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Tunable-Sensitivity Flexible Pressure Sensor based on Graphene Transparent Electrode

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Abstract

Tunable-sensitivity and flexibility are considered as two crucial characteristics for future pressure sensors or electronic skins. By the theoretical calculation model, we simulated the relationship curve between the sensitivity and PDMS pyramids with different spacings, and found that the spacing of pyramids is a main factor to affect the sensitivity of the capacitance pressure sensor. Furthermore, we fabricated the capacitance pressure sensors using graphene electrodes and the PDMS pyramid dielectric layers with different spacings. The measurement data were consistent with the simulation results that the sensitivity increases with the spacing of pyramids. In addition, graphene electrode exhibits prefect flexibility and reliability, while the ITO electrode would be destroyed rapidly after bending. These graphene pressure sensors exhibit the potential in the application in the wearable products for monitoring breath, pulse, and other physiological signals.

Keywords: graphene electrodes, pressure sensor, tunable sensitivity, flexibility.

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1. Introduction

Recent years, flexible pressure sensor has attracted an amount of interest, owing to increasing demand over the last decades. Flexible pressure sensors could work as human’s skin, and have potential in a lot of application fields, including robotic arms,[1-4] flexible electronic devices,[5-9] wearable devices,[10-14] health monitoring[15-18], human-machine interaction[19-21]. These pressure sensors have different sensing mechanics, including capacitive [11, 20, 22], resistive[21, 23, 24]，piezoelectric[25, 26] and so on. Among them, capacitive sensors have unique advantages in terms of stability, low power consumption and fast response. At present, scientists have been able to improve the sensitivity mainly by microstructure of electrode and dielectric layer.[4,27] Yong taek Hong reported that silver nanowires was covered on PDMS substrate with ripple structures as an electrode[27], and these pressure sensors achieved a high sensitivity (3.8KPa^{-1}). Later, they improved the sensitivity to 9.9 KPa^{-1} by the spacing structure of dielectric layer. [28] In 2011, the group of Zhenan Bao fabricated a sensitive and stable flexible pressure sensors using the pyramid structures of Polydimethylsiloxane (PDMS) as a dielectric layer,[29] and studied the relationship between compressibility of microstructure and the side wall angle, space of pyramids.[30] The sensitivity of optimized pyramid structural pressure sensor was two orders of magnitude higher than the sensor without any structure. However, for different applications, the pressure sensors with different sensitivity are also necessary. Owing to the geometric nonlinearity of microstructure dielectric layer, the relationship between capacitance and pressure become complicated. Hence, a systematic and comprehensive analysis is necessary for understanding the relationships between the sensitivity and structures of dielectric layers.

Here, we simulated the relationship curve between the sensitivity and PDMS pyramids with different spacings. It was found that the spacing of pyramids is a main factor to affect the sensitivity of the pressure sensor, and revealed the mechanism of the fact that sensitivity decreased with pressure. Furthermore, the PDMS pyramid dielectric layers with different spacing were used to fabricate the pressure sensors with graphene film as electrode. The measurement data was consistent with above simulation result. Hence, the sensitivity of pressure sensors could be adjusted by the spacing of pyramids.

In addition, graphene electrodes exhibited excellent mechanical property. It was found that the sensor with indium tin oxide (ITO) electrode would be destroyed rapidly after bending, but the sensor with graphene electrode could work stably after bending 50 times. The sensor with graphene electrode will be more suitable to apply in flexible electronic products than that with ITO electrode.

