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Flexible and Stretchable Electrodes for Wearable Energy Storage Devices

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Abstract. Wearable electronic devices have been developing rapidly. Whether it is health monitoring, electronic skin, or smart wearables, people have higher requirements for the high performance and safety of these devices. Flexible and stretchable electrodes are a key component of wearable energy storage devices, so they have become a research hotspot in the field of new energy materials. This review first introduces the design concepts and preparation methods of flexible and stretchable electrodes, then summarizes the research progress of several types of stretchable electrodes, including carbon-based, metal-based, and conductive polymer-based ones. Finally, it discusses their specific applications in stretchable supercapacitors, stretchable lithium-based batteries, and wearable sensors. The article also analyzes the main challenges currently faced by such electrodes and provides an outlook on future development directions, hoping to offer some reference for the research and production of related devices.

Keywords: micro-nano materials, stretchable structure, stretchable electrode, stretchable supercapacitors, wearable sensor

1. Introduction

Wearable electronic devices are now commercializing rapidly and undergoing continuous technological upgrades in fields such as health monitoring, electronic skin, and smart wearables. This has led to relatively high demands on the supporting energy storage devices. Specifically, these devices need to have good flexibility, stretchability, high performance, and sufficient safety. Typical wearable energy storage devices mainly include supercapacitors [1] and lithium-based batteries. Moreover, an intelligent wearable device usually consists of several parts, including a sensing module, an energy storage module, a control module, and a flexible encapsulation. Among these, the core of the energy storage module is the flexible and stretchable electrode [2]. Such an electrode has to meet several conditions at the same time: high electrical conductivity, good mechanical stretchability, stability under repeated deformation, decent electrochemical activity, and good interfacial compatibility with the flexible substrate.

Traditional rigid electrodes are mostly made of hard metals or ceramic-based materials. A major drawback of such materials is their mechanical brittleness and poor deformability. When subjected to external forces like stretching, bending, or twisting, their conductive networks are prone to breakage, and their electrochemical performance drops sharply. Therefore, they cannot adapt to the

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close-fitting use scenarios required by wearable devices. At present, the core challenges and technical bottlenecks in the development of flexible and stretchable energy storage devices are concentrated in several areas: it is difficult to synergistically improve the electrical conductivity and stretchability of electrode materials; the structural stability of the conductive network under complex deformation is insufficient; the interfacial bonding force between the electrode and the flexible substrate is weak; and the cost and compatibility of large-scale preparation processes remain problematic [3]. Hence, developing high-performance flexible and stretchable electrodes has become key to promoting the practical application of wearable energy storage devices.

2. Design and preparation of flexible and stretchable electrodes

The preparation of flexible and stretchable electrodes relies on the intrinsic properties of the conductive materials as well as structural design. Based on differences in preparation principles, the methods can be divided into two main categories: physical methods and chemical methods [4]. The core goal is to construct a continuous conductive network that can maintain integrity under mechanical deformation while also taking electrochemical performance into account.

2.1. Physical methods

Physical methods are the mainstream approach for preparing flexible and stretchable electrodes. Their core idea is to combine electrical conductivity with stretchability through physical deformation, filler composite, or macrostructural design, without changing the chemical composition of the materials, which makes the process highly compatible. Commonly used conductive materials [5] include liquid metals, carbon-based materials, metal oxides, and conductive polymers. Among these, carbon nanotubes have a high aspect ratio [6] and excellent electrical conductivity [7]. Graphene [8] has an ultra-large specific surface area and high mechanical strength [9]. Carbon nanofibers have a good porous structure and deformation toughness [10, 11]. Metal oxides such as manganese dioxide and ferric oxide have high electrochemical activity, which can increase the energy storage capacity of electrodes. Conductive polymers such as poly(3,4-ethylenedioxythiophene) (PEDOT) and polyaniline (PANI) are both flexible and conductive [12], and they can be easily compounded with flexible substrates .

