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Advances in bio-based wearable flexible sensors

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Amidst rising environmental consciousness and the proliferation of wearable technology, bio-based wearable flexible sensors, crafted from biomass, biodegradable, and biocompatible materials, have emerged as a promising field. These sensors are lauded for their eco-friendliness and comfort, positioning them at the forefront of industry trends. The increasing number of research papers in this field has become an indicator of their industrial potential, so it is time for us to summarize the recent research progress on bio-based wearable flexible sensors. This review begins with an exploration of bio-based materials, categorizing them into natural polymers and synthetic polymers and highlighting key examples utilized in sensor fabrication. We then outline various sensor mechanisms and fabrication techniques, evaluating their distinctive features. The work proceeds to enumerate the applications of these sensors, emphasizing their utility in health monitoring, exercise tracking, and the realm of smart clothing. The paper concludes by identifying current challenges and proposing an outlook for future research directions. The aim of this paper is to synthesize the latest research results on these innovative sensors and provide insights to facilitate their wider applications. At the same time, expanding the development potential of bio-based materials enables a win-win situation between the scale-up of biomass applications and the preparation of wearable flexible sensors.

Green foundation

1. Bio-based materials offer an eco-friendly, biodegradable, and biocompatible option, supporting sustainable development and shaping materials science trends. Traditionally used for chemicals and fuels, biomass now extends to wearable sensors, enhancing its utility and development prospects.
2. Wearable sensors, made from flexible materials, improve functionality across healthcare and environmental monitoring. Bio-based materials provide a stable, comfortable, eco-friendly, biodegradable, and biocompatible solution, compensating for the shortcomings of traditional materials.
3. Bio-based sensors will expand beyond health and smart clothing to aerospace, human-computer interactions, and e-skin. We have summarized material selection, preparation methods, and application scenarios to guide future research.

1. Introduction

Sensors, as vigilant sentinels in the realm of monitoring, are devices designed to measure, transmit, and transform collected data into readable signals following specific patterns. Currently, the traditional sensors are fabricated from rigid materials, which often falter when it comes to softness, comfort, and ductility. These attributes deviate significantly

from the evolving demands of the market. In contrast, flexible sensors utilize materials that possess a degree of softness and bendability, enabling them to detect pressure and contour variations of objects with enhanced utility and a more extensive range of applications.^{1–5}

Combining the benefits of flexible sensors and wearable device design, the emergence of wearable flexible sensors has taken a significant leap forward.^{6–10} This design philosophy aims to ensure a comfortable wearing experience while achieving precise bio-signal monitoring. Characterized by their adaptability, real-time monitoring capabilities, accuracy, and portability, these sensors have found utility in various domains, including healthcare, environmental monitoring, health management, and human-computer interactions.^{11–14} In the healthcare sector, wearable flexible sensors are adept at monitoring vital signs and physiological parameters such as heart

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rate, blood pressure, and blood glucose levels, offering crucial data support for medical diagnosis and treatment.^{15–19} In the realm of environmental monitoring, these sensors are indispensable for tracking key parameters such as temperature, humidity, ultraviolet radiation intensity, concentrations of harmful substances, and ambient light levels. They empower users to make well-informed decisions, stay updated on environmental conditions, and implement proactive measures to mitigate risks associated with conditions like sunburn and skin cancer.^{20–23} Furthermore, with the development of the smart city concept, wearable flexible sensors also provide technical support for its rapid realization. In the realm of human-computer interactions, these sensors are paving the way for the development of touch screens, gesture recognition systems, and smart home solutions, thereby ushering in an era of more intuitive and supple human-computer interactions. For instance, sensors integrated into gloves or wristbands can enable gesture-controlled smart home management.^{24–31} Notably, wearable flexible sensors can be incorporated into smart wearable devices, such as smart watches and fitness bands, which are designed to track user movement and health.^{32–38} However, there is an imperative need to enhance the stability of their signals and to expand their application on a larger scale.

In addition, the conventional composition of wearable flexible sensors often used, such as conductive materials, nano-materials, conductive fibers, *etc.*, is not only pervasive but also problematic due to their environmental impact and contribution to electronic waste. These materials often fall short in terms of biocompatibility and are ill-suited for prolonged contact with the skin.^{39–42} In contrast, bio-based (including biomass, biodegradable and biocompatible) materials, derived from biomass like plants, animals, and microorganisms, offer a more stable, comfortable, eco-friendly, biodegradable, and biocompatible alternative.^{43–51} They align well with the contemporary thinking of sustainable development and are poised to shape the future development trend of materials science.⁵² At the same time, biomass can usually be made into materials to be used, such as chemicals, fuels, *etc.*, but this precisely limits the applications of biomass. The emergence of bio-based wearable flexible sensors has led to broader utilization of biomass as a material and increased the development potential of biomass applications. Consequently, the exploration of bio-based materials for the fabrication of wearable flexible sensors has garnered significant research interest. In addition, there is a burgeoning array of finished products featuring wearable flexible sensors crafted from bio-based materials. By searching the Web of Science, for the keywords 'bio', 'wearable', 'flexible' and 'sensor', a remarkable exponential growth in research and development within this sector over the past two decades can be noted. Furthermore, the surge in published articles within the last five years has reached a zenith, garnering extensive global interest (Fig. 1). For example, Pal and co-workers prepared a bio-based sensor that specifically detects dopamine and ascorbic acid using biodegradable filipin protein as a carrier.⁵³

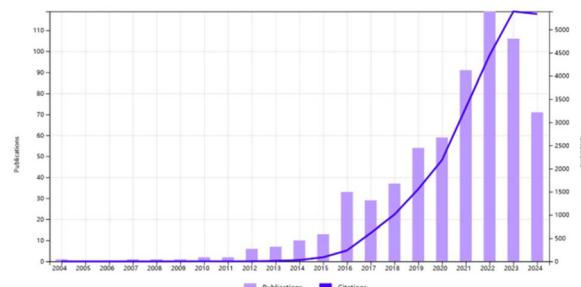


Fig. 1 Statistics of the last decade of research on bio-based wearable flexible sensors.

In this review, we provide a systematic summary and critical analysis of the current research into bio-based wearable flexible sensors (Fig. 2). The review encompasses an overview of the materials used, including natural polymers like alginate, chitosan, gelatin, cellulose, *etc.*, and synthetic polymers such as poly(hydroxylalkanoates) (PHA), poly(lactic acid) (PLA), poly(caprolactone) (PCL), *etc.*, the operational mechanisms (piezoelectric, piezoresistive, and capacitive sensing), and the preparation methods (printed, braided, and coated wearable flexible sensors). Additionally, we summarize their current applications in health monitoring, exercise tracking, and smart clothing, while providing comments on the present state and prospective evolution of this domain. This review aims to offer a reference for the large-scale application of bio-based wearable flexible sensors.

2. Selection of bio-based materials

The selection of materials for crafting wearable flexible sensors is a critical factor influenced by the intended application, inherent characteristics, and the associated production costs.

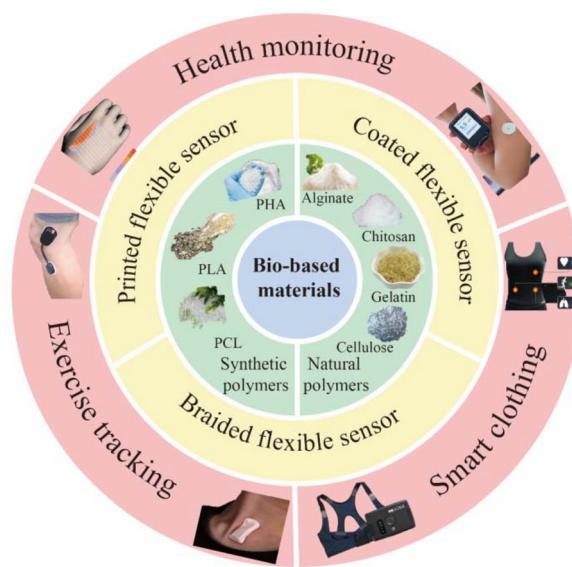


Fig. 2 Materials, preparations, and applications of wearable flexible sensors from bio-based materials.