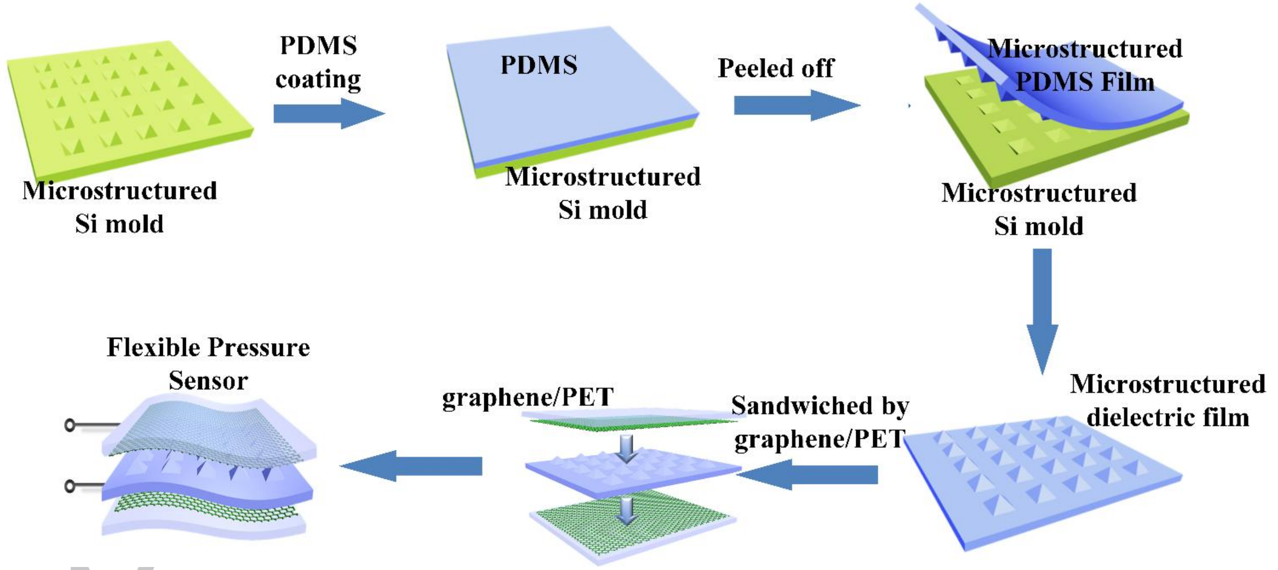

Fig.1. the preparation processes of PDMS dielectric layer with micro-pyramid structures and a pressure sensor based graphene electrodes.

2. Experimental

Figure 1 illustrate the preparation process of dielectric layer and flexible pressure sensor. First of all, the silicon (Si) mold with micro-pyramids structure was fabricated by lithography and etchant (KOH: IPA: H_2O). The photoresist we used was S1806 and the crystal orientation of silicon was <100>. The etch rate of silicon was 0.175μm/min. Owing to the etching direction of silicon, the angle of pyramids is fixed, so the heights of pyramids increase with the side length linearly. Then, PDMS was coated onto the Si mold, and was cured under 70 °C for about 3 hours. The PDMS film is easy to be demolded because the surface energy of PDMS film was very low. After PDMS film was cured, we used tweezers to nip the corner of PDMS film and peeled it off from Si mold to obtain the micro-pyramid structure dielectric layer. Further, the graphene film was grown on the copper foils via chemical vapor deposition method and the CVD temperature for graphene film growing was 1050 °C. Graphene deposition rate was one layer per 15 minutes. We could control the thickness of graphene film by CH_4 concentration.[30] Graphene was transferred onto PET film as the top and bottom electrodes, owing to the stable electrical performance on the deformable substrate under bending. The ITO on PET was purchased from SIGMA-ALDRICH and its number was 639303. Finally, the sandwich structural sensor was assembled and packaged by optically clear adhesive (OCA) glue. In addition, the capacitance of the pressure sensor was measured by Keithley 4200A parameter analyzer (4200A-SCS). We linked our device to the Keithley 4200A parameter analyzer and used capacitance-voltage measurement unit to measure the capacitance of our device. Then We putted the different weights on the device and monitored the change of capacitance.

The morphology of micro-structured silicon mould and dielectric layer was directly characterized by field-emission scanning electron microscopy (SEM, JSM-7800F). The capacitance of pressure-sensitive sensor were measured by 4200A-SCS.