Physical methods mainly include coating, sputtering, infiltration, soaking, and 3D printing [13] on elastic substrates. Typically, the active conductive materials are uniformly dispersed in a suspended ink, which is then coated or printed onto elastic substrates such as silicone rubber or polyurethane. Alternatively, the flexible substrate can be immersed in a casting solution, followed by spin coating to form a conductive film with uniform composition and good interfacial adhesion. For example, in Figure 1,one study used multi-material 3D printing (two-photon polymerization, 2PP) to composite multi-walled carbon nanotubes (MWCNTs) and PEDOT:PSS into a silicone rubber (PDMS) elastic matrix. The resulting flexible electrode retained 65% of its electrical conductivity at 50% tensile strain, with no significant degradation in electrochemical performance. After 3000 stretching cycles at 10% strain, no delamination occurred between the electrode and the PDMS substrate, and key indicators such as electrochemical impedance and specific capacitance (≈667 F/g) remained stable [14]. Meanwhile, physical structural design can be used to construct stretchable structures such as wavy, island-bridge, fabric-based, spiral, and folded types. These structures use geometric deformation to offset external tensile forces, thereby avoiding damage to the conductive network. For instance, fabric-based woven structure electrodes [15] rely on the inherent stretchability of the fabric [16] to achieve conductive stability under repeated cyclic deformation.

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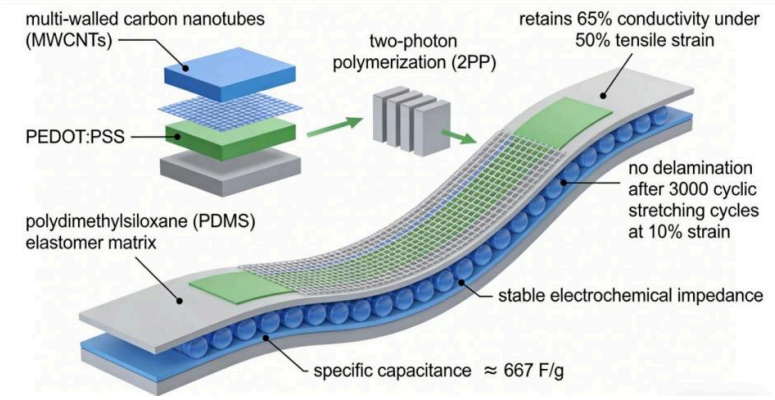


Figure 1. Multi-material 3D printing preparation and performance characterization of MWCNTs/PEDOT:PSS/PDMS stretchable composite electrodes

2.2. Chemical methods

Chemical methods mainly optimize stretchability and electrochemical performance by regulating the chemical composition [17] and molecular structure of the materials [18], or by constructing chemically bonded composite systems . These methods are suitable for the flexible modification of intrinsically rigid conductive materials and the construction of functional conductive networks. Common approaches include hydrogels, surface modification, and bond-energy bonding. For hydrogel preparation [19], conductive nanomaterials are chemically crosslinked with a polymer hydrogel network to produce a hydrogel electrode that combines high stretchability, water retention, and conductivity, which can meet the close-fitting needs of flexible wearables. Surface modification [20] involves chemical grafting or functional group modification of the conductive material or flexible substrate to improve the interfacial bonding force between the two and reduce interfacial delamination during deformation. Bond-energy bonding uses covalent bonds or coordination bonds to combine conductive materials with a flexible polymer matrix [21], forming a stable chemically crosslinked conductive network that enhances the cyclic deformation stability of the electrode.

A well-structured paper follows a clear hierarchy of sections, with different levels of headings to organize content logically. Generally, a conference paper may include the following sections: *Introduction, Literature Review, Methodology, Results, Discussion, Conclusion, and References*. Fourth- and lower-level headings should not be used in your paper. Each section heading should be numbered according to the formats defined in Table 1. Note that the styles for headings will automatically add numbers for each section. Use sentence case for the section headings. Only the first word should be capitalized, and the rest should be lowercase except for proper nouns, abbreviations, and acronyms.

3. Stretchable electrodes

Electrodes are the core component of energy storage devices [22], directly determining key performance indicators such as conductivity, energy storage capacity, and cycle stability. The material selection and structural design of stretchable electrodes are central to their adaptation for wearable energy storage devices [23]. Currently, the mainstream stretchable electrode materials in research fall into three major categories: carbon-based materials, metal-based materials [24], and conductive polymer-based materials [25]. All of these have achieved stretchability modification and performance optimization through physical or chemical methods.

