Microporous Induced Fully Printed Pressure Sensor for Wearable Soft Robotics Machine Interfaces

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Soft robotics machine interfaces are desirable for facilitating handling of objects in soft robotics applications. High-speed responses of the interfaces are crucial for achieving statement conversions in novel robotics systems. Herein, a novel scheme for synthesizing a functional ink for producing fully printed soft pressure sensors that are highly responsive for detection of an applied vertical force is presented. The sensor consists of carbon nanotubes and polymeric soft materials, and achieves good response characteristics because of the microporous sensing layer. The fabricated sensor shows high performance for detecting forces with a high-speed response. Novel wearable robotics machine interfaces for the printed soft sensor and a soft robotic hand are fabricated to facilitate object manipulation. The artificial sensor for a switching system demonstrates successful gripping and release of an object when controlled by a switch. Further, these findings show that the switching performance of the sensor is suitable for the machine interface for switching applications. This implies that the fabrication of a sensing system for remotely controlled soft robots is possible. Thus, high-sensitivity printed devices for tactile machine interfaces in the form of a wearable e-skin are experimentally demonstrated.

1. Introduction

Soft polymer-based resistive-type mechanical sensors responsive to external mechanical stimulation have attracted much attention for applications in replicating human skin and soft robotics interfaces. For enhancing the human-like cutaneous sensation in robotics systems, highly sensitive mechanical soft sensors are the most important factor. Printing methods for fabricating the sensors are attractive due to the low device cost and several functionalities can be provided by using solvent-based systems. Resistive-type sensors rely on a simple operation principle, where electrons flow through conductive materials, such as nanocarbons and metal fillers. In particular, pressure-sensitive devices typically contain elastomers and conductive materials, and are widely used to fabricate flexible resistive-type mechanical sensors. One of the approaches for increasing the pressure sensitivity is to introduce a porous structure into the sensing layer. In recent years, sponge-like structures have been produced, including electrically conductive sponges, polyurethane-based sponges, and carbon–Ag hybrid sponges which have suitable properties for use in robotics interface applications. Moreover, previous studies have realized reverse-micelle-induced porous pressure sensors by printing aqueous carbon ink, which were intended for human–machine interfaces. Their findings showed that the size of the micropores in the sensing layer affects the sensitivity. Therefore, porous structures could be candidates for the development of highly sensitive soft sensors.

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by applying it to a robotics machine interface as a wearable switch for a soft robot gripper. This study offers a general platform for the development of next-generation robotic interfaces and e-skins for soft robotics sensing and actuation applications.

2. Results and Discussion

2.1. Sensor Fabrication

2.1.1. Ink Formulation

Figure 1 shows schematic illustrations of the fabrication procedure of a functional ink and the printing processes. Figure 1a shows the ink-synthesis schemes used in this study. The ink for the sensor was prepared by ultrasonication of 1.43 g hexane as a main solvent (Tokyo Chemical Industry) and 20.0 mg of 4-dodecylbenzenesulfonic acid (DBS) surfactant (model number: 44198, Merck) for 5 min. Next, 1.30 g of polydimethylpolysiloxane (PDMS) as a silicon elastomer (Sylgard 184, Dow Chemical Co.) and 71.5 mg multiwalled carbon nanotubes (MWCNTs) (aspect ratio: 100, model number: 659258, Merck) were mixed. After ultrasonication for 5 min, 350 mg of pure water was added and the mixture was stirred for 30 min. Then, 0.130 g of a stiffener for the PDMS was added to the ink and stirred for 5 min. Finally, vacuum evacuation was performed for 10 min. The viscosity of the ink was \( \approx 800 \text{ mPa} \), which was measured by a viscometer (Type-B, EKO Instruments Co.).

2.1.2. Device Fabrication

The printed pressure sensor consisted of electrodes and a sensing layer. Figure 1b shows the fabrication processes for the printed electrode using the dispensing system. Figure 1c shows the sensing layer being produced using the blade-coating system. The electrode was printed using the PEDOT:PSS [poly(3,4-ethylenedioxythiophene)-poly(4-styrenesulfonate)] (Clevios SV4 STAB, Heraeus) conductive polymer. The sensing layer was fabricated using a glass or 50 \( \mu \text{m} \) thick poly(ethylene naphthalate) (PEN; Q65HA, DuPont) substrate. A cross-linkable poly(4-vinyl-phenol) (PVP; 436224, Sigma-Aldrich) solution consisting of a mixture of PVP and melamine resin (418560, Sigma-Aldrich) with 1-methoxy-2-propyl acetate (Kanto Chemicals 01948-00) as the solvent was spin coated onto the substrate as the planarization layer. Next, PEDOT:PSS was printed on the planarization layer using the dispenser system (350PC, Musashi Engineering) to form the electrodes. The interelectrode distance was 1 mm. The electrode was annealed at 150°C for 30 min and had a thickness of \( \approx 500 \text{ nm} \). PEDOT:PSS can realize a lower resistive electrode by printing process. Moreover, we used this material for the printed electrodes because of good wettability and high conductivity. The detailed characteristics of the electrode are shown in the Supporting Information: Figure S2 (conductivity changes) and S3 (electrode distance), Supporting Information. Figure 1d shows the formed printed electrodes and sensing layer using several printing processes. Further, a 300 \( \mu \text{m} \) thick sensing layer was formed using blade coating by applying the ink through a printing mask, followed by annealing at 120°C for 2 h.