Traditionally, the fabrication of flexible sensors has relied on three primary categories of materials, including metals, carbon-based materials, and inorganic materials.⁵⁴ While these options exhibit superior conductivity and stability, which are pivotal for enhancing sensor sensitivity, they are not without their drawbacks. Complex preparation processes, high costs, and inadequate comfort during wearing pose significant challenges, which are further compounded by their environmental impact.^{55–58}

In response to these limitations, bio-based sensor manufacturing has become a clear trend. This paradigm shift has prompted researchers to explore innovative avenues in materials science, as highlighted in the discussion that follows. The focus is on two types of bio-based materials – natural polymers (alginate, chitosan, gelatin, and cellulose) and synthetic polymers (PHA, PLA, and PCL) – emphasizing their potential to redefine wearable sensor technology.

2.1. Natural polymers

Natural polymers represent a significant class of degradable polymers, prized for their renewability and non-toxicity.⁵⁹ Among the plethora of natural polymers, alginate, chitosan, gelatin, cellulose,⁶⁰ hemicellulose,⁶¹ silk fibroin,⁶² and lignin⁶³ stand out for their abundance and cost-effectiveness, with each polymer offering distinct benefits. In this review, we specifically focus on alginate, chitosan, gelatin, and cellulose due to their unparalleled contributions to sensor performance.⁶⁴ Below, we provide a concise description of these four materials.

Alginate, predominantly sourced from brown algae, stands out for its abundant availability and good flexibility. This natural polymer facilitates the creation of bio-based sensors that can conform to various shapes and movements, thereby enhancing their versatility. Its compatibility with other materials makes it an excellent candidate for combining with others, expanding its applicability.⁶⁵ For instance, Wang and co-workers developed a graphene-based flexible temperature bio-based sensor that leverages alginate's ability to disperse graphene nano-platelets (GnPs) by reducing van der Waals forces to create a bio-based sensor that can adhere closely to the human skin and monitor temperature variations effectively (Fig. 3A).⁶⁶

Chitosan, derived from the deacetylation of chitin, is a poly-saccharide that offers superior flexibility and wettability, as well as antimicrobial properties. These characteristics make it an ideal material for bio-based sensor fabrication, particularly when combined with other substances to augment the piezoelectric effect.⁶⁷ Hosseini and co-workers developed a glycine-chitosan-based pressure bio-based sensor that not only monitors pressure but also demonstrates enhanced flexibility and piezoelectric properties, with the chitosan component playing a crucial role in controlling the crystalline selectivity of glycine and improving sensor performance. This bio-based sensor exhibits an output voltage of 190 mV at a pressure of 60 kPa, with a sensitivity of 2.82 ± 0.2 mV kPa⁻¹, showing its potential in biological diagnostics and wearable technology (Fig. 3B).⁶⁸

Gelatin, a natural polymer derived from hydrolyzed animal collagen, is valued for its sensing capabilities. Its hydrophilic nature makes it a popular choice for composite materials (Fig. 3C).⁶⁹ Wang and co-workers exploited gelatin's properties to develop carbon microtubes (CMTs)/KCl/gelatin composite, harnessing its conductive and hydrophilic attributes to monitor temperature and pressure.⁷⁰ Furthermore, gelatin's ability to form a gel structure positions it as a key material in the fabrication of flexible bio-based sensors. Wang and co-workers demonstrated this by creating self-powered wearable sensors that capitalize on the interaction forces and hydrogen bonding between gelatin and tannic acid, resulting in a sensor with an impressive stretchability of 1600% and self-healing capabilities.⁷¹

Cellulose, the principal constituent of plant cell walls, is abundant in cotton and hemp plants, and is also synthesized by certain bacteria and algae. This versatile material boasts a range of favorable properties, including high mechanical strength, relative thermal stability, biocompatibility, low cost, and environmental sustainability, making it a popular choice for the fabrication of wearable flexible sensors. For instance, Su and co-workers capitalized on the abundance of oxygen-containing functional groups in bacterial cellulose (BC), which confers exceptional hydrophilicity and mechanical strength due to hydrogen bonding (Fig. 3D).⁷² By combining BC with Ti₃C₂Tx nanosheets, they developed thin film pressure sensors capable of detecting not only conventional human body movements such as finger pressing and nodding but also subtle laryngeal muscle movements as well as discerning voice tones.

2.2. Synthetic polymers

Synthetic polymers, crafted through chemical synthesis and related methods, are recognized for their biocompatibility and biodegradability, offering a viable alternative to traditional materials (e.g. metals, carbon-based materials, and inorganic materials) in the realm of wearable bio-based sensors. These polymers can be eventually degraded into water and carbon dioxide within natural environments, aligning with the principles of environmental sustainability. They have been extensively utilized across diverse sectors, including pharmaceuticals, food science, and environmental protection. While a variety of synthetic polymers have been developed, this review specifically highlights PHA, PLA, and PCL owing to their exceptional biocompatibility, biodegradability, flexibility, stability, and favorable processing characteristics. These materials can be effectively combined with conductive substances to create high-performance composites, which have found extensive application in the development of wearable flexible sensors.

PHA, alternatively synthesized by microorganisms, is a biodegradable polymer with innate antibacterial properties, effective against bacterial cell walls and viral cell membranes (Fig. 3E).⁷³ Sha and co-workers developed a PHA-based hydrogel sensor for attachment to the human body, capable of accurately tracking motion while inhibiting bacterial growth. This hydrogel demonstrated remarkable efficacy against *E. coli* (killing rate: 98%) and MARSA (killing rate: 100%), showcasing

