

Graphene Composite Based Flexible Sensors for Wearable Applications

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Abstract:

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In recent years, flexible wearable sensors have received widespread attention for their potential application value in motion monitoring, healthcare and human-computer interaction. Graphene, as a two-dimensional zero-bandgap semi-metallic material, is an ideal material for the preparation of wearable flexible sensors due to its high carrier mobility, mechanical flexibility, and biocompatibility. Based on target species and performance requirements, researchers utilize graphene and its redox products to form carbon nanocomposites by combining them with polymers, metals and other substrates through specific preparation methods. This helps to increase the number of active sites and functional groups of modified graphene, achieving dispersion and functionalization of graphene. Such materials can effectively solve the current problems of low sensitivity and poor sustainability faced by pure graphene flexible sensors in the wearable field and promote the maturity of next-generation flexible electronic products in diversified directions. This review starts from the types and classic preparation methods of graphene composites. Then it elaborates the principles and applications of existing pressure/strain, biological, and humidity graphene composites based wearable flexible sensors. Finally, the paper comprehensively summaries the problems and challenges at the present stage and proposes the future development trends.

Keywords: Graphene composite, flexible sensor, wearable

1. Introduction

A sensor is an electronic device that recognises target signals and converts them into an electrical or other forms of signal output, which helps to facilitate mes-

sage processing at a terminal. With the innovation of the Internet of Things (IoT), machine learning and other technologies and people's rising demand for functionality, comfort and reliability of sensors in healthcare, the market has put forward requirements

for wearable sensors ranging from flexibility, lightweight, high sensitivity, low energy consumption, biocompatibility to chemical stability.

Since Gaim et al. transformed graphene from theory to reality by using the mechanical exfoliation method [1], graphene and its derivatives, which have the advantages of large specific surface area, low resistance to electro-thermal conduction, good light transmittance, and strong affinity for materials, have attracted extensive interests from researchers all over the world for application in wearable flexible sensors. Building wearable flexible sensors based on graphene composites can effectively solve the problems of traditional material based sensors, such as poor stability, high manufacturing cost, weak permeability, and poor flexibility. As a result, it has great development potential in the field of human physiological signal detection. Liang et al. [2] showed that the application of graphene-polymer composites can significantly enhance the self-healing and mechanical properties of flexible sensors. Ren et al. [3] have successfully developed a graphene-based wearable artificial throat, which overcomes problems including the insensitivity of traditional artificial vocal cords. Yang et al. [4] developed a flexible strain sensor based on graphene oxide reinforced nanocomposite (NC) hydrogel, which is capable of sensitively detecting all kinds of minute physiological activity in extreme environments. Xing et al. [5] produced a flexible sensor that employs laser-induced graphene (LIG) technique and succeeded in precisely and quickly recording the physiological state of the pilot by it during the experiment.

This paper reviews the main mechanisms and latest applications of graphene-based wearable flexible sensors from the basic properties of graphene composites. Firstly, the types of graphene composites and their classic preparation methods are introduced. Subsequently, the major theory and achievements of graphene composite based flexible sensors in pressure/strain, biology, and humidity fields will be summarized. Finally, combined with the current wearable composite material and sensors breakthroughs and shortcomings, this paper concludes in detail the open issues and challenges of wearable graphene composite flexible sensors, and comprehensively look forward to the development direction of graphene composite flexible sensors in wearable devices.

2. Classification and Preparation of Graphene Composites

2.1 Types of Graphene Composites

Graphene composites can be broadly summarised into

three categories, graphene polymer composites, graphene inorganic nanocomposites, and metal-based graphene composites. In graphene polymer composites, graphene is uniformly dispersed in the polymer substrate's interstices, thereby improving the agglomeration of graphene and mitigating the drawbacks of traditional polymer materials, such as poor electrical conductivity, weak heat resistance, and low tensile strength. Graphene-based inorganic nanocomposites utilise inorganic nanoparticles to modify graphene sheets or use graphene to encapsulate inorganic nanoparticles, which enable both reduction of internal interference and synergistic effects. Metal-based graphene composites are able to enhance the material's friction resistance, corrosion resistance, etc., performance through growing graphene on the metal surface. These advantages allow them to have greater application potential in various fields including aerospace.

