Conductive Film with Flexible and Stretchable Capability for Sensor Application and Stealth Information Transmission

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Abstract Flexible and wearable strain sensors for human-computer interaction, health monitoring, and soft robotics have drawn widespread attention to promising applications in the next generation of artificial intelligence devices. Here, we reported a kind of novel and simple sensor based on layer-by-layer (LBL) method. Carbon nanotubes (CNTs) layer provides high ductility and stability in the process of tension sensing, while silver layer provides low initial resistance and fast reflecting in the process of tension sensing. LBL method ensures the uniformity of the conductive layer. The sensor has superior sheet resistance of 9.44 $\Omega$/sq., high elongation at break of 104%. For sensing capability, the sensor has wide reflecting range of 60%, high gauge factor (GF) of 1000 up to 60% strain, fast reflecting time of 165 ms. Excellent reliability and stability have also been verified. It is also worth mentioning that the entire process does not require any expensive equipments, complicated processes or harsh experimental conditions. The above features provide an idea for large-scale application of flexible stretchable sensors.

Keywords Strain sensor; Flexibility; Silver films; Carbon nanotubes; Human motion monitoring

INTRODUCTION

With the development of smart materials\cite{1}, stretchable strain sensors which can convert mechanical deformation into readable resistive or capacitative signals are widely utilized in human activity recognition\cite{2,3}, health monitoring\cite{4}, and wearable display\cite{5,6,7,8}. For example, human-computer interaction\cite{9} records small skin deformation caused by speech and large deformations for health assessment monitoring, such as bending/straightening body joints to analyze motion patterns. The sensor can be categorized into piezoelectric type\cite{11}, resistive type\cite{12,13,14} capacitive type\cite{15,16} and the others on the basis of different working mechanisms. Compared with other kinds of sensors, resistive sensors have the advantage of simplicity and reliability and can be used for signal readout and feasible device manufacturing, which makes it possible for large-scale applications. There are various requirements for the performance of strain sensors, such as low initial resistance, high elongation, high stability, good fatigue resistance and so on. These properties are closely related to the material composition and structure of the sensor.

The key to improving the comprehensive performance of the sensor is suitable conductive materials and preparation methods\cite{17}. CNTs are promising materials for highly stretchable strain sensors because of their great aspect ratio and excellent flexibility\cite{18,19,20,21}. Metal nanomaterials are widely used in the field of flexible electronics because of their excellent electrical and mechanical properties\cite{22}, including silver nanowires\cite{23}, gold nano meshes\cite{24,25}, silver nanoparticles\cite{26}, silver films\cite{27}, and so on. However, conductive carbon materials and metal nanomaterials both have their own limitations. The high resistance of conductive carbon materials hinders its development in the field of flexible devices, while the limited stretchability properties of metal nanomaterials also restrict their application in related fields. How to combine the properties of conductive carbon materials and metal nanomaterials and give full play of their respective advantages to make up for their disadvantages remains challenging.

In this work, CNTs and silver layer were combined and a kind of sensor with excellent comprehensive performance was obtained. The synergistic application of metal nanomaterials and carbon nanomaterials in flexible stretchable sensors has broad prospects for performance complementarity and optimization.

EXPERIMENTAL

Materials

Multi-walled carbon nanotubes (MWCNTs) (diameter, about 10–30 nm; length, about 10–30 $\mu$m; the content of $-COOH$ is about 1.55 wt%) with a purity of over 90% were acquired from Chengdu Organic Chemistry Co., Ltd. $\text{AgNO}_3$ was obtained from Chengdu Organic Chemistry Co., Ltd.

Experimental (Experimental details are available in the Supporting Information.)
China Pharmaceutical Group Chemical Reagents Co., Ltd.; NH$_3$·H$_2$O (25%−28%), was purchased from Tokyo Chemical Industry Co., Ltd. Glucose and lysozyme were obtained from J&K Scientific Ltd. SYLGARD184 was purchased from Dow Corning. Chitosan was purchased from Qingdao BZ Oligo Biotech Co., Ltd. All the materials were directly used without further purification.

