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Flexible Electronic Devices Based on Two-Dimensional Material/Polymer Composites

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Abstract

The new generation of electronic devices is incorporating mechanical flexibility into their design, a trend that demands large-area, ultra-thin functional materials that are both energy-efficient and economically viable. The emergence of two-dimensional materials has ushered in a new era of integrated, flexible technology, surpassing the capabilities of traditional semiconductors. Furthermore, 2D materials are highly valuable for advancing a wide range of emerging fields, including wireless health monitoring systems, flexible bioelectronics, and origami-inspired unconventional device architectures, all of which are suitable for challenging applications. This review examines the technological progress and fundamental aspects of 2D material-based flexible devices. This article will systematically summarize the applications of these devices in various areas. Subsequently, this paper focuses on elaborating the applications of such devices from three core perspectives: flexible sensors, flexible displays, and bio-integrated soft electronics. These devices offer promising prospects in fields such as health monitoring, human-machine interaction, and medical diagnosis. Finally, the paper discusses current challenges and their solutions.

Keywords

flexible electronics, two-dimensional material, polymer composites

1. Introduction

Over the past few decades, with the rise of technologies such as the internet, big data, and brain science, humanity has entered the era of intelligence, gradually creating a demand for electronic devices with mechanical flexibility. Consumers not only pursue stronger performance but also increasingly expect devices to be lighter, bendable, and foldable [1]. As a result, flexible electronic devices have gained significant attention in recent years. However, these electronic devices require specific materials and properties, as they must be integrated into various surfaces or folded and rolled to create new forms.

Although traditional silicon-based semiconductor materials exhibit excellent performance, their rigid, brittle nature makes it difficult to meet the requirements of flexible devices. At this point, two-dimensional materials with exceptional electrical, optical, and mechanical properties have become an excellent platform for the development of flexible electronic products. Common two-dimensional materials, such as graphene, are widely used as electrode materials in flexible optoelectronics and wearable sensors due to their outstanding mechanical flexibility, thermal conductivity, and optical transparency. However, standalone two-dimensional materials also have drawbacks, such as insufficient mechanical strength. Combining two-dimensional materials with polymer materials offers the advantages of both: excellent mechanical flexibility and conductive and sensing functionalities [2], thereby strongly promoting the development of next-generation flexible electronic devices.

Flexible electronic devices based on two-dimensional material-polymer composites have demonstrated their potential in various areas, particularly in health monitoring and human-machine interaction. For instance, they can be used to fabricate sensors. Compared to sensors developed with bulky materials, two-dimensional material-based sensors offer multiple advantages. Two-dimensional materials possess excellent flexibility, enabling stable operation even under various deformation processes. They can even conformally contact rough surfaces, such as skin, and the strong adhesion at the interface allows the sensor to attach to the target for extended periods [3]. Furthermore, they can monitor physiological signals such as heart rate, body temperature, and bodily fluids in real time, providing health support and helping drive the development of wearable devices toward lighter, more comfortable designs.

This article aims to review the technological progress and fundamental status of flexible devices based on two-dimensional material-polymer composites, systematically summarize the current state of such devices in fields such as health monitoring and human-machine interaction, and finally analyze the main challenges faced in current research along with future development directions.

2. Introduction to Two-Dimensional Material and Polymer Composite Systems

2.1 Commonly Used Two-Dimensional Materials

Two-dimensional materials are materials confined to atomic-level thickness in one dimension, typically only a few atomic layers or even a single atomic layer. Due to their excellent electrical, optical, and mechanical properties, they have become a research hotspot in the field of flexible electronics. Currently, the commonly used two-dimensional materials include the following:

Graphene is the earliest discovered and most classic two-dimensional material. As a zero-bandgap semimetal, it is characterized by its honeycomb lattice of carbon atoms and exhibits outstanding electron transport, thermal conductivity, mechanical strength, and biocompatibility. Graphene-based flexible wearable sensors have been used to detect various physical, chemical, and physiological parameters, including pressure, temperature, pH, cells, DNA, and proteins, and have been widely applied in flexible electronics and wearable devices [4].

