Proceedings of the 3rd International Conference on Signal Processing and Machine Learning
DOI: 10.54254/2755-2721/4/2023373

A review of flexible wearable sensors

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Abstract. With the improvement of science and technology, wearable sensor technology has been widely used in health management, human-computer interaction and many other fields, becoming a mainstream research direction. Moreover, it has a good application prospect in various industries. In this paper, the performance requirements of wearable sensors are analyzed, and the future direction of wearable sensors is discussed in combination with the development of polymer for flexible sensors on flexible substrates and conductive materials. Based on the limitations of current technology, the optimization of wearable sensor technology mainly focuses on the improvement of sensing element materials and the accuracy of collected signals.

Keyword: Flexible wearable sensors, Flexible sensors, Electronic skin technology, Signal acquisition, Compliance materials.

1. Introduction

As an emerging industry with rapid development recently, wearable technology has been widely used in human health monitoring, health management, disease prevention, artificial skin, human-computer interaction and other fields. There are many types of wearable devices, such as pressure sensors, strain sensors, gas sensors, for detecting respiratory humidity, heart rate, body temperature, and other physiological information [1]. With the improvement of science and technology, flexible electronic equipment has gradually become a hot research direction. Traditional electronic devices such as silicon, germanium, gallium arsenide and other inorganic semiconductor materials are often rigid and unable to meet the demand of deformation. On the contrary, flexible wearable devices have good application prospects due to their excellent flexibility, good malleability and portability.

In recent years, people have been studying stretchable sensors with high sensitivity, short response time, and wide sensing range. However, traditional sensors are facing many problems, such as narrow sensing range, low sensitivity, complex manufacturing process, single function and low wearing comfort, which limit their potential for accurate detection of medical signals and promotion to practical applications. Therefore, it is of great practical significance to develop multifunctional flexible wearable devices with excellent transmission sensitivity. At present, there are three trends in the development of flexible sensors: (1) Development and application based on novel nano-sensing materials. For example, the development of nanomaterials such as low-dimensional nanomaterials and new two-dimensional carbonitride MXene can make the sensor tend to be high-performance, miniaturized, integrated, and low-cost. (2) The adoption of new preparation processes. The development and application of micro and nano technologies such as wet spinning and 3D printing will promote the industrialization of new

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sensors in the future. (3) The development of new intelligent sensors. The multi-function of future sensors can be realized through the innovation of new principles and bionic technology.

This paper introduces the performance requirements of flexible wearable sensors, summarizes the research progress of polymer used in flexible sensors on flexible substrates and conductive materials in recent years, and prospects for their application.

2. Performance of wearable electronic devices

2.1. Flexibility
Traditional electronic equipment is generally made of metal. With hard and rough surface, they cannot be closely contacted with soft human tissue, which is the key influencing factor of the inability to accurately transmit physiological signals between the human body and electronic equipment. As a flexible polymer material, conductive hydrogel has a broad application prospect in the field of flexible wearable devices due to its good water retention, biocompatibility and mechanical tunability [2].

2.2. Electrical conductivity
Electrical conductivity is one of the key properties of wearable devices. Wearable devices can be roughly divided into two categories according to their functions: one is smart wearable devices based on sensor applications, which need to be connected to terminals such as smartphones to analyze, manage and display data. The other is intelligent wearable devices that support human-computer interaction [3]. Both the transmission from the human body to electronic devices and the supply of more stable power to electronic devices are inseparable from the support of electrical conductivity. Common materials for wearable devices include carbon nanotubes, metal nanowires, organic semiconductors, and other conductive materials.

2.3. Sensitivity
Wearable electronic devices are often used to monitor various physiological signals and human activities. How to improve the sensitivity of the device is the focus of current wearable sensor research. Elastomers can be combined with conductive materials such as graphene, carbon black, carbon nanotubes, and metal nanoparticles, conductive polymers, ion-conducting materials, etc., to create high-sensitivity sensors.

2.4. Data security
In the development and data collection of wearable devices, the security of user data privacy is often ignored for the sake of commercialization, which has also become an obstacle to the further expansion of the application of wearable devices. Due to the ease of data collection by wearable devices and the lack of encryption in data transmission, the database of the server may have security vulnerabilities. It can result in security risks for a large number of different data information of users recorded through sensors [4].