**Preparation of Sensor**

The preparation process of sensors is shown in Fig. 1. Preparation of CNTs layer: After dispersing CNTs in absolute ethanol, the mixture was blended by ultrasonication for 1 h, and then the stable CNTs dispersion was obtained by standing for 1 h. Then, the dispersion was slowly injected into a Petri dish containing deionized water. It could be seen that CNTs formed a film on the surface of the liquid. After that, the foamed polyester sponge was slowly inserted from the edge of the Petri dish into the deionized water dripped with CNTs dispersions. It was clear that the CNTs layer moved in the opposite direction as a whole, and was continuously compacted to become thicker.

Preparation of silver layer: First, AgNO$_3$ was dissolved in deionized water. After the solute was completely dissolved, 2.5% NH$_3$·H$_2$O was added drop by drop. Glucose and lysozyme were dissolved in deionized water, respectively, followed by ultrasonication. Then, the above solution was mixed in a Petri dish. After 10 h of reaction at room temperature, the silver film floated on the water surface.

Manufacturing of sensitive and flexible sensors: First, the mixture of PDMS and crosslinking agent (16:1) was stirred for 20 min, and then placed in a Teflon mold with a dumbbell shape. Second, the above mixture was degassed in a vacuum dryer at room temperature for 30 min to remove bubbles. Then, the PDMS mixture was solidified at 80 °C for 2 h. The CNTs film was transferred onto the cured PDMS surface by horizontal transfer method before dried in an oven to remove absolute ethanol and deionized water, which made the CNTs film adhere tightly to the polymer. The above process was repeated 5 times to obtain the uniform CNTs layer. PDMS sample with CNTs layer was immersed in the solution of chitosan (0.4 g in 100 mL of 1% acetic acid) to increase the adhesion. By alternating transfer of CNTs film and silver film, a conductive layer with excellent comprehensive properties was obtained. After the conducting layer was air-dried, the wires were connected to both ends of the conductive layer as the electrode. Finally, PDMS with the same composition was poured on the top of the sample for encapsulation.

**Characterization and Measurement**

Scanning electron microscopy (SEM) was conducted on JSM-IT300LV (NEC Corporation). Thermogravimetric analysis (TG) was conducted on SDT-600 (TA Instruments) at 10 °C/min. X-ray diffraction (XRD) profiles were recorded on a high resolution X-ray polycrystalline diffractometer (XRD-6100, Shimadzu, Japan), the scanning rate was set as 3 (°)/min. Raman spectra were analyzed with a Renishaw Raman Spectrometer in Via Reflex (diode laser wavelength 532 nm), the scanned wavenumbers ranged from 100 cm$^{-1}$ to 3000 cm$^{-1}$. For the attenuated total reflection (ATR) mode, a Silver Gate, Nicolet6700 Fourier transform infrared (FTIR) spectrophotometer with a resolution of 4 cm$^{-1}$ was used to study the surface functional group of the samples, the scanned wavenumbers ranged from 600 cm$^{-1}$ to 3000 cm$^{-1}$. Sensor resistance measurements were performed on an Ucetech (UC2868A) digital multimeter, and the stretcher used for strain-resistance testing was performed on IMADA CO., LTD. MX2-500N. The sensor currents were measured on a Zennium-E electrochemical workstation (ZAHNER, Germany). The tensile testing of the strain sensor (extension speed: 10 mm/min) was performed by an electronic tensile testing machine CMT4503 MTS SYSTEMS (China) Co., Ltd.