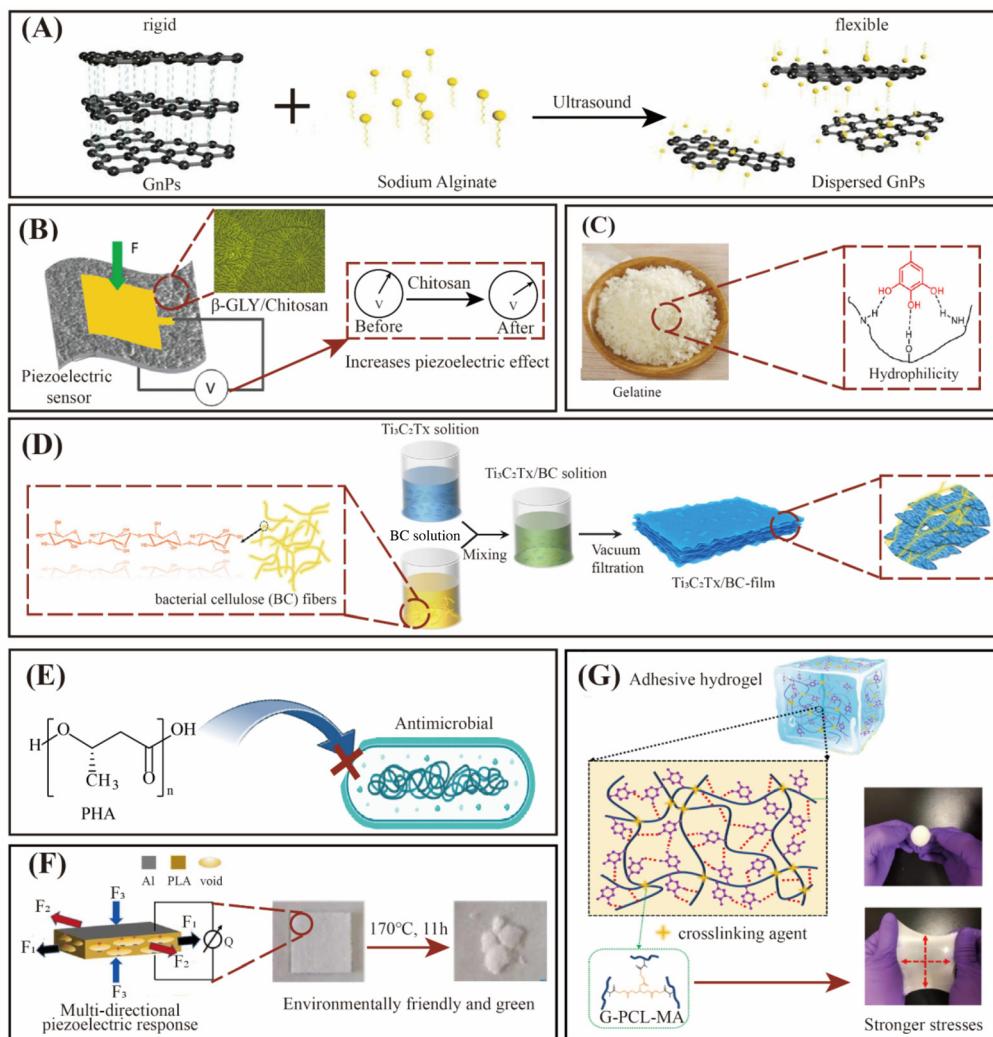


Fig. 3 (A) Alginate can increase flexibility in combination with other substances. Reprinted with permission from ref. 66. Copyright (2019) Springer. (B) Chitosan increases the piezoelectric effect of sensors. Reprinted with permission from ref. 68. Copyright (2020) American Chemical Society. (C) Hydrophilicity of gelatine. (D) Cellulose is hydrophilic and combines with other substances to increase mechanical strength. Reprinted with permission from ref. 72. Copyright (2022) American Chemical Society. (E) The molecular formula of PHA and its antimicrobial properties. (F) Sensors made from PLA are environmentally friendly and green. Reprinted with permission from ref. 76. Copyright (2022) Elsevier. (G) PCL can be used as a crosslinker to increase sensor stress. Reprinted with permission from ref. 82. Copyright (2021) Elsevier.

its potential in prolonged sensor applications, particularly for conditions like rheumatoid arthritis monitoring.⁷⁴

PLA is an aliphatic polyester noted for its affordability, availability, and accessibility. Its molecular structure, featuring hydroxyl and carboxyl groups, facilitates complete environmental degradation through hydrolysis and microbial action.⁷⁵ Additionally, PLA is easy to hydrolyse. On the one hand, the ester chains in PLA can react with water molecules to produce smaller lactic acid molecules; on the other hand, hydrolytic enzymes secreted by fungi and bacteria can break down PLA into hydroxy acids for further degradation. The easily hydrolyzable characteristics have been capitalized upon by Ma and co-workers, who developed a green piezoelectric sensor from PLA. This sensor, designed for attachment to the human body, effectively monitors bodily movements such as finger bending

and foot motion, leveraging PLA's lightweight, flexibility, and superior sensing capabilities. Moreover, its biodegradability allows for complete degradation under mild conditions (in deionized water at 170 °C for 11 h), exemplifying its eco-friendly nature (Fig. 3F).⁷⁶ The porous structure, flexibility, and hydrophilic nature of PLA have also been harnessed by Li and co-workers, who developed a bio-based humidity sensor. By integrating pleated graphene nanosheets, which are sensitive to moisture, they created a sensor capable of monitoring human respiration and skin humidity with high sensitivity.⁷⁷

PCL is a biodegradable polyester with a slower degradation rate compared to PLA, making it particularly suitable for applications where prolonged stability is required.⁷⁸ PCL is internally connected by ester bonds and is susceptible to ester exchange reactions during application and can be modified

into various cross-linkers.^{79–81} Zhang and co-workers exploited the ester cross-linking reactivity of PCL to develop a sensor with enhanced fatigue resistance and mechanical characteristics (stress: 50.2–72.4 kPa, strain: 700–1140%, Young's modulus: 8.6–14.8 kPa, and toughness: 16.4–53.6 kJ m^{−3}), suitable for a broad range of human activity monitoring (finger bending, frowning, *etc.*) (Fig. 3G).⁸²

3. Mechanism of operation and common preparation methods

3.1. Operational mechanism of sensors

Sensors function based on distinct mechanisms, broadly categorized into piezoelectric, piezoresistive, and capacitive sensing.⁸³

Piezoelectric sensors harness the piezoelectric effect, transforming mechanical stress or strain into quantifiable electrical voltage.⁸⁴ Upon the application of external pressure or deformation, these sensors undergo a change in charge distribution across the sensing element, thereby generating a voltage proportional to the applied pressure or deformation.⁸⁵ Characterized by their high sensitivity and the absence of a requisite power source, these sensors swiftly produce electrical signals in response to minute changes, making them ideal for monitoring subtle human movements, such as pulse and finger flexion.⁸⁶

Piezoresistive sensors parallel the operating principle of piezoelectric sensors, yet they detect mechanical changes by the alteration of material resistance upon deformation. This resistance change is subsequently transduced and detected by an electrical circuit.⁸⁷ Noted for their real-time monitoring capabilities and high sensitivity, piezoresistive sensors are commonly employed in medical applications and human-computer interactions.⁸⁸

Capacitive sensors have gained prominence for their rapid response times, minimal power consumption, and robustness against temperature fluctuations.⁸⁹ Comprising electrodes and a dielectric material, these sensors operate on the principle that capacitance varies with the electrode separation or overlapping area. Capacitance (C) is defined by the following equation:⁹⁰

$$C = \frac{\epsilon A}{d}$$

where ϵ is the permittivity of the dielectric material, A is the area of electrode overlap, and d is the distance between the electrodes. When subjected to an external force, the capacitance alters due to changes in the distance or area between the plates, while the dielectric constant remains constant. This mechanism endows capacitive sensors with greater stability and durability, as they lack moving parts that could potentially lead to damage.

3.2. Common classification of preparation

3.2.1. Printed flexible sensors. Printed flexible sensors typically utilize the flexible material in the sensor as the printing substrate. Lithography and various printing techniques are commonly employed in their fabrication. Lithographic printing, while precise, demands stringent conditions and complex

processes, leading to higher production costs. Conversely, direct printing methods are more cost-effective and straightforward, making them suitable for the large-scale production and ongoing development of flexible sensors.^{91–95}

Printing technologies are typically categorized into template-less printing, which includes 3D printing and inkjet printing, and template printing, such as screen printing. 3D printing constructs three-dimensional objects layer by layer, offering a high degree of customization. For instance, Fortunato and co-workers used 3D printing to fabricate piezoresistive pressure sensors for monitoring heartbeats before and after aerobic exercises. These sensors exhibit high sensitivity, with a maximum sensitivity of 0.088 kPa^{−1} under pressures below 10 kPa, and 0.24 kPa^{−1} under pressures exceeding 80 kPa, respectively.⁹⁶ Despite its advantages, 3D printing is relatively slow, limiting its widespread adoption.