3. Research progress
Generally speaking, a flexible wearable sensor is composed of three parts: polymer matrix, sensing element and conductive interface [5], as shown in Figure 1.

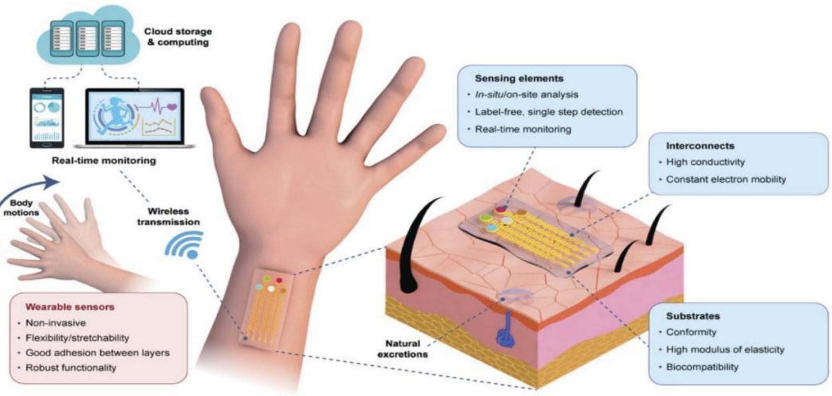

Figure 1. Wearable sensor composed of matrix, sensing element and conductive interface [5].

3.1. Flexible matrix

Due to the direct contact with the skin, the polymer matrix usually needs to have a certain degree of comfort, high elastic modulus and good biocompatibility. Traditional flexible pressure sensors are often based on air-impermeable rubber or dense plastic films with poor moisture permeability, which can easily make the skin uncomfortable. In order to obtain skin-like fit and high stretchability to improve comfort, flexible thermoplastic [7] polymers such as polycarbonate (PC), Ethylene phthalate (PET), polyimide (PI), polylactic acid (PLA) and other materials, elastomers such as polydimethylsiloxane (PDMS), polyurethane (PU), styryl Elastomers (SEBS/SBS), etc.

3.1.1. Flexible thermoplastic polymers. PET and PLA have high mechanical strength, but the Young's moduli of these materials are high, yet they do not have tensile resilience. PI has excellent mechanical and bending properties, corrosion resistance, high temperature resistance and high tensile strength (up to 100MPa), so it is often the best choice for the preparation of flexible bendable pressure sensors [7].

Guo et al. made a flexible wearable pressure sensor with a sandwich structure by sandwicsing a porous MXene impregnation layer between a biodegradable PLA sheet and a PLA sheet coated with an interdigital electrode [8]. This sensor has high sensitivity, wide detection range (10.2 Pa - 30 kPa), fast response time (11ms), low power consumption (8-10W), great reproducibility (over 10 000 cycles), and excellent degradability.

In addition, flexible wearable sensors can also be used to manufacture electronic skin and predict the potential health status of patients, proving their application potential in personal health monitoring, clinical diagnosis and artificial skin. Zhan et al. [9] used PI instead of PDMS as the substrate to avoid the decrease of electrode conductivity during testing and treatment, considering the good adhesion between the interdigitated electrodes and PI. They impregnated single-walled nanotubes (SWNTs) into facial tissue and sandwiched them between bare PDMS flakes and PI flakes with interdigitated electrodes to fabricate a flexible wearable pressure sensor. This sensor has a sensitivity of 1.3kPa-1 in the range of 35 to 2500Pa and 2.2kPa-1 in the range of 2500 to 11700Pa. The sensor is cheap to prepare and easy to mass-produce. At the same time, the sensor has low preparation cost, so it is easy to carry out large-scale production.

3.1.2. Elastomeric polymers. PDMS has wide temperature range, good transparency and air permeability, high resilience, hydrophobic and acid and alkali corrosion resistance. In addition, the preparation method of PDMS is simple and it is easy to be combined with conductive materials, making it a common matrix for flexible sensors. PU has extremely high tensile strength, and the carbamate groups in its molecular chain can be cross-linked, so it is easy to control its performance.