**RESULTS AND DISCUSSION**

**Morphology**

Fig. 2 reveals the SEM images of PDMS/CNTs, PDMS/silver and...
PDMS/CNTs/silver samples. The uniform distribution of the conductive layer on the micro-nano scale can be observed, which shows that they are barely damaged in the process of transfer and encapsulation. CNTs and silver layer were transferred onto the surface of PDMS to form good conductive networks. The top-view SEM images of PDMS/CNTs, PDMS/silver and PDMS/CNTs/silver conductive films are presented in Figs. 2(a)−2(c). In Figs. 2(a) and 2(b), we can see the top conductive layer of the conductive film. It can be seen that the silver layer has the structure of microcracks, and CNTs can be seen from the cracks of silver layer from Fig. 2(c). Fig. 2(d) shows the cross section of the PDMS/CNTs/silver sensor. The intertwined CNTs are expected to connect the micro-cracks between the silver films. Because of the low viscosity of uncured PDMS, it is easy to occupy the space between CNTs and silver layer for encapsulation. As a result, almost all CNTs and silver layers are adhered and embedded in the cured PDMS substrate, which leads to the strong peeling resistance of the conductive layer. Pouring and in situ curing of encapsulation layer give the conductive network the same deformation as the PDMS substrate. Benefiting from these aspects aforementioned, rapid and efficient recovery of electric properties as well as deformation could be expected.

**Mechanical Properties**

In the application of strain sensor, the mechanical properties are vital significant. The types of conductive layers have effects on the mechanical properties of sensors. The tensile curve at a rate of 10 mm/min is shown in Fig. 3(a). All sensors exhibit approximately the same tensile strain behavior overall. With the change of the conductive layer composition, the stress-strain curves of the sensors are different. The tensile strength and elongation at break of neat PDMS are the highest, which are 345 kPa and 104.76%, respectively. The mechanical properties of PDMS/CNTs/silver layer are similar to those of neat PDMS (Fig. 3b). These results are due to the fact that the CNTs layer

![Fig. 2](image)

**Fig. 2** Surface SEM images of (a) PDMS/CNTs, (b) PDMS/silver and (c) PDMS/CNTs/silver sensor. (d) Combination of conductive layer and substrate.

![Fig. 3](image)

**Fig. 3** The stress-strain behavior of neat PDMS and sensors with various conductive layers: (a) tensile behavior, (b) ultimate elongation (unfilled) and tensile strength (stripe filled). (c) Photos of PDMS/CNTs/silver sensor in different application scenarios.
and silver layer are physically attached to the cured PDMS surface, and the mass fraction of conductive material is rather small, which barely has influence on the molecular structure and aggregation structure of PDMS.

In addition, the various deformations that the PDMS/CNTs/silver sensor may have in daily applications are shown in Fig. 3(c). The above results indicate that the sensor has capability to cope with compound deformation, which lays a foundation for its application in intelligent equipments.

As shown in Fig. 4(a), the stress-strain relationship tends to be stable after the first stretching/releasing cycle in the ten-consecutive stretching/releasing processes with a strain of 10%. Reversible tensile strain curves show that PDMS/CNTs/silver sensor has excellent mechanical stability during stretching/releasing. As shown in Fig. 4(b), in the process of continuously increasing strain for tension-recovery (from 10% to
60%), it can be observed that the shape of hysteresis loops on the stress-strain curve is similar. In general, the hysteresis effect of the sensor is not obvious, which is beneficial to the real-time reflecting.

**Thermal Stability and Crystallization Properties**

TG (a) and DTG (b) curves of neat PDMS, PDMS/CNTs, PDMS/silver and PDMS/CNTs/silver are shown in the Fig. 5. After heated to 800 °C in the nitrogen atmosphere, PDMS, PDMS/CNTs, PDMS/silver and PDMS/CNTs/silver retained 42.2 wt%, 42.3 wt%, 52.1 wt% and 52.3 wt% of the original weights, respectively, indicating that most of the Si—O bonds are broken and oxygen functional groups are removed. At the same time, it can be estimated that the weight content of the silver layer in the PDMS/silver and PDMS/CNTs/silver is about 10%. The TG curves also show that the initial thermal degradation temperature of the four samples is about 260 °C. The maximum degradation peaks of neat PDMS and PDMS/CNTs are 504 and 520 °C, respectively, while those of PDMS/silver and PDMS/CNTs/silver are 540 and 532 °C, respectively. The results show that both CNTs layer and silver layer can improve the thermal stability of the sensor slightly.