3. Simulation and analysis

The pyramid structure was divided into many pieces of square sheets with the same thickness. At certain height, the distribution mapping of pyramids could be confirmed so that we could calculated the capacitance of this interval via linear elastic theory. Then we could get the capacitance of the whole pressure sensor by accumulating the capacitance of every interval along Z direction. The equation of compressed sensor's capacitance was derived in supporting information, and was given as follow:

$$C' = \frac{1}{\sum_{i=n_3}^{i=n_1} \frac{1}{\frac{n_2 \cdot \varepsilon_r \cdot (x_i + \Delta x_i)^2}{4\pi k \cdot (dh - \Delta l_i)} + \frac{\varepsilon_0 \cdot (S_{PET} - n_2 \cdot (x_i + \Delta x_i)^2)}{4\pi k \cdot (dh - \Delta l_i)}}} \quad (1)$$

Here, C' is the capacitance under the pressure, ε_r is relative dielectric constant, x_i is the length of each sheet which is resulted from dividing pyramids, Δx_i is the increment of the sheet's length, S_{PET} is the area under the pressure, dh is the height of each sheet.

At First, the compressed heights of the sensor with different spacings under different pressure were simulated. In Figure 2a, the compressed height increased with the pyramids' spacings under the same pressure. Owing to the increase of the spacing and the same pyramids' sizes, the density of pyramids would decrease. Hence, every pyramid would suffer larger force, and the compressed height would also increase. In Figure 2b, capacitance's change ($\Delta C/C_0$) increased with the pyramids' spacing, and the slope of curve decreased with the pressure.

In addition, the relationship of sensitivity and pressure was calculated by the derivative of the function of relative capacitance-pressure, and the relationship curve was shown in Figure 2c. The sensitivity decreased with pressure, which was resulted from the geometry of pyramids. When the pyramid suffered a small pressure, the tip of pyramid was not completely compressed and was easily compressed, which led to capacitance change rapidly. When the pyramid suffered a large pressure, the tip of pyramid was completely compressed and the stressed area expanded. Then compressed length increased slowly and the capacitance increased slowly too. When the pyramids were compressed completely, the change of the slope would stop. So the sensitivity of sensor decreased with pressure.

These simulation results show that the main factor, which have a great influence in the sensitivity of sensor, is spacing of the pyramids. Owing to the geometry of pyramids, the sensitivity decreased sharply in the range of small pressure and decreased slowly in the range of large pressure.

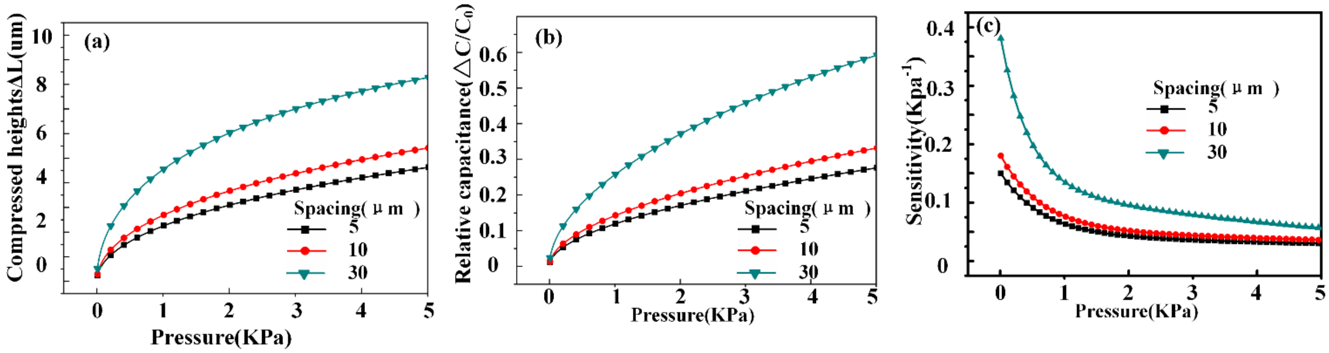

Fig.2. predicted relationships of (a) compressed height-pressure; (b) relative capacitance-pressure, and (c)sensitivity-pressure with different pyramids' spacings (the length of the pyramid is 15 μ m).