Gong et al. [10] took MXene as the conductive material and made a sandwich structure substrate with thin gold nanowires impregnated tissue paper between PDMS sheet and patterned interdigital electrode array, which was finally assembled to prepare a flexible wearable pressure sensor with high sensitivity. Under the action of external pressure, compression deformation makes more gold nanowires contact with the interdigital electrode, so that the contact resistance between the tissue paper and the interdigital electrode decreases. The sensor has a sensitivity of 1.14kPa-1, a minimum detection limit of 13Pa, a response time of 17ms, and it can withstand more than 50,000 cycles. It can be used for real-time monitoring of blood pulses.

Song et al. [11] deposited a gold film on PU, and then used PET to cross-fix the end and center of the sensor to change the local stress distribution, preparing a new biaxial strain sensor. The sensor has high permeability, good biocompatibility, low cost, and it can withstand 140% stretch, but the detection range is limited.

3.2. Sensing element

Sensor elements can build a stable sensor network to convert external stimuli into electrical signals. Such elements are generally conductive materials dispersed in the matrix by surface adsorption or uniform blending. It can be divided into [6] three categories: carbon-based active materials, metal-based active materials and conductive polymer-based active materials. The Figure 2 shows common conductive fillers and flexible substrates used in flexible wearable electronic devices [12].

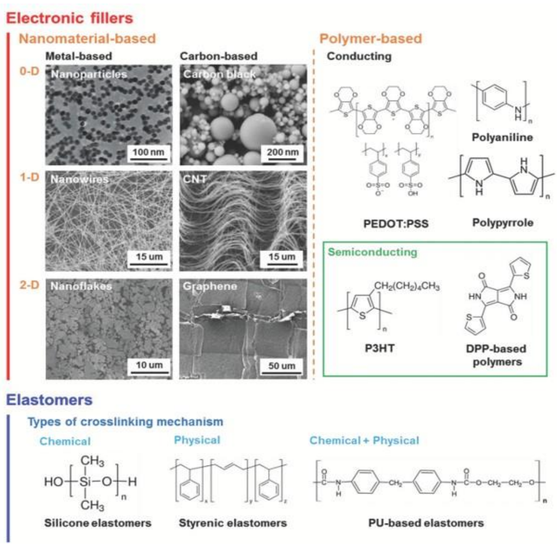

Figure 2. Common sensing elements and flexible matrix of wearable flexible sensors [12].

Metal nanomaterials have good electrical conductivity, such as gold, silver, nickel, and other rare metals, which can also form composites with polymers and fibers. Although metal nanoparticles can be used to produce highly sensitive flexible sensors, the tensile capacity and detection range of the sensors are limited due to their limited chemical stability [13].

Carbon nanomaterials are important materials for flexible wearable sensors, such as graphite, graphene, carbon fiber and carbon nanotubes. For a long time, carbon-based nanomaterials have played an important role in sensing components due to their advantages, such as excellent electrical conductivity, flexibility, chemical and thermal stability, low weight, easy functionalization and mass production [14].

The conductivity of conductive polymer materials will change with the change of conductive network in polymer matrix. When subjected to external stimuli such as temperature, pressure, strain and so on, the internal conductive network will change, which is manifested as the change of resistance externally. Common conductive polymer active materials are mainly represented by polyaniline (PANI), polypyrrole (PPy) and poly 3, 4-ethylenedioxythiophene (PEDOT) [7]. The sensor can be prepared by filler blending, layer assembly, surface growth and other methods combined with the flexible matrix.

DOI: 10.54254/2755-2721/4/2023373

Excellent charge transport capacity often requires high crystallization. However, the semi-crystalline properties of polymers limit the electron transport capacity of conductive polymer materials. The stability and conductivity of these materials need to be improved. Yonggang Wang et al. [15] constructed a flexible wearable pressure sensor by clamping the carbonized organometallic frame (MOF) material and polyurethane sponge (C-MOF/PANIF@PU) composed of polyaniline fiber between the breathable fabric and the fabric with interdigital electrode. The experimental results show that the pressure sensor has high measuring range (60kPa), high sensitivity (158.26kPa-1), fast response (22ms)/ recovery (20ms), good air permeability and excellent repeatability (15000 cycles). Atalay A et al. [16] designed and synthesized a highly stretchable fiber-silicone capacitive flexible sensor with conductive knitted fabric as the electrode and silicone rubber as the dielectric material, which was used for human joint detection and integrated into gloves for hand motion tracking. The capacitance of the sensor can be changed with the change of gesture, showing good tracer motion ability.

