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A Paper-Carbon Nanotube Based Wearable Pressure Sensor for Physiological Signal Acquisition and Soft Robotic Skin

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Abstract

Wearable and flexible pressure sensor is essential to the realization of personalized medicine through continuously monitoring an individual's state of health and also the development of highly intelligent robot. A flexible, wearable pressure sensor is fabricated with novel SWNT/tissue paper through a low-cost and scalable approach. The flexible, wearable sensor showed superior performance with concurrence of several merits, including high sensitivity for a broad pressure range and ultralow energy consumption level of ($\sim 10^{-6}$ W). Benefited from the excellent performance and ultra-conformal contact with uneven surface, vital human physiological signal (such as radial arterial pulse, muscle activity at various positions) can be monitored in real-time and in situ. In addition, the pressure sensors could also be integrated into robotic artificial skin that could sense the force/pressure and also the distribution of force/pressure on the artificial skin.

Keyword: Wearable; Pressure sensor; Physiological signal; Carbon nanotube; Robotic skin

Introduction

Wearable sensors are devices that can be worn or mounted on human skin to continuously and closely monitor an individual's activities, without interrupting or limiting the user's motions, and thus are of great importance for various potential applications in future, including electronic skins,¹⁻⁵ real-time physiological information acquisition,⁶⁻⁹ human-machine interfacing,¹⁰⁻¹³ and energy harvesting.¹⁴⁻¹⁷ For most of practical applications, pressure sensors with a high sensitivity in low-pressure regime such as subtle pressures induced by tiny activities such as gentle touch, heartbeat, and respiration, are highly desired. The real-time acquisition of human vital signs is essential to the realization of personalized medicine through continuously monitoring an individual's state of health. The soft electronic skin with tactile sensibility is also of great importance for the development of highly intelligent robots.

Recently, a wide range of nanomaterials, including nanowires,^{15, 18-22} carbon nanotubes (CNTs),^{11, 23-25} polymer or carbonized fibers,²⁶⁻²⁷ and graphene²⁸⁻³³ have been used to fabricate novel flexible pressure sensors. To date, majority of the reported works have realized the sensing by utilizing piezocapacitive,^{2, 17, 34-35} piezoelectric,^{13, 15, 36-38} triboelectric,^{12, 14} and piezoresistive^{11, 18, 20, 27, 30, 39-40} effects. Compared to other types of pressure sensors, flexible piezoresistive sensors, which consist of conformable substrates and compliant conductive materials, can detect applied pressure or mechanical force by giving a change in current or resistance, and thus attract a great deal of attention.

Generally, in order to obtain a high sensitivity and a low detection limit, microstructures flexible substrates are used for the fabrication of pressure sensors. For example, polydimethylsiloxane (PDMS) films with microstructures are widely used as the flexible substrates to integrate with active materials. It was reported that the pressure sensors with pyramidal microstructures on PDMS substrates presented sensitivity of about 30 times higher than that of devices fabricated with planar substrates.² However, fabrication of microstructures usually is rather complicated, costly, and time-consuming. In another aspect, the selection of active materials is also vital to achieve a high sensitivity and a low detection limit. Normally, low dimensional conductive materials such as CNTs, metal nanowires, some other conductive nanofibers and graphene could offer high sensitivity.^{18, 27, 40-41} However, most of metal nanowires are susceptible to oxidization in ambient environment, while noble metal nanowires are stable but costly. Some conductive nanofibers such as carbonized nanofibers are difficult to be prepared and handled during the device fabrication. On the contrary, CNTs have excellent chemical stability in ambient environment and could be produced in large scale and at low cost by well-established chemical vapor deposition process.⁴²⁻⁴⁵ Another stringent requirement on wearable sensors is the low power consumption, considering that fabrication of flexible power source with high power density is still a challenge.

