Flexible and Transparent Pressure Sensor Based on CNTs Film Microstructure

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Flexible and transparent pressure sensor based on CNTs film microstructure

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Abstract

In recent years, capacitive flexible pressure sensors have been widely studied in electronic skin and wearable devices. The traditional capacitive pressure sensor has a higher production cost due to micro-nano machining technology such as lithography. This paper presents a flexible transparent capacitive pressure sensor based on a PDMS/CNT composite electrode, simple, transparent, flexible, and arrays without lithography. The sensitivity of the device has been tested to 0.0018 kpa⁻¹ with a detection range of 0-30 kPa. The sensor is capable of rapidly detecting different pressures and remains stable after 100 load-unload tests.
1. Introduction

With the development of intelligent and flexible electronic technology, it is expected that robots can respond to external stimuli just like human beings, and e-skins can help robots understand the "feelings" of human beings. In recent years, research on electronic skin has become a hot topic in flexible electronics technology. As a new type of electronic device that imitates human skin in its sense of external pressure, flexible pressure sensor has a broad application prospect in tactile perception, biomedical equipment, health monitoring, human-computer interactions and wearable devices.

According to its working principle, the pressure sensor can be divided into three types, namely, capacitive, resistance, and piezoelectric. Compared with the other two kinds of capacitive pressure sensors, it has the advantages of high sensitivity, wide detection range, good dynamic response, simple device structure, and flexible pressure sensor parts are generally made flexible by integrating organic polymers or inorganic materials into flexible substrates. Common flexible electrode materials for flexible pressure-sensors include CNTs, graphene, silver nanoparticles, metal nanowires, etc. These materials are typically mixed and filled with corresponding flexible substrates, such as PDMS, styrene-butadiene-styrene block copolymer, polyimide (PI), and conductive fabrics, etc. in the form of dopants. For instance, Zhang T et al. used polydimethylsiloxane (PDMS) silicone rubber to imitate the microstructure of silk and carbon nanotubes as electrodes in 2014, to obtain a stress-sensor with excellent performance. For example, Li et al. used silver nanowires, graphene and polyamide nanofibers to form a flexible pressure-sensor. This pressure-sensor has an ultra-high sensitivity (134 kPa-0.15 kPa) with a minimum detection limit of 3.7 Pa. Hence, it can be used
to detect the pulse rate of the human body and the vibration of the vocal cord during speech. In order to obtain a sensor with excellent performance, a lot of research has been done on the device structure besides the choice of flexible materials. By introducing some unique structures, the sensitivity of the device can be significantly improved. It has been proved that the sensitivity of the pressure sensor can be effectively improved by using the nanometer or micron structure of the dielectric layer. For example, Yunsik Joo et al. [23] used wavy microstructures in their work. After the dimethylsiloxane plates were treated with ultraviolet light and oxygen, these were stretched and subsequently recovered to form wavy PDMs. Additionally, there are micro-structure pressure sensors manufactured by the MEMS process [24]. As the sensor is loaded, the microarray is compressed. With the gradual increase of stress, the soft effect of the small structure slowly disappears, and the contact area reaches the maximum. This phenomenon leads to a decrease in sensor sensitivity. Some researchers have noticed a variety of microstructures in nature. Su B et al. [25] was inspired by Mimosa and designed a touch-sensitive pressure sensor. Although the introduction of composite electrode and microstructure improved the sensitivity of flexible pressure sensors, its manufacturing process was relatively complex, and the manufacturing cost was relatively high.

This paper presents a flexible transparent capacitive pressure sensor based on PDMS/CNT composite electrode prepared by a simple process. The sensor uses structured PDMS as the microstructure layer. The composite electrode of large-area carbon tube and PDMS prepared by vapor deposition can realize a large-area sensor array. With a sensitivity of 0.0018 kpa^{-1} and a detection range of 0-30 kPa, the sensor can quickly detect different pressures and remains stable after 100 load-unload tests Principles and Materials.
2. Principles and Materials

The flexible pressure sensor designed is shown in Figure 1, which adopts a sandwich structure. The dielectric layer is PDMS with a microstructure. With the purpose of have both transparency and high sensitivity, the composite electrode of PDMS/CNTs is used. The capacitance value of the capacitive pressure sensor can be calculated by the capacitance formula of the parallel plate capacitor:

\[
C = \frac{A \varepsilon_r \varepsilon_0}{D}
\]

where \(\varepsilon_0\) is the vacuum dielectric constant; \(\varepsilon_r\) is the relative dielectric constant; \(A\) is the relative area of the upper and lower plates; \(D\) is the distance between the upper and lower plates. As per Equation (1), the capacitance value \(C\) is determined by three parameters: \(\varepsilon_r\), \(A\) and \(D\). The capacitance value can be altered by changing \(D\), \(A\) and \(\varepsilon_r\) to improve the performance of the sensor.

**Figure 1.** Schematic illustration and the profile display of the pressure sensor. Microstructure PDMS was used as the dielectric layer, carbon nanotube film as the electrode, and PDMS as the flexible substrate.

