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Flexible and stretchable sensors for wearable medical monitoring

Ruiyang Liu[1, *]

[1]Harbin Engineering University, Heilong Jiang, China

*Corresponding author: 2021080724@hrbeu.edu.cn

Abstract:

With the rapid development of medicine in contemporary society, wearable and flexible healthcare devices will attract great attention as emerging medical electronics. Flexible and stretchable sensors, as an important component of many wearable healthcare devices, will allow real-time monitoring of daily health data for diagnosis and prediction of early stages of diseases, providing a great boost to modern medicine. This paper reviews the development of wearable flexible sensors and the latest research progress, including flexible sensors based on conductive nanocomposite hydrogels, graphene, and Janus membranes, analyzes the principles and characteristics of flexible sensors of different technological routes, and discusses the challenges and opportunities of flexible sensors. Achieving accurate real-time data transmission and analysis for flexible sensors, as well as low power consumption, is a major challenge. Flexible sensors will become increasingly important in personalised medicine, health monitoring, non-invasive diagnostics and telemedicine. They will be a tool for more convenient, accurate and efficient health management.

Keywords: flexible sensors; medical monitoring; health care.

1. Introduction

In recent decades, flexible electronics have rapidly advanced across various fields, such as human-computer interaction, medical monitoring, point-of-care testing, environmental monitoring, and electronic skin. One of the most prominent areas of research is continuous healthcare monitoring, which offers significant advantages over traditional medical practices. Unlike conventional methods that require repeated sampling and analysis, continuous healthcare monitoring allows for the simultaneous tracking of multiple physical and chemical signals, enabling early detection and long-term patient observation. This approach transforms medical diagnostics by offering comfort, portability, remote operation, timely feedback, and other benefits that enhance patient care. Monitoring physiological parameters such as heart rate, respiratory rate, blood pressure, body temperature, and body composition provides valuable insights into an individual's health, surgical outcomes, and treatment effectiveness. Flexible electronic sensors, including pressure, temperature, electrophysio-

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logical, and electrochemical sensors, are widely used for tracking health indicators. Wearable sensors, although less precise than implantable ones, are easier to implement due to lower biocompatibility requirements, reducing risks like infection associated with surgery. Additionally, wearable sensors are easier to maintain, replace, and upgrade, making them ideal for non-invasive, daily health monitoring. This makes them a convenient, cost-effective, and user-friendly solution for continuous health tracking.

This review examines recent advances in flexible wearable sensors for medical applications, offering an analysis and outlook. Firstly, a concise overview of recently successfully demonstrated flexible wearable sensors is presented, including conductive nanocomposite hydrogel-based flexible sensors, graphene-based flexible sensors, and Janus membrane-based flexible sensors. The fundamental principles are introduced, and applications are analysed. Finally, the paper summarises the principal challenges and opportunities for flexible wearable sensors, and identifies that advances in wearable flexible sensors are crucial for the development of the medical field.

2. Conductive nanocomposite hydrogels based flexible sensors

Extensive research has investigated the potential of stabilized hydrogels made from nanopolymer matrices as replacements for traditional rigid sensors in flexible wearable applications [1]. A critical factor in enhancing the performance of conductive nanocomposite hydrogels is the effective dispersion of nanofillers within the polymer matrix. Incorporating nanofillers into hydrophilic polymer networks has been shown to significantly enhance the mechanical properties of these hydrogels. Additionally, the inclusion of various nanofillers can endow conductive hydrogels with unique characteristics, expanding their range of practical applications. The combination of different conductive nanomaterials with other nanofillers and conductive polymers has been extensively studied, as shown on Fig.1.

