A Flexible Resistive Strain Sensor Based on Mixed Carbon Nanomaterials

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Abstract. With the development of flexible electronic technology, flexible strain sensors are playing an increasingly important role in human motion detection and medical health. It is still an important research topic to consider the sensitivity and stability of flexible strain sensors in practical applications. This work describes a new type of flexible resistive strain sensor that uses a new and simple method to embed sensitive materials on the surface of a flexible substrate. The flexible strain sensor has both excellent stability and good flexibility. The sensor has a gauge factor of 5.875 in the strain range of 0-5%, and a gauge factor of 4.252 in the strain range of 10%-120%. The sensor remains stable over 1000 repetitions. The sensor can be used to detect human movements, including large-scale movements of human limbs and small-scale bending of finger joints. Because of its simple manufacturing process and excellent characteristics, it has shown great potential in medical health and smart wearable devices.

1. Introduction
With the development of society and the improvement of people's living standard, flexible strain sensors[1,2] have been widely concerned and studied due to their applications in human movement detection[3] and medical health[4]. Resistive strain sensors have become the mainstream of flexible strain sensors because of their low cost and little influence by environmental factors. However, in order to realize the detection of human movement, the flexible resistive strain sensor should have sufficient stability, high sensitivity, high resolution and large measuring range.

Because of its extremely high aspect ratio, the conductivity of Carbon Nanotubes (CNTs) mainly depends on direct contact, and the tunneling effect only occupies a small part. This makes flexible sensors made of CNTs as conductive materials maintain greater linearity, but it is difficult to detect small deformations and has low sensitivity[5]. Carbon black (CB) is a classical zero-dimensional carbon conductive material with low gravity, low cost and good conductivity. Since the electron tunneling process is highly sensitive to the distance between CB, the flexible sensor based on CB has a high sensitivity and resolution, but it is difficult to achieve a large measurement range and a high linearity[6]. CB and CNTs were mixed to construct a conductive network with different sensing behaviors, which was richer than a single nanomaterial. The mixed carbon nanomaterial compensated for the limitations of each material while maintaining the advantages of each material, thus effectively improving the sensitivity of flexible resistive sensors[7].

Polydimethylsiloxane (PDMS) is a stable and reliable flexible material, which has been widely used in flexible electronic products due to its advantages such as low cost, excellent elastic performance and easy processing. However, the low adhesion between PDMS and the sensing material will lead to poor performance and poor repeatability of the sensor. One solution is to mix

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PDMS and sensitive materials together to form mixed materials[8], but this will lead to the decline of PDMS elastic performance, affecting the linearity and measurement range of the sensor. Chen da[9] proposed that MWCNTs multilayer film was deposited on PDMS substrate by layer-by-layer (LBL) self-assembly method. The sensor produced has high sensitivity, but the process of LBL self-assembly method is relatively complex, making it difficult to realize mass production and large-scale application of the sensor.

CB and MWCNTs were mixed as sensitive materials in this work. The conductive layer of CB-MWCNTs was inlaid on the surface of PDMS by a simple method, and PDMS was coated on the dry CB-MWCNTs layer. Under the influence of gravity, PDMS solution would be immersed in the dry CB-MWCNTs layer, and the sensitive material would be inlaid on the surface of the flexible substrate after heating and curing by PDMS solution. In this way, CB-MWCNTs were integrated into PDMS to ensure the high reliability and durability of the sensor without affecting the elastic performance of PDMS. After testing, the flexible resistive strain sensor made by this method has the advantages of good elasticity, excellent sensitivity and high durability.

2. Materials and method

2.1. Materials
In this work, MWCNTs (Nanjing Xianfeng Nano Material Technology Co., Ltd.) are used as conductive materials, with purity of 95%, length of 10-30 μm, and carboxyl content of 2.0 wt%. CB of the model BP2000 was purchased from Cabot Corporation, Boston, Massachusetts, USA. PDMS was used as the matrix material (Sylgard 184; Dow Corning Corp., Gales Ferry, Connecticut, USA).

2.2. Sensor fabrication
First, 2wt% CB solution, 2wt% MWCNTs solution and 1 wt% CB and MWCNTs solution were respectively prepared with anhydrous ethanol. In order to disperse the carbon nanomaterials better, the solution was put into the ultrasonic machine and shaken by ultrasonic for 30 min. After standing for two hours, the supernatant was taken out to remove the bottom precipitation. The treated solution is evenly dripped onto the smooth glass sheet and dried in a heating box. A dry conductive layer of carbon nanomaterials is formed on the surface of the glass sheet. The main PDMS agent and curing agent were mixed at a ratio of 10:1. After 20 min of mechanical stirring, the mixture was put into the empty chamber for 20 min of vacuum to remove the bubbles in the mixture. The PDMS solution was then spun onto the glass sheet at a speed of 300 rpm. The sample was put into a heating box and heated at 70°C for 2h, and then the solidified PDMS film was removed from the glass sheet. At this time, the conductive layer was also embedded on the lower surface of PDMS and removed together. The manufacturing process of composite film is shown in Figure 1(a). The composite PDMS film was cut into strips of 4 cm long and 1 cm wide as the main body of the sensor, and copper wire was fixed
at both ends of the strip PDMS as wires with copper foil tape. The sensor made is shown in Figure 1(b).

![Figure 2](image)

Figure 2. (a) Cross-sectional SEM image of composite film; (b) diagram of tensile evolution mechanism.

