Low-cost Graphene-based Flexible Strain Sensor with a Novel Transfer Method

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ABSTRACT

Flexible strain sensors, as one of the important members of intelligent electronic devices, have attracted a lot of attention in areas such as structural health monitoring, smart wearable devices, and electronic skin. In structural health monitoring, the sensor is required to be able to adapt to the complex structure surface, and has a high sensitivity so that the micro cracks can be found in time. At present, the traditional metal strain sensors have short plates with large rigidity and low sensitivity, prompting the exploration of new strain sensitive mechanisms and materials. Graphene, as a novel material, has been continuously explored and found its good flexibility and sensitive strain piezoresistive effect. The sensor made of graphene has high sensitivity, but the high production and transfer cost of high-quality thin-layer graphene limits its application. In this paper, we report a new flexible strain sensor based on thick CVD graphene which is produced by a low-cost graphene film transfer strategy. A 150nm-thick nickel-based CVD graphene film was used as a strain-sensitive material, the poly (dimethylsiloxane) (PDMS) that was not completely dried was used to transfer graphene, and the dried PDMS was considered as the flexible substrate, which simplified the process and reduced costs. A tensile strain test with a range of up to 10% was performed and the sensor achieved a gauge factor (GF) of 130 under high strain. The conductive paths model based on the percolation network theory fit the experimental date accurately. Through the experiments above, we have obtained a low-cost graphene flexible strain sensor, which is able to monitor micro cracks and it has a suitable sensitivity and range. The sensor has important reference significance for the future research of complex structural health monitoring system, and has great potential value in the field of commercial applications.

1. Introduction

With the development of new materials and new structural technologies, industrial designs such as complex structures and intelligent deformation structures have emerged. Traditional structural health testing techniques are challenged. At present, there is an increasing demand for strain detection of complex and variability structures. However, conventional strain sensors such as metal foil sensors, semiconductor sensors, piezoelectric wafer active sensors (PWAS)\(^1\) and fiber Bragg grating (FBG) sensors\(^2\) have limitations in installation, flexibility, and ductility\(^3\). Therefore, Nano technology-based sensors have become one of the main research direction in the field of advanced sensing technology of structural health monitoring.\(^4\)

In recent years, Nano technology-based sensors have developed rapidly. For flexible strain sensors, researchers have conducted many explorations on emerging nanomaterials. Inpil Kang et al.\(^5\) used a

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carbon nanotube polymer material to form a piezoresistive strain sensor, which is useful for detecting large strains and cracking, and can reduce the number of channels of data acquisition needed for the health monitoring of large structure. Sang-Ha Hwang et al.\[6\] used free-standing carbon nanomaterial hybrid sheets to fabricate the strain sensor, which consist of multi-walled carbon nanotubes (MWCNTs), exfoliated graphite nanoplatelets (xGnPs) and nanographene platelets (NGPs). Besides carbon nanotubes, silver nanowire\[7-8\] is another popular material. Morteza Amjadi et al.\[9\] reported highly flexible, stretchable and sensitive strain sensors based on the nanocomposite of silver nanowire (AgNW) network and PDMS elastomer in the form of the sandwich structure. Chan-Jae Lee et al.\[10\] fabricated a transparent (>90% at 550nm wavelength), stretchable (up to 100%), and sensitive (gauge factor (GF) of 30 at 100% strain) strain gauge by depositing an encapsulated crack-induced Ag nanowire (AgNW) network on a hydroxylated PDMS film.

Graphene flexible strain sensor is another research hotspot. Wang Yi et al.\[11\] transferred the graphene film onto a pre-stretched PDMS substrate and released it to obtain a graphene corrugated structure, which achieved a strain detection range of more than 30%. Xiao Li et al.\[12\] produced a CVD graphene network structure and proposed the principle of resistance-strain change of the conductive network. Xinming Li et al.\[13\] fabricated a large area graphene nanosheet network film by convection self-assembly method. Yueyi Jiao et al.\[14\] used CVD graphene with a thickness of about 105 nm as a sensitive material and a liquid metal as electrodes to make a strain sensor with a GF of about 1.51 and a test range of 9%. Until now, there are relatively few studies on the use of thick nickel-based CVD graphene as a sensitive material for strain sensor. The strain sensor based on such graphene film has the advantages of relatively low cost and simpler fabrication process, which are valuable for exploration and research.