Transition metal dichalcogenides (TMDs), commonly represented by the chemical formula MX_2 (where M denotes a transition metal and X denotes a chalcogen atom), are also a crucial class of two-dimensional materials. Examples include tungsten disulfide (WS_2) and molybdenum disulfide (MoS_2). In contrast to graphene's zero-bandgap nature, TMDs exhibit tunable band gaps. They have wide band gaps exceeding 1 eV and excellent electrical properties, making them highly promising two-dimensional materials for wearable sensors. Among them, atomically thin MoS_2 nanosheets exhibit exceptional flexibility, biocompatibility, and electrical properties, making them among the most promising materials in the TMD family and widely used in physical, chemical, and biological sensors [5].

Transition metal carbides/nitrides (MXenes) are an emerging class of two-dimensional materials [6]. MXenes exhibit excellent electrical conductivity, mechanical flexibility, chemical stability, and good hydrophilicity. Through appropriate surface modification techniques, the performance of MXenes can be significantly enhanced, further expanding their application potential in wearable sensors. As an emerging member of the two-dimensional material family, MXenes have attracted widespread attention for their remarkable electrical conductivity, high pseudocapacitance, robust mechanical properties, and excellent hydrophilicity. These characteristics make them highly promising materials for wearable energy devices.

2.2 Commonly Used Polymer Matrices

Polymer materials possess excellent flexibility, bendability, and mechanical strength. In composite systems, they can significantly enhance the fatigue resistance and mechanical stability of the overall structure. Depending on the requirements, polymers with different characteristics can be selected as the matrix.

PET substrates. PET is a low-cost, recyclable, semicrystalline thermoplastic polymer. It exhibits good toughness, relatively low gas permeability, high visible-light transmittance, low water-vapor permeability, and acceptable chemical resistance. Therefore, it is widely used in flexible solar cells and flexible batteries [7]. However, its poor heat resistance limits its application in high-temperature processes.

PI substrates. PI exhibits excellent chemical stability, mechanical properties, and good heat resistance. It can withstand temperatures up to 400°C and can be used for extended periods in environments above 300°C. However, traditional PI films have a significant drawback: the formation of strong intramolecular and intermolecular charge transfer complexes (CTCs), which causes the films to appear yellow or brown. This severely limits their use in devices that require high optical transparency, such as flexible displays. Therefore, improving the transparency of PI remains a hot topic in current research.

PDMS substrates. PDMS is an elastomer with excellent performance and is currently one of the most commonly used flexible substrate materials. Its elongation can reach 160-180%, and it possesses good thermal stability [7]. Furthermore, PDMS exhibits excellent transparency and biocompatibility. Due to its low Young's modulus, it can withstand significant stretching and bending, making it highly suitable for flexible electronic devices such as wearable sensors.

2.3 Preparation Methods of Composite Materials

Combining two-dimensional materials with polymers can leverage the advantages of both. Currently, the main preparation methods are as follows:

Solution mixing is the most widely used technique for fabricating graphene–polymer composites. The core procedure involves dissolving graphene and the polymer in a suitable solvent, forming a homogeneous blend via ultrasonication or vigorous stirring, and subsequently removing the solvent to yield the final composite material. This approach is straightforward, requires no complex equipment, and is amenable to large-scale industrial production [8].

Melt blending involves mixing two-dimensional materials with the polymer matrix at high temperatures. Uniform dispersion of the fillers within the matrix is achieved through intense mechanical shearing, which also enhances the interfacial interaction between the fillers and the matrix. This method does not require organic solvents, making it more environmentally friendly and cost-effective. However, high temperatures and high shear forces may cause damage to two-dimensional materials.

In-situ polymerization is a method where a polymer monomer solution is mixed with two-dimensional materials, followed by the addition of an appropriate amount of catalyst. The polymerization reaction is then initiated under suitable conditions, including temperature and potential, thereby forming the composite material. This method enables more uniform dispersion of the two-dimensional materials within the matrix, thereby maximizing their modification effect [8].

