosthetic device.

Figure 2. Soft robotic finger strain measurement: a) Finger interphalangeal region with the strain distribution during the bending process, obtained by FE analysis with a maximum strain of 10 %, and b) experimental method used to record the strain of the interphalangeal regions.

Figure 3. Spray coated MWCNT-SBS sensors: a) surface and b) cross-section of the sensor. Inset shows a detail of the filler among the polymer matrix.

Figure 4. FTIR spectra obtained for the soft finger substrate and for the MWCNT/SBS printed sensors.

Figure 5. Electrical behavior of the MWCNT-SBS sensor printed on the surface of a TPU soft finger: a) Current-Voltage data recorded at different temperatures and b) evolution of the sheet resistivity with temperature.

Figure 6. Simultaneous measurement of the change in resistance in the different phalanges, when the soft robotic finger was flexed: a) a series of 15 bending cycles with 30 s interval between each cycle, b) electrical behavior of the sensor placed in the proximal phalange, and

the corresponding mechanical actuation, c) Strain sensors hysteresis obtained at a bending rate of 8.6 mm.s^{-1} , and d) Evolution of the GF for the three strain sensors recorded at different bending rates of the soft finger matrix.

Figure 7. Resistance change during the bending of the soft TPU finger during more than 10,000 cycles, recorded simultaneously for the three phalanges at a bending rate of 8.6 mm.s^{-1} .