The sensing performance of a sensor can be evaluated by certain key parameters, including sensitivity, sensing range, and response time. High sensitivity requires that the conductivity of the microstructured sensing layer can change significantly even under small deformation, while wide sensing range requires that the sensing layer can continue to conduct electricity under large deformation [17]. However, high sensitivity and wide detection range are often incompatible, which greatly limits the application of flexible sensors. Therefore, the balance and promotion of the two has become the focus of research.

4. Modification methods

4.1. Compound modification

Comprehensive utilization of the advantages of various materials has always been a major purpose and advantage of composite materials. In order to further enhance the conductivity and sensitivity of the sensor, researchers have tried to use other conductive materials such as carbon nanotubes (CNTs) to form composites with the sensor, or choose to prepare effluent gel to prepare three-dimensional conductive network to increase the porosity of the conductive filler, so as to increase the conductive channel. All of these improve the conductivity of the sensor to a certain extent. In addition, in order to enable the sensor to simultaneously possess properties such as wide detection range, high durability, high cycle times, degradability, self-healing and capability, conductive fillers can also be compounded with other materials with special properties themselves, making the durability of the sensor improve qualitatively

Seyedin et al. [18] prepared Ti3C2Tx MXene/polyurethane (PU) composite fibers by wet spinning technology. Using isopropanol and acetic acid as coagulants, the MXene/PU composite fiber obtained by wet spinning technology perfectly inherits the good tensile properties of PU and the excellent electrical conductivity of MXene, so that the prepared fiber sensor has up to about 152 % strain, and has ultra-high sensitivity (GF≈12900). This proves that PU can be combined with MXene to prepare a composite material to enhance the conductivity of the sensor.

4.2. Microstructure modification

Researchers have found that coating or filling the microstructures with conductive materials is an effective strategy to improve the sensitivity of sensors and reduce their response time, since microstructures lead to more stress contacts and strain concentration. In addition, microstructures such as cracks, folds, overlaps and sandwich structure can be constructed according to the bionics principle to broaden the detection range and improve the sensitivity of the sensor.

Tang et al. [19] fabricated a flexible piezoresistive sensor by combining controlled graphene nanowall wrinkles (GNWs) with PDMS. The sensitivity of the sensor is 59.00 kPa-1 in the range of 0-2kPa and 4.84 kPa-1 in the range of 2-20kPa. The response time is 6.9ms and the minimum detection limit is 0.2Pa. Compared with other sensors of the same type, it shows higher sensitivity, lower detection limit and faster response speed, which proves that the folded surface can be separated microstructure

DOI: 10.54254/2755-2721/4/2023373

can improve the sensitivity of the sensor. However, the preparation process of this method is complex and it is difficult to put into mass-production.

Chaoyang Li et al. [20] prepared a multi-level hemispherical microstructure array using flexible polydimethylsiloxane (PDMS) transfer on an acrylonitrile-butadiene-styrene (ABS) template with a hierarchical hemispherical structure array. The PDMS thin film (Hi-PDMS) was fabricated by spraying graphene on the microstructure to obtain the sensing layer. Finally, the sensing layer and the interdigital electrodes embedded with silver nanowires were assembled into a piezoresistive flexible pressure sensor. The preparation process is shown in the figure below. As shown in the experiment. The sensor has a high sensitivity of 15.4kPa-1 in the linear sensing range of 0-200kPa, a low detection limit of 16Pa, a fast response time of 20ms and a high stability that can withstand 7500 cycles of pressure. It has been successfully applied to detect human motion signals such as voice, pulse, gesture, etc., and has shown good application prospects in the fields of human health monitoring and electronic skin.

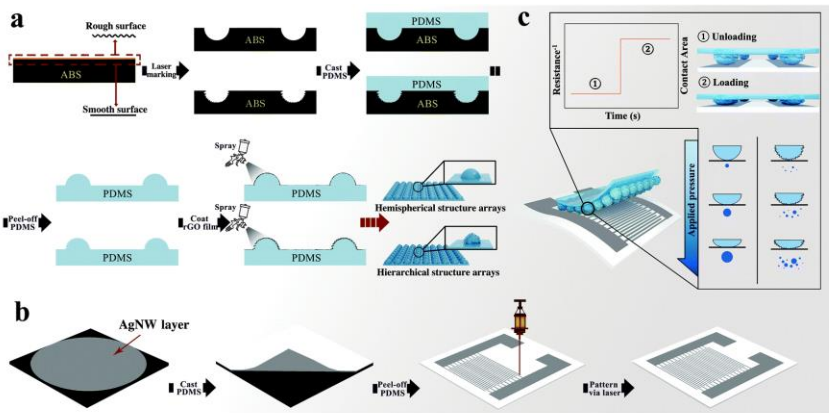

Figure 3. Fabrication process diagram of flexible sensor with surface multistage hemispherical microstructure [20].