3. Structure and working principle

Figure 2(a) shows the scanning electron microscope (SEM) diagram of the cross section of PDMS/CB-MWCNTs composite film. It can be seen from the figure that the sensitive layer of CB-MWCNTs occupies only a small part of the whole composite film and will not have an evident impact on the elastic performance of the whole composite film. The introduction of CB particles can fill the gap of MWCNTs network and improve the electrical conductivity of composites. The evolution mechanism of the conductive network under tension is shown in Figure 2(b). When the composite material is deformed, the destruction and reconstruction of the conductive network coexist, and the destruction effect is much greater than the reconstruction effect when it is stretched. When stretching in a small range, the first changes are the disconnection of contact between CB and CB and the disconnection of contact between CB and MWCNTs, and the conductivity of the conductive network changes significantly, which makes the sensor has a greater sensitivity in the low strain range. When the tension gradually increased, the contact between MWCNTs and MWCNTs was destruction, and the conductivity of the conductive network was further changed, which made the sensor still have resistance changes in a large tensile range.

4. Results and discussion

4.1. Introduction of the test tools

The pressure gauge for measuring the dynamic performance of the sensor was ZQ-990A (Dongguan Zhiqu Precision Instrument Co., Ltd., Dongguan, Guangzhou Province, China), with a measurement range of 0-50 N and a force resolution of 0.01 N. The instrument for measuring the resistance was a desktop LCR meter (TH2826; Changzhou Tonghui Electronics Co., Ltd., Changzhou, Jiangsu Province, China).

![Figure 3](image)

Figure 3. (a) Comparison diagram of tensile properties of composite films; (b) Response curve of the resistance variation of the different sensors; (c) resistance response curve with micro strain.
4.2. Performance test of sensor

Figure 3(a) shows the tensile characteristic curves of the film with CB-MWCNTs embedded on the surface of PDMS, the film made by CB-MWCNTs as a hybrid filler doped in PDMS, and the pure PDMS film. The elastic modulus is defined as the proportional coefficient of the stress-strain relationship. It can be seen that the composite film made by CB-MWCNTs doping has the highest Young's modulus, and the pure PDMS film has the lowest Young's modulus. This may be because carbon nanofillers will reinforce the PDMS and increase its Young's modulus. The Young's modulus of the CB-MWCNTs embedded on the PDMS surface is closer to the pure PDMS film, and the conductive layer of CB-MWCNTs has little effect on the Young's modulus of PDMS. Figure 3(b) shows the stress-strain test curve of a sensor made of CB, MWCNTs, and CB-MWCNTs as conductive materials. The sensitivity of the sensor is evaluated by calculating the gauge factor (GF), 

\[ GF = \frac{\Delta R / R_0}{\varepsilon} \]

The sensor with CB as the sensitive material has a GF of 6.954 in the low strain range of 0-5%. This is because CB is a zero-dimensional material, and the small deformation of the composite material will cause the destruction of the conductive network. The sensitivity of the sensor with MWCNTs as the sensitive material in the low strain range of 0-5% is 3.192, because MWCNTs are one-dimensional carbon nanomaterials with a large aspect ratio, and the conductive network does not change significantly in the low strain state. The CB-MWCNTs composite material has a sensitivity of 5.875 in the low strain range of 0-5%, which is between the single CB and the single MWCNTs flexible sensor. When the strain range is between 10% and 120%, the GF of the CB-MWCNTs composite material is 4.252, which is significantly greater than that of the sensors containing only CB and MWCNTs materials. Figure 3(c) shows the resolution of the CB-MWCNTs sensor. It can be seen from the figure that the sensor also has a significant resistance change to a small strain of 0.1%. In the small strain range of 0-1%, the sensitivity of the sensor can reach 0.77. Figure 4(b) shows the different tensile speeds of the sensor for testing, and the strain is 10%. It can be seen that the resistance change of the sensor at different stretching speeds can remain stable. Figure 5(a) is the repeatability test of the sensor under 5% strain and 20% strain respectively. The total number of repetitions of the sensor exceed 1000 cycles. It can be seen from the figure that the resistance change rate tends to stabilize after the initial downward drift. It shows that the sensor can maintain good stability for a long time under different strain amplitudes. A cycle period is selected from Figure 6(a) (repeatability graph) to show the hysteresis of the sensor. As shown in Figure 6(b), the hysteresis of the sensor under dynamic pressure is 10.5%.

![Figure 4. Response curve of sensor resistance under different stretching speed.](image)
Figure 5. (a) The resistance response curve of the sensor is repeated for 400 cycles under 5% strain; (b) the resistance response curve of the sensor is repeated for 200 cycles under 20% strain; (c) dynamic hysteresis of sensor cyclic stretch release.

Figure 6. Monitoring of human motions using sensors: (a) finger bending; (b) wrist bending; (c) elbow bending; (d) walking, running and squatting.

4.3. The application of sensor
Figure 6(a) shows a finger bending test using a sensor. As shown in the figure, the sensor can produce different resistance changes for different degree of finger bending. This sensor can detect the difference between small angle bending and large angle bending. In addition, sensors are placed at the arms, wrists and knees to detect human movement, as shown in Figure 6(b)-(d). These large-scale motion changes cause large resistance changes in the sensors. It can be seen that the sensor still has a good detection effect for large motion. The sensor can effectively distinguish the change of motion Angle of different joints. These results have important reference value for human health.

5. Conclusion
The synergistic effect of CB and MWCNTs can effectively improve the sensitivity of the sensor. The conductive layer of CB-MWCNTs was inlaid on the surface of PDMS membrane by a new manufacturing process. The manufactured sensor has high sensitivity, great resolution as well as good
elasticity and stability. Due to its excellent mechanical and electrical properties, the sensor can detect the subtle movements and large movements of the human body, which has great potential in medical health and intelligent wearable technology.

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