In addition, one-dimensional graphene fibres can be used instead of graphene layer to synthesize graphene composite fibre materials with the specific substrates by domain-limited hydrothermal methods. Because of enhanced interfacial forces or changes in fibre structure, this new material has high tensile strength and toughness while ensuring high electrical conductivity. Similarly, three-dimensional graphene aerogel composites are also highly prized for their low density, high porosity, and malleability. All of the above materials regard graphene as an ideal reinforcing phase or synergistic phase, resulting in targeted enhancement of the mechanical, electrical, and chemical properties of the composites.

2.2 Typical Preparation Methods for Graphene Composites

2.2.1 Chemical vapor deposition method

Chemical Vapor Deposition method (CVD) refers to the decomposition of carbon source precursor through high temperature and the deposition of graphene on the surface of the substrate material directly in a protective gas (e.g., argon) atmosphere. This method is commonly used in the preparation of metal-based graphene composites and graphene composite fibre materials. The carbon source precursor can be a gaseous hydrocarbon, liquid, or solid carbon source, while the substrate material is generally selected from metals, ceramics, and nanodevices.

The CVD method consists of four main stages, warming, substrate heat treatment, graphene growth and cooling. Main process of the CVD is settling carbon atoms generated by high temperature cracking to the surface of the substrate through the carrier gas at first. Then apply temperature variations to lead substrate material molecules to form condensation nuclei on the carbon atom aggregation

group's surface. Finally, collect films formed in the continuous growth and degradation of the nucleus.

The CVD method is suitable for the preparation of large-area, high-quality graphene films. Elements such as graphene composites domain size, morphology, defects, number of layers can be adjusted by controlling the growth conditions, which is conducive to meet different environments and purposes of sensors. Furthermore, the CVD technique can be used to grow graphene on a specific substrate, while it can effectively enhance the physical and chemical properties of the composite material itself. The disadvantages of the CVD are demanding preparation environment and complex process, eliminating the possibility of its large-scale industrial production.

Bai et al. [6] used the CVD method to grow graphene in situ on the surface of copper foam. By this method, graphene is uniformly dispersed in the copper matrix, and the interfacial bonding between graphene and copper matrix is enhanced. By analysing Raman plots of the composites after 15 minutes of deposition, the researchers confirmed that the CVD method yielded large, monolayer, ordered graphene films on the surface of copper foam. Liu et al. [7] used the CVD method to deposit graphene films directly on inner aperture walls of photonic crystal fibre (PCF), and achieved in-situ full-coverage growth of graphene films with different thicknesses and homogeneities on the PCF outer surface and inner aperture wall. Ouyang et al. [8] came to the conclusion that the CVD method could further improve the corrosion resistance of the composites by effectively resolving the issues of poor wettability and easy agglomeration between graphene and metal during the synthesis of metal-based graphene composites.

2.2.2 In-situ polymerisation method

To address the challenges of graphene polymer composites in terms of dispersion of the reinforcement as well as interfacial bonding, the popular approach is to introduce graphene into the polymer matrix to form strong interfacial interactions at the expense of cohesive interactions between graphene sheets. This class of methods are mainly divided into in-situ polymerisation, melt blending and solution blending method.

In the in-situ polymerisation method, first of all, graphene or modified graphene is blended with monomers or pre-polymers, with appropriate amount of initiator added. Next, the polymerisation is initiated by intense heat or radiation, and variables (e.g., pressure, temperature) are adjusted to control the length and quantity of polymer chains form on the surface of the carbon material radicals. Meanwhile, generated polymer chains can effectively prevent graphene films from aggregating.

The in-situ polymerisation method is characterised by the ability to link graphene with monomer polymers or oligomers via covalent bonds, showing a distinct layer structure under scanning electron microscopy. Compared to the non-covalent modification such as the $\pi-\pi$ conjugation and the weak interaction force of non-chemical bonding, covalent modification enables graphene to form a unitary network with the polymer. This structure ensures the filler particles are uniformly dispersed in the polymer matrix [9]. Hence, the nanocomposites prepared by the in-situ polymerisation method have large internal interaction forces, enhancing the mechanical flexibility significantly, which is conducive to stress transfer. Although the approach is capable of improving the performance including the carrier mobility of composites, problems such as the impact of functionalisation and the difficulties of processing cannot be avoided.