(a) preparation of conductive PDMS films with surface multistage hemispheric microstructures and with hemispheric structures,
(b) preparation of interdigital electrodes,
(c) schematic diagram of flexible pressure sensor with multi-level hemispherical microstructure and sensing process of piezoresistive flexible pressure sensor

Inspired by the crack-type sensing system of spider legs, Guo et al. [21] used PU sponge as the framework to self-assemble and grow rGO and conduct in-situ reduction. By growing polyaniline through in-situ polymerization, researchers prepared a layered micro-structure flexible wearable pressure sensor combined with microcrack and interlocking array. Its microstructure is shown in the Figure 4. The sensor can achieve pressure detection in the range of 27 Pa-25 kPa, with response/recovery time 22ms/20ms, high sensitivity and ultra-sensitive load sensing (25mg feather), excellent repeatability (over 10,000 cycles), It can be assembled as an intelligent electronic skin to draw pressure distribution curves and predict static simulated tremor in Parkinson's patients.

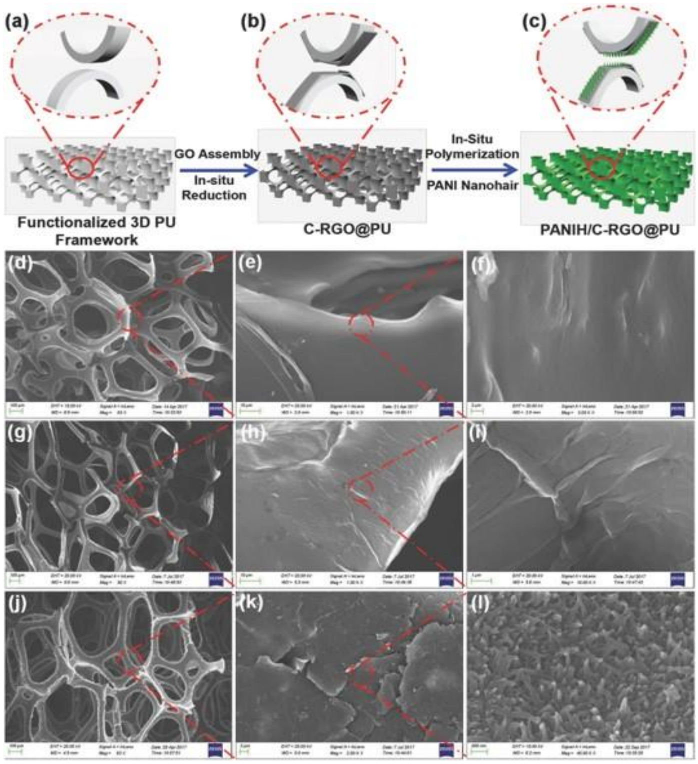

Figure 4. Microstructure of C-RGO@PU flexible sensor.[21].

(a-c) coating RGO on 3D PU network framework to prepare C-RGO hierarchical synergistic microstructure, and in-situ polymerization on C-RGO sheet to prepare tunable interlocking PANIH array,
(b-f) SEM image of pure PU sponge framework,
(g-i) C-RGO slices on 3D PU network backbone,
(j-l) Nanoscale PANIH/C-RGO@PU with PANIH-coated C-RGO sheets

5. Conclusion

This paper summarizes the required properties of wearable electronic devices, such as flexibility, conductivity, sensitivity, detection range, data security. The research progress of flexible wearable sensors in recent years is reviewed, and feasible modification schemes are proposed. Wearable sensors still have a long way to go in pursuit of further improvements in sensitivity and sensing range, while reducing costs and expanding production. The future development trend of wearable sensors may focus on further improvements in electronic skin technology and improving the accuracy of collecting signals. This paper mainly analyzes the current performance and technical points of wearable electronic devices by means of a review. In the follow-up, the detection and simulation of sensor flexibility will be combined with the current leading experimental methods.

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