Inkjet printing uses fine nozzles to deposit ink onto a substrate, resulting in high print quality and compact equipment suitable for various media (Fig. 4A). Won and co-workers took full advantage of this technology and reported a flexible bio-based humidity sensor based on cellulose nanofibres (CNF) and silver nanoparticles (AgNP). CNF provided satisfactory humidity sensing performance in the bio-based sensor. The bio-based sensor's electrode pattern was created by reverse offset printing of silver nano-ink onto the interdigitated electrode (IDE), followed by inkjet printing of the CNF-AgNP composite onto the IDE to form the sensing layer. This process enabled the sensor to exhibit enhanced sensitivity to humidity changes, with response and recovery times of 4 s and 43 s, respectively, over a 35% to 70% RH range.⁹⁷ Inkjet printing also allows for the production of thin-film sensors with higher sensitivity and quick response times, making them highly portable. Zhang *et al.* developed a transparent polyester (PE) film for monitoring human sweat, demonstrating a rapid 95% response rate within 100 s (Fig. 4B).⁹⁸

Template printing encompasses methods like screen printing, embossing, aniline printing, and intaglio printing. Screen printing is particularly popular due to its versatility and cost-effectiveness. The basic principle of screen printing is the transfer of ink to a substrate through a screen. The stencil can be used to design any pattern, and unwanted areas are blocked to prevent the ink from being deposited on the substrate. The ink is first projected evenly onto the mesh and then a blade is moved to diffuse the ink into the openings of the stencil (Fig. 4C). Carbon black (CB) is a typical printing material due to its excellent conductivity.⁹⁹ Tachibana and co-workers developed a printable flexible sensor from CNFs and CB using screen printing. CNFs facilitated uniform mixing with CB, forming an ink suitable for screen printing electrodes onto polyethylene naphthalate (PEN) films (Fig. 4D). The sensor operates on the principle of CNF swelling upon water absorption, which modulates the resistance by disrupting the conductivity of CB. This mechanism enables the sensor to monitor human respiration and fingertip moisture levels effectively.¹⁰⁰ In the meantime, Wang and co-workers prepared a deep eutectic solvent (DES) and used it, along with benzophenone

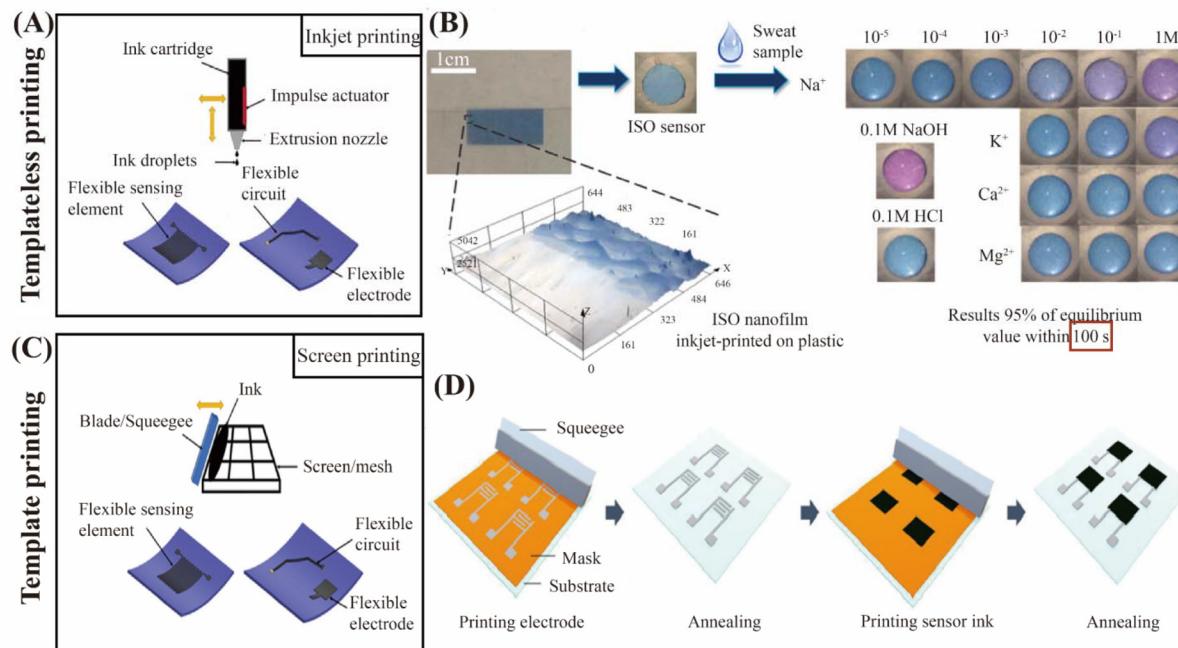


Fig. 4 (A) Schematic diagram of inkjet printing. Reprinted with permission from ref. 36. Copyright (2022) Royal Society of Chemistry Advances. (B) A new strategy for detecting sweat using inkjet printing technology. Reprinted with permission from ref. 98. Copyright (2020) American Chemical Society. (C) Schematic diagram of screen printing. Reprinted with permission from ref. 36. Copyright (2022) Royal Society of Chemistry Advances. (D) Schematic diagram of the preparation of printed flexible sensors using the annealing process. Reprinted with permission from ref. 100. Copyright (2022) American Chemical Society.

none (BP), diphenylamine (DP), polydimethylsilane (PDMS), and CB, to create an ink paste for template printing. After printing the electrodes onto a polyethylene glycol naphthalate substrate, the PDMS was cured, and the DES was removed, yielding flexible electrodes.¹⁰¹ These bio-based sensors can monitor both static and dynamic forces, such as human finger movements, and have potential applications in the development of smart gloves and soft robotics.¹⁰²

3.2.2. Braided flexible sensors. Braided flexible sensors represent an innovative class of sensors that capitalize on the principles of resistance or capacitance change to monitor pressure and other characteristic quantities. The superior flexibility of the materials used allows for the fabrication of sensors in various shapes, thereby increasing the sensing area and enhancing performance.^{103–106}

Inspired by spider webs, Liu and co-workers developed a sensor resembling a spider web using a graphene braid (Fig. 5A). This bio-based sensor is adept at monitoring human physiological signals.¹⁰⁷ In a similar vein, Pan and co-workers prepared a composite yarn for flexible sensors by *in situ* polymerization of polypyrrole (PPy) on poly(dopamine) (PDA)-braided composite yarns (BYs), creating a unique braided structure. This is done as follows: First, wind the polyethylene terephthalate (PET) onto a spool for spinning and feed it into the knitting machine to prepare BYs-PDA; then mix all materials at 0 °C for 2 h to produce BYs-PDA-PPy (Fig. 5B). The subsequent formation of a cauliflower-like PPy layer on the BYs surface resulted in a sensor with an extensive strain range

(strain gauge factor of 50.8 for 0–40% and 29.7 for 40%–105%), capable of monitoring a wide array of human movements and even serving as an electronic heating device in wearable applications, thus expanding its potential uses.¹⁰⁸ Concurrently, Chen and co-workers have developed an unobtrusive and customizable capacitive strain sensor utilizing braided electronic wires made from cotton fibers. Given that cellulose is the primary component of cotton fibers, this confers upon the sensor excellent flexibility and significantly reduces production costs, heralding its potential for mass industrial production. Furthermore, the versatility in the design and application of these electronic wires suggests that this innovation could soon find practical applications in everyday life, thereby advancing the field of human–computer interaction.¹⁰⁹

While these sensors possess considerable potential, their sensing speed and stability have not yet attained the standards necessary for certain high-demand applications. To address this, Zheng and co-workers prepared cotton fabric-based strain sensors (CFSSs) by encapsulating cotton fabric with PDMS. The incorporation of cellulose in this sensor significantly enhances its flexibility, enabling the creation of an interpenetrating structure that dramatically reduces the sensor's response time to a mere 90 ms (Fig. 5C).¹¹⁰ Zhang and co-workers fabricated a braided bio-based flexible wearable sensor with a mesh layered structure by immersing a commercial cotton fabric in a single-walled carbon nanotube (SWCNT) dispersion (Fig. 5D). The cellulose within the cotton fabric