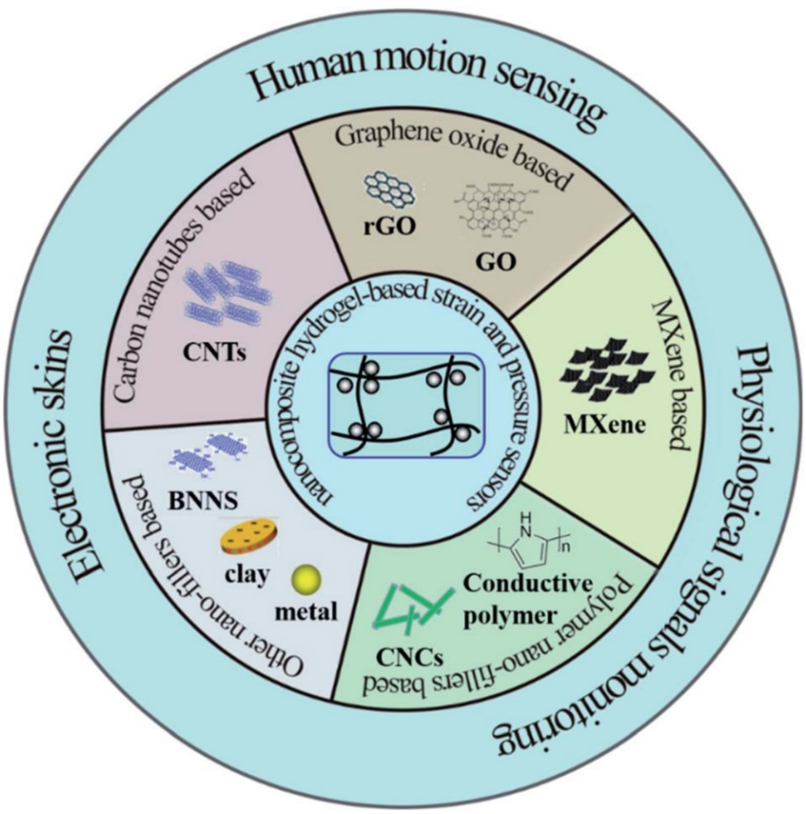

Fig. 1 Schematic illustration of five categories of nanocomposite hydrogel-based sensors and their applications [2].

Hydrogels are three-dimensional polymer networks composed of either natural or synthetic materials, known for their high water content, tunable mechanical properties, excellent biocompatibility, and superior electrical conductivity. These characteristics make them highly suitable for applications in sensors and actuators. Unlike rigid elastomers, their high moisture content facilitates the transport of chemical molecules and conductive substances, boosting conductivity. Moreover, the polymer structure can be customized to closely match the elastic modulus of biological tissues, resembling human skin, which helps reduce mechanical mismatches. Ensuring biocompatibility is essential for applications such as health monitoring and human-computer interaction, as it improves the interface between the hydrogel and biological tissues, reducing the risk of inflammation.

The self-healing ability of hydrogels is another notable feature that enhances their durability, reliability, and lifespan [3]. In electronic devices, combining self-healing with conductivity is crucial. Self-healing hydrogels can repair damage, restoring their structure and function. While the development of these materials is still in its early stages, many systems have been created using reversible covalent interactions, such as disulfide bonds, acylhydrazone bonds, phenylborate bonds, diborate bonds, Diels-Alder reactions, and Schiff bases. Noncovalent interactions, like metal coordination, hydrogen bonding, host-guest interactions, hydrophobic interactions, and π-π stacking, have also been explored. Hydrogel-based sensors show superb promise for use in implantable devices.

The challenge for conductive hydrogels is achieving enough electrical conductivity while maintaining their key properties, enabling precise and efficient signal capture and transmission. Additionally, conventional hydrogels often lack optimal mechanical properties, requiring improved designs for practical use in flexible wearable sensors. Since pure hydrogels generally lack the necessary strength and toughness, dual-network structures or the addition of nanomaterials have been developed to enhance their durability. Most hydrogels are non-conductive, so constructing conductive hydrogels for flexible electronics remains a priority. Conductive hydrogels function through either ionic or electronic mechanisms, with ions or electrons moving through conductive channels. The most effective method for imparting conductivity is through nanocomposites [4]. For instance, Karimzadeh et al. reviewed the properties and strain-sensing efficiency of PVA nanocomposite hydrogels, along with the challenges of strain sensor hydrogels [5]. Park et al. reviewed advances in nanocomposites, focusing on materials, strategies, applications, and limitations. With their biocompatibility and ability to bind biomolecules (e.g., enzymes, antibodies, DNA), hydrogels are ideal for detecting biomarkers in bodily fluids, such as glucose, lactate, and pH levels.

