Advanced three-dimensional graphene-based piezoresistive sensors in wearable devices

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Abstract. The piezoresistive pressure sensor made with a three-dimensional (3D) structure as the core material is considered a potential precision sensor for wearable devices because of its unique advantages of simple preparation principle, low power consumption, stable performance and high signal acquisition efficiency. It has attracted the attention of many scientific researchers who are committed to realizing the high intelligence and informatization of personal devices. After the rapid development in the 21st century, this technology has made many breakthroughs and showed strong application potential in high-tech fields such as human motion detection, health monitoring and electronic skin. However, this technology is still a long way from full commercial mass production. This paper introduces the unique advantages of the 3D graphene-based piezoresistive sensor in intelligent wearable devices, summarizes several production methods and applications of this 3D graphene-based piezoresistive sensor, explores the future development trend and application prospect of this technology, and discusses the challenges and prospects of piezoresistive pressure sensor based on three-dimensional graphene.

1. Introduction

With the development of science and technology and the continuous progress of information technology, modern society puts forward higher and higher requirements for the intelligence and informatization of personal equipment[1]. At present, flexible wearables such as coats, hats, gloves and headphones are developing in intelligence and integration. At the same time, more consumers require that they be connected with information devices such as mobile phones and computers so that wearables can have more functions to make life more comprehensive or meet special needs. However, the materials for manufacturing such equipment shall have special properties such as high capacitance, high sensitivity, high comfort, high safety and high scalability. Therefore, flexible wearable intelligent devices have become a research hotspot in portable energy, sensors, artificial intelligence, and the human body. This research has developed rapidly in the 21st century. Piezoresistive sensors made of 3D graphene have shown good application prospects in electronic skin, motion recording and health management. Gao[2] et al. studied the flexible wearable sensor to recognize the contour, soft and hard, material, surface temperature and other information of the captured object, which can effectively improve the intelligence level. Graphene is an excellent material for making this kind of intelligent equipment. It has attracted researchers’ attention because of its good flexibility, conductivity, capacity, strength, environmental

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friendliness and processability. Rajan et al. reviewed the development of graphene composite electrodes for supercapacitors[3]. The piezoresistive sensor has become the first choice for wearable devices because of its simple structure, reliable performance, high sensitivity, fast response, convenient miniaturization and miniaturization, and no external components. A wearable piezoresistive sensor based on three-dimensional graphene is considered a potential flexible sensor because of its simple preparation principle, low power consumption, stable performance and high signal acquisition efficiency. To sum up, exploring the relationship between the structure, performance, production process and various influencing factors of 3D graphene-based piezoresistive sensors has become an important topic of intelligent equipment. With researchers’ exploration, many achievements have been made in this technology, but it still takes time to popularize commercial and cheap production fully.

2. Unique advantages of graphene piezoresistive sensor

2.1. Potential 3D graphene
Graphene is a simple material of carbon. An sp² hybrid orbital connection separates each carbon atom, and these carbon atoms are closely packed to form a two-dimensional honeycomb lattice structure of the monatomic layer, as shown in Figure 1. Because graphene has a unique microstructure, it shows many excellent physical properties. The thickness of single-layer graphene is only 0.335 nm, its light transmittance is as high as 97.7%, Young's modulus is 1 TPA, and its strength is very high. Because of excellent flexibility and conductivity, graphene is considered ideal for preparing flexible conductive materials[4].

Figure 1. Structure of graphene

The preparation methods of graphene can be roughly divided into two main methods: top-down (TD) and bottom-up (BU). TD manufacturing usually starts with large materials and obtains smaller products that meet the requirements by removing unnecessary materials. BU starts from a smaller unit and grows into a larger functional structure. How to choose a suitable method to prepare 3D graphene is an important problem. BU is the main exploration direction at present. It has the advantages of good product quality and controllable size, but it is prohibitive because of its high cost and complex transfer process. The goal of scientists is to achieve low-cost, large-scale and high-quality graphene production. Jing Zhang et al. [5] explored a method for preparing high-yield graphene (GRZ) by ball milling and liquid phase stripping (LPE), which is of great significance for the commercialization of graphene.