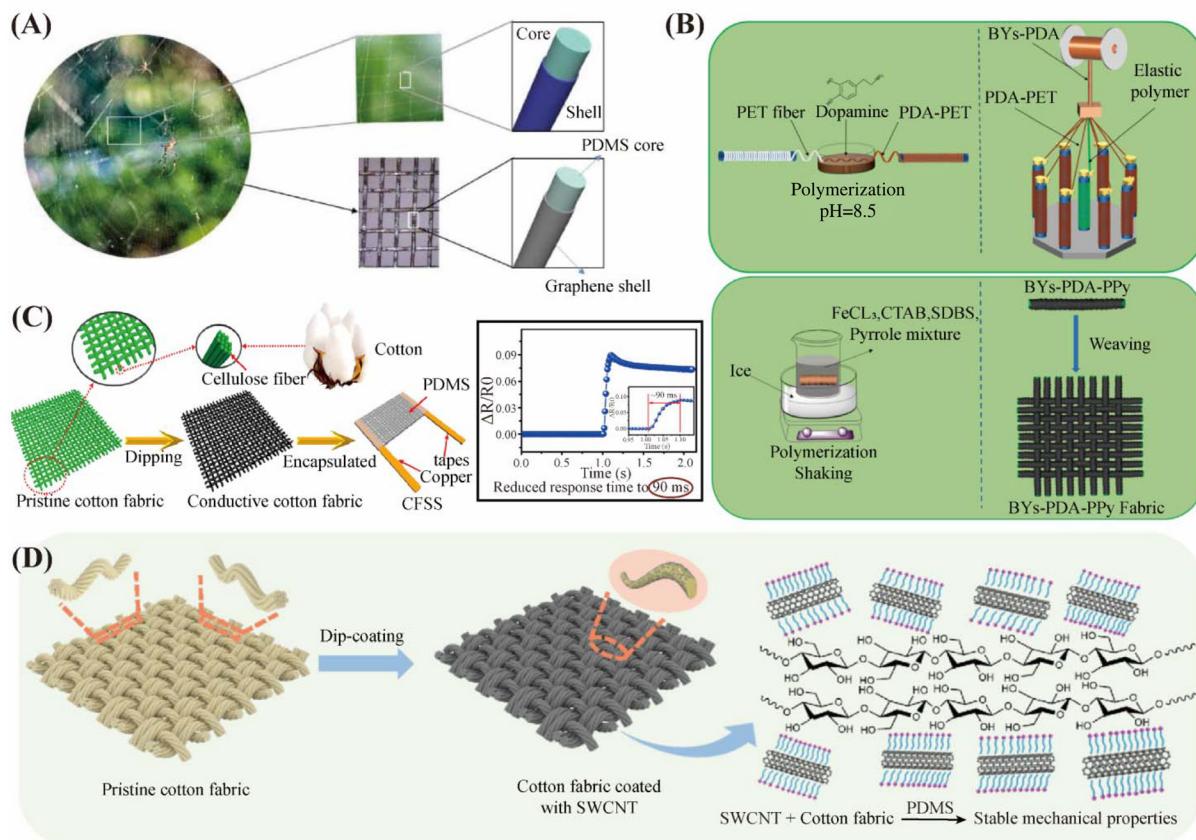


Fig. 5 (A) Illustration of a braided flexible sensor inspired by a spider web. Reprinted with permission from ref. 107. Copyright (2019) American Chemical Society. (B) Preparation of BYs-PDA-PPy. Reprinted with permission from ref. 108. Copyright (2019) American Chemical Society. (C) Schematic illustration of the structure of pristine cotton fabric and the preparation process of the CFSS. Reprinted with permission from ref. 110. Copyright (2020) American Chemical Society. (D) Schematic diagram of a woven flexible wearable sensor with a mesh layered structure fabricated using the dip-coating method. Reprinted with permission from ref. 111. Copyright (2022) American Chemical Society.

exhibits excellent adsorption properties towards the dispersion, which contributes to the fabrication of wearable flexible sensors characterized by a low detection limit of 0.1% and an extensive linear range spanning from 0 to 150%. This design not only allows for the monitoring of a variety of movements but also ensures stability of the structure. The sensor's distinctive mesh-layered structure allows it to endure multi-directional forces and deformations, demonstrating remarkable stability over 50 000 cycles at a 40% tensile strain.¹¹¹

3.2.3. Coated flexible sensors. Coated flexible sensors are a class of sensors that leverage coating technology to fabricate devices capable of monitoring various physical quantities such as pressure, temperature, and humidity. These sensors typically comprise a flexible substrate and a conductive coating, allowing for the detection of changes in capacitance or resistance. Dip coating is primarily achieved by completely immersing an object in the coating material (Fig. 6A). The dipping time depends on the type of coating and the desired coating thickness. The resulting bio-based sensors are not only thinner and lighter but also more discreet, making them highly suitable for wearable applications compared to other sensor types.^{103–115}

Chen and co-workers introduced an innovative process for enhancing the functionality of flexible fibers. They proposed a method that involves dip-coating the fibers with active substances followed by chemical nickel plating (Fig. 6B). This technique significantly improves the electrical conductivity of the composite fibers, which can be tailored for various sensing applications.¹¹⁶ Building upon this approach, Zhang and co-workers utilized a cracked cellulose nanofibril/silver nanowire (CA) to coat polyurethane (PU) sponges, creating CA-PU sponges with controlled conductive and mechanical properties (Fig. 6C). Cellulose is distinguished by its high modulus, and the incorporation of cellulose nanofibers (CNF) enhances the dispersion of silver nanowires (AgNWs) while promoting robust adhesion between the conductive layer and the polyurethane (PU) framework. By adjusting the number of impregnation and coating cycles, they were able to create sensors that form cracks in the conductive CA layer, thereby increasing the effective contact area. Detection limits as low as 0.2%. These sensors were effectively used for monitoring human body movements such as vocalization and bending.¹¹⁷ Qu and co-workers successfully developed a PCL-based fiber with thermoelectrically responsive memory characteristics using the

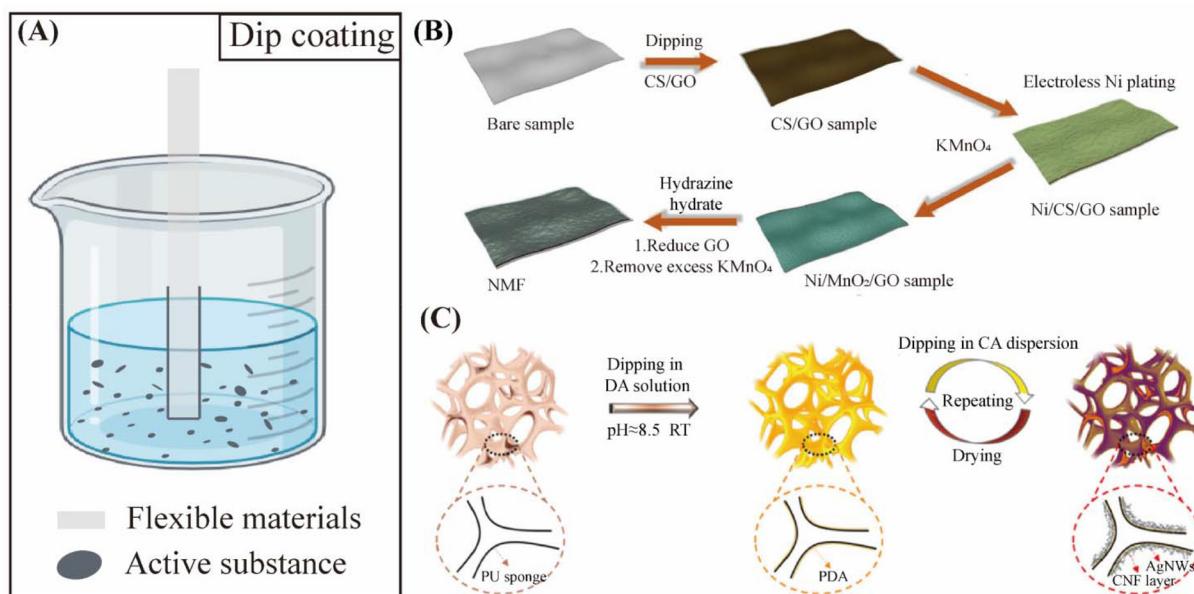


Fig. 6 (A) Schematic of the manufacturing principle of the dip-coated sensor. (B) Preparation of flexible sensors by chemical nickel plating after dip-coating of fibers with an active substance. Reprinted with permission from ref. 116. Copyright (2019) American Chemical Society. (C) Schematic illustration for the preparation of conductive CA@PU sponge. Reprinted with permission from ref. 117. Copyright (2019) American Chemical Society.