3. Graphene-based flexible sensors

The quality of flexible sensor components largely lies on the inherent characteristics of the conductive materials used. Currently, the most commonly used conductive materials in flexible sensors are metals, conductive polymers, and carbon-based materials. Metallic materials and nanosheets are frequently used in flexible sensor production [6]. Conductive polymers, though employed in flexible sensor manufacturing, tend to have lower conductivity and stability than metals. Despite their advantages, these materials also present challenges in producing high-quality flexible sensors. Carbon-based materials are noted for their high conductivity, flexibility, thermal properties, biocompatibility, and affordability. As such, carbon materials are expected to be highly effective in the development of flexible sensors, shown on Fig.2.

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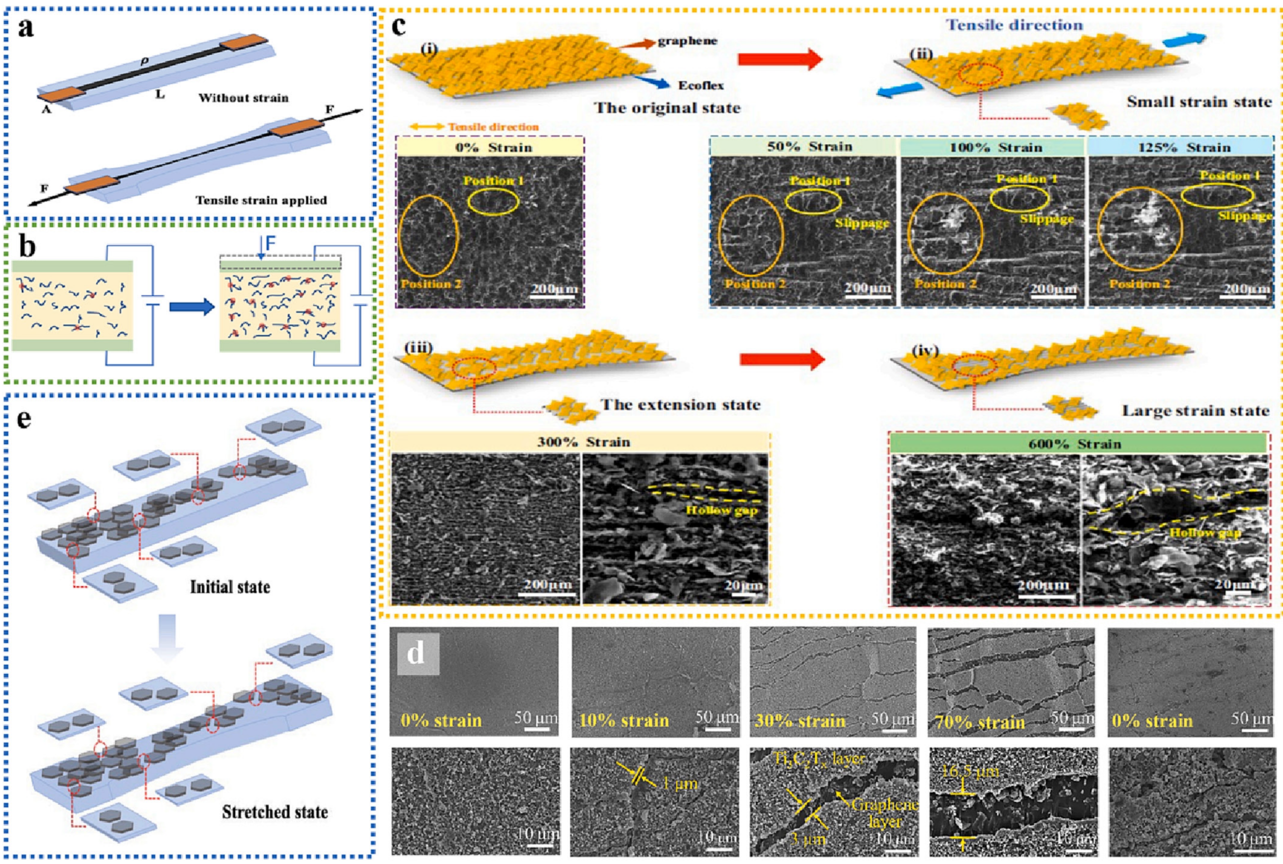

Fig. 2 Sensing mechanisms of stretchable graphene-based strain sensors. a) Geometrical effect. b) Piezoresistive effect. c) Disconnection mechanism. d) Crack mechanism. e) Tunneling effect [7].