Both CNTs and silver films were characterized by X-ray diffraction to reveal their crystallization. The double peaks reflecting the (100) and (002) planes of CNTs are observed at 2θ=26.2° and 40.2°.[23] For silver film, a striking and sharp peak at 2θ=38.1° is attributed to the (111) plane. What is more, four weak peaks at 2θ=44.3°, 64.4°, 77.4°, and 81.5° reveal the (200), (220), (311), and (222) planes of silver film.[27] All peaks of the silver film are independent and sharp, indicating that the silver film has high crystallinity. The peaks of CNTs are relatively noisy, indicating that the CNTs film is not highly crystalline. The XRD profiles of PDMS/silver layer with different concentrations of lysozyme are shown in Fig. 5(c), and the diffraction peaks of silver are shown in (i)–(iv). When lysozyme concentration is 0.7, 0.4, and 0.2 mg/mL, the particle size of silver nanoparticles in the silver layer is 33.7, 35.7, and 48.2 nm, respectively. However, when the silver layer with lysozyme concentration of 0.2 mg/mL is superimposed on the CNTs layer, the particle size of the silver layer is 31 nm. These data show that the lower the lysozyme concentration, the smaller the size of silver nanoparticles. At the same time, the lamination process with CNTs layer has little effect on the crystallization of silver nanoparticles.

**Chemical Structure**

The attenuated total reflectance IR (ATR-IR) spectra of neat PDMS, PDMS/CNTs, PDMS/silver and PDMS/CNTs/silver are shown in Fig. 6(a). A band (C=O stretching vibration) of CNTs appears at 1701 cm$^{-1}$. For PDMS/silver and PDMS/CNTs/silver, I-band of amide near 1665 and 1642 cm$^{-1}$ can be observed, which is caused by the existence of a small amount of amino acid components in silver layer. Slight redshift corresponds to the formation of hydrogen bonds between $-\text{NH}_3^+$ on silver and...
COOH on CNTs.

The Raman spectra of neat CNTs and PDMS/CNTs are shown in Figs. 6(b) and 6(c), respectively. For PDMS/CNTs, G band at 1588.1 cm\(^{-1}\) (associated with crystalline sp\(^2\) carbon) and D band at 1352.3 cm\(^{-1}\) (related to defects or heteroatom doping) are shown \((I_D/I_G=0.897)\). Compared with neat CNTs with \(I_D/I_G=0.951\), it can be seen that the process of preparing the PDMS/CNTs has little effect on the structural integrity of CNTs.

In Fig. 6(d), Raman spectral deconvolution analysis reflects the propensity of hydrophobic tryptophan (Trp) and histidine (His) residues on the silver layer,\(^{27}\) which coexists with the D-band and G-band (green peaks) of CNTs.

**Electrically Conductive Property**

The sheet resistance of PDMS/CNTs/silver sensor with various of CNTs/silver layers is illustrated in Fig. 7(a). PDMS/CNTs has a semi-conducting property, and the sheet resistance is about 4500 \(\Omega/\text{sq}\). A remarkable decrease in sheet resistance \((147 \Omega/\text{sq})\) by approximately 30 times of magnitude is observed for the PDMS/CNTs/silver with only one silver layer transferring. The sheet resistance of the sensor decreases with increasing numbers of CNTs/silver layers. This phenomenon can be understood as that with the increase of CNTs/silver layers overlap, the conductive network becomes more perfect, forming additional new conductive path. When the CNTs/silvers layers are over four layers \((\text{C/S/C/S})\), the sheet resistance reaches a relatively stable value, indicating that a perfect conductive network has been formed, and the increase of CNTs/silver layers thereafter can hardly affect the sheet resistance. It is noteworthy that the sheet resistance of seven-layer conductive network is higher than that of four-layer conductive network, probably because the contact resistance of copper wire-conductive silver pasting to silver layer is much smaller than that of CNTs layer. Therefore, the PDMS/C/S/C/S \((9.44 \Omega/\text{sq})\) was selected for further research.