In this paper, the multilayer nickel-based CVD graphene film was used as sensitive material. A strain sensor with high sensitivity was prepared by a dry-transfer-post-etching process. The measurement range of the sensor is approached to 10%, and GF is from 60 to 130. Tensile test experiment was carried out. The conductive paths model based on the percolation network theory was established to illustrate resistance-strain characteristics of the kinds of material which crack under strain. The model fitted the experimental data accurately. The feasibility of the sensor in structural health monitoring, robot motion capture, human body information perception, complex structure deformation detection and other applications is proved.

2. Experimental methods

2.1 Structure and materials of sensor

The sensor mainly includes three parts: flexible substrate and package, graphene film, and electrodes (Fig.1a).

![Figure 1](image_url)

**Figure. 1** (a) Structure diagram of the Sensor. (b) The Raman spectrum results. (c) The optical micrograph of graphene.
The PDMS (Dow Corning Sylgard 184) was used as flexible substrate and package material. Its base fluid and curing agent are mixed in a volume ratio of 15:1. A custom-made nickel-based CVD graphene film (produced by Shenzhen JingGe Nano Technology Co. Ltd.) was used, whose thickness was about 100-200 nm (about 300-600 layers). The Raman spectrum results are shown in Fig.1b, and the optical micrograph of its surface is shown in Fig.1c. Conventional conductive silver paste is applied as electrodes. In this paper, the size of the graphene film on the sensor is about $5mm \times 2.5mm$.

2.2 Fabrication Process

The preparation process of the graphene flexible strain sensor mainly consists of three parts: preparation of flexible substrate, transfer of graphene, production of electrodes and packaging. As shown in Fig.2.

2.2.1 Preparation of flexible substrate (Fig.2a)
The PDMS base fluid and the curing agent were mixed well for 30 minutes and evenly spread on a plastic plate with a thickness of about 0.6 mm. Let stand for 10 minutes to remove air bubbles and baked at 70°C for 20 minutes to obtain a semi-cured PDMS substrate with strong viscosity.

2.2.2 Transfer of graphene (Fig.2b-g)
The nickel-based graphene film was cut together with the nickel film to a desired size. The cut nickel-based graphene was gently placed on the semi-cured PDMS substrate, where graphene was not bonded tightly to the PDMS substrate. The PDMS and graphene films were carefully covered with a transparent PET film, and the PET film was gently pressed along the edges of the graphene to remove bubbles between the graphene and the PDMS substrate. Then the PET film was slowly peeled off, and the PDMS substrate was further cured by baking at 70°C for 60 minutes to complete the dry transfer of the graphene film. The prepared nickel-based graphene/cured PDMS was then etched in a ferric...
chloride solution for 4 hours to remove nickel. After the etching, it was repeatedly washed with DI water and air-dried, then baked at 80°C for 5 minutes to remove the residual moisture to obtain a graphene/PDMS structure. After these process, the graphene film was in close contact with PDMS substrate.

2.2.3 Production of electrodes and packaging (Fig.2h-j)
A small amount of liquid conductive silver paste were carefully coated on the surface of the graphene film as electrodes, and the copper wires were placed. The silver paste dried naturally at room temperature. The graphene film, electrodes, wires, and substrate were fully covered with PDMS whose thickness is about 0.4 mm. Let stand for 10 minutes to remove air bubbles and then baked for 90 minutes at 80°C. Finally, carefully remove the sensor from the production device.

Compared with other fabrication methods, the dry-transfer-post-etching process has the advantages of simple, fast, and cheap price, which means a superior commercial potential.