impregnation method, capitalizing on PCL's inherent shape memory capabilities. In the experimental process, the PCL composite fiber underwent an impregnation-drying cycle, which involved immersing it in a MWCNT dispersion for 1 min followed by drying at room temperature for 30 min. This method yielded a memory-like fiber that exhibits rapid response and stable cyclic performance. Additionally, the composite fiber boasts a self-healing efficiency of 86%, with the healing process achievable within 600 s. It demonstrates a swift response time of 187 ms and exceptional cycling stability over 2000 cycles. These composite fibers hold great potential for applications in wearable devices, stretchable electronics, and soft robotics.¹¹⁸

4. Typical applications

4.1. Health monitoring

Bio-based wearable flexible sensors have emerged as a pivotal area of research within the medical industry, leading to the development of numerous innovative products. These sensors are celebrated for their straightforward structure, lightweight design, compact size, and rapid technological advancements. With the development of science and technology, these sensors have faster sensing speeds and shorter response times. Such features make them ideal for monitoring human health, including vital signs and body index monitoring.^{119–128}

Natural polymers exhibit significant advantages in biocompatibility and biodegradability, making them highly suitable for health monitoring applications that require real-time tracking. Their reduced potential to elicit immune responses or

inflammation means they are gentler on human skin and tissues, ensuring safety for prolonged use. For instance, Liu and co-workers have developed a hydrogel sensor composed of sodium alginate and starch, capable of real-time sleep breathing monitoring. This hydrogel demonstrates outstanding mechanical strength and long-term stability across a broad temperature spectrum (–20 to 40 °C). It also features a wide operational range (0–1119%), high conductivity (8.55 S m^{−1}), and excellent linearity ($R^2 > 0.99$), making it ideal for wearable electronics. The incorporation of sodium alginate and starch not only helps dissipate energy through deformation but also ensures that these components are biorenewable and biocompatible, mimicking the physiological and mechanical attributes of human skin. Consequently, when this hydrogel sensor is applied to the skin, it maintains its biocompatibility, minimizes the risk of immune reactions and inflammation, and is non-irritating to the human body.¹²⁹ Yao and co-workers revolutionized traditional medical testing by developing a non-invasive wearable dual-electrode blood glucose sensor. This sensor employs reverse iontophoresis (RI) to extract glucose from interstitial fluid (ISF) and monitors glucose concentration amperometrically (Fig. 7A). The two-electrode glucose sensors demonstrated sensitivities that varied between 1.39 and 1.81 $\mu\text{A } \mu\text{M}^{-1} \text{ cm}^{-2}$, with a remarkably low limit of detection for glucose at 0.06 μM . This non-invasive sensor is in line with the current shift towards minimally invasive health monitoring methods, providing results that are highly congruent with those obtained from commercial blood glucose meters. Both electrodes of the sensor are composite textiles, which can be attached to human skin for semi-continuous monitoring of human blood glucose. At the same

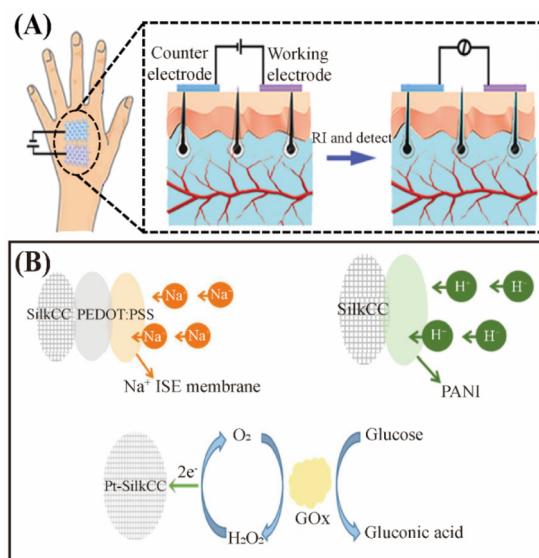


Fig. 7 (A) Structure and working principle of the two-electrode non-invasive blood glucose sensor. Schematic to show extraction of ISF by the RI process. Reprinted with permission from ref. 130. Copyright (2021) Elsevier. (B) Schematic diagram showing the working mechanisms of Na^+ , pH, and glucose sensors. Reprinted with permission from ref. 132. Copyright (2023) Elsevier.

time, because of its simple structure, it can also be used in conjunction with other wearable devices, as an important sensing element of joint devices, to jointly monitor human body indicators and manage human health.¹³⁰

Sweat, being more accessible than other bodily fluids and rich in health-related biomarkers, presents an alternative medium for health monitoring. Wang and co-workers have developed a wearable flexible sensor utilizing chitosan to detect cortisol levels in human sweat. Cortisol is a key indicator of psychological stress and overall health in everyday life. The sensor capitalizes on chitosan's porous structure to efficiently collect sweat, leveraging its adhesive qualities and an 80% swelling capacity to offer an economical alternative to microfluidic systems. This innovative method broadens the detection range to 10^{-3} to 10^{-4} nM, with a sensitivity of $189.2 \text{ nA lg(nM)}^{-1}$. This non-invasive, high-sensitivity device introduces a novel pathway for monitoring mental stress and aiding clinical diagnosis, thereby enhancing the tracking of human physiological conditions.¹³¹ Zhang and co-workers expanded the potential of sweat analysis further by developing a wearable sensor capable of simultaneously detecting multiple sweat constituents and indicator, including Na^+ , pH, and glucose (Fig. 7B). This sensor, ingeniously fabricated from all-fabric materials, has shown remarkable capability in collecting up to $0.2 \mu\text{L}$ of sweat with high sensitivity, thereby expanding the potential applications of wearable sensors.¹³²

4.2. Exercise tracking

Bio-based wearable flexible sensors are revolutionizing not only the monitoring of human physiological indicators but

also the detection of movement status, providing invaluable data for exercise tracking. Synthetic polymers offer better benefits in this aspect of motion tracking. Synthetic polymers provide a range of adjustable mechanical properties and can be tailored through the polymerization of monomers. This process allows researchers to design specific structures by carefully choosing the right monomers and polymerization conditions. Such design flexibility permits the customization of physical and chemical attributes to suit a wide array of applications. Furthermore, the degradability of these polymers offers a solution to the environmental issues posed by the electronic waste generated from rigid sensors. A prime example of Yang and co-workers, who employed PHA and other materials to develop ion-conducting hydrogels for use in wearable flexible sensors. These sensors are adept at detecting various human activities with both stability and sensitivity (Fig. 8). The impressive sensitivity and fatigue resistance of these sensors can be attributed to the physical cross-linking *via* hydrogen bonding, a characteristic feature of PHA.¹³³