3. Applications of Flexible Electronic Devices

Flexible electronic devices fabricated on ultra-thin platforms, owing to their shape-adaptive capabilities, demonstrate significant technological potential for smart applications. Two-dimensional material/polymer composites, owing to their excellent flexibility and outstanding electrical, optical, and mechanical properties, exhibit broad application prospects in multiple fields. This chapter introduces these flexible electronic devices by function and application, focusing on three categories: flexible sensors, flexible displays, and biointegrated soft electronics.

3.1 Flexible Sensors

Compared with sensors developed using bulky materials, two-dimensional material-based sensors offer multiple advantages. For example, two-dimensional materials possess exceptional flexibility, enabling stable operation even under various deformations. Furthermore, two-dimensional materials can achieve conformal contact with rough surfaces, such as human skin, and the strong adhesion at the interface enables the sensor to attach to the target for extended periods. Additionally, their transparent and lightweight nature enables comfortable daily use without discomfort [1].

3.1.1 Flexible Gas Sensors

Flexible wearable gas sensors are crucial for real-time monitoring in various fields, including food management, environmental monitoring, healthcare, and safety protection. As layered materials with extremely high effective surface areas, two-dimensional materials offer significant advantages for flexible gas sensing applications. Such sensors can detect harmful gases at room temperature [9].

Two-dimensional materials (including transition-metal dichalcogenides, black phosphorus, graphene, etc.) stand out for their high specific surface area, high carrier mobility, and intrinsic flexibility. The working principle of this type of gas sensor is based on the chemiresistive mechanism. When gas molecules come into contact with the surface of the two-dimensional material, the material's conductivity or resistivity changes as charge transfer is modulated by the gas's electron affinity, altering the material's carrier concentration and, consequently, its resistance. Benefiting from their extremely high specific surface area, even trace amounts of gas molecules can be detected. Various two-dimensional materials, such as graphene and transition-metal dichalcogenides (TMDs), also exhibit good mechanical stability, making them ideal candidates for the development of flexible gas sensors. Therefore, recent progress has enabled their integration into various wearable devices for real-time chemical sensing applications.

Notably, TMD-based sensors can detect not only inorganic gases but also identify volatile organic compounds. This makes TMD-based sensors versatile tools for industrial safety, health applications, and environmental monitoring.

3.1.2 Flexible Photodetectors

Photodetectors are key components that convert optical signals into electrical signals. They have been widely applied in various fields such as imaging, sensing, communication, and environmental monitoring. Two-dimensional materials, with their atomically thin nature, stable surfaces, and layer-tunable optical and electronic properties arising from quantum confinement effects, have emerged as promising candidates for next-generation flexible optoelectronic devices. By employing suitable device architectures, excellent mechanical flexibility can be achieved in two-dimensional material-based devices. The working principle of photodetectors is based on the photoelectric effect, in which electrons are excited by the absorption of photon energy, generating a measurable photocurrent or photovoltage. Among the numerous two-dimensional materials, graphene stands out: its high carrier mobility and broadband light absorption [11] make it highly promising for the development of ultra-broadband flexible photodetectors and an ideal choice for transparent electrodes in applications such as flexible touch panels.

3.2 Flexible Displays

Flexible displays represent another important application direction for flexible electronic technology, encompassing flexible touchscreens, flexible displays, and more. Such devices require high optical transparency from the materials. In the development of flexible displays, organic light-emitting diodes (OLEDs) have emerged as the most suitable candidates due to their advantages, including thinness, light weight, and flexibility. Their characteristics, including no need for a backlight, high brightness, and lower power consumption than traditional liquid crystal displays, have led to their use in foldable OLED displays for smartphones and notebooks by renowned display manufacturers such as Samsung and LG. Flexible touchscreens serve as important interfaces for human-computer interaction, and their core component is the flexible, transparent, conductive electrode.