In 2004, a significant advancement in graphene production occurred when Geim and colleagues introduced mechanical exfoliation [8]. This breakthrough greatly enhanced graphene output, and with the development of various fabrication techniques, its use in sensors became vital, initially focusing on rigid sensors. As research progressed, more attention was drawn to graphene's stretchability, which held great promise for use in flexible sensors. This led to a fusion of flexibility and rigidity properties, ultimately enabling the creation of electronic skin templates and marking the beginning of integrated sensors. In recent years, flexible sensors have become more refined, driven by innovative applications of graphene composites and structures, often through interdisciplinary collaborations. These sensors now have a wide range of applications, including healthcare and firefighting, with the energy sector also increasingly adopting custom flexible sensors [9].

The incorporation of graphene fibers in the production of flexible sensors presents numerous benefits, such as lightweight construction, durability, adaptability, and flexibility. Researchers continue to explore methods to enhance the performance of these fibers. The elongated shape of fiber materials allows for seamless integration with graphene nanomaterials, which improves the overall quality of sensors. For example, Zhang et al. developed a single-fiber strain sensor with a silk fiber core and graphite flake sheath, leveraging silk's natural flexibility and biocompatibility. This sensor operates within a 15% strain range, has a sensitivity of 14.5, and maintains stability across 3000 cycles, offering a cost-effective and eco-friendly solution. Li et al. created a dual-coaxial strain sensor using a polyurethane yarn core and a graphene-polyvinyl alcohol sheath, providing high elasticity, thermal stability, and adjustable electromechanical properties, with sensitivity levels of 28.5 and 86.9. On the other hand, Souri et al.'s natural fiber yarn sensor had lower stretchability (60%) and faced sensitivity limitations due to the viscoelastic properties of Ecoflex. Wang et al. introduced a sophisticated strain sensor featuring an optical fiber structure made from polydimethylsiloxane (PDMS) and graphene, capable of measuring strain via optical loss, with a high tensile strength of 150% and water resistance, making it suitable for robotics and wearable technology. Ma et al. devised a sensor using graphene-coated glass fibers embedded in PDMS, offering a straightforward approach for assessing flexible devices.

Furthermore, Li et al. pioneered conductive fabrics made from reduced graphene oxide through chitosan electrostatic self-assembly. Graphene-based flexible sensors hold great promise in areas such as wearable electronics, health monitoring, biomedical engineering, and smart materials. Future research will focus on improving scalability, durability, and practical stability [10].

4. Janus Membrane-Based Wearable pH Sensor

Wang et al. developed a breathable Janus membrane-based pH sensor that is self-adhesive on the skin, thereby facilitating long-term stable skin pH monitoring, as shown on Fig.3 [11]. An ultrathin hydrophobic polyurethane (PU) - polydimethylsiloxane (PDMS) porous substrate was combined with a hydrophilic polyvinyl alcohol (PVA) - polyacrylamide (PAA) porous nanofiber layer to form a self-adhesive Janus membrane for stable pH monitoring. The PU-PDMS porous substrate, due to its fiber-reinforced porous membrane structure, has an ultrathin thickness of 700 nm ~ 1.8 μm and a 78.5 ± 8.0 μJ/cm² self-adhesion, as well as exhibiting good water transport, hydrophobicity and breathability. The successful monitoring of skin pH for 7.5 hours demonstrated the feasibility of continuous long-term pH monitoring without compromising comfort.