However, hydrogels can be sensitive to environmental factors like pH, temperature, and humidity, which may lead to performance instability. In high-moisture environments, such as during swimming or bathing, they risk excessive swelling and structural degradation, limiting their practical application in daily life. To address the challenges in sensor technology, Guo and co-workers developed a highly sensitive, flexible, and biodegradable pressure sensor using PLA. This innovative wearable transient pressure sensor was constructed by sandwiching biodegradable PLA sheets with fork-guided electrode patterns between nanosheets, fully leveraging the biodegradable and structure tunable characteristics of PLA as a synthetic polymer. The resulting wearable pressure sensor boasts high sensitivity, a low detection limit of 10.2 Pa, a fast response time of 11 ms, and excellent reproducibility over 10 000 cycles, all while being biodegradable. These fabricated pressure sensors can be attached to human skin to detect human movements.¹³⁴ Additionally, Sui's team invented a wearable flexible pressure sensor based on PLA, a widely used piezoelectric polymer known for its good mechanical and processing properties. By capitalizing on PLA's degradable and structurally tunable nature, the team incorporated barium titanate into PLA to synthesize a composite with a high output voltage of 22.57 V and high sensitivity of 0.176 V kPa^{-1} .¹³⁵

4.3. Smart clothing

Bio-based wearable flexible sensors, characterized by their ultra-thin and highly flexible substrates, are redefining the concept of smart clothing with their lightweight and adaptable form factors. These attributes allow for their integration into various garments, enabling a new generation of attire that is both functional and comfortable. For instance, these sensors can be incorporated into socks or shoe soles to detect changes in plantar pressure during activities like walking or running, providing valuable data for gait analysis and injury prevention. Similarly, they can be embedded into masks to monitor breathing in real-time, offering potential health benefits and

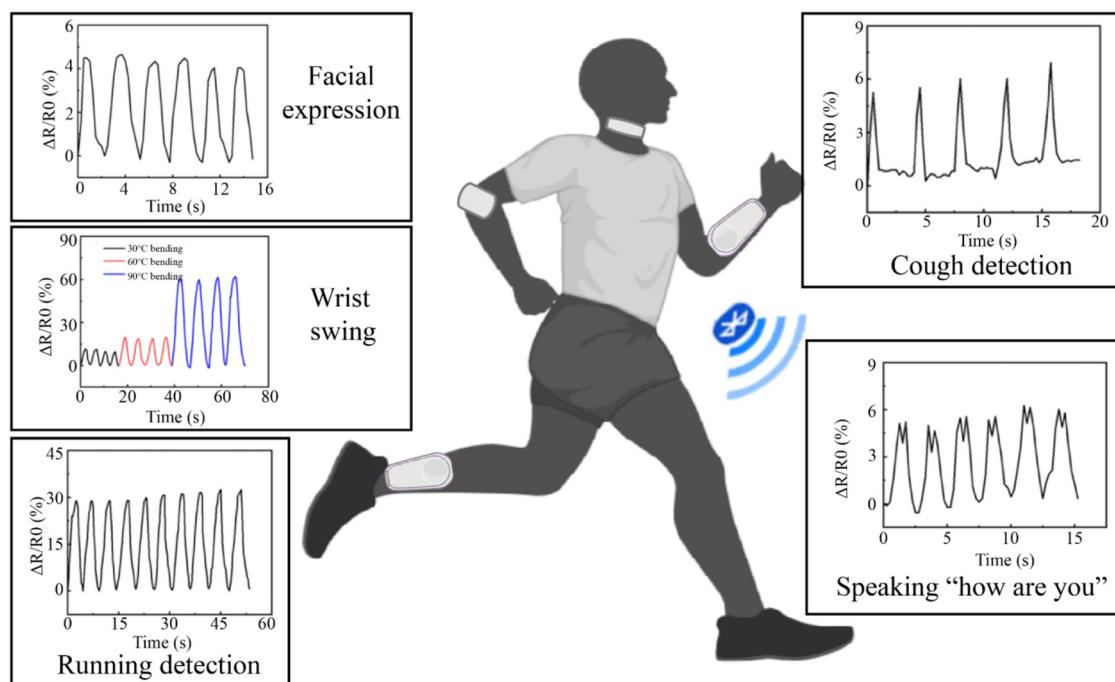


Fig. 8 Wearable flexible sensors detect a person's coughs, facial expressions, wrist swings at different bending, running, and speaking "how are you". Reprinted with permission from ref. 133. Copyright (2021) Elsevier.

enhancing user safety. Given the wide array of finished products and the multifunctional nature of smart clothing, it is essential to carefully choose the most suitable bio-based material tailored to a specific object or intended application. Ryu and co-workers have pushed the boundaries of wearable sensor technology by utilizing a thermoplastic PLA with a thermal drawing process (TDP) to create highly flexible temperature sensors. These sensors exhibit excellent sensitivity in the 25–45 °C range and can be integrated into gloves or other clothing to monitor human body temperature, providing real-time health data (Fig. 9A).¹³⁶ Concurrently, Liu's team has developed a flexible sensor with a high strain coefficient, featuring a cracked structure, which has been ingeniously incorporated into a data glove. This innovative glove is constructed from a biodegradable and elastic gelatin composite layered on a fabric substrate, boasting impressive durability over 6000 cycles and a rapid response time of just 60 ms. The gelatin has been specially modified to achieve a high gauge factor and superior stretchability. Furthermore, the glove is capable of recognizing nine distinct gestures through machine learning, with an exceptional gesture mapping accuracy of up to 99.6%.¹³⁷

However, Zhao and co-workers have developed a capacitive flexible sensor using electrostatic spinning technology, featuring a natural elastic substance, thermoplastic polyurethane (TPU), as a dielectric layer. This sensor, when attached to the wrist, can monitor pulse in real-time, offering health tracking capabilities. Using this technology, the sensor can be braided into gloves, creating a "piano glove" that enables real-time piano playing (Fig. 9B).¹³⁸ The gloves'

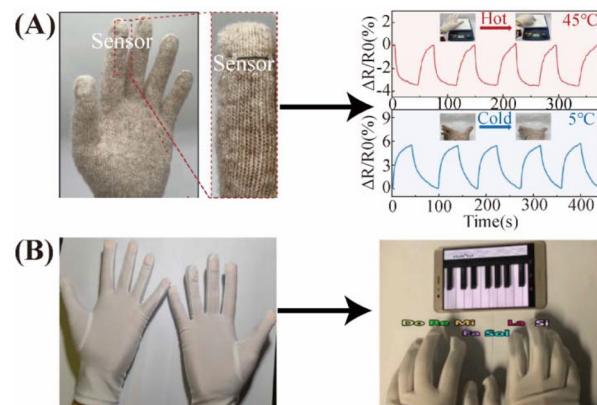


Fig. 9 (A) Image of the fiber temperature sensor sewn onto the tip of a hand glove. Temperature response of the fiber sensor to repetitive touch on a hot (45 °C) or cold (5 °C) object. Reprinted with permission from ref. 136. Copyright (2023) Springer Nature. (B) Photograph of the data glove with ten independent capacitive sensors. Demonstration of playing the piano based on the data glove and displaying in the mobile app. Reprinted with permission from ref. 138. Copyright (2020) American Chemical Society.

sensors correspond to musical notes "do-re-mi-fa-so-la-si" and the high notes "do-re-mi", allowing individual note recognition and integration with smart devices for a seamless musical performance.

This innovative application not only facilitates new possibilities in human-computer interaction but also showcases the potential of wearable sensors to revolutionize the way we interact.

