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Multi-material Additive Fabrication of a Carbon Nanotube-Based Flexible Tactile Sensor

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Abstract

Additive fabrication use for the development of sensing devices with complex geometries is increasing constantly. Novel polymer-based materials with conductive properties are among the most investigated materials for soft electronics. In this study, we developed a fully additive-fabricated flexible contact pressure sensor using multi-material printing. The sensing part is a three-dimensional disc printed by extruding a multi-walled carbon nanotube-polymer composite using a laboratory-scale direct ink writing (DIW) process. The DIW process repeatability was studied, and it shows highly repeatable printing characteristics of the sensors. The pressure sensor response under compressive load was also investigated, showing that the sensor exhibited high sensitivity over a wide range of pressure and sensitivity to small increments of compressive force. The dynamic response of the sensor was also studied for different loading conditions where it showed good dynamic characteristics, and the sensor response-recovery time was 0.02 s.

Keywords Printed electronics · Conductive polymers · Direct printing · Piezoresistive sensors

1 Introduction

Microelectromechanical systems (MEMS) based technology has been the main fabrication method for sensors over decades. Most conventional MEMS-based sensors include silicon substrates, and they usually involve complex fabrication steps in relatively expensive environments [1]. On the other hand, printed electronics gained notice in recent years both in the academic community and the industry due to the advantages that they offer, such as the production of electronics and sensors on mechanically flexible substrates and fewer fabrication steps which makes it a simpler and

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more cost-effective fabrication process [2]. The most used printing techniques can be classified into two categories of contact and non-contact printing, which depend on whether the printing heads are in contact with the substrate or not [3]. There is an emerging technique called direct ink writing, a non-contact printing method, which is an additive fabrication method that uses inks with a wide range of viscosities, inorganic, organic materials, and biomaterials with no thermal treatment required for the extrusion. The extrusion force can be pneumatic or via mechanical pistons or Archimedes screws [4]. As printing techniques are in continuous development, materials with promising properties for electronic applications have also to follow the trend of the printing technologies. Piezoresistive composites are receiving a lot of interest in electronics, in particular for tactile sensing applications [5–7], and carbon-nanotubes are considered to be good candidates for developing flexible electronics [6, 8]. Works by Zhang et al. [9] demonstrated the fabrication of a flexible resistive humidity sensor by printing carbon nanotubes using screen and gravure printing processes. Michelis et al. [10] developed CNT-based resistive strain sensors using inkjet printing on a flexible substrate. In literature, three-dimensional fabrication techniques for flexible electronics are still not fully developed. Most applied techniques are two-dimensional printing methods such as screen printing, pattern transferring, and spray coating [11–14]. Inkjet printing which is considered a three-dimensional fabrication method, is often used to fabricate two-dimensional structures [10, 15–17]. Another concern is that it is most common to use one type of material for the printing process. However, multi-material additive fabrication can offer design freedom and opportunities to integrate complementary and functional materials with compositional and geometric complexity. For example, electrically conductive structures can be obtained by combining conductive materials with built materials to create conductivity in specific areas without additional joining process or assembly [18].

In this regard, we demonstrate the design and fabrication of a tactile sensor that is fabricated using a threedimensional printing process under ambient conditions via a combination of nanocomposite and silver ink extrusion on a flexible substrate. We demonstrate the printing process capability to obtain highly repeatable structures with minimal geometrical errors. The developed three-dimensional sensor performance was tested under static and dynamic pressure loading with different frequencies, and the sensor showed good performance over a large measurement span.

2 MWCNT-PDMS Material

2.1 MWCNT Ink Formulation

MWCNTs (Industrial grade, NanoLab, Waltham, MA, USA), particles length of 5-20 µm, diameter 10-30 nm, and purity > 85%) were used for the preparation of the nanocomposite. The multi-walled carbon nanotube (MWCNT) particles were dispersed and sonicated in a sufficient amount of Isopropyl Alcohol IPA (>99%, Daejung Co., Ltd., Siheung, Korea) for 30 min intervals to avoid damaging the CNT particles. Then 20 wt% of methyl group-terminated PDMS (MEP) (Sigma-Aldrich, St. Louis, MO, USA) was added and further sonicated. Finally, 80 wt% of PDMS prepolymer (Sylgard 184 Silicone Elastomer kit, Dow Corning, MI, USA) was mixed and sonicated with the solution before adding the PDMS cross-linker with a mixing ratio of 10:1 after complete evaporation of the solvent. To observe the quality of dispersion of the CNT particles in the PDMS matrix, scanning electron microscopy (SEM) was performed. Figure 1 is a high magnification SEM image of a cross-section of the MWCNT-PDMS material. As see, although there are still a small amount of CNT agglomerations, the distribution is overall uniform. The bright filament-like substances in the SEM image indicate connected CNT.