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3.1. Carbon-based materials

Carbon-based materials are among the most widely used materials for stretchable electrodes [26]. They offer excellent electrical conductivity [27], high chemical stability [28], a large specific surface area [29], and good mechanical toughness. Moreover, they can be efficiently modified through both physical and chemical methods. In terms of physical methods, one study used an electrolyte-mediated bonding approach to blend high-density graphene paper with an elastomer substrate. By utilizing the mechanical instability of the material, the graphene paper was induced to self-assemble into a self-organized wrinkled structure, successfully producing a high-performance carbon-based stretchable electrode. This electrode achieved not only a uniaxial tensile strain of 300% but also an area tensile strain of up to 800%, far exceeding the required 100% tensile performance. Its electrochemical performance was also excellent, with a specific capacitance of 196 F/g, which is more than three times that of traditional carbon nanotube-based stretchable electrodes. After 1000 tensile-relaxation cycles, the electrode showed no structural damage, and its electrochemical performance did not degrade significantly. A solid-state supercapacitor constructed based on this electrode could operate stably under large deformation conditions, providing a classic design paradigm for the simple and scalable preparation of high-stretchability carbon-based flexible electrodes [30]. As for chemical methods, carbon-based materials can be chemically crosslinked with polymer hydrogels through hydrothermal synthesis or chemical vapor deposition, or carbon nanomaterials can be modified with functional groups to improve their compatibility with flexible substrates. In one study, a graphene film was grown in situ on a fabric substrate using chemical vapor deposition (CVD), achieving a tight integration of the graphene with the fabric's woven structure, thus successfully preparing a graphene/fabric composite electrode. This electrode combined the inherent stretchability of the fabric's woven structure with the high conductivity of graphene. After multiple stretch-relaxation cycles, the interface between the graphene and the fabric remained stable, with no obvious delamination or structural damage, and the electrochemical activity was well maintained. This provides an efficient and feasible technical path for electrode preparation in the field of wearable flexible electronics [31].

3.2. Metal-based materials

Metal-based materials have intrinsically high electrical conductivity [32] and are an ideal choice for preparing high-rate stretchable electrodes. They mainly include metal nanowires, metal thin films, and liquid metals. Among these, liquid metals, because they have no fixed shape and can maintain continuous conductivity under deformation [33], have become a research focus for metal-based stretchable electrodes. For rigid metal materials such as metal nanowires and metal thin films, physical structural designs (e.g., island-bridge structures, wrinkled structures) or chemical coating modifications [34] are often used to solve the problem of easy fracture under stretching. Liquid metals, on the other hand, are mostly prepared into flexible and stretchable electrodes through physical methods such as microencapsulation or blending with elastic polymers [35].

3.3. Conductive polymer-based materials

Conductive polymer-based materials are soft materials that inherently possess a certain degree of flexibility and stretchability [36]. They also exhibit high electrochemical activity, and their conductivity and mechanical properties can be tuned through chemical methods such as doping and copolymerization [37]. They are therefore a new type of electrode material suitable for wearable

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energy storage devices. The mainstream conductive polymers include PEDOT, PANI, and polypyrrole (PPy). These can be grown in situ directly on a flexible substrate via chemical polymerization to form a conductive film that bonds tightly with the substrate [38]. Alternatively, they can be blended with elastic polymers through physical mixing to improve their stretchability [39]. For example,in Figure 2, one study used electrochemical polymerization to grow a polyaniline (PANI) nanoflower structure in situ on the surface of polyurethane (PU) nanofibers, successfully preparing a core-shell structured stretchable conductive composite electrode. This system fully combined the high elasticity and high stretchability of the polyurethane fiber matrix with the high specific capacitance and high electrochemical activity of polyaniline. The composite electrode maintained stable electrochemical performance even under stretching, effectively solving the problems of high brittleness and poor cycling stability of pure polyaniline, and could be directly applied to the construction of flexible supercapacitors [40].