Table 1 Summary of bio-based wearable flexible sensors

| | | Advantages | Disadvantages | Scope of applications | Ref. |
|----------------------|--------------------------|--|--|--|------------------|
| Bio-based materials | Natural polymers | Good molecular structure; chemical properties | Relatively high cost; vulnerable to environmental impacts | — | 59–61 |
| | Synthetic polymers | Adjustable characteristics; low-cost | Simple structure and single-function | — | 62–64 |
| Preparation methods | Printed flexible sensors | Low cost; lightweight and portable | Low accuracy | Mainly suitable for health monitoring and exercise tracking in daily life | 91–93 |
| | Braided flexible sensors | Highly comfortable; breathable | Low sensitivity; high environmental impact | Preparation of smart clothing | 103–106 |
| Typical applications | Coated flexible sensors | High conductivity; high sensitivity; more accurate results | High cost | Ideal for disease diagnosis and areas with a high demand of monitoring results | 112–114 |
| | Health monitoring | Highly comfortable; can be monitored in real-time; easy to operate | Accuracy needs to be improved; range issues | Elderly care; chronic disease surveillance management | 139–142 |
| | Exercise tracking | Real-time monitoring; data analysis | Accuracy is subject to external influences; privacy breaches | Health management; sports training; rehabilitation activities | 143–146 |
| | Smart clothing | Personalized experience; versatility; human–computer interaction | Difficult to maintain and clean; high cost | High-risk workforce; fashion periphery | 137, 138 and 147 |

act with our environment, promoting advancements in both healthcare and entertainment.

5. Summary and outlook

This review focuses on the advances of bio-based wearable flexible sensor materials in three critical aspects: preparation materials, preparation methods, and application scenarios. For a detailed overview, Table 1 encapsulates the advantages, limitations, and application scope of these sensors.

The materials for these sensors are primarily categorized into natural and synthetic polymers. Natural polymers are increasingly utilized due to their intricate molecular structure and chemical properties. Synthetic polymers, with their tunable properties, have paved the way for an expanding array of materials, further diversifying the options available to researchers and engineers.

The fabrication methods of these sensors typically involve printing, weaving, and coating. With these preparation methods, wearable flexible sensors can be prepared as thin films, arrays, various patterns, *etc.* Printed flexible sensors are cost-effective and well-suited for mass production, offering a lighter, more portable option that caters to the everyday health monitoring and motion tracking needs of the general public. The braided flexible sensor adopts textile technology and can be used to create comfortable wearable sensors, making them ideal for smart clothing applications. Meanwhile, the coating can make wearable flexible sensors have higher conductivity, improve the sensitivity of the sensor, and monitor the results more accurately, but the cost is higher, suitable for disease diagnosis and the field of higher requirements for monitoring results.

The versatility of bio-based wearable flexible sensors is underscored by their broad utility in health monitoring, exercise tracking, and smart clothing. These sensors are not only adept at monitoring vital signs like pulse and heartbeat but can also employ various media for the non-invasive surveillance of physiological markers such as lactic acid and glucose levels. Their application extends to tracking fine motor movements like wrist bending and throat swallowing, as well as to the creation of temperature-regulated attire and interactive smart devices that enhance the human–computer experience.

While the advancement of bio-based wearable flexible sensors has indeed made significant strides and accomplishments, the journey toward widespread adoption is fraught with challenges that demand innovative solutions. A key issue is the trade-off between sensor functionality and comfort. Many existing sensors are not sufficiently pliable or comfortable for prolonged use. This shortcoming underscores the imperative to investigate novel substrate materials that enhance both the flexibility and comfort of wearable sensors, thereby improving their wearability.

Moreover, for these bio-based sensors to be viable for large-scale deployment, it is essential to harness materials that are not only readily available and facile to process but also cost-effective. Simplifying sensor construction could lead to reduced manufacturing and processing expenses, enhancing the economic viability of such technologies.

Furthermore, the current range of wearable flexible sensors often suffers from a limited scope of functionality. To address this, there is a pressing need to ramp up research and development efforts aimed at creating multifunctional sensors capable of monitoring a series of physiological parameters simultaneously. Such an integrated approach could significantly amplify the utility and appeal of wearable sensors.

The potential of wearable flexible sensors could be further harnessed by integrating them with intelligent devices, fostering interdisciplinary collaboration, and expanding their application scenarios. Personalized services, tailored to cater to the unique requirements of individuals, could also be pivotal in enhancing user acceptance and satisfaction.

The significance of bio-based wearable flexible sensors is twofold. Firstly, in contrast to traditional petroleum-based materials, the adoption of bio-based materials enhances environmental friendliness and sustainability. This aligns with the global demands for environmental conservation and sustainable development, thereby addressing the challenge of electronic waste (e-waste) disposal. Moreover, the majority of bio-based materials are biodegradable, meaning that when these sensors reach the end of their life cycle, the resulting waste can decompose naturally over time. This characteristic prevents the long-term environmental pollution typically associated with traditional e-waste, thus aiding in the resolution of e-waste management issues.

Secondly, bio-based sensors have already found extensive application in specific domains and are poised for broader utilization in the future. Currently, they are employed in sports to accurately monitor athletes' exercise and health data. With advancements in science and technology, smart bracelets have become capable of tracking exercise metrics such as calorie consumption, step count, and step frequency, as well as assessing the wearer's mood, sleep quality, stress levels, and heart rate. The industry has seen significant growth in bio-based sensors, which are now commercially available. Looking ahead, the applications of bio-based sensors are expected to expand beyond health monitoring, exercise tracking, and smart clothing to encompass other sectors, including aerospace, human-computer interaction, and electronic skin (e-skin).

In the realm of aerospace, astronauts can wear bio-based sensors to reliably collect physiological data over extended periods, thereby facilitating the monitoring of their health. Given the unique challenges of the space environment, where the recycling and disposal of space debris are particularly arduous, bio-based materials present an ideal solution due to their excellent biodegradability and the absence of post-use space debris. These sensors not only assist astronauts and ground personnel in acquiring vital body data to ensure the astronauts' safety and well-being but also foster advancements in remote monitoring and diagnostic technologies within aerospace medicine. For instance, the data garnered enable ground medical teams to conduct precise assessments of individual astronaut differences and to administer personalized medical care, thereby enhancing the relevance and efficacy of medical support. Consequently, bio-based sensors hold immense potential for applications in the space sector.¹⁴⁸

In the field of human-computer interaction, bio-based sensors have the potential to significantly enhance both the

depth and breadth of engagement. Owing to the excellent biocompatibility of bio-based materials, these sensors can be attached to the scalp or even implanted within the brain for extended durations without eliciting an immune response. Moreover, their superior flexibility allows them to conform seamlessly to the contours and movements of the brain, avoiding any additional pressure or discomfort. These sensors are designed to capture the subtle electrical signals emitted by the brain and, through sophisticated signal processing and interpretation techniques, translate these signals into commands that are comprehensible to computers. This capability paves the way for more advanced and intuitive human-computer interaction technologies.¹⁴⁹

In the realm of e-skin applications, bio-based sensors hold tremendous potential for use as e-skin for robots. The biocomfort of bio-based materials makes them ideal for modifying robots with e-skin capabilities. These sensors can significantly enhance a robot's sensory acuity, allowing it to detect the strength and reactions of human touch. This capability enables the robot to modulate its own movements with greater gentleness and safety, ensuring a more delicate and secure form of interaction. Additionally, this electronic skin can cater to human aesthetic and psychological preferences. By giving robots a more human-like or creature-like appearance with a soft, lifelike texture, the e-skin improves the robot's appeal and aesthetics, making it more readily accepted by people.¹⁵⁰

Ultimately, the absence of a standardized system for wearable flexible sensors is a hurdle that the scientific community and industry stakeholders must collaboratively surmount. The establishment of unified standards and protocols is crucial to ensure the reliability, interoperability, and safety of these sensors, paving the way for their broader integration into various aspects of daily life.

Author contributions

W. Zhao, J. Yan, and J. Sun directed the perspective and revised the manuscript. Z. Zhang and B. Feng carried out the literature collection. Z. Zhang organized and wrote the manuscript. Z. Zhang and W. Zhao drew the schemes and checked over them.

Data availability

This review article does not involve the generation or analysis of new data. All data referenced and discussed in this review are available from the original published studies, which are cited in the text. Therefore, a data availability statement is not applicable.

Conflicts of interest

There are no conflicts to declare.

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