Different two-dimensional materials play distinct roles in flexible displays due to their varying characteristics:

Graphene-based transparent conductive electrodes were the first system to be extensively studied. Researchers have transferred CVD-grown monolayer graphene onto PET substrates to fabricate flexible transparent conductive electrodes. These electrodes achieve a light transmittance of up to 97.7% and a sheet resistance of approximately 100-300 Ω/sq, and can operate normally under a bending radius of 5 mm. Their advantages include extremely high optical transparency and excellent mechanical strength, making them a preferred choice for transparent electrodes [11].

MXene-based electrochromic devices have garnered widespread attention in recent years. Researchers have utilized the two-dimensional layered structure and excellent metallic conductivity of $Ti_3C_2T_x$ MXene to prepare flexible electrochromic films. These films can undergo a reversible change from transparent to dark blue within seconds upon application of a voltage, with a modulation of light transmittance exceeding 40%. After 1000 bending cycles, the performance shows no significant degradation, enabling fast electrochemical response, making them suitable for electrochromic applications [12].

Transition-metal dichalcogenide-based light-emitting devices demonstrate the potential of two-dimensional materials for luminescence. Researchers have fabricated flexible electroluminescent devices by combining a monolayer of MoS_2 as the light-emitting layer with h-BN as an encapsulation layer. Monolayer MoS_2 has a direct bandgap, with photoluminescence efficiency significantly higher than that of multilayer MoS_2. These devices can operate under bent conditions, although their luminescence efficiency is currently relatively low [13].

In summary, the application of two-dimensional materials is evolving from single-material usage towards heterogeneous integration.

3.3 Bio-Integrated Soft Electronics

Bio-integrated soft electronics refer to flexible electronic devices capable of tight integration and coordinated operation with biological tissues. In recent years, advancements in materials production, device fabrication, and flexible circuits have enabled significant progress in soft bioelectronics for human medical monitoring and disease diagnosis. The emergence of two-dimensional materials has further advanced this field. This section summarizes the progress of bio-integrated flexible sensors based on two-dimensional materials in detecting the following two types of physiological information.

3.3.1 Skin-Attachable Chemical Biosensor

Skin-attached chemical biosensors are flexible devices that can conform to the skin surface for real-time monitoring of novel biomarkers in body fluids such as sweat, saliva, tears, and interstitial fluid. They play a significant role in health monitoring and disease prevention. These devices achieve tight adhesion to the skin primarily through intermolecular forces or interfacial covalent bonds, and adapt to skin deformations via flexible substrates to ensure interfacial stability. They function by capturing biomarkers (such as glucose, lactic acid, electrolyte ions, etc.) and converting them into electrical signals. To date, extensive efforts have been devoted to exploring two-dimensional materials for diverse wearable sensors, enabling the detection of multiple biological signals from different regions of the human body.

3.3.2 Skin-Attachable Physical Sensor

Skin-attachable physical sensors are a class of flexible devices that can conform to the skin surface for real-time monitoring of human physical signals such as pressure, strain, temperature, and humidity. To ensure that wearable devices maintain effective sensitivity when subjected to stress, their design must also consider mechanical performance requirements. Among these, pressure and strain sensors are the most common types of skin-attachable physical sensors. They typically use two-dimensional materials (such as graphene, MXene) or polymer composites as the sensitive layer, achieving highly sensitive detection of physiological activities like human motion, pulse, and respiration through changes in the resistance or capacitance of the material under deformation.

Overall, flexible electronic devices based on two-dimensional material/polymer composites are leveraging their advantages to drive technological advancements in fields such as wearable devices, human-computer interaction, and biomedicine, continuously promoting development in related areas.

4. Challenges Faced and Future Development Directions

4.1 Main Current Challenges

4.1.1 Limitations in Material Performance

Currently, the mechanical exfoliation method can only provide small-area, high-cost samples, while scalable methods such as chemical vapor deposition (CVD) suffer from issues like non-uniform layer thickness, grain boundary defects, and residual wrinkles. These defects severely impact the mechanical and electrical properties of the materials, making it difficult to meet the stringent requirements of flexible electronics for material uniformity and consistency [1]. Furthermore, many two-dimensional materials are highly sensitive to environmental conditions and struggle to remain stable under ambient conditions, directly limiting the long-term stability of devices.