Malang et al. [10] compared the in situ polymerization method with the conventional solution polymerization method during the preparation of graphene/polyimide (PI) composites. The researchers found that the in-situ polymerisation method can effectively inhibit the agglomeration of oxygen-containing functional groups and make the surface of the composites smoother. Therefore, the mechanical and electrical properties of the materials were effectively enhanced. It was experimentally determined that when the content of reduced graphene oxide (rGO) was 1.0 wt%, the tensile modulus increased by 68.8% to 132.5 MPa compared with that of pure PI. If the rGO content is raised to 3.0 wt%, the electrical conductivity is increased by 8 orders of magnitude compared with pure PI. Wang et al. [11] synthesised uniformly dispersed polypyrrole/graphene composites and polybenzimidazole graphene composites by in-situ polymerisation. The scholars demonstrated that in-situ polymerisation made graphene/polymer composites as lubrication additives revealed good friction reduction and anti-wear effects by controlled experiments with a four-ball friction tester.

3. Application Analysis

3.1 Flexible Pressure/Strain Sensor

Different human body parts experience different levels of pressure, such as skin stretching, voice pressure (generally less than 1 Pa), eye pressure (generally less than 10 kPa), and jugular venous pressure (generally less than 100 kPa) [12]. Therefore, accurate recognition of different pressures generated by different physiological activities will undoubtedly have broader applications in the fields of electronic skin devices, human movement health monitoring, and expression recognition. Wearable flexible pres-

sure/strain sensors are typically attached to the surface of human bodies to measure pressure or deformation, and convert it into a corresponding electrical signal for transmission. Yet the graphene composites flexible pressure/strain sensors have the advantages of large dynamic piezoelectric range of detection, low detection limits, strong stretchability, small size and mass, and good biophilicity, compared with traditional wearable sensors.

Graphene composites based flexible pressure/strain sensors are usually classified into capacitive, piezoelectric, piezoresistive, friction electric and other types according to different principles. Different types of pressure/strain sensors are suitable for different detection environments. For example, capacitive pressure/strain sensors are capable of detecting small static forces, while piezoelectric pressure/strain sensors are widely used to monitor dynamic mechanical changes in real time. Among them, the flexible piezoresistive type sensor has the characteristics of technological simplicity, low cost, fast response, good sensitivity, etc., which has a promising development prospect and high exploitation potential.

Current graphene composite piezoresistive flexible pressure/strain sensor mechanisms are categorised into four primary points.

Firstly, because of external pressure or deformation, the graphene lattice structure is distorted, and the electron transport efficiency near the energy band structure and the Fermi energy level is changed according to the piezoresistive effect. As a result, the average hole rate is reduced, which lowers the conductivity.

Secondly, the general effective media (GEM) theory is used to explain the relationship between the resistance of the composite material and the conductive filler material, which is given in the following equation [13].

$$(1-\phi)\left(\frac{\frac{1}{\sigma_i^\omega}-\frac{1}{\sigma_m^\omega}}{\frac{1}{\sigma_i^\omega}+\frac{1-\phi}{\phi_c}\frac{1}{\sigma_m^\omega}}\right)+\frac{\phi\left(\frac{1}{\sigma_h^\omega}-\frac{1}{\sigma_m^\omega}\right)}{\frac{1}{\sigma_h^\omega}+\frac{1-\phi}{\phi_c}\frac{1}{\sigma_m^\omega}}=0 \quad (1)$$

Where the conductive filler material's volume concentration is presented by ϕ . The seepage threshold is indicated by ϕ_c . The morphological parameter of the composite is denoted by ω . The conductivity of the composite material with an unformed conductive network is represented by σ_i . The electrical conductivity of the conductive filler material at ϕ is indicated by σ_m . And the conductivity at which the saturated formed conductive network occurs is indicated by σ_h [12].

When the conductive filler material in graphene composites is in the seepage threshold, the pressure of the exter-

nal environment will cause the volume of the conductive filler material to expand and the distance of the conductive particles to decrease. The changes in turn will make the conductive network transformed from a stable stage to a saturated stage, and the conductivity will be increased.

Thirdly, if the concentrations of conductive filler materials are at lower stage, based on the tunnelling effects, thermal vibrations generated by pressure or deformation can excite electrons to cross the potential barriers of thinner insulating polymer layers. This causes the formation of electric currents, which in turn increases conductivity.