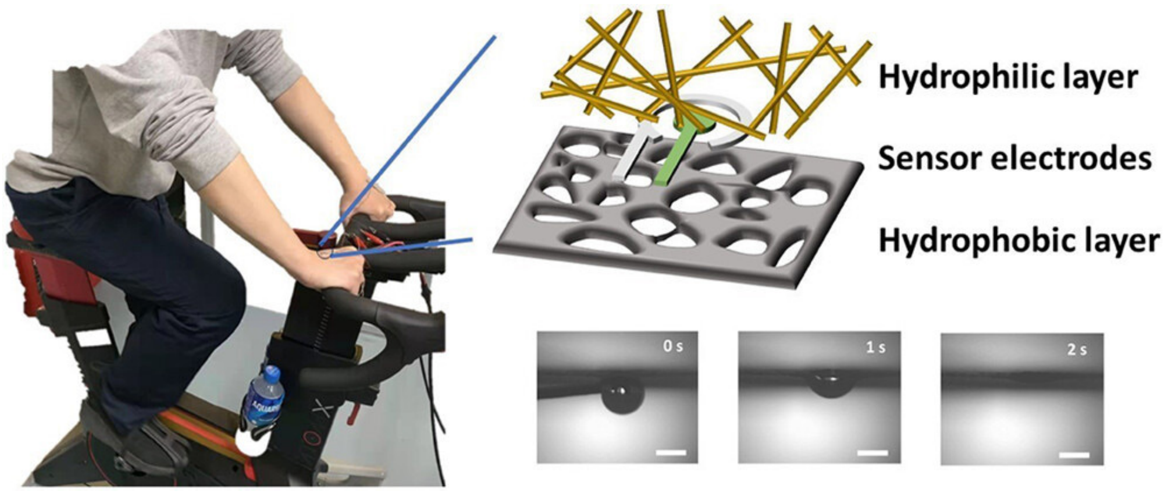

Fig. 3 Janus Membrane-Based Wearable pH Sensor [11]

The preparation and characterisation of each layer of Janus membrane were conducted as discrete processes. The substrate layers were designed to exhibit a porous structure, hydrophobicity and an ultra-thin thickness. The dip-coating method was employed in order to fulfil the requisite specifications. It was demonstrated that dip-coating polyurethane (PU) nanofibres in a hexane-diluted polydimethylsiloxane (PDMS) solution resulted in the formation of a non-porous film with a thickness of several hundred nanometres. In order to further elucidate the solution absorption properties of Janus films, it is evident that the hydrophobic PU-PDMS substrate must exhibit sufficient pore density for water transport and a suitable water contact angle. An increase in the extraction rate and a reduction in the film thickness result in the PDMS becoming insufficiently thick to maintain a continuous morphology at specific locations on the membrane, thereby causing the formation of pores. As the rate of dip coating was decreased, the water contact angle remained within a similar range due to the thinning of the PDMS film and the increase in the pore ratio. The alteration in contact angle may be associated with a partial transition between the Cassie-Baxter and Wenzel states. From the SEM images taken at an angle of tilt, it can be observed that the porous films prepared at a dip-coating rate of 20 mm/s exhibited a thickness range of 700 nm to 1.8 μm. Please refer to Supporting Information 1 for a detailed analysis of the impact of dip-coating speed. The adjustment of the dip-coating speed enabled the hydrophobic PU-PDMS substrate to achieve the ultra-thin porous structure required for the formation of Janus membranes.

Another layer of the Janus membrane is a hydrophilic porous layer, formed by electrostatically spun hydrophilic PVA-PAA nanofibers. It is typical for hydrophilic materials with hydroxyl groups, such as PVA, to exhibit hydrophilicity, which results in macroscopic hydrophilicity. Conversely, the hydroxyl groups are readily attracted to water molecules, forming hydrogen bonds that lead to polymer chain breaks, resulting in a fully water-soluble material (Wang et al.). The objective was to create an insoluble hydrophilic material by cross-linking PVA and PAA. The hydroxyl groups of PVA and the carboxylic acid groups of PAA can form a cross-linking structure, forming a carbonyl group. As the number of hydroxyl groups de-

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creases, hydrophilicity tends to decrease and water stability increases. The hydrophilicity and water stability of the nanofiber layer were balanced by a ratio of 7.3:2 between PVA and PAA, with an electrostatic spinning time of 4 hours. The water contact angle of the hydrophilic nanofiber layer was observed to be 13°. To gain further insight into the chemical modification of PVA, Fourier transform infrared spectroscopy (FTIR) measurements were employed to observe the structure of PVA. In comparison to hydrophilic PVA nanofibres, there was a notable decrease in the intensity of the hydroxyl peak and an increase in the carbonyl peak. The fibre density increased in line with the extension of the electrostatic spinning time.