**Strain Sensing Behavior**

The functional relationship between relative resistance \((\Delta R/R_0)\) and tensile strain is revealed in Fig. 7(b). Obviously, the two are positively related at each step. This phenomenon can be explained by the change of contact conditions of CNTs/silver layer network, such as disconnection, destruction and reorganization. A diagram of these steps in the CNTs/silver conductive layer is demonstrated in Fig. 7(c). Under 60% strain, the accurate sensing effect of the PDMS/CNTs/silver sensor can be shown. At the beginning of the tensile process, the silver layer gradually breaks down and the CNTs layer becomes sparse. The above process leads to the destruction of the conductive path. When the elongation of the sensor reaches 16%, the relative resistance has a sudden change. After the conductive network is completely separated, the application range of the sensor

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**Fig. 7** (a) Sheet resistance of PDMS/CNTs/silver sensor with varying numbers of CNTs/silver layers. (b) Gauge factor of PDMS/CNTs/silver sensor under different strains. (c) Schematic design of conductive layers under reversible tensile strain. (d) Tensile fatigue performance of PDMS/CNTs/silver sensor.

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reaches the maximum limit. This great strain sensing behavior of PDMS/CNTs/silver sensor can effectively identify the external strain.

Gauge factor (GF) is a significant parameter for exploring the sensitivity of strain-resistance sensors, which has practical significance for the application of sensors. The GF is defined as:

$$GF = \frac{dY}{dX}$$

(1)

where dY and dX refer to the relative resistance change of sensors and the applied strain. The resistance-tensile strain process can be analyzed in three steps (A, B, C), which is revealed in Fig. 7b. In step A (under 16% strain), resistance increases slowly with strain. The PDMS/CNTs/silver sensor can competently contribute to accurate work, thanks to the reversibility of the CNTs/silver conductive network in the process of resistance-tensile strain. The GF in step A is 38.6. Then, GF rises to 667 in step B (16% to 33% strain) and to 1000 in step C (over 33% strain), illustrating good sensitivity of PDMS/CNTs/silver sensor under large strain. In step B, the silver layer has been broken in the whole area and could only reduce the resistance in the local area of the CNTs network. In step C, the conductive path becomes thinner. The high sensitivity under all strain is attributed to the combination of the CNTs/silver layer.

The fatigue test of the PDMS/CNTs/silver sensor was also performed. As shown in Fig. 7d, during 1300 reversible resistance-tensile strain (10 mm/min), the resistance changes quite little. After several cycles of damage and reconstruction, the conductive network maintains its initial stage. The resistance usually reaches a constant value at the set strain and returns to the initial value after the strain is recovered. The excellent durability and fatigue resistance of the PDMS/CNTs/silver sensor can be demonstrated by the above experiments, thanks to the good adhesion between the conductive layers and the PDMS matrix.

Fig. 8(a) plots the relative resistance between 0% and 10% strain and the strain versus time. Resistance is positively related to the strain and tends to be linear. When no strain occurs, resistance decreases due to poor contact and separation of CNTs/silver conductive network. During the residence period, the reflecting of the sensor changes quite little. When the strain is released from 30% to 0%, the relative resistance is restored due to the reconstruction of the CNTs/silver contact joint. Due to the elasticity of PDMS matrix, the novel adhered and embedded structure is introduced. In the recovery process, the relative resistance can almost return to the original value. The experimental results show that PDMS/CNTs/silver sensor has high tensile properties and enviable stability, and can be used to monitor small and large movements.

Typical current voltage (I-V) curves of PDMS/CNTs/silver sensor under different tensile strains are shown in Fig. 8f. The enviable linear relationship between voltage and current shows that no matter what strain is applied, PDMS/CNTs/silver sensor has good Ohmic behavior. The slope of the I-V curve is the real-time resistance of the sensor under the corresponding tensile state. Obviously, it decreases with the increase of strain. This result is consistent with the above, indicating that the relative resistance increases when a larger strain is applied.