Fig. 1 Representative cross-sectional SEM image of MWCNT-PDMS composite

2.2 Sensor Design and Fabrication

The flexible piezoresistive sensor was fabricated using a multi-material additive fabrication approach where two additive manufacturing processes were used: Fused filament fabrication (FFF) and direct ink writing (DIW). The sensor consisted of three layers: a flexible substrate, a printed silver ink electrode, and a 3-dimensional MWCNT sensing layer.

The laboratory-scale direct ink writing process was used to extrude the MWCNT ink to print the sensing part of the pressure sensor. The same process was also used to extrude the silver ink to print the electrode layer. The FFF process, on the other hand, was used to fabricate the mold for the flexible Polydimethylsiloxane (PDMS) substrate.

The mold for the PDMS substrate was printed on a desktop 3D printer Cubicon Single (3DP-110F) (Cubicon Co., Ltd, Gyeonggi-do, South Korea). The extrusion parameters were as follows: the nozzle diameter was 0.4 mm, the extrusion temperature was 240 °C, and the bed temperature was 115 °C. The layer height was 0.2 mm, and the wall thickness was 1.2 mm with 50% infill density and 100% flow rate using acrylonitrile butadiene styrene (ABS) material.

To fabricate the substrate layer, PDMS was mixed with the cross-linker at a 10:1 ratio, and the mixture was then degassed in a vacuum chamber to eliminate air bubbles and was cured at 80 °C until fully hardening, after which it was peeled off the mold.

The silver ink electrode layer was then printed on top of the PDMS substrate. The electrode consisted of a single layer extruded using the direct ink writing process, with a feed rate of 900 mm/min and with a low dispensing pneumatic pressure (25 kPa) as the silver ink viscosity was very low. For the DIW extrusion, the nozzle diameter was 0.26 mm, and the stand-off distance (SOD) was 0.2 mm.

The MWCNT sensing layer was printed on top of the electrode layer. It consisted of a 3-dimensional disc with



Fig. 3 3D printed contact pressure sensor. A Shows the design of the sensor. B Photograph of the printed sensor

1 mm height and 10 mm diameter and had five layers. DIW was also used to fabricate the sensing part. The MWCNT-PDMS nanocomposite was extruded from a Polypropylene barrel through a 0.45 mm precision nozzle. The barrel was mounted on a 3-axis stage and connected to the pneumatic dispenser via a Teflon tube (10 mm inner diameter and 1 m length). The compressed air was delivered through the tube from an air compressor which was controlled by a pneumatic dispenser. The dispensing pressure was 300 kPa, the feed rate was 200 mm/min, and the SOD was 0.2 mm. Figure 2 shows the laboratory-scale DIW system used to extrude the sensing layer and the electrode layer. After the extrusion, the MWCNT-PDMS disc was put in a dry oven for thermal treatment to evaporate the residue

	Line height	Line width	Line length
Nominal	0.50 mm	2.00 mm	40.00 mm
Mean $(\overline{\overline{X}})$	0.51 mm	2.00 mm	39.85 mm
$SD(\overline{S})$	0.02	0.03	0.05



Fig. 4 Microscope photographs of printed MWCNT- PDMS lines

solvent and harden the structure. Figure 3 shows the design of the sensor (A) and the obtained result (B).

3 Results and Discussion

3.1 Direct Writing Process Control

The printing quality for inks is mainly affected by the nozzle diameter, feed rate, dispensing pressure, and SOD. The values of these parameters are determined according to the ink viscosity, which can vary from batch to batch. Therefore, it is crucial to control the printing process to obtain reproducible and repeatable structures. In addition, for accurate characterization of sensors performance, all the printings have to be repeatable under the same conditions.