As shown in Figure 2, when the pressure is applied on the surface of the capacitive sensor, the dielectric layer and the electrode of the flexible material compress and deform to a definite extent. This deformation results in the decrease in the distance between the electrode layers. At the same time, the air in the coarse porous composite electrode is extruded as a result of
compression deformation, and the $\varepsilon_r$ increases accordingly. The relative area between the
electrode layers also increases due to the deformation of the microstructure. Under the joint
action of these three parameters, the sensitivity performance of the sensor is greatly improved.

**Figure 2.** Schematic illustration of the working principle.

Carbon nanotube films were obtained by continuous fabrication of meter-scale single-walled
carbon nanotube films [26] reported in our previous work and collected on the filter membrane.
The pressure sensor fabrication process is shown in Figure 3. Initially, the PDMS primary agent
and curing agent were prepared into a solution in the proportion of 9:1, and the solution was
stirred using a magnetic mixer at 1500r/min for 10min to mix evenly. Then the silicon wafer
coated with hydrophobic monolayer was used as the rigid substrate. The prepared PDMS was
poured onto the silicon wafer. After suspending the coating, the silicon wafer was heated at 75℃
for 180 minutes. After peeling, the transparent and flexible PDMS with uniform thickness was
obtained. The PDMS can easily form covalent bonds with silicon, and the hydrophobic
monolayer can guarantee that PDMS does not adhere to the silicon wafer during stripping. The
conductive CNT on the filter membrane was transferred to the PDMS stripped from the silicon
wafer via imprint method to form the PDMS/CNT composite electrode. However, this step also
requires the copper foil to be extracted from the CNT through the conductive silver glue to
facilitate the subsequent testing. In order to ensure the transparency and flexibility of the device,
we used PDMS with microstructure for the dielectric layer. The PDMS solution was poured on
the sandpaper as the substrate. The PDMS dielectric layer with microstructure was obtained by stripping after solidification. The assembly of the sensor adopts the sandwich structure. The composite electrode obtained in the above steps and the dielectric layer overlapped up and down to form the "top electrode - dielectric layer - bottom electrode".

**Figure 3.** Procedure for fabricating the sensor

### 3. Results and discussion

The morphology and characterization of CNT and PDMS are shown in Figure 4.

**Figure 4.** The morphology and characterization of the PDMS and CNT films: (a) and (b) show the PDMS microstructure under light microscope and atomic force microscope. (c) and (d) show the CNT film under a light microscope and atomic force microscope.
Sensor sensitivity is defined as Equation 2:

\[ S = \frac{\Delta C}{C_0 \times \Delta P} \]  

where \( \Delta C = C - C_0 \) is the capacitance change; \( C \) is the capacitance value at the corresponding pressure; \( C_0 \) is the initial capacitance value; \( \Delta P \) is the change in relative pressure. According to Equation (2), sensor sensitivity is the slope of the curve in Figure 5.

![Figure 5. The capacitance-pressure curve of the sensor.](image)

It can be seen that in the range of 0-10 kPa, the sensor's sensitivity reaches 0.0018 kPa\(^{-1}\), and the detection limit of the sensor is 0-30 kPa\(^{-1}\). The sensor's sensitivity can be divided into three parts, with the highest sensitivity in the range of 0-10 kPa. At this point, the air in the microstructure and the porous electrode is emptied with the increase of pressure, and the contact area \( A \) between the \( \varepsilon_r \) and the upper and lower plates increases, whereas the spacing \( D \) decreases. The combination of these three factors results in the maximum sensitivity of the device at the given time. With the increase in pressure, the air is exhausted, and the \( \varepsilon R \) does not change. The dielectric layer of the microstructure becomes the critical factor affecting the
sensitivity of the device. At this point, with the increase in pressure and the change in microstructure, the distance $D$ between the upper and lower plates of the device gradually decreases. The device can still maintain a high sensitivity (0.000848 kPa$^{-1}$). When the pressure gradually increases to the point where the elastic effect of the microstructure is exhausted, the device sensitivity drops to its lowest (0.000273 kPa$^{-1}$).

Device stability is also one of the performance indicators of pressure sensors. Figure 6 shows that the sensor was repeatedly loaded/unloaded at 10K Pa 100 times at a frequency of 4MHz to test its repeatability. The test results show that the sensor has decent repeatability stability.

![Figure 6. Durability test of the sensor.](image)

Figure 7 shows the dynamic response of the device. The test results show that the sensors can rapidly detect different pressures and good dynamic response and stability when applied with 5 kPa, 10 kPa and 15 kPa loads, respectively.
Figure 7. Dynamic response of the sensor.