2.2. 3D piezoresistive sensor
According to the research of Lu et al., the piezoresistive pressure sensor is mainly composed of three parts: active material, flexible substrate and conductive electrode. The active material is made of various conductive materials, which is also the main part of the sensor. Therefore, graphene can be used as active material in the sensor. However, as a pressure sensor, the component structure often bears large stress, which puts forward higher requirements for the mechanical properties of materials. The deformation capacity of a two-dimensional (2D) structure is far less than that of a 3D structure, and the
two-dimensional pressure sensor cannot work well under a large deformation rate. Therefore, 3D structural pressure sensors with high compressibility and flexibility have become more popular. Sponge and foam are two common 3D flexible substrates[6].

2.2.1. Working principle of the piezoresistive sensor. A piezoresistive sensor is an electronic component that converts the pressure on the equipment into an electrical resistance signal. Compared with other pressure sensors (such as capacitive and piezoelectric sensors), the piezoresistive sensor has the advantages of simple structure, small volume and low power consumption. It is very suitable for wearable devices, so it has attracted the attention of researchers.

The following equation can explain the working principle of piezoresistive sensor:

\[ R = R_e + R_a \]  

For a given sensor, \( R_e \) is a constant, and the change of \( R_a \) is the main source of electrical signal change. Since the resistance \( R \) of the bulk material can be described as the following equation:

\[ R = \rho \frac{L}{A} \]  

where \( \rho \) is the material's resistivity, \( L \) and \( A \) are the length and area, respectively[7].

Figure 2 shows the basic structure diagram and current working principle of the semiconductor piezoresistive sensor.

![Figure 2. Semiconductor-piezoresistive-pressure-sensor section (left) and circuit schematic (right)](image)

2.2.2. Unique advantages of piezoresistive sensor based on graphene

2.2.2.1. High sensitivity. According to the above formula, it can be seen that the lower the modulus of the material, the more sensitive the sensor is because a greater degree of deformation can be carried out in the low modulus material under a given pressure. Sensitivity is defined as the slope of the electrical signal pressure curve. The electrical signal includes capacitance, resistance, current or voltage signal, which generally measures the current strength. Even if the applied pressure is very low for high sensitivity sensors, it can also feedback to obvious electrical signals. Although 2D graphene has a high tensile modulus, 3D graphene structure can well meet this condition. The piezoresistive sensor prepared from 3D graphene material with porous structure has the significant advantages of low density and high sensitivity.

2.2.2.2. Large detection range. The sensing range of most high-sensitivity sensors is very small. For the existing technical conditions, how to balance the sensitivity and sensing range is still an important topic. Graphene has excellent conductivity so that its electrical signal will not decay too much when conducted in a large range. Therefore, graphene is an excellent sensor active material, especially when graphene is made into a sensor with a layered microstructure. This advantage will be more obvious if the sensor array[8].

2.2.2.3. Performance stability. Graphene with a 3D structure has excellent elastic recovery ability. Because the piezoresistive sensor is often extruded and deformed, the material inevitably accumulates
plastic deformation. After a while, the accuracy of the sensor will be significantly reduced. 3D graphene-based piezoresistive sensor can significantly improve this defect.

2.2.2.4. Safety and environmental protection. Due to the excellent piezoresistive properties of semiconductor materials, most piezoresistive sensors take semiconductor materials such as silicon as the core. Some semiconductor materials are toxic. Similarly, the production of various semiconductor compounds may also cause environmental pollution. In contrast, graphene is a simple carbon substance with no harmful chemical properties, a higher affinity with organisms, and higher stability suitable for wearable devices.