4. Results and Discussion

In order to verify above simulation results, PDMS micro-pyramids dielectric films with different spacing were fabricated via lithography, etching, coating and solidification, as shown in Figure 1. In Figure 3a-c, the micro-pyramids moulds and PDMS copies are uniform with the same size. The cross-section SEM image shows the highly parallel rows of pyramids with the same height.

The curve of relative capacitance-pressure with different spacings was obtained by the tests of loading pressure. In Figure 4a, the change of relative capacitance increases with the spacing under the same pressure. The increase of the spacing causes the increase of period, and every pyramid will suffer bigger pressure. So the sensitivity will increase with spacing. And it is clear that the change trend of relative capacitance with different spacing is similar to the simulation result, as shown in Figure 2.

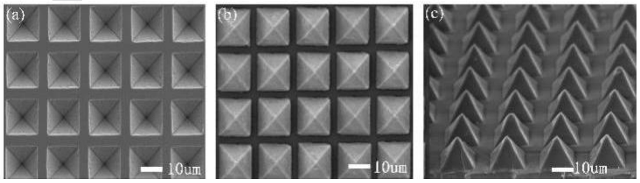

Fig.3. (a)Top-view SEM image of silicon mold; (b) top-view and (c) side-view SEM images of the patterned PDMS film with pyramids features.

Furthermore, the relationship of the relative capacitance -pressure with different spacings was nonlinear fitted and got a function. We calculated the relationship of sensitivity-pressure via the nonlinear fitted function, and the relationship curve was shown in Figure 4b. Under the same pressure, the sensitivity increased with the spacing of microstructure, which was similar to the simulation result. Figure 4c reveals the relationship between the sensitivity and different number of pyramids. It is observed that the sensitivity dropped with the number of pyramids. This curve further demonstrates that the sensitivity of the sensor extremely relied on the density of pyramids.

The real-time relative capacitance response curve of the sensor was measured by loading and unloading the pressure. Figure 4d shows that rapid rising edge of about 50 milliseconds reveal fast response to loading the pressure of 5Pa. Meanwhile, the relative capacitance signals could return to original level if the pressure was unloaded, exhibiting the prefect recoverability. Hence, the flexible pressure sensor could rapidly respond to the pressure with controllable sensitivity, highlighting the potential in dynamic force monitoring.