3. Conclusion

A capacitive flexible pressure sensor based on carbon nanotube film has been fabricated. PDMS and carbon nanotube films were used as a composite electrode to construct a microstructure dielectric layer in the form of a sandwich structure device. The device with a pressure detection range from 0 kPa to 30 kPa is obtained through a simple process. The maximum sensitivity of the pressure sensor is 0.0018 kPa\(^{-1}\), with good stability after 100 load-unloading cycles and improved ability to quickly detect different pressures. In addition, the sensor can be fabricated over a large area. The flexible pressure sensor reported in this paper has excellent performance. It is expected to be applied to electronic skin and flexible wearable devices.

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References

[1] M.L. Hammock, A. Chortos, B.C. Tee, J. B Tok, Z. Bao, *Adv. Mater.* 2013, 25, 5997.
[2] X. W. Wang, Z. Liu, T. Zhang, *Small.* 2017, 13, 1602790.
[3] T.Q. Trung, N.E. Lee, *Adv. Mater.* 2016, 28, 4338.
[4] X. Sun, J. Sun, T. Li, S. Zheng, C. Wang, W. Tan, J. Zhang, C. Liu, T. Ma, Z. Qi, C. Liu, N. Xue, *Nano-Micro Lett.* 2019, 11, 57.
[5] Z. Wang, C. Dong, X. Wang, M. Li, T. Nan, Xi. Liang, H. Chen, Y. Wei, H. Zhou, M. Zaeimbashi, S. Cash, N.X. Sun, *npj Flexible Electron.* 2018, 17, 2978.
[6] P. Huang, Y.Q. Li, X.G. Yu, W.B. Zhu, S.Y. Nie, H. Zhang, J.R. Liu, N. Hu, S.Y. Fu, *ACS Appl. Mater. Interfaces,* 2018, 10, 11197.
[7] R. Guo, X.L. Wang, W.Z. Yu, J.B. Tang, J. Liu, *Sci. China: Technol. Sci.* 2018, 61, 1031.
[8] N. Manikandan, K. Sriram, S. Muruganand, K. Balakrishnan, Sebastain, J. Sivasankaran, V. kumar, A. Suresh. *Adv. Sci. Lett.* 2017, 23, 1875.
[9] Y. Wang, X. Wu, D. Mei, L. Zhu, J. Chen. *Sens. Actuators, A,* 2019, 297, 111512.
[10] B. Wang, A. Facchetti, *Adv. Mater.* 2019, 31, 1901408.
[11] M.H. Medagedara, T.T. S Peiris, N.D. *Instrumentation,* 2020, 7, 36.
[12] C. Wang, K. Xia, H. Wang, X. Liang, Z. Yin, Y. Zhang, *Adv. Mater.* 2019, 31, 1801072.
[13] O. Kanoun, A. Bouhamed, R. Ramalingame, J.R. Bautista-Quijano, A. Al-Hamry, *Sensors,* 2021, 21, 341.
[14] Z. Jing, Q. Zhang, Y. Cheng, J. Chao, D. Zhao, Y. Liu, W. Jia, S. Pan, S. Sang. *J. Micromech. Microeng.* 2020, 30, 085012.
[15] X. Zhao, W. Xu, W. Yi, Y. Peng, *Sens. Actuators, A,* 2019, 291, 23.
[16] X. Zhou, Y. Zhang, J. Yang, J. Li, S. Luo, D. Wei. *Nanomaterials,* 2019, 9, 496.
[17] W. Li, Y. Zhou, Y. Wang, Y. Li, Li. Jiang, J.i Ma, S. Chen, *Macromol. Mater. Eng.*
2020,305,1900736.

[18] Z. Q. Duan, T. P. Li, K. Yao, Z.L. Xuan, Y.T. Liu, Micro Nano Lett. 2016,12,0578.
[19] H. Xu, L. Gao, Y. Wang, K. Cao, X. Hu, Li. Wang, M. Mu, M. Liu, H. Zhang, W. Wang, Y. Lu, Nano-Micro Lett. 2020,12,1.
[20] M. Fortunato, I. Bellagamba, A. Tamburrano, M. S. Sarto. Sensors, 2020,20, 4406.
[21] M. Jian, K. Xia, Q. Wang, Z. Yin, H. Wang, C. Wang, H. Xie, M. Zhang, Y. Zhang, Adv. Funct. Mater. 2017, 27, 1606066.
[22] X. Li, Y. J. Fan, H. Y. Li, J. W. Cao, G. Zhu, ACS Nano, 2020,14, 9605.
[23] Y. Joo, J. Byun, N. Seong, J. Ha, H. Kim, S. Kim, T. Kim, H. Im, D. Kim, Y. Hong, Nanoscal, 2015, 7, 6208.
[24] B. Liang, W. Chen, Z. He, R. Yang, Z. Lin, H. Du, Y. Shang, A. Cao, Z. Tang, X. Gui, Small, 2017,13,1702422.
[25] B. Su, S. Gong, Z. Ma, L.W. Yap, W. Cheng, Small, 2015,11,1886.
[26] B.W. Wang, S. Jiang, Q.B. Zhu, Y. Sun, L. Jian, P.X. Hou, S. Qiu, Q.W. Li, L. Chang, D.M. Sun, Adv. Mater. 2018, 30,1802057.