In this paper, we proposed a novel strategy to fabricate a flexible and wearable pressure sensor by impregnating single wall carbon nanotube into tissue paper (SWNT/tissue paper) and sandwiching them between a bare PDMS sheet and a polyimide (PI) sheet patterned with interdigitated Au electrodes. Here, we used PI instead of PDMS as substrate to avoid the degradation of electrodes conductivity during the testing and handling, considering the excellent adhesion between electrodes and the PI substrate.⁴⁶ Both SWNT and tissue paper are abundant resources or easy to be obtained. The fabrication process is facile and almost no chemical involved. A typical sensitivity of 2.2 kPa^{-1} could be achieved in a wide range of 35-2500 Pa, and a sensitivity of 1.3 kPa^{-1} in the range of 2500-11700 Pa, which is comparable with the record of organic transistor pressure sensors reported recently.³⁴ Notably, the active elements, SWNT/tissue paper could be easily fabricated at low-cost and large-scale. In addition, our SWNT-based pressure sensors is able to detect gentle touch from human

body with high sensitivity, enabling them to sense some vital physiological signals of human body at a ultra-low power consumption (power consumption could be as low as $\sim \mu\text{W}$). The pressure sensors could also been integrated into soft skin that could position the site and even the magnitude of pressure applied onto a robot. We believe that the SWNT/tissue paper based pressure sensor, which is fabricated through a low cost and facile process combined advantages of high sensitivity, versatility and low power consumption, will have many potential applications in next generation artificial skins and wearable electronics.

Experimental Section

Sensor Fabrication The device fabrication process is schematically shown in Figure 1. The first step is the preparation of the sensing material. The water-based SWNT dispersion is purchased from Chengdu Organic Chemical and was diluted to a concentration of 0.25 mg/mL. Tissue paper was cut into 10 mm $\times$ 10 mm pieces and then immersed into the SWNT dispersion for 5 s and then annealed at 110 °C for 10min to fully evaporate the water. After one cycle of coating and drying, the resistance of 10mm $\times$ 10mm tissue paper could reach a value of $\sim 12.6 \text{ k}\Omega$. The PDMS layer was prepared by mixing base gel and the curing agent (Sylgard 184 Silicone Elastomer) at the weight ratio of 15:1. The mixture was mixed well and then degassed for half an hour before being poured onto glass slide. Then the mixture was cured at 80 °C for 2 h. After curing, the PDMS film was peeled off from glass slide and cut into pieces with proper size. The thickness of the PDMS sheet was controlled to be around 300 μm . The interdigitated electrodes were fabricated by depositing Ti/Au (10 nm/80 nm) onto a 25 μm thick polyimide (PI) sheet mounted on PDMS sheet by electron beam evaporation through a metal shadow mask. The size of the interdigitated electrode is 11 mm $\times$ 11 mm. The interspacing between the adjacent fingers is typically 0.5 mm, with the width of interdigitated electrodes of 0.2 mm. Then, wearable pressure sensors were assembled by stacking the active SWNT/tissue paper and a layer of PDMS in sequence onto electrodes patterned PI/PDMS substrate. Soft PDMS layer could well adhere to the bottom PI layer and seal the tissue paper/SWNT with good strength. Then two 0.1 mm thick Aluminum (Al) wires were bonded to the electrode pads with silver conductive epoxy for electrical connection. Finally, the whole device except the Al wires was encapsulated by PDMS.

Device Test The $I-t$ characteristics of the pressure sensors were collected by an electrochemical workstation (CHI 760D) at an operating voltage of 0.1 V. The shortest sampling period is 20 ms which is determined by the electrochemical workstation. To evaluate the response of sensor to applied force, a thin glass square plate with weight of 0.1 g and dimension of 11 mm $\times$ 11 mm (corresponding to a “base pressure” of 8.2 Pa with the area of 121 mm²) was first covered onto the PDMS layer to uniformly distribute the pressure to the whole device and also avoid the adhesion between the PDMS surface and the loading.²⁷ The pressure induced by the glass plate is defined as “base pressure” for the devices, which corresponds to an initial current (I_o) of 2.6 μA at a bias voltage of 0.1 V.