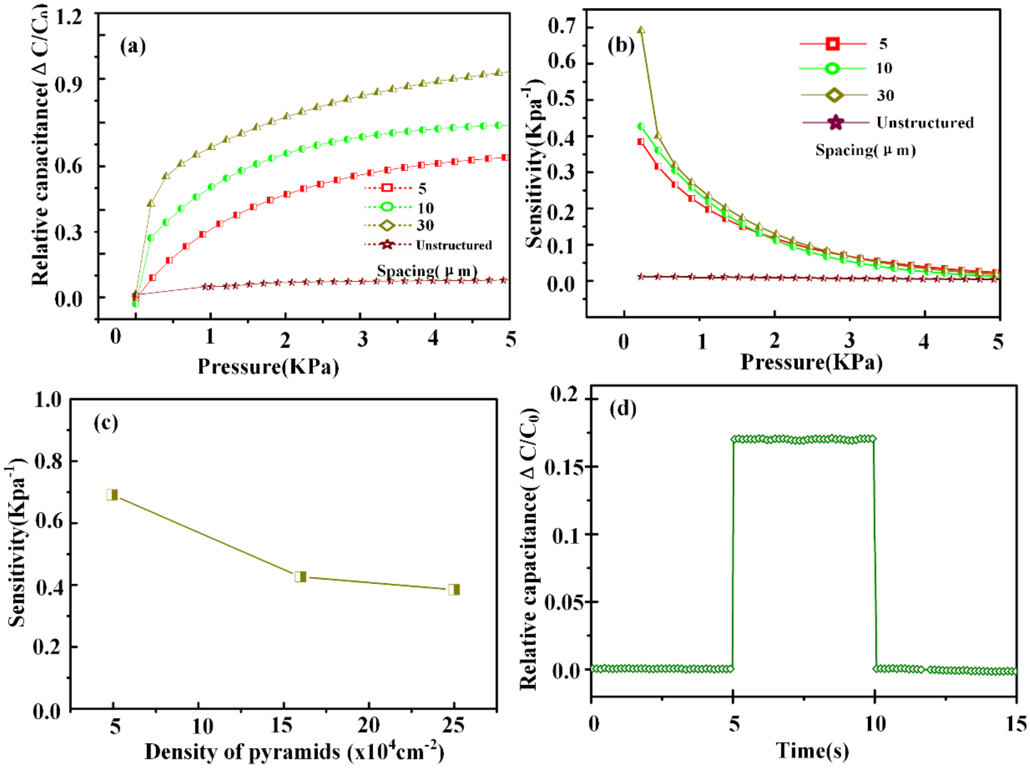

Fig.4. the relationships of (a) relative capacitance-pressure and **(b)** the sensitivity-pressure with different spacings between pyramids (the length of the pyramid is 15 μm); **(c)**the sensitivity of sensors with different density of pyramids;**(d)** the response time of the flexible pressure sensor, whose length is 15 μm and spacing is 5 μm (under the pressure of 5 Pa).

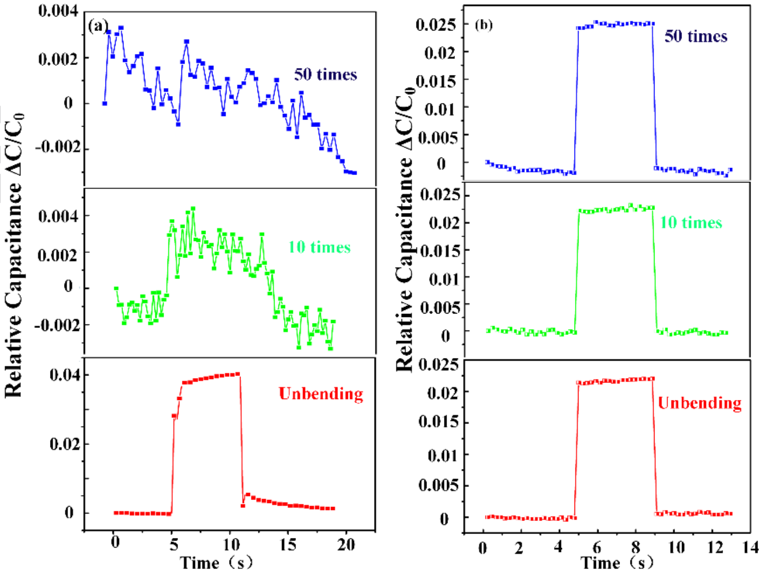

Fig.5. (a) Response of sensor with ITO electrode after bending 0,10,50 times, respectively; **(b)** response of sensor with graphene electrode after bending 0,10, 50 times, respectively.