Table 1 summarizes the measurements of five samples of linear sensors that were tested in terms of geometrical features (height, length, and width). The printed lines had 0.5 mm height, 2.0 mm width, and 40.0 mm length. Each characteristic was measured three times using a digital microscope (OSM-U, Dongwon Systems, Seoul, South Korea) and calibration software (ITPlus4.0) as shown in the images in Fig. 4, and the mean and the standard deviation of the measurements were calculated. The obtained results show that the DIW printing process is highly repeatable for samples fabricated and measured under the same conditions.

3.2 Contact Pressure Sensor Performance

To test the printed MWCNT-PDMS composite as a contact pressure sensor, wires were connected to both sides of the silver electrode and attached with copper tape and Kapton tape to minimize contact errors. The sensor was then placed under a push-pull force gauge (SH-200, SUN DOO Instruments, Wenzhou, China) where a compressive load was applied (up to 60 kPa), and the change in the electrical resistance of the sensor was recorded with a digital multimeter (DMM6500, Keithley Instruments, Solon, OH, USA). Figure 5 is a schematic representation of the test and measurements setup. When the CNT nanocomposite is under compressive stress, the interconnection and the spacing between the CNT particles change, which leads to a change in the material electrical resistance [19]. The increase or decrease of the value of the resistance is related to the aspect ratio of the fillers. Figure 6 shows the sensor response to the exponential increase in the compressive load. The senor was sensitive to small increments (of the magnitude of 0.6 kPa), and a more significant decrease of the electrical resistance was observed with the drastic increase of the applied force. Moreover, the sensitivity was calculated as the change in the resistance relative to the base resistance divided by the applied load, according to Eq. (1), and is 0.14 kPa^{-1} .

The sensor response time and recovery time were also determined according to the recorded response of the sensor to the static application of the pressure presented in Fig. 7. The sensor showed a response time of 8 ms and a recovery time of 20 ms. The graph also shows that the sensor has a good performance even at very high pressure (300 kPa).



Fig. 5 Test and measurements setup





Fig. 6 Sensor response to force loading

$$S = \left| \frac{\Delta R_{R_0}}{p} \right| \tag{1}$$

3.3 Dynamic Performance

Dynamic performance of a sensor refers to the response time after the sensor input changes.

To test the dynamic performance of the tactile sensor, cycles of loading and unloading of the force were applied with a frequency of 0.5 Hz (Fig. 8) and 0.25 Hz (Fig. 9) at a speed of 30 mm/min for each of the loading and the unloading phases. For a loading frequency of 0.5 Hz, the applied force oscillated between 0 kPa (unloading phase) and 350 kPa (loading phase) and the values of $\Delta R/R_0$ oscillated between 0.8 and 1.5.

For a loading frequency of 0.25 Hz, the applied pressure was between 0 and 200 kPa; the sensor response, therefore,



Fig. 7 Step response of the sensor to the applied pressure



Fig. 8 Dynamic response of the sensor for cyclic loading at 0.5 Hz



Fig. 9 Dynamic response of the sensor for cyclic loading at 0.25 Hz

was between 0.7 and 1.4. These results indicate that the sensor shows good performance when varying the pressure loading conditions, and the frequency of the response signal of the sensor was 0.5 Hz and 0.25 Hz, respectively, for 0.5 Hz and 0.25 Hz pressure loading frequencies. After processing the sensor's output signal using Matlab R2021a, we obtained that the delay between the loading–unloading input signal and the sensor response signal for both loading frequencies, over a sampling duration of 80 s, was 0.02 s. The sensor exhibited fast response and recovery, and the baseline resistance was quickly recovered once the push–pull gauge tip was removed from the sensor.

4 Conclusion

In this paper, the static and dynamic response of a multimaterial 3D printed carbon nanotube-based sensor was studied. The sensor is composed of three layers which were obtained using a combination of additive fabrication processes. The sensing layer is a 3-dimensional MWCNT-PDMS disc printed on top of a silver ink electrode. The sensor showed a high sensitivity for a wide range of applied pressure and a short response/recovery time for the dynamic loading of the pressure. Compared to the state of the art of carbon nanotube-based sensors, the sensing part is a 3-dimensional structure fabricated with five layers, and the structure was able to withstand the cyclic compressive stress without showing any significant degradation in the performance or deterioration of its shape.

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