3. Graphene-based pressure resistance sensor preparation method and application

3.1. Selection of materials
The choice of sensor material has an important impact on its performance. Although traditional strain sensors have been widely used, they also have inherent limitations, such as poor scalability[9, 10], one-way sensing[11]. With the development of polymer nanocomposites, a new stretchable strain sensor based on their preparation is expected to solve the above limitations. Graphene nanochips (GNP) have good conductivity and high Yang's modulus. When the substrate is stressed, the material is not easily damaged so that a stable conductive network can be formed. The combination of conductive fillers of different dimensions (e.g., carbon nanofibers and graphene nanochips) can significantly improve sensor performance, sensitivity and effective linearity. However, the disadvantages are also obvious; in the preparation process, conductive fillers are not easy to disperse, the need for surface modification treatment with dispersants.

3.2. Synthesis process
In the development of stretchable strain sensors, the general research of nanocarbon material and two-dimensional graphene material has made a significant change in the performance of strain sensors[12]. As shown in Figure 3, the following image shows the growing use of nanocarbon materials and graphene in sensors over the past decade.

![Figure 3. Trends in graphene-based strain sensors[13].](image-url)
Sensor design mainly involves two aspects of choice, on the one hand, is the selection of materials, conductive fillers are commonly used nanocarbon materials and two-dimensional graphene, they have good conductivity, can build different dimensions of material joining (such as carbon nanofibers and graphene nanofibers); On the other hand, the selection of material preparation process, can improve the sensor sensing performance by designing different structures. The preparation methods commonly used in stretchable compound strain sensors are mainly divided into the following (Figure 3): liquid-phase mixing method, chemical vapor deposition method, a coating method, filtration method, a rotary coating method, weaving method, die molding, electrostatic assembly method, etc. The preparation methods for several typical materials are described below.

3.2.1. Liquid phase mixing method. Sensors with high tensile properties are obtained by mixing conductive nanomaterials with elastomers in the liquid phase and then covering them with polymer nanocomposites as substrate materials. This preparation method effectively solves the problem of uneven dispersion in the material mixing process and can form a more stable structure (Figure 4a).

3.2.2. Chemical vapor deposition method. Surface modification of conductive filler graphene to improve the interface compatibility between the substrate and conductive filler can improve the dispersion of conductive fillers and better build a conductive network.

3.2.3. Printing technology. With printing technology to print conductive inks onto existing elastomers, this method solves the problem of uneven solution dispersion during dispersion (Figure 4f).

3.2.4. Filtering method. Filtering elastomers into the conductive structure of nanomaterials to form resistive strain sensors or capacitive strain sensors can significantly improve the strain tensile range of the sensor, improve sensitivity and stability, and greatly help improve performance.

3.2.5. Electrostatic spinning technology. The formulated polymer solution or melt is sprayed in a strong electric field and then covered with conductive fillers, with the advantage of a uniformly mixed binding with good tensile properties (Figure 4b).

3.2.6. Weaving method. By modifying the fibers to make them easy to process, they are prepared into the desired structure, and then a highly sensitive sensing structure is obtained by adding conductive fillers (e.g., surface silver plating, internal implantation of conductive fibers, etc.) (Figure 4c).

3.2.7. Press molding. Mix the polymer with the conductive filler, then heat to remove excess moisture, pressurized-molding-release. The advantages are simple operation, sample preparation cycle is short, but there are some problems, such as uneven distribution conductive fillers, internal defects, etc. (Figure 4d).

3.2.8. Electrostatic assembly method. Flexible and high-performance strain sensors can be created by simply self-assembling carbon nanoparticles layer by layer on polymers such as polyurethane sponges. The alternating assembly of carbon nanotubes (CNT) and graphene nanochips (GNPs) helps build a complete conductive network. Because the synergies between CNT and GNP significantly improve sensor sensing performance, sensor composites with a greater strain detection range and higher linearity are obtained (Figure 4e).