2.2. Wearable Robotics Machine Interfaces

The system was constructed using the printed sensor, a reference resistance, control computer, and the soft robot hand. The output voltage was used as a voltage divider circuit. The reference

![Figure 1](https://www.advancedsciencenews.com)
resistance and initial resistance of the sensor were 1.0 kΩ. A voltage of 1.0 V was applied using a multifunctional output DC power supply (GPE-4323, Texio Technologies), where the generated signal was transferred to the control computer. After signal processing using Microsoft Visual Studio software, the on/off commands were transmitted to a solenoid valve for controlling the gripper, where the off/on states of the gripper corresponded to air pressures of 0 and 30 kPa, respectively.

2.3. Sensor Performance

Figure 2 shows the surface morphologies and electric properties of the sensing layer made from our novel functional ink as a function of the added water content (0–1400 mg). The layers were annealed at 120 °C for 2 h after printing. The top row of Figure 2a shows surface photographs of the layer. There were no significant changes with different water contents. The middle row shows cross-sectional laser microscopy images of the layers to evaluate their thickness. The uniform films should provide mechanical stability under applied pressure. The measured thicknesses were all around 500 μm and did not depend significantly on the weight of added water. In the bottom row, surface scanning electron microscopy (SEM) images of the layers are shown. The addition of water to the ink clearly produced a microporous structure in the sensing layer. Moreover, the diameter of the pores depended on the added water content. The measured diameters were <0, 1, and 3 μm and 5 mm, for the addition of 0, 350, 700, and 1400 mg of water, respectively. The measured diameter 0 means no pores. There was correlation between the size of a pore and the layer flatness. These results show that the added weight of water distinctly affected the formation of micropores. Figure 2b shows the resistances of the sensing layer as a function of the added water weight. The average resistance values for three samples are plotted. Lower water content in the ink resulted in more highly conducting films. Higher water content resulted in larger pores that increased film resistance.

Figure 2. Evaluation of the surface morphologies and electric properties of the sensing layer made from the MWCNT-based functional ink. All measurements were made after a 120 °C annealing process. a) (Top row) Photographs of the surfaces of the layers (defined by the red dashed lines) prepared with various weights of added water (0–1400 mg). (Middle row) Cross-sectional images of the layer obtained using a laser microscope. (Bottom row) Surface SEM images of the layers. The yellow arrows indicate nanopores, which varied in size depending on the amount of added water. b) Electrical resistance of the films as a function of the added water weight, measured over a distance of 2.0 mm. The average resistance values for three samples are plotted. c) Schematic illustration of the formation of pores during annealing.
In this work, the percolation resistance was $\approx 1 \, \text{k}\Omega$ (Figure S1, Supporting Information). The addition of 350 mg of water was considered optimal for achieving both high sensitivity and flatness. Figure 2c shows a schematic diagram showing the formation of pores during annealing of the layer. The fabrication method evenly distributes MWCNTs in the viscous media and forms micelles, which are surrounded by water droplets. Before annealing, the surfactants exist in the matrix and solvents in the statement of water micelle coating. After annealing, the water micelles evaporate, and then the micropores generate in the sensing layer.\textsuperscript{[15,27]}

Figure 3 shows the morphology of the sensing layers prepared using the functional ink. Here, we evaluated the influence of annealing temperature on the sensing layers. The samples were annealed at either 60, 90, or 120 °C for 2 h in ambient air. All the measurements were made using the ink with 350 mg of added water. Figure 3a shows surface photographs of the layers prepared using different annealing temperatures. At all temperatures, flat layers were produced. However, at 60 °C, as isolation point (the layer that does not include MWCNTs) due to the PDMS was generated, which was not observed for the samples annealed at 90 and 120 °C. Figure 3b shows magnified photographs of the area indicated by the dashed line in Figure 3a. Again, an obvious isolation point was observed for the sample annealed at 60 °C, which was not observed for those annealed at 90 and 120 °C. Because of low-temperature annealing, it seems that the isolation point was generated due to aggregation of MWCNTs in the layer. Figure 3c shows the measured resistance of the layers as a function of annealing temperature. The average resistance values for three samples are plotted. Annealing at 60 °C resulted in a film with a resistance of $\approx 500 \, \text{\Omega}$. In this case, due to aggregation of MWCNTs, the resistance was lower than the percolation value. In contrast, the samples annealed at 90 and 120 °C showed resistance values around 1 k\Omega due to the lack of the isolation spot. These findings show that the ink can produce high-performance sensing layers with a microporous structure, where the optimal processing conditions were the addition of 350 mg of water and annealing at 120 °C.