The response to applied pressure was performed by gently loading objects of 0.42 g, 0.78 g, 1.45 g, 2.30 g, 3.50 g, 7.6 g, 14.0 g, 26.7 g, 31 g, 41.4 g, 64.0 g, 105.4 g and 146 g onto the glass cover, the corresponding pressure are 35 Pa, 65 Pa, 120 Pa, 190 Pa, 290 Pa, 630 Pa, 1200 Pa, 2200 Pa, 2500 Pa, 3400 Pa, 5200 Pa, 8500 Pa and 11700 Pa respectively. The mechanical durability of the sensor was tested under around 2.0 kPa pressure applied by a shaker vibrator at a constant frequency of 5 Hz. The electrical signal was collected by a source measurement unit (SMU, Agilent B2902A) at sampling periods of both 1 ms and 10 ms.

For long term test, the current signal in pressure sensor was collected by the electrochemical workstation which is connected to computer. To demonstrate their capability of sensing human vital

physiological signals, pressure sensors were mounted to the artery vessel at the wrist, and on throat with a hansaplast plaster (Figure S1). The devices could easily recognize the artery pulse wave and also muscle activities on fingers and throat. The muscle activity at the wrist caused by finger movement was monitored with the hand open/close. The tongue movement could also be detected by monitoring the muscle motion at the throat. To demonstrate the soft robotic skin based on the pressure sensors, 16 pressure sensors were assembled into a 4×4 array. The pressure was applied by gently touching the pressure sensors with fingers. The current change in the sensing matrix was observed with a microcontroller (Arduino Mega 2560, 16 Analog input ports) connected to computer. The transient current was measured by the voltage drop across a series resistor.

Results and Discussions

The device fabrication process is as schematically illustrated in Figure 1. The top and bottom PDMS layers could not only seal the whole device but also provide mechanical support for subsequent handling and testing. After assembly, the PDMS packed device is quite mechanically robust. The most critical part in this pressure sensor is the active material which is made of SWNT and tissue paper composite. Due to the uniform dispersion of SWNT, the SWNT could impregnate the tissue paper and adhere firmly to the fibers in tissue paper after water evaporation, as shown in Figure 2a-b. Both optical microscopy and scanning electron microscopy were employed to examine the morphology of SWNT coated tissue paper.

Figure 1. Schematic illustration of the fabrication process of a pressure sensor. a) The SWNT were onto a tissue paper through a dip-coating process; b) SWNT/tissue paper was assembled onto Au electrodes on PI substrate, PDMS layers were used to seal the device and also provide mechanical support; c) A pressure sensor was mounted to a human wrist for heart pulse sensing.

As illustrated in Figure 2a, after evaporation of water, the color of tissue paper turned from white to light gray. Figure 2b shows a low magnification SEM image of tissue paper, only very thick fibers (~10 μm) could be observed, with increasing of magnification, smaller tissue fibers (~100 nm) could be observed, as shown in Figure 2c. After coating of SWNT, the SWNT adhere firmly to the tissue paper, which is confirmed by SEM image in Figure 2d. The interdigitated electrodes were deposited on PI/PDMS stack through a metal shadow mask, considering the good adhesive between Au and PI film. In addition, the large tensile strength of PI film could also protect the electrode and avoid the conductivity degradation caused by stretching of PDMS. Figure 2e-f show the optical images of the assembled device, the pressure presents excellent flexibility. The sheet resistance of paper sheets decreased to around 12.6 k Ω after decoration by SWNT, which is conductive enough to be active layer in our pressure sensor. The resistance of SWNT/tissue paper could be affected by the concentration of SWNT dispersion. The relationship between SWNT dispersion concentration and

sheet resistance was plotted in Figure S2. It is observed that the average sheet resistance decreases from 45 k Ω to 0.6 k Ω with the concentration of SWNT dispersion increasing from 0.15 mg/mL to 1.1 mg/mL. The sheet resistance should be carefully selected for an optimized device performance. On one hand, if the sheet resistance is too low (in case of 1.1 mg/mL SWNT dispersion), the energy consumption of the pressure sensor would increase, which causes thermal effect and degrades the performance for long term work; on the other hand, the SWNT/tissue paper prepared with low SWNT concentration (e.g. 0.15 mg/mL SWNT) will have large variation in resistance value, as shown in Figure S2, resulting in a low reproducibility of the pressure sensor. Thus, 0.25 mg/mL SWNT dispersion was chosen to prepare the SWNT/tissue paper with an average resistance of 12.6 k Ω .