For wearable devices or products, flexibility is an important property. All components, including dielectric layer and electrode, need to be flexible. Graphene was considered as an ideal candidate of next-generation transparent electrode. Hence, we fabricated a pressure senor using graphene as electrode, and compared the pressure sensors with ITO and graphene electrodes by push-pull machine, as shown in Figure 5.[31] The bending radius was 0.5mm. After bending 0, 10, 50 times, the pressure response performances of sensors with ITO electrodes were measured. Figure 5a shows that the relative capacitance response decreases rapidly after bending, and there is little response after bending 10 times, which results from the brittle property of ITO electrode.[32] On the contrary, the sensor with graphene electrode could work stably in bending test. After bending 10 or 50 times, the relative capacitance response curves were almost coincided with each other, as shown in Figure 5b. In Figure S1b, no crack could be observed after bending, revealing perfect flexibility of graphene electrode. In Figure S1c, no crack but graphene grains could be observed in the photograph of graphene film. Figure S1d shows that after being bended 50 times, graphene film has no obvious cracks. In addition, we painted silver electrode to both end of ITO electrodes and graphene electrodes. After bending ITO electrodes and graphene electrodes 1000 times, we use Keithley 7510 to test the resistance of the electrodes. The resistance of ITO increase sharply and graphene perform stably, which could be observed in FigureS2. Hence, the sensors with graphene electrodes possess both high sensitivity and excellent bendability.

In order to verify high performance of flexible sensors for detecting weak physiological signals, we fabricated a pressure sensor using graphene electrodes and PDMS dielectric layer with the spacing of 30 μm. In Figure 6a-b, pulse waveform could be detected, when the flexible sensor was attached onto the wrist. Each period stood for a vasodilation and contraction. The frequency of pulse could be observed in the Figure 6a. and we could diagnose a subject's physical condition. Figure 6c-d shows that the pressure sensor also could be used to test the breath signal when facing and breathing. The periodic flow of breath would produce the periodic pressure on the surface of the sensor, and our pressure sensor could detect impulsive force from breath. More interestingly, this sensor could distinguish if the mouth is open when breathing. When mouth is open, the waveform has a shrill peak.

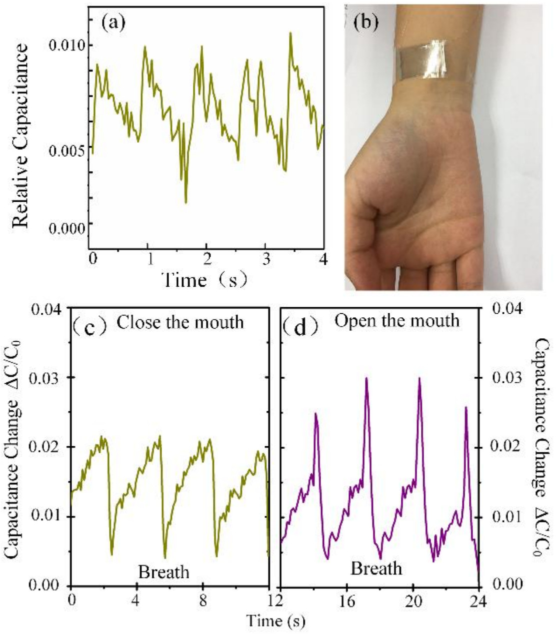

Fig.6. (a) Relative capacitance response of graphene sensor to the pulse; **(b)** the photograph of flexible sensors for pulse test. **(c)**breath waveform by nose; **(d)**breath waveform by nose and mouth.

5. Conclusion

In summary, the main influential factor of microstructural capacitive pressure sensor's sensitivity was studied by simulating the relationship between capacitance and pressure. The sensitivity of the microstructural sensor increased with the spacing of pyramids and decreased with pressure dramatically because of the geometry of pyramids. This flexible sensor could rapidly respond to the loading of the pressure with the response time of about 50 milliseconds. Moreover, these sensors exhibit the potential in the monitoring of breath, pulse, and other physiological signals. Excellent flexibility and reliability provide the possibility in the application for wearable products.

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Highlights

1. A capacitive pressure sensor with high-sensitivity and high-speed was proposed.

2. The sensitivity could be adjusted via the density of PDMS micro-pyramids.

3. The pressure sensor with graphene electrode exhibits prefect flexibility.

4. The flexible pressure sensor could monitor weak physiological signals.

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