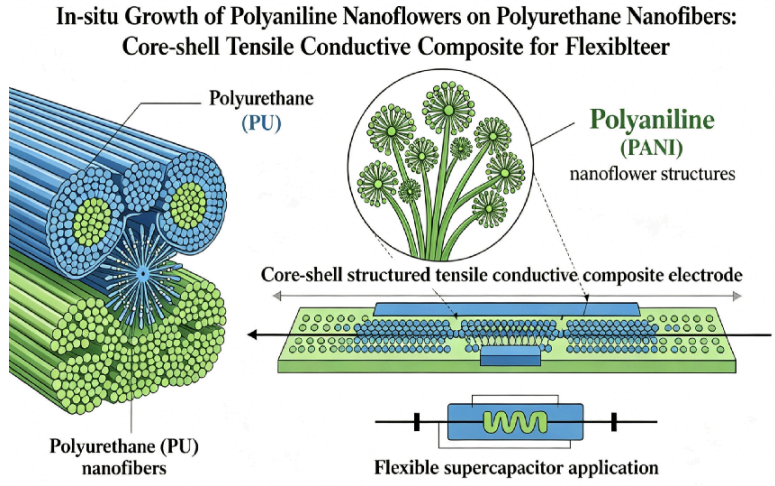


Figure 2. In-situ growth of PANI nanoflowers on PU nanofibers and structure of core-shell stretchable composite electrode

4. Applications of stretchable conductors in energy storage devices

The core application scenarios for flexible and stretchable electrodes are wearable energy storage devices and flexible sensors. Relying on their excellent mechanical deformability and electrochemical performance [41], they solve the problem that traditional rigid electrodes cannot adapt to the close-fitting, dynamic deformation requirements of wearable devices. Currently, they have been used in applied research and performance validation in stretchable supercapacitors, stretchable lithium-based batteries, and various types of flexible wearable sensors [42].

4.1. Stretchable supercapacitors

Supercapacitors have the advantages of fast charge/discharge rates, long cycle life, and high power density [43], making them ideal energy storage devices for wearable equipment. Stretchable supercapacitors, by incorporating flexible and stretchable electrodes, achieve stable energy storage under deformation [44, 45]. Most stretchable supercapacitors currently under research use carbon-based, conductive polymer-based, or composite stretchable electrodes as the electrode material [46], combined with gel electrolytes and elastic encapsulation materials, to construct diverse structures such as planar and fiber types, adapting to different wearable scenarios. Among the strategies for

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constructing stretchable electrodes, carbon-based and conductive polymer composite systems show outstanding advantages. For example, a fiber-type stretchable supercapacitor designed based on a carbon nanotube/hydrogel composite system can withstand multi-dimensional complex deformations such as bending, twisting, and stretching. It maintains a capacitance retention of over 85% at 100% tensile strain and can be further woven into smart wearable fabrics, effectively realizing the integration of energy storage and wearable functions, thus providing an important design concept for wearable flexible energy storage devices [47].

4.2. Stretchable lithium-based batteries

Lithium-based batteries have high energy density [48] and are the core energy storage devices for meeting the long-endurance needs of wearable devices [49]. The research and development focus of stretchable lithium-based batteries is to solve the problems of high electrode brittleness and electrolyte leakage in traditional lithium batteries [50] through the design of stretchable electrodes. Currently, stretchable lithium-based batteries mostly use structurally designed stretchable electrodes (e.g., wavy or island-bridge type metal-based/carbon-based composite electrodes), combined with flexible gel electrolytes and elastic encapsulation materials, to achieve a certain degree of stretchability while maintaining the battery's energy density . Research has shown that through the synergistic design of structure and materials, stretchable lithium-ion batteries have achieved an effective tensile strain of 50%. They can maintain stable charge/discharge efficiency and capacity retention during repeated dynamic stretching, effectively solving the problem of traditional energy storage devices easily failing under deformation. This provides a feasible solution for high-energy-density, high-mechanical-adaptability energy storage systems for wearable applications [51].

4.3. Sensors

In wearable sensors, flexible and stretchable electrodes mainly serve as the sensitive layer [52]. They utilize changes in resistance or capacitance of the electrode under deformation to monitor human physiological signals and motion states. The main types are strain-resistance sensors and pressure-capacitance sensors [53]. Strain-resistance sensors are based on the linear change in resistance of the stretchable electrode with tensile deformation, enabling monitoring of dynamic deformations such as joint movement and muscle contraction [54]. Pressure-capacitance sensors use a stretchable electrode and a flexible dielectric layer to construct a parallel-plate capacitor structure. The change in dielectric layer thickness under external pressure causes a capacitive response, allowing high-sensitivity and high-precision detection of weak physiological pressure signals such as pulse and blood pressure.

5. System-integrated flexible and stretchable energy storage devices

As wearable electronic devices become more integrated and miniaturized, a single stretchable energy storage device can no longer meet their needs [55]. Therefore, system-integrated flexible and stretchable energy storage devices have become a development trend. The core idea is to integrate stretchable electrodes, energy storage devices, sensing modules, control modules, and flexible encapsulation into one system, achieving the multi-functional synergy of "sensing-energy storage-regulation."