The following diagram summarizes some of the methods used to make sensors:
Fabrication of organic-inorganic nanocomposite strain sensors: (a) schematic diagram of a cellulose/RGO strain sensor made by mixing GO with multi-carbon nanotubes[14]; (b) coating carbon nanotubes with polyurethane through electrostatic spinning, followed by polydimethylsiloxane (PDMS) modification to obtain a 500% high-strain sensor[15]; (c) electroless silver-plated sensor based on cotton/spandex blended fabric, resulting in high sensitivity with measured gauge factor up to 26.11[16]; (d) schematic diagram of a multi-carbon nanotube (MWCNTs)/silicon fluid (SF)/PDMS sensor prepared by compression molding method[17]; (e) diagram to illustrate the structure of the strain sensor assembled from carbon nano/ polyurethane foam; (f) MWCNTs/PDMS composite films prepared by extrusion[18].

More studies are made of graphene as a filling material strain sensor, with relatively high conductivity and stretchability, so attention has been focused on[19-21]. The graphene and PDMS are mixed with a new type of conductive nanomaterial, and by chemical vapor deposition, a graphene fabric is attached to or embedded in PDMS to make a stretchable strain sensor, whose sensing mechanism includes crack generation and expansion evolution[22-24].

3.3. Synthesis process
Stretchable sensors are also widely used in medical devices, human electronic skin implants and so on. Due to the advantages of graphene materials themselves (low Yang's modulus, good biocompatibility and high light transmission), graphene materials are ideal for wearable and implantable biomedical devices compared to conventional metal and semiconductor materials. Because of their unique characteristics, such as ultra-flexibility, wearability (sensors can be woven into clothing due to the toughness of the fiber), minimal intrusiveness (carbon fiber size is generally in microns, and therefore less invasive in the human or biomedical field) and tissue adaptability. With the development of electronic skin, flexible, stretchable and wearable sensors are widely used in human-machine interfaces, soft robots, health monitoring, virtual reality and entertainment technology. Figure 5 shows the specific applications of sensors in human health detection, electronic devices, human-computer interaction, electronic fabrics and other fields.
Figure 5. Emerging applications of tensile strain sensors in wearable physical sensors[25]
4. The graphene-based pressure sensor in wearables

As for 3D graphene wearable pressure-resisting sensors, their applications include software robots, human-machine skin, etc. [26-28]. Among them, human motion detection, health monitoring and electronic skin have attracted the attention of scientists because they could provide the basis for the next generation of artificial intelligence.

4.1. Human motion detection

Graphene-based 3D wearable pressure sensors need to sense both tiny and powerful pressures to detect human movement. It is quite challenging because large movements such as joint bending > 20 kPa, while tiny movements such as speaking or pulsating can be less than 1kpa. The method of constructing a multi-level microstructure has proved to be feasible after many attempts. Recently, Dong et al. synthesized a hybrid 3D rGO/polyaniline sponge (RGPS) structure and used it to detect human motion[29]. Under the synergy of rGO and PANI, an ultra-soft flexible sensor based on RGPS is obtained. As shown in Figure 6, the sensor shows high sensitivity to detect speech, swallowing, opening, blowing, crying, breathing, and squatting. The tunable sensitivity from 0.042 to 0.152 kPa\(^{-1}\), the broad working range (0–27 kPa), the fast response time (96 ms) and the excellent durability (> 9,000 cycles) make it ensures the ability in detecting both tiny and big motions. In fact, many literatures have reported on the excellent performance of 3D graphene piezoresistive sensors in detecting human activities, confirming the great advantages of 3D graphene in this field[30-33].

Figure 6. (a-b) Photographs of the twisted and folded RGPS. (c) Current signal when speaking "I AM". Current response upon (d) swallowing, (e) mouth-opening, (f) blowing, (g) crying, (h) breathing and knee squatting–arising process. (Reprinted with permission from References[29]).

4.2. Health monitoring

3D graphene sensors can be used for human health detection after real-time monitoring of human movements. However, the exploration of a new non-destructive detection technique to monitor human health has been the pursuit of researchers[34, 35]. Obviously, this is not easy because pulse and blood vessels are hidden under the skin, and the signal is so small that it is not easy to detect, as shown in Fig.
12a. To this end, Chen et al. reported a new approach to fabricate the graphene-based pressure sensor with 3D microstructures in 2019[36]. As shown in Fig. 12b, the environmentally friendly solution and self-assembly methods were used to obtain the ultra-large interracially self-assembled graphene (ISG) film. The 3D sensor is established after the ISG film is transferred to the micro mode 3DMS elastomer and then annealed, providing excellent performance for high sensitivity in detecting pulses and wrist bending. As is clearly shown in Figure 7c-f. This work can be seen as a breakthrough in this field, with graphene-based pressure sensors with an unprecedented high-tuning sensitivity of 1.04 to 1875.5 kPa⁻¹, with a wide detection range of 1 to 40 kPa.