4.1.2 The Dual Challenge of Performance and Integration Density

High contact resistance between metal electrodes and two-dimensional materials degrades device response speed and increases power consumption. On flexible substrates, the mechanical and electrical stability of the contact interface is more difficult to maintain, further accelerating performance degradation. Simultaneously, the mechanical properties of many flexible devices gradually deteriorate over time. During use, flexible devices undergo various deformations such as stretching, compression, and twisting. This can easily lead to cracks and delamination between the two-dimensional material and the polymer substrate, resulting in performance degradation or even device failure.

4.1.3 Industrialization Challenges

The preparation of high-performance two-dimensional materials often involves complex synthesis processes and intricate handling procedures. This keeps the production costs of two-dimensional materials high, making it difficult for them to compete effectively with conventional silicon-based or organic materials on cost, severely restricting their large-scale industrial applications.

4.2 Future Development Directions

4.2.1 Precision Synthesis and Controlled Growth

By precisely regulating key parameters such as the concentration of metal precursors and growth temperature, the uniformity of two-dimensional materials can be effectively improved. The use of epitaxial growth techniques can significantly reduce grain boundary defects and enhance carrier mobility. Growing two-dimensional materials directly onto flexible substrates such as PET and PI can avoid material loss during the transfer process. Exploring methods like low-temperature CVD can effectively prevent damage to the substrate caused by high-temperature processes, thereby expanding the application scenarios of the materials.

4.2.2 Device Structure and Performance Optimization

Using two-dimensional metal electrodes to replace traditional metals can effectively reduce contact resistance, improve device response speed, and lower power consumption. Methods such as in-situ doping or charge transfer doping can precisely regulate carrier concentration, achieving device balance and optimization. Designing special structures like wrinkle-type or wave-type configurations to release mechanical stress can significantly enhance mechanical durability. Encapsulation with elastic polymers can protect two-dimensional materials from mechanical damage and environmental interference.

4.2.3 Industrialization Ecology

To effectively reduce production and comprehensive costs for the industry, the construction of industrial ecology needs to be improved along two dimensions: by optimizing precursor material and equipment costs and by establishing a comprehensive material recovery and recycling system, production waste and comprehensive costs can be reduced. Focus on research breakthroughs in degradable technologies, using degradable polymer materials such as PVA as substrates, paired with compatible semiconductors, to achieve rapid device degradation and ultimately attain the goals of sustainable development and green ecological construction.

5. Conclusion

This article reviews the research progress in flexible electronic devices based on two-dimensional material-polymer composites. Two-dimensional materials (such as graphene, MXenes, and transition-metal dichalcogenides) are ideal functional materials for flexible electronics due to their excellent electrical, optical, and mechanical properties. Polymer matrices (such as PDMS, PI, and PET) endow devices with good flexibility and mechanical support, and their combination leverages the distinct advantages of both components.

In terms of applications, this article specifically summarizes three main categories of such devices: flexible sensors (including gas sensors and photodetectors), flexible displays, and bio-integrated soft electronics (skin-attached chemical biosensors and skin-attachable physical sensors). These devices offer significant potential in fields such as health monitoring, human-computer interaction, and medical diagnosis.

However, numerous challenges with these devices remain unresolved, including high equipment costs, insufficient device integration density, and inadequate long-term stability. To address these issues, researchers are exploring solutions from multiple directions, including structural design and interface engineering. Meanwhile, innovations in non-traditional dielectric materials (such as ion gels, two-dimensional crystalline materials, and polymer-based dielectrics) offer new pathways to overcome the performance bottlenecks of conventional materials. These materials can achieve superior electrical performance while maintaining the mechanical flexibility of devices, laying an important foundation for the transition of flexible electronics from laboratory research to industrial applications.

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