Figure 4 shows the sensing performance of the printed pressure sensors as a function of the applied pressure. Figure 4a shows a photograph of a fabricated sensing device with a PEDOT:PSS printed electrode. Figure 4b shows surface SEM images of the printed sensing layers with a random distribution of nanopores in the layer on the printed electrode. The right-hand-side image shows a magnified view of the area indicated by the dotted line in the left surface SEM image. There was no difference of the surface condition in the printed sensing layer between on the electrode and substrate. The conductive MWCNT component and polymeric soft matrix are both visible. Figure 4c shows photographs of the sensitivity measurement setup used to characterize the sensors with a mechanical compression indenter. The resistance changes of the sensing layer at different applied pressures are shown in Figure 4d. For evaluating the pressure sensitivity of the printed vital sensor, the sensitivity response was measured by applying 25–130 kPa pressure. The resistance values were measured using a digital multimeter. A linear resistance curve in response to the applied pressure was observed. Figure 4e shows the resistance changes as a function of applied pressure for sensors with different substrates (a glass and plastic substrate). Figure 4f shows a magnified plot of a part of Figure 4e around

![Figure 3](https://example.com/figure3.png)

**Figure 3.** Morphological evolution of the printed sensing layer with increasing annealing temperature (60–120 °C). a) Surface photographs of the layers annealed at 60, 90, and 120 °C. b) Magnified photograph of the surface profiles shown in (a) for measurement of the nanopore diameters. c) Resistance as a function of the annealing temperature. The average resistance values for three samples are plotted.
the 20 kPa pressure range. Our sensor with the PEN substrate could sense a lower pressure strain indicated of 5 kPa. The sensor on the PEN substrate showed a wider dynamic range than that on the glass substrate due to the smaller Young’s modulus of the PEN, which corresponds to high sensitivity at low pressure. These values are considered reasonable for a printed soft sensor. Figure 4g shows the mechanical reliability of the sensor against continuous application. The pressure applied by the indenter was 25, 50, and 130 kPa. The higher pressure showed the bigger resistance change. Moreover, our sensor showed stable electrical performance during this test. In particular, stable measurement of a low applied pressure (e.g., 25 kPa) is important for soft robotics interface applications. Therefore, our soft sensor is thought to be a good candidate for a wearable switch for soft robotics. Figure 4h shows the long-term stability examined by continuous loading and unloading at 50 kPa of applied pressure. No significant reduction in the output was observed after applying pressure cyclically for over 600 cycles, which confirms the long-term stability of the sensor.

2.4. Wearable Robotics Machine Interface

One of the promising applications of the printed soft sensor is a switching system for a soft robotic hand, as shown in Figure 5. The human–robot interface is one of the important technologies against realization of a remote-control technology. The gripper grasped an object with an inflation pressure of 30 kPa. The required inflation pressure depends on the handling object. In this case, we can grasp a fragile object such as a strawberry with 30 kPa inflation. Figure 5a shows a schematic of our system for a wearable robotics machine interface consisting of the printed sensor, a reference resistor, a control computer, and the soft robot hand. The output voltage of the sensing signal was used as a voltage divider in the circuit. The reference resistance and initial resistance of the sensor were 1.0 kΩ. The applied voltage was 1.0 V, and the sensing signal was transferred to the computer. After signal processing using a program written in the C language, the command was transmitted to a solenoid valve for on/off control. For the off state of the gripper, the air pressure was 0 kPa, while the on state applied 30 kPa.
attached to human skin on the index finger of a volunteer using a skin-compatible adhesive patch (Nexcare, 3M), as shown in Figure 5b. When the volunteer closed their finger, the switch turned on and moved the soft robot hand. Figure 5c shows a demonstration of an automatic gripping system with wearable switch actuation. After actuation of the wearable switch, the gripper closed when an air pressure of 30 kPa was applied (gripper closed). Figure 5d shows the measured response time from the signals obtained during the action shown in Figure 5b. The response time (off to on) was only 70 ms to move from the baseline to the stable voltage level. These findings show that the performance of our printed sensor is suitable for use in machine interfaces such as a switch. A soft robotics experiment using our remote-control system with the soft wearable switch is shown in Figure 5e. The corobot automatically moved upward and downward with respect to the object, while the grasping of the gripper is controlled using the switch. When the corobot moves toward the object, the gripper can grasp the object by turning the switch on. After grasping and moving upward, the switch is turned off, and the gripper releases the object.

3. Conclusion

We fabricated a fully printed soft sensor using functional soluble materials, such as MWCNTs, PDMS, and a surfactant, which could measure and detect low pressure with a fast response. The sensors showed high performance in terms of the resistance change range, mechanical stability, and sensing ability. We performed grasp control in real time with our novel switching system. The system can be used for various tasks where air pressure can be used to control devices, thus demonstrating its potential for various practical soft robotics applications. These outcomes show that the switch performance of our printed sensor is suitable for a switch in machine interfaces. This allows the fabrication of a sensing system with remote-control capability suitable for soft robotics applications. We experimentally demonstrated high-sensitivity printed devices for tactile machine interfaces as wearable e-skin.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.
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Conflict of Interest

The authors declare no conflict of interest.

Keywords

carbon nanotubes, microporous sensing layer, printed pressure sensors, soft robotics interfaces

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