The design of system-integrated devices relies on the versatility and compatibility of flexible and stretchable electrodes. Through integrated preparation processes, the same stretchable electrode can

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serve simultaneously as the electrode for the energy storage device and the sensitive layer for the sensor, achieving functional integration and miniaturization of the device [56]. At the same time, in Figure 3,flexible gel electrolytes and elastic encapsulation materials are used to ensure the mechanical stretchability and operational safety of the entire integrated system. Current research has achieved the integrated combination of a stretchable supercapacitor and a strain sensor. Such a device can simultaneously monitor human motion signals and store its own energy, requiring no external power source, thus meeting the portability needs of wearable devices [57]. The key challenges for system integration include interfacial matching between different modules, suppression of signal interference, and the integration of scalable preparation processes.

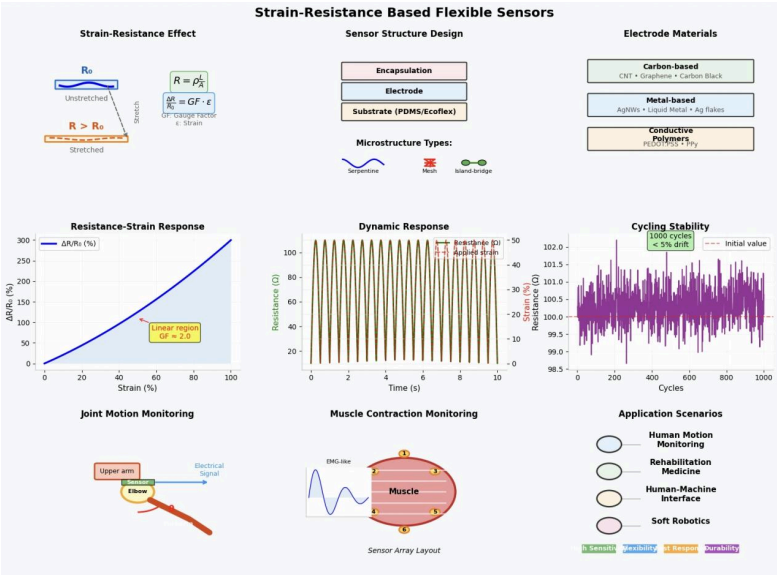


Figure 3. Flexible and stretchable electrodes for wearable physiological signal monitoring sensors

6. Conclusion

This review has summarized the research progress on flexible and stretchable electrodes for wearable energy storage devices. From the perspective of preparation methods, physical methods are simple and highly compatible, currently serving as the mainstream approach. Chemical methods, on the other hand, enable the tuning of intrinsic material properties. Combining the two has become an important direction for improving electrode performance. Regarding material systems, carbon-based, metal-based, and conductive polymer-based materials each have their own advantages, and composite design has become a key strategy to solve the difficulty of synergistically achieving high conductivity and high stretchability in single materials. Furthermore, flexible and stretchable electrodes have been widely applied in stretchable supercapacitors, lithium-based batteries, and wearable sensors, and the development of system-integrated devices offers a new path for the integrated development of wearable electronics.

Currently, flexible and stretchable electrodes and their supporting energy storage devices still face many core challenges. The synergistic optimization of electrical conductivity, stretchability, and electrochemical performance of electrode materials still needs breakthroughs. The long-term stability of the conductive network and device structure under complex dynamic deformation is insufficient. The interfacial bonding force between the electrode and various functional layers is weak. Moreover, large-scale, low-cost preparation processes are not yet mature.

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Future research directions can focus on the following aspects. First, develop new micro-nano composite conductive materials to achieve intrinsically high conductivity and high stretchability through molecular design and structural regulation. Second, innovate stretchable structural designs, drawing on bionic principles, to develop new structures that adapt to multi-dimensional complex deformations and improve the cyclic deformation stability of electrodes. Third, optimize interface design and preparation processes to enhance the interfacial bonding force between functional layers, and develop low-cost, scalable printing, weaving, and other preparation processes. Fourth, accelerate the development of system-integrated devices to achieve a high degree of integration of energy storage, sensing, control, and other functions, while improving practical performance such as water resistance and breathability, thereby promoting the industrialization and commercial application of flexible and stretchable energy storage devices.

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