Figure 2. a) Optical image of a bare tissue paper and a SWNT/tissue paper; b) SEM image of tissue paper at low magnification; c) SEM image of tissue paper at high magnification; d) SEM image of SWNT/tissue paper; e) Optical image of an assembled pressure sensor wired with aluminum wires; f) Optical image of a bent pressure sensor.

The pressure sensor was applied with different static loading to test its sensitivity to external pressure, as shown in Figure 3a. The current response of the device was defined by the ratio of change in current (ΔI) and current under base pressure (I_0). With increasing static pressure, the current response also increases. It is obvious that, 35 Pa pressure could already produce a response on this pressure sensor. Once the pressure increases to 65 Pa, the response is well-defined and repeatable, indicating that the pressure detection limit is in the range of 35-65 Pa. The sensitivity (S) of the pressure

sensors is defined as $S = \delta(\Delta I/I_0)/\delta P$. Similar to most of the reported pressure sensors,^{18, 27, 40, 47-48} the plot for sensitivity is composed of more than one regions. The current change ΔI almost linearly increases with pressure change (δP) in the range of 35-2500 Pa with the sensitivity S_1 of 2.2 kPa⁻¹, while with further increasing the pressure to the range of 2500-11700 Pa, the sensitivity S_2 decreases to 1.3 kPa⁻¹.

Figure 3. Sensitivity of the pressure sensor to pressure. a) Current response to increased pressures under static loading and unloading, the inset shows the current response at low pressure range of 35-65 Pa; b) The current response against the applied pressure; c) The durability test under a constant pressure (~2.0 kPa) applied by a shaker vibrator at a frequency of 5 Hz; d) An enlarged view of the part of the $I-t$ curve in c).

The mechanism could be explained as follows: soft tissue paper has porous and rough surfaces with hairy SWNT, the amount of SWNT bridging interdigitated electrode pair depended on the external pressure applied. Once exerting an external force, a small compressive deformation of tissue paper enabled more SWNT to contact with the interdigitated electrodes, leading to more conductive pathways. This produced an increase in current when a fixed voltage of 0.1 V was applied. Upon the removal of external pressure, both PDMS and tissue paper recovered to their original shapes, reducing the number of SWNT bridging the electrode pairs, causing a decrease in current. With increasing pressure, more compressive deformation would be induced, leading to more SWNT bridging the finger electrodes and more conductive pathways. The high sensitivity of the pressure sensor could be understood considering that a smaller number of SWNT bridging interdigitated electrodes corresponds to a higher relative increase of SWNT bridging interdigitated electrodes while a certain pressure is applied. Meanwhile, the improved contact between individual SWNT should also contribute to the sensor performance.

The durability of the pressure sensor was then tested on a shaker vibrator under a pressure of about 2.0 kPa at frequency of 5 Hz, as displayed in Figure 3c, the pressure sensor could easily detect the

contact between the sensor and the shaker at a frequency of 5 Hz. Figure 3d shows an enlarged view of a part of the $I-t$ curve in Figure 3c. The current response time depends on both pressure loading/unloading frequency (Figure S3) and also sampling periods of the equipment. At pressure loading frequency of 5 Hz, the response time could reach 40 ms, as shown in Figure S3. The well-defined signal of current response indicates the high response speed of the pressure sensor to applied pressure. Another merit of this pressure sensor is the ultralow power consumption, the pressure sensor could operate well at an operation voltage of 0.1 V and the current is at the order of 10^{-5} A in magnitude, and thus the energy consumption is around 10^{-6} W. The performance of flexible piezoresistive pressure sensors reported recently has been summarized in Table 1. The performance of this pressure sensor is comparable to most of the best reported results in literature.

Table 1. Summary of performance of flexible piezoresistive pressure sensor reported recently.