2.3 Test system

The experimental device consists of a micro-movement stage, voltage divider circuit, and data acquisition system (Fig.3). The copper wire is connected with the test circuit through soldering. The voltage value of the power supply $V$ and the value of the voltage divider of the fixed resistor $U_t$ were collected through the data acquisition system, and calculate the resistance value of the graphene strain sensor $R_g$ according to the series voltage divider equation: Eq.1.

$$R_g = \left( \frac{V}{U_t} - 1 \right) R_s$$

3. Results

3.1 Tensile test

The sensor to be tested was stretched from the initial length (0% strain) to a certain length, and kept it stretched until the resistance stabilized. Then the strain was released and the sensor was moved to the
initial length waiting for the next test cycle. During the whole process, the resistance was monitored and recorded in real time.

When the strain is less than 10%, the resistance change ratio is relatively stable. After repeated testing, the sensor is quite sensitive during 0-10% strain. Meanwhile, the resistance can drop back to the initial value when the strain totally released, which shows a good performance of operational stability. The relationship between the stable resistance change ratio after stretching and the magnitude of strain is shown in Fig.4a.

The GF is usually used to describe the sensitivity of strain sensor, which is calculated as Eq.2.

\[ GF = \frac{\Delta R}{R_0} \frac{1}{\varepsilon} \]  

Where \( \Delta R \) is the resistance change ratio, \( R_0 \) is the initial resistance, and \( \varepsilon \) is the strain. The GF of our sensor can be obtained as approximately 60-130 (Fig.4b).

When the strain was small (less than 1%), the GF showed a downward trend, falling from about 110 to 60. The experimental data (Fig.4c) shows that the reaction of the resistance is rapidly. It has a good repetitive stability, whose deviation of the resistance value at the 0% strain is within 2%.
In the case of large strain (1%-10%), the GF of the sensor slowly rises from 60 to 130 (Fig.4b). The resistance recovery performance kept well (Fig.4d), whose deviation of the resistance at the 0% strain is within 3.5%.

### 3.2 Structure-reconfiguration of the CVD graphene film

For the new sample has not been stretched, the stable resistance value at the 0% strain position rises from 12.3 ohms to 37.43 ohms as the maximum strain of the test cycle increases (Fig.5). Once the strain in the test cycle achieved 10%, the resistance will be maintained at about 40 ohms when the strain released to 0%.

![Figure 5](image.png)

**Figure. 5** 0% strain position resistance of sensor has not been stretched and sensor has been stretched.

![Figure 6](image.png)

**Figure. 6** The optical micrograph of graphene under different strain. (a-e) The optical micrographs of sensor has not been stretched under 0%, 2.5%, 5.0%, 7.5%, 10.0% strain. (f-j) The optical micrographs of sensor has been stretched under 0%, 2.5%, 5.0%, 8.0%, 10.0% strain.

Through observing optical micrographs of the stretching process of the graphene film on the sensor (Fig.6), it was found that with the maximum strain of the test cycle increased, the macroscopic crackles began to show up on the surface of the new sample (Fig.6a-e), which was the first time to achieve such
strain. The original structure of the graphene film was destroyed, resulting in that the graphene fragmentations cannot return to their initial position after the strain recovered to 0%. The generation of crackles lead to the increase of resistance at initial length and the size of crackle was varying with the strain loading and releasing, resulting in the change of resistance. These phenomenon can be summarized as structure-reconfiguration of CVD graphene film.

4. Discussion

It can be found from previous experiments that the basic principle of the strain sensor we made is the generation and size change of the material crack. At present, there are relatively few studies on the strain sensitivity mechanism of such thick CVD graphene, and it is valuable to explore. The sensor works under 10% strain, it is more practical to study the strain-resistance theoretical model of the sensor which has been stretched to 10% strain.

4.1 Model derivation

The strain-sensitive mechanism of graphene films mainly includes quantum tunneling theory\textsuperscript{[15-16]} and percolation network\textsuperscript{[17-20]}. The quantum tunneling theory is based on the underlying quantum effect, which interprets the strain-resistance effect as a result of strain causing a change in the tunneling barrier. The theory starts from the microscopic effect and analyzes the macroscopic resistance change law. It is mainly used for the analysis of physical and chemical properties of ultra-thin two-dimensional materials.