Fourthly, the deformation of sensor's own material leads to fracture of the flexible matrix with lower Young's modulus and higher elongation. Expansion of internal cracks in conductive materials results in reduction of the contact area, disruption of the sensor's electronic pathway, and decline in the conductivity [14].

However, graphene composites flexible piezoresistive pressure/strain sensors all have certain measurement limits, because excessive pressure and strain can destroy the graphene hexagonal structure and cause irreversible plastic changes. Overall, there is a current trend towards e-fabrics [15], three-dimensional architectures, and hydrophobic properties for graphene composite flexible pressure/strain sensors.

Kaidarova et al. [16] patterned conductive porous graphene electrodes based on laser-induced graphene technology (LIG), using a CO_2 infrared laser acting directly on a $125 \mu m$ commercial polyimide (PI) film. Protective polymethylmethacrylate (PMMA) coating was applied over the LIG graphene as well. The average linear sensitivity of the LIG flexible pressure sensor was determined to be 1.23×10^{-3} kPa, with a standard deviation of $\sigma \pm 0.005 \times 10^{-3}$ kPa. Furthermore, the LIG sensor maintained excellent stability in 15,000 times load cycle experiments ($>100h$). The researchers also discovered that animal cells maintained high cell viability ($>90\%$) after 24h of exposure to the LIG sensor by using the coulometric assay (alamarBlue cell viability assay) and confocal microscopy. The result proves that the sensor has good biocompatibility. These advantages have been shown to help graphene hold great potential in wearable areas such as pulse detection and gait analysis.

Cao et al. [17] prepared vertical graphene-polydimethylsiloxane (VG-PDMS) flexible pressure sensors by plasma-enhanced chemical vapour deposition method (PECVD), which has the benefits of catalyst-free and transfer-free. As depicted in Fig. 1, copper inter-finger electrodes were plated on standard-size flexible mica paper. After full coverage of VG growth was achieved in the substrate above, dual-layer PDMS packaging was carried

out. The sensor has outstanding high-temperature resistance and bending flexibility, whilest achieving high sensitivity (4.84 kPa^{-1}) and an ultra-wide pressure monitoring range (0~120 kPa), which gives direction to the development of flexible electronic skin in the future.

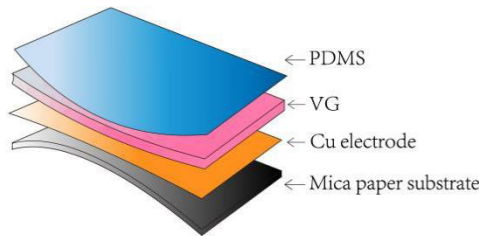


Fig. 1 Structure of flexible vertical graphene based pressure sensor [17]

3.2 Flexible Biosensors

Flexible biosensors can convert biological signals into electrical signals for transmission and expression. The principle of the sensors is split into two main kinds. One is to directly regulate the charge channel on the contact surface with the biological analyte. The other is to indirectly use the grid as an electrostatic gating to amplify the electrical effect of source leakage current, which holds higher sensitivity than the former. This type of sensors usually detects objects including enzyme reactions, immune reactions, and molecular recognition. They are usually highly flexible and biocompatible, which allows them to perform real-time monitoring without damaging biological tissue [18].

The breathability and stability of traditional flexible biosensors show deficiencies in complex and extreme scenarios [19]. Whereas graphene composites based wearable bioelectronics is superior in portability and constancy due to the features of graphene, such as two-dimensional structure, non-toxicity, and compatibility with solid supports. Additionally, these sensors are capable of labelling-free detection. Thus, they have huge potential for applications in real-time human monitoring, digital health and AI medicine.

Wang et al. [20] created a type of copper nanowires (CuNW) based reduced graphene oxide cotton fabric (CF) composite for flexible non-enzymatic glucose sensors. The experimenters verified the short response time and high sensitivity of the sensor by cyclic voltammetry. In addition, the CuNW/rGO/CF sensor demonstrated excellent selectivity, reproducibility and stability in controlled experiments. These benefits showed reliable sensing performance in detecting glucose levels in human sweat and serum.

Wei et al. [21] designed a protein GB1 mutant with two amino acids mutated in the α helix structure and combined it with graphene material. It has been shown by Molecular Dynamics Simulations that experimenters can preserve the protein conformation and control its orientation on the graphene surface by adjusting small changes in the protein sequence. This provides new ideas for the wider application of graphene protein composite based flexible biosensors in human body.