Materials/Structures	Mechanism	Operating Voltage (V)	Sensitivity (kPa ⁻¹)	Response time (ms)	Detection limits (Pa)
rGO wrapped PVDF Nanofiber films ³⁰	Piezoresistive	1	3.1(10-60 Pa) 15.6(20-60 kPa)	5-7	1.2
Carbonized Silk Nanofiber Membrane	Piezoresistive	0.1	34.47(0.8-400 Pa) 1.16(400-5000 Pa)	16.7	0.8
PDMS/Graphene ⁴¹	Piezoresistive	1	8.5 (<2000 Pa)	40	1
CNT/Graphene, Microstructured PDMS ⁴⁸	Piezoresistive	0.03	19.8(<600 Pa) 0.27(600-6000 Pa)	16.7	0.6
rGO/PU sponge ⁴⁹	Piezoresistive	—	0.26 (<2000 Pa)	—	9
CNT/Ag NPs/sponge ⁵⁰	Piezoresistive	1	2.21 (2.24-11 kPa) 9.08 (11-61.8 kPa)	—	—
CNT/PDMS microdome arrays ⁵¹	Piezoresistive	—	15.1 (<500 Pa)	40	0.2
PEDOT:PSS/PUD microarrays ⁵²	Piezoresistive	0.2	56.8 (<200 Pa)	—	13
Cu NW/PVA ⁵³	Piezoresistive	—	0.036 (<4000 Pa)	—	400
Sparkling Graphene Block ⁵⁴	Piezoresistive	0.1	229(0-0.12k Pa) 26.9(0.4-1k Pa)	—	10
rGO foam ⁵⁵	Piezoresistive	—	15.2 (<300 Pa)	—	165
SWNT/PDMS microstructure ⁴⁰	Piezoresistive	2	1.8 (<300 Pa)	<10	0.6
rGO/PDMS microstructure ⁵⁶	Piezoresistive	1	5.53 (<100 Pa) 0.01(100-1000 Pa)	0.2	1.5
SWNT/cotton thread ⁵⁷	Piezoresistive	—	0.0156(<2500 Pa)	—	500
Carbon black/PU sponges ⁵⁸	Piezoresistive	—	0.068 (<2300 Pa)	<20	17
MOFs/PI ⁵⁹	Piezoresistive	1	6.25 (<1500 Pa)	<10	0.73
PPy/PDMS ⁶⁰	Piezoresistive	—	0.03~17(<1 kPa)	—	2
Inter-locking patterned PPy@PVA-co-PE/POE ⁶¹	Piezoresistive	2	1.24 (<150 Pa)	—	1.3
Au NW/Tissue paper ¹⁸	Piezoresistive	1.5	1.14(<5000 Pa)	<17	13
SWNT/Tissue paper(this work)	Piezoresistive	0.1	2.2(35-2500 Pa) 1.3(2500-11700 Pa)	35-40	<35

Due to its high sensitivity and fast response speed, the pressure sensor could be used to monitor the human physiological signals in real-time. The pulses of arterial blood pressure could be read out accurately. According to the measurement results of a male adult (male, 34 years old, 170 cm height,

65 kg weight), a typical radial arterial pulse waveform with two or three peaks in one cycle pulse and 68 beats per minute is shown in Figure 4a. For a healthy young man, a typical radial arterial pulse waveform was constituted of two distinguishable peaks (P_1 and P_3) and a late shoulder (P_2) in between, as shown in Figure 4b. The P_1 and P_2 correspond to the early and late systolic peaks pressure, respectively, while the P_3 usually showed up in the diastole region.⁶² The line shape is known to be caused by blood pressure from the left ventricle contracts and reflective wave from the lower body. We can also derive a very important parameter commonly used for health monitoring, namely the radial augmentation index $AI_r = P_2/P_1$,⁶² which is a characteristic value for arterial stiffness and highly related to the age of people. From a series of waveforms measured with our SWNT/tissue paper pressure sensor, the radial augmentation index AI_r is calculated to be about 0.65, which is a characteristic value for a healthy adult male.

Figure 4. Real-time monitoring of human physiological signals. a) Detection of the physical force applied by arterial heart pulse on the wrist, the heart beats 68 times per minute; b) The enlarged view of one cycle of the radial arterial pulse waveform; c) Responsive curve of the pressure sensor mounted on the wrist to monitor muscle activity during the hand open and grip; d) Responsive curve of the pressure sensor mounted onto throat to monitor muscle activity caused by tongue movement.