The theory of percolation network is based on macroscopic statistics, and it is believed that strain causes changes in the conditions under which conductive paths are formed in the sensitive material. Mainly used for mechanism analysis of conductive composites.

During the CVD growth process, the graphene nanosheets are connected to each other and stacked together, and a plurality of graphene nanosheets composed the film. They are relatively brittle and are susceptible to breakage under macroscopic strain conditions. The strain loading process causes the graphene nanosheets to slip and the gap to become large. Macroscopically, it is reflected in the generation of graphene fragments and the increase in the size of crackles.

Due to the nanosheets internal structure and the macroscopic crack under strain, it is difficult to find enough evidence for the quantum tunneling theory. The gap of the crack is larger than the range in which the quantum tunneling effect occurs, and the experimental data indicates that the characteristics of the change in the resistance of the sensor are different from those of the tunneling effect. However, it can be considered that the size variation of cracks on the graphene film changes the original conditions of conductive paths, which is similar to the idea of the percolation network. Therefore, the mechanism of such a strain sensor based on the fracture of a conductive material can be attempted to be explained by the theory of percolation network.

According to the percolation network theory, it is assumed that the electrical resistance of graphene is uniformly distributed, and the average resistance of the graphene fragmentations is a constant value. Then, the number of fragmentations between the electrodes and the number of conductive paths become factors of this relationship. The total resistance can then be described by\textsuperscript{[21]}

\[
R = \frac{(L-1)R_m + LR_c}{S} \approx \frac{L(R_m + R_c)}{S} \tag{3}
\]

Where \( R \) is the total resistance of the sample, \( R_m \) the resistance between two adjacent platelets, \( R_c \) the resistance across one platelet, \( S \) the number of conductive paths, and \( L \) the number of platelets forming one conducting path.
As the graphene film cracks during the stretching process, the number of conductive paths will change, assuming that the relationship between $S$ and strain $\varepsilon$ is Eq.4\textsuperscript{[22]}

$$S_\text{tot} = S_0 \exp[-(\alpha \varepsilon + \beta \varepsilon^2 + \gamma \varepsilon^3)]$$

(4)

Where $S_0$ is the number of conductive paths at the initial state, $\alpha$, $\beta$, $\gamma$ are parameters to be determined. Take the average value of $L$, $R_m$, $R_c$, and the conductive paths model (Eq.5) can be obtained as

$$\ln \frac{R}{R_0} = \alpha \varepsilon + \beta \varepsilon^2 + \gamma \varepsilon^3$$

(5)

4.2 Fitting results of stretched sensor.

For samples have been stretched, since many crackles have been generated, the change in the number of conductive channels can be considered as one phase during the stretching process. The model fitting results are shown in Fig.5b, which fit the experimental data accurately. The fitting parameters: $\alpha = 49.26$, $\beta = -380.51$, $\gamma = 1551.73$, and the Residual Sum of Squares (RSS) = 0.06746. The fitting line proves that the conductive paths model is able to describe the resistance-strain phenomenon of the stretched thick CVD graphene strain sensor precisely.

![Figure 7](image)

**Figure.** 7 Experimental data and fitting result of the conductive paths model.

5. Conclusions

In this paper, we report a method for preparing a graphene-based flexible sensor by dry-transfer-post-etching process. Using the incompletely cured PDMS has the characteristic of strong viscosity, the complete transfer of the graphene film is realized, and a thick CVD graphene flexible strain sensor is obtained, which has a range up to 10%, GF from 60 to 130, a minimum detectable strain of 0.1%, and good repeatability. The conductive paths model based on the percolation network theory and structure-reconfiguration of graphene under strain is proposed. By fitting the experimental data and observing optical micrographs, the mechanism of the strain-resistance change process of the sample are described accurately. The sensor can be attached on complex structure surface due to its flexible
substrate and adaptable sensitive material. We believe that the low-cost flexible strain sensors prepared in this way have great application prospects in structural health monitoring, human health monitoring, complex structure deformation detection and other field of flexible strain sensing.

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References and Footnotes