**Human Motion Monitoring**

PDMS/CNTs/silver sensor exhibits the merits of good flexibility, high sensitivity, rapid reflecting and durability, which has application prospect in the comprehensive detection of physiological movement. The application of PDMS/CNTs/silver sensor for physiological movement monitoring is shown in Fig. 9. We pasted the sensor on several parts of the human body to detect various physiological movements of the human body. In Fig. 9(a), repeated finger bending and straightening resulted in a typical relative resistance change of the sensor. In the process of finger bending, the resistance of the sensor also increased. These two processes were carried out at the same time. When the finger straightened again from the bending state, the resistance of the sensor also returned to the initial value. It is found that the bending reflecting curve of the same action is not exactly the same, which means that PDMS/CNTs/silver sensor can detect subtle differences in several similar actions, which once again verifies its sensitivity.

In addition to large-scale motion detection, PDMS/CNTs/silver sensor also has the ability to monitor small-scale motion. PDMS/CNTs/silver sensors were connected to the laryngeal node and eyebrow tail to monitor the expression induced micro muscle movement, as shown in Figs. 9b and 9c. The changes in the resistance of the sensor correspond to the stretch of the facial muscles caused by the cheek bulge, and blinking and swallowing can be accurately recorded. It is worth noting that different expressions produce different signal patterns, because each expression will lead to various movements of facial muscles. The significant differences among these relative resistance signals reveal that the sensor can be used as an expression recognition device. From these
applications, it can be seen that the PDMS/CNTs/silver sensor has good stability and repetitive motion in human body monitoring, which is achieved by the synergistic effect of the clever combination of the CNTs layer and the silver layer.

**Morse Code Transmitting**

PDMS/CNTs/silver sensor can also be used to transmit international Morse code because of its rapid reflecting and highly reliable piezoresistive reflecting. The code encodes 26 English letters as short message numbers and long signal standard sequences called "dots" and "dashes". When we pressed the sensor intermittently with our fingers in the way of generating a telegram, we would get a series of electrical signals, which is a process of transmitting secret information, as shown in the Fig. 10(a). The finger could alter the CNTs/silver layer conducting network reversibly by pressing device, so as to increase the relative resistance. After removing the pressure from the finger, the conductive network was reconstructed under the traction of PDMS, which could completely recover to the initial value of low resistance. The key of information transmission is the sensitivity

Fig. 8 (a) The reversible resistance-tensile strain curves (10 mm/min). (b) The reflecting time of the PDMS/CNTs/silver sensor. (c,d) Resistance changes versus strain of 5%, 10%, until 60% at a rate of 10 mm/min. (e) Relative resistance reflecting from 0% to 30% and then returned. (f) Ohmic behavior under different strains (0% to 20%) for the PDMS/CNTs/silver sensor.
of the sensor, which could clearly distinguish various parts of the information. Through the drawing process of relative resistance time, the three processes can be clearly distinguished, thus representing “point”, “dash” and “space”. The experimental results show that the sensor responds quickly and accurately, which can be used to transmit Morse code.

**CONCLUSIONS**

In a word, we studied a unique method for manufacturing a tensile sensor with the synergistic effect of the clever combination of the CNTs layer and the silver layer. CNTs layer provides high ductility and stability in the process of tension.
sensing, while silver layer provides low initial resistance and fast reflecting in the process of tension sensing. Unique conductive network endows PDMS/CNTs/silver sensor with superior sheet resistance of 9.44 Ω/sq, wide strain applied range of up to 60%, good sensitivity (GF 1000 up to 60%), rapid reflecting time of 165 ms and great durability (1300 cycle tests). PDMS/CNTs/silver sensor can be used to accurately monitor the large and small movements of human body in real time. The results show that the strain sensor developed by us has broad development potential in intelligent flexible equipments. Our work provides a valuable method for the development of human motion monitoring sensors with excellent comprehensive performance.

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