Due to high flexibility of this sensor, it could be mounted to any position of human body to study the muscle activity. We further use this sensor to monitor muscle motion caused by finger movement (Figure 4c) and tongue movement (Figure 4d). For monitor finger movement, the sensor is attached on the wrist, which has the muscle to control movement of the finger. For example, when the hand opens or closes, the finger movement could readily cause the muscle to straighten or loosen, changing the pressure applied onto the pressure sensor, as shown in Figure 4c. This is extremely useful in curing muscle damage and even for physical training. The pressure sensor was also attached to the throat to study the muscle motion caused by tongue movement. It is well known that the tongue's stretching and rolling could cause the motion of muscle on the throat. And the motion of the muscle on the throat could also apply a mechanical force onto the pressure sensor. As shown in Figure 4d, the tongue movement could be well monitored by our pressure sensor mounted on the throat. If the

tongue keeps stationary in the mouth, the current also keeps at a constant value, while the motion of tongue could drive the motion of muscle on the throat and apply a pressure on the pressure sensor, thus significantly increase the current in the pressure sensor. Currently, the relationship between current response pattern and tongue motion has not been established and studied, which should be done in the next step.

Figure 5. A simple demonstration of pressure sensor array as soft robotic skin. a) Optical image of the 4×4 pressure sensor array; b-d) Response of the sensor array to the touch of one, two, and three fingers, respectively.

A soft skin with capability of sensing force, pressure, and temperature is one of the most important components of an intelligent robot. Here, we demonstrate a prototype of such soft skin that could position the site and even the magnitude of pressure applied onto a robot. As a proof of concept, a sensing area was constructed by assembling 16 pressure sensors into a 4×4 pixel sensory array, as shown in Figure 5a. This sensor array could readily detect the touch by other objects (see Movie 1 in supporting information). Figure 5b-d show that the skin could well differentiate the positions and number of fingers pressing the sensing area. When the fingers touched the sensors in the sensing area, the sensor matrix would give a fast response which to be shown on monitor, and the addresses of sensors that were touched could be also positioned (see Movie 1 in supporting information). The response speed of the skin is determined by the response speed of individual pressure sensor in the array. The sensory array could also realize large-area pressure sensing, and successfully detect the shape of object lying on it, as shown in Figure S4 (supporting information). Figure S4a shows a M6 hex key (26 g, average ~ 520 Pa on each sensor) lying on the sensor array, it covers 4 sensors in the array; Figure S4b shows the corresponding pressure pattern detected by the sensor array. This large-area pressure sensing capability is extremely important for practical robotic-skin application.

Conclusions

In conclusion, we fabricated flexible, wearable pressure sensors based on novel SWNT/tissue paper through a simple dip-coating and heat treatment process. High performance flexible, wearable pressure sensors could be facilely fabricated by stacking SWNT/tissue paper onto electrodes on PI substrates and encapsulating with a PDMS layer. Due to the unique structure of SWNT interconnection in the tissue paper, the sensor showed superior performance with concurrence of several merits, including a high sensitivity of 2.2 kPa^{-1} in pressure range of 35-2500 Pa and of 1.3 kPa^{-1} in pressure range of 2500-11700 Pa, an ultralow energy consumption ($\sim 10^{-6} \text{ W}$), and a rapid response time. Based on the excellent performance and the ultra-conformal contact of the sensor on uneven subject, high-fidelity human physiological signal (such as radial artery pulse, muscle motion at various positions) can be monitored in real-time and in situ. Soft robotic skin was also demonstrated by integrating pressure sensors into array which could simultaneously monitor the pressure and also positions of pressure applied on the skin. We believe that the SWNT/tissue paper-based pressure sensor, which is fabricated from abundant tissue paper and SWNT aqueous dispersion through a low cost and scalable process combined advantages of high sensitivity, versatility and low power consumption, holds great prospect in developing low-cost artificial skins and wearable electronics.

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Table of Content

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