The Janus membrane's permeability allows for the unrestricted passage of gases through one side of the membrane, thereby ensuring the stability of the sensor in situations where a gaseous environment must be detected. This is of particular importance for real-time, non-invasive environmental monitoring. The incorporation of a pH-sensitive material, such as polymers or nanomaterials with acid-base responsive properties, into the Janus membrane enables the sensor to respond to pH fluctuations in the surrounding environment. To illustrate, certain materials alter their electrical properties or colour in response to fluctuations in pH, thereby facilitating the detection and quantification of pH.

5. Disscussion

Wearable flexible sensors for medical applications have a large amount of potential applications in healthcare, making real-time monitoring of various physiological signals and supporting personalized medicine, remote monitoring possible. However, despite the great promise of this technology, it still faces a series of challenges in its application, while also presenting many opportunities for innovation.

Medical-grade sensors must be highly accurate to ensure the reliability and accuracy of monitoring results. For example, when monitoring data such as electrocardiograms, oxygen saturation, and blood glucose, small deviations in accuracy may affect diagnostic results. Flexible sensors, when attached to human skin, may be affected by factors such as sweat and skin movement, which may reduce the accuracy of the signal. In practice, flexible sensors are susceptible to external factors such as electromagnetic interference, temperature drift, and human movement, resulting in signal distortion or increased noise, which can adversely affect critical medical monitoring. And flexible sensors usually need to be attached to the human body for a long time, especially in chronic disease monitoring, which requires good long-term stability of the material. The durability of the material and the maintenance of its performance during repeated bending, stretching, and compression is a challenge. Prolonged use may cause fatigue, damage, or material deterioration of the sensor, affecting its monitoring capabilities. Flexible sensors are in direct contact with human skin and therefore need to have good biocompatibility to prevent sensitization, skin irritation, or inflammation. Additionally, the breathability and water resistance of the sensor is an important factor in improving comfort and avoiding infections.

The effective analysis and interpretation of data in order to extract meaningful information while avoiding the inclusion of redundant data represents a significant challenge in the field of data processing. Furthermore, the analysis of data in real time is of paramount importance for the prompt formulation of medical decisions. The realisation of real-time data transmission and analysis of flexible sensors also represents a significant challenge. It is of great importance that flexible sensors are designed with low power consumption in order that they may be used over extended periods of time. The operational requirements of medical sensors frequently necessitate continuous functionality for extended periods, which places considerable demands on the sensor's power consumption. The reduction of energy consumption while maintaining high sensitivity represents a significant technical challenge.

6. Conclusion

Despite the challenges, compared to traditional invasive medical monitoring methods, wearable devices based on flexible sensors are able to perform biosignal monitoring, such as biomarker detection in sweat and tears, in a non-invasive manner. This method greatly reduces pain and discomfort and is suitable for long-term monitoring applications.

In comparison to traditional medical devices, flexible sensors are lightweight and small enough to fit snugly against the skin, thereby providing a more comfortable wearing experience. The thin-film structure of the sensor can be integrated into wearable devices, such as smart clothing and patches, without affecting the patient's daily activities. Furthermore, the sensor can be integrated with a variety of sensing functions, including pressure, temperature, humidity, and biochemical detection, enabling comprehensive monitoring of multiple health indicators. This integrated design optimises the size and complexity of the device, providing users with a more comprehensive health management tool.

The challenges associated with the use of wearable flexible sensors in medical applications can be broadly categorised into four main areas: accuracy, durability, power management and data processing. However, with the ongoing advancement of materials science, electronic technology and data processing technology, these issues are being progressively addressed. The role of flexible sensors in personalised medicine, health monitoring, non-invasive diagnosis and telemedicine is set to become increasingly significant. They will facilitate more convenient, accurate and efficient health management tools.

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