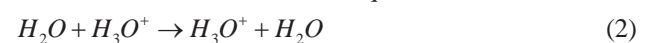
Giwan Seo et al. [22] developed a graphene transistor-based COVID-19 field effect transistor (FET) flexible biosensor. It can detect spiny proteins of SARS-CoV-2 virus with a limit of detection (LOD) of 1 fg/mL. The researchers utilized this sensor to detect antigenic proteins of the SARS-CoV-2 virus in nasopharyngeal swabs and clinical samples. The result shows that the new sensor is responsive, accurate and does not require pre-processing at low viral concentrations, confirming the potential for clinical wearable application.

3.3 Flexible Humidity Sensors

Flexible humidity sensors use variations in physical or chemical characteristics of the humidity-sensitive material, such as capacitance, resistance, and spectral properties, to determine the humidity of the surrounding air or breath. The sensor is suitable for a variety of applications such as human respiratory rate monitoring, non-contact sensing, and package opening status detection [23]. Although humidity sensors based on active materials such as aluminium trioxide have achieved mass production, the rigidity of these materials limits the development of wearable humidity sensors [2].

At first, graphene was considered to be a sensitive material for next-generation humidity sensors due to its low cost and ease of fabrication. Recently, with the purpose of enhancing the sensitivity and comfort of humidity sensors, graphene oxide (GO) and reduced graphene oxide (rGO) have been commonly used to replace graphene. These graphene derivatives are rich in hydrophilic groups and cavities, demonstrating strong hydrophilicity.

The mechanisms of composites based on these two materials for humidity sensors are generally recognized in two ways. Under low humidity conditions, GO and rGO absorb water molecules into cavities through hydrophilic groups on the surface, reducing positive charge carriers and increasing resistance. As for high humidity conditions, however, according to the chain reaction principle of the Grottuss mechanism, the equation is as follows.



Water molecules are continuously ionised on the electrode surface, releasing a large number of protons. These pro-

tons change the dielectric constant of the original conducting material, thus reducing the resistance in the end.

Zhang et al. [24] developed flexible LC humidity sensors based on GO/Mxene (Two-Dimensional Transition Metal Carbides and Nitrides, as in Fig. 2, rich in pleated) composites. The researchers plotted the correlation curves of resonance frequency and intensity through the humidity test platform. Through experimental data, the sensor showed high sensitivity, stability, and response speed within the humidity range of 20%~95% RH, holding potential in human respiration monitoring test.

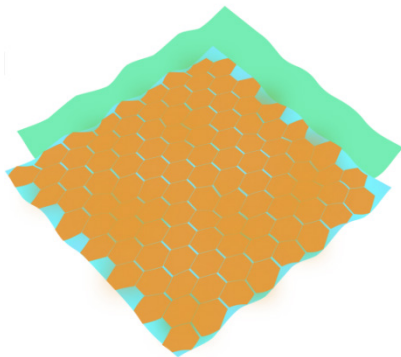


Fig. 2 Schematic structure of Mxene flexible film [25]

4. Challenge and Future

4.1 Overview of Current Issues and Bottlenecks

To begin with, the question about graphene composite based wearable flexible device in the field of materials is divided into two main parts. Firstly, at present, the industrial production process of high-quality graphene materials is complicated and expensive. Thus, the persuasive power in large-scale commercial application is low. Secondly, studies by some relevant scholars have shown that graphene's properties such as oxidative stress, sharp edges, hydrophobicity and adsorption may cause certain damages to biological cells in contact conditions [26]. Consequently, the potential hazards of graphene for long term effects to the human body are unclear.

Afterwards, the issue of energy supply for graphene composites based flexible sensors is a hot topic of academic discussions nowadays. Graphene cannot cycle sufficiently during charging and discharging process and has poor stability. Moreover, current flexible batteries cannot provide the power needed for Integrated Sensing Systems. As a result, external battery for flexible sensors is still the mainstay of the market. However, the shape of traditional rigid batteries is not conducive to the development of flexible sensors in the direction of miniature compatibili-

ty. The use of rigid batteries also introduces the soft-hard interface instability that makes it unsafe to operate as a non-stand-alone system. Commercial batteries with high power, high capacity, cycle stability, and good stretchability are desperately needed.