![Figure 7. (a) Schematic of detecting arterial pulse on the skin surface. (b) Schematic of the manufacturing process of 3D PDMS-Gr. (c-d) 3D PDMS-Gr-based sensors are used for pulse and blood pressure detection. (e) The pulse signals recorded under different applied pressure. (The details of the waveform are shown in i and ii. The statistics of the current change under different voltage is shown in iii.) (f) A real-time pulse signal measured during the wrist bending process. (Reprinted with permission from References[36]).](image)

4.3. Electronic skin

Natural skin integration with flexibility, high sensitivity, rapid response, good self-healing and so on inspired researchers to explore skin-like sensors[37, 38]. Self-healing, which can efficiently repair itself after mechanical injuries[39], is pivotal when fabricating electronic skin. Liu et al. synthesized a bionic tactile proanthocyanin/reduced graphene oxide/polyvinyl alcohol (PC/rGO/PVA) hydrogel with the
ingenious using of PC/rGO composite and PVA-borax hydrogel[40]. The PC/rGO/PVA hydrogel-based electronic skin in Figure 8a has good conductivity. In this composite hydrogel, PVA-borax is responsible for improving biocompatibility and provides self-healing function. The addition of borax makes up for the poor elasticity of PVA[41]. As Figure 8b-c further shows, the key structure on the real skin can be found in the hydrogel, which shows good compatibility with the finger skin. In the synergy between PC/rGO and PVA-borax, mixed hydrogels show the ability to squeeze and stretch, such as sensing skin epidermis, mood changes such as happiness and sadness, accompanied by speech recognition. Applying electronic skin to the fingers of prosthetics can give the soft robot important skin functions and provide new ideas for the expansion of artificial intelligence. This suggests broad prospects in the field of wearable electronic skin and bionics.

Figure 8. (a) Schematic diagram of the preparation process of the PC/rGO/PVA hydrogel. (b) Schematic diagram of the nerve-like PC/rGO and the construction details of each part. (c) Schematic and digital photos of the ultra-compliance electronic skin. (d-f) The as-prepared PC/rGO/PVA hydrogel is used to control a smartphone. (Reprinted with permission from References[40]).

5. Conclusions
This paper mainly summarizes the research progress of graphene-based pressure-resisting strain sensors and deepens and expands the analysis around the material selection, synthesis process and structural design, performance improvement, and specific sensor application. At present, relevant researchers have carried out a lot of fruitful research work on the structural design, material selection and performance optimization of sensors, and summarized and prospected the flexible nanocomposite sensors by reviewing the latest research progress in this field:
(1) Graphene-based sensors have more advantages in aeronautical component health monitoring than traditional metals and semiconductor materials. Because of its high conductivity and ductility, the performance of the prepared sensor has been greatly improved.

(2) Graphene-based pressure-resisting strain sensors need to consider the selection of conductive fillers and substrates, structural design, preparation process and use of the environment and other aspects of the needs. In addition, the material, structure, and connection status of the internal conductive network of the sensor have a very important influence on the sensor's sensing performance.

(3) Graphene-based sensors in the human body motion detection, health detection, electronic skin have broad prospects for development, but there are many problems to be overcome. Signal-to-noise ratio (SNR) and selectivity are important issues that cannot be ignored when prepared 3D graphene sensors are used to detect human health. The production of large graphene-based 3D wearable sensors requires both low cost and low power consumption. Integrating the advantages of high sensitivity, wide detection range, excellent durability and low detection limits is also a challenge for wearable sensors.

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