Furthermore, solving bottlenecks in wireless communication for sensors is imminent. Flexible sensors often rely on external Bluetooth modules for wireless communication with other devices, which lacks convenience. While radio frequency (RF) wireless antenna technology has reached a certain level of success in wearable sensors [27], the problems of poor signal accuracy, prominent crosstalk effects of different stimuli, strong interference from the external environment, and significant signal drift effects remain severe. Current graphene-based flexible sensors are technically challenged to integrate with traditional hard interface circuits as well.

In addition, there are numerous issues in other areas that need to be taken into account and resolved. The development of multifunctional integrated sensors is slow. The permeability of composite materials needs to be improved. The smart IoT technology is short of arithmetic support. Graphene composites and wearable flexible sensors lack unified measurement standards.

4.2 Exploration of Solutions and Development Directions

Aiming at the corresponding problems, the author puts forward a series of suggestions that can be referred to by relevant researchers.

In terms of difficulties from material basic properties, firstly, in order to lower the cost of raw materials, it is advised to combine the practical challenges of large-scale production with laboratory preparation methods, and optimize the structural design and dispersion process of graphene composites. Secondly, to address the potential health threatening of carbon nanomaterials in wearables, researchers may consider employing protective layer encapsulation to avoid direct contact. Nonetheless, the protective material needs to ensure its own permeability and stability to maintain the normal operation of the sensor.

Additionally, in face of the challenges of providing and storing electricity, scientists can explore the feasibility of energy harvesting for environmental energy such as friction or bioelectricity based on existing self-powered flexible sensors. It may help ensure the sensors themselves are supplied with a stable and sufficient energy, and improve the convenience of wearable use.

Furthermore, in the fields of wireless communications and the IoT, two ideas can provide scientists with inspirations. One way is to try new materials and routes to design an-

tennas, such as graphene-assembled film (GAF) and near-field communication (NFC) technology [28]. The other way is to upgrade sensor electronics and computing technology, making full use of convolutional neural networks, multi-intelligence bodies, and other related technologies to improve the detection and control mechanism. Developing integrated data analysis, processing systems based on quantum algorithms, and breaking through the obstacle of multifunctional flexible sensor signal blending are highly recommended as well.

The study also suggests the relevant scientists fully develop the application scope of multidimensional architecture-modified graphene and derivatives such as rGO and GO based on specific scenario needs. Meanwhile, according to the latest research trends, enhancing the sensitivity and stability of the sensors themselves through, e.g., non-linear synergistic strategies [29], graphene nanoribbons, has a good potential for application.

Moreover, researchers can focus on the development of flexible sensors degradable, self-cleaning, electromagnetic shielding, and other practical functions. These features can effectively resist the interference of external factors, maintain their own equipment status, and protect the environment.

Finally, from a macro point of view, enhancing direct communication and co-operation between cross-disciplines and cross-industries, and improving the mode of sharing talents and resources in the scientific research are very crucial propositions. They deserve the attention of more and more research departments.

5. Conclusion

Graphene and its derivatives have attracted the interest of researchers worldwide for their excellent electrical, thermal, optical, chemical, and mechanical properties. Flexible sensors have been used as core electronic devices for the development of medical and healthcare businesses. With the rapid development of composite modification technology and preparation crafts, graphene composites have been shown to have great advantages, such as diversification and functionalisation in the field of wearable flexible sensors. This review covers the various types of graphene composites and their preparation techniques, as well as the mechanism and applications of graphene-based pressure/strain, biological, and humidity flexible sensors in the wearable field.

It is found that the action mechanism and optimisation ideas for wearable flexible sensors based on graphene composites are well established. These sensors have been shown in specific experiments to be sensitive, flexible, lightweight, rigid, inexpensive, chemically stable, and bio-

philic. Although the development of graphene composites based wearable flexible sensors currently encounters technical barriers in the areas of material preparation, energy supply, wireless communications. Via the use of process optimisation, technology integration, and interdisciplinary field collaboration, the prospects for the development of graphene-based flexible sensors are still immensely promising.

This paper is not comprehensive in terms of preparation methods and applications, and the analysis of the current problems and methodological recommendations are rather general. However, this study has changed the situation of lack of concise, logical, and comprehensively analysed review articles on related topics in academia. This review strongly argues that researches on the application of graphene composites based wearable flexible sensors in various fields is bound to attract more and more attention along with the continuous development of graphene materials and improvement of sensor technologies in the future.

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