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Biomaterials for reliable wearable health monitoring: Applications in skin and eye integration

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ABSTRACT

Recent advancements in biomaterials have significantly impacted wearable health monitoring, creating opportunities for personalized and non-invasive health assessments. These developments address the growing demand for customized healthcare solutions. Durability is a critical factor for biomaterials in wearable applications, as they must withstand diverse wearing conditions effectively. Therefore, there is a heightened focus on developing biomaterials that maintain robust and stable functionalities, essential for advancing wearable sensing technologies. This review examines the biomaterials used in wearable sensors, specifically those interfaced with human skin and eyes, highlighting essential strategies for achieving long-lasting and stable performance. We specifically discuss three main categories of biomaterials—hydrogels, fibers, and hybrid materials—each offering distinct properties ideal for use in durable wearable health monitoring systems. Moreover, we delve into the latest advancements in biomaterial-based sensors, which hold the potential to facilitate early disease detection, preventative interventions, and tailored healthcare approaches. We also address ongoing challenges and suggest future directions for research on material-based wearable sensors to encourage continuous innovation in this dynamic field.

1. Introduction

As chronic diseases increasingly threaten global public health, the need for timely monitoring of individual well-being grows, with wearable technologies emerging as a powerful solution that provides real-time insights and opens new healthcare avenues [1–4]. These systems are capable of continuously capturing physical data, assessing health status, and providing personalized medical insights through advanced data analysis [5–7]. Key innovations in commercial wearable technology for health monitoring include: i) advanced algorithms for fitness optimization [8], ii) enhanced sensor accuracy for real-time chronic condition management [9], iii) improved metrics for assessing sleep patterns [10], and iv) sophisticated biometric systems for stress analysis [11]. These advancements focus on elevating the precision and applicability of health monitoring tools in both consumer and clinical settings [12].

The efficacy of wearable healthcare devices critically hinges on the incorporation of advanced biomaterials, crucial for their optimal function and seamless interaction with the human body [13,14]. These materials are meticulously selected for their biocompatibility, lightness, breathability, and ability to detect and respond to stimuli, making them ideal for wearable technologies [15–20]. A principal goal in this field is the seamless integration of these materials with human tissues, especially skin and eyes, to ensure consistent and reliable data capture [21–23].

Skin, the body's largest organ, serves as a multifunctional protective barrier, playing a role in regulating body temperature, sensing external stimuli and protecting against harmful chemicals [24]. It is composed of three primary layers: the epidermis, dermis, and hypodermis. The epidermis is the outermost layer, responsible for the skin's barrier function [25]. Beneath it, the dermis provides structural support with its collagen and elastin fibers, which contribute to the skin's elasticity [26].

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These properties allow the skin to conform to movements and return to its original shape after deformation, making elasticity and stretchability critical factors in the design of wearable sensors. Below the dermis lies the hypodermis, which acts as an insulator, regulating body temperature and cushioning against shock [27].

The eyes, responsible for vision, are highly sensitive organs that require precise protection. The surface of the eye is covered by the cornea, a transparent, avascular tissue that plays a key role as both a structural barrier and in protecting the eyes from infections [28]. Any biomaterial designed for ocular use must have oxygen permeability to avoid corneal hypoxia (oxygen deficiency), which can lead to discomfort or long-term damage [29]. Furthermore, materials used for contact with the eye, such as in contact lenses or ocular sensors, must be flexible to accommodate the natural blinking motion [30]. Skin and eye are exposed to a variety of external factors, including mechanical stress or temperature fluctuations, affecting the performance of wearable devices [31,32].

Therefore, the integration of biomaterials with both skin and eyes for wearable devices demands thorough consideration of several critical factors to ensure long-term durability. Self-healing capabilities are particularly important, enabling the material to recover from minor damages caused by environmental factors or mechanical stress, thus extending the device's lifespan and maintaining its functionality over time [33]. Washability is another key aspect, as it allows the device to be cleaned regularly without degrading the material, ensuring both hygiene and sensor effectiveness [3,13]. Flexibility is crucial to accommodate natural movements, whether from the skin during daily activities or from the eye's constant blinking, in order to maintain comfort and consistent sensor performance [1,13]. Permeability is also essential, as it helps prevent sweat accumulation on the skin and allows for adequate oxygen exchange to the cornea, reducing discomfort and the risk of sensor malfunction [1,3,13].

This integration necessitates materials that are both physically and chemically compatible with human tissue and capable of operating effectively under physiological conditions unique to various environments. These environments may present challenges such as temperature fluctuations, moisture exposure, and mechanical stresses [34–36]. Integrating biomaterials with skin and eye tissues has proven challenging, further complicated by these demanding conditions [37–39]. Biomaterials are prone to tearing from user mishandling and may malfunction under harsh external conditions, leading to failure [38,39]. Addressing these challenges involves incorporating features like self-healing capabilities, freeze resistance, washability, and flexibility, all crucial for strong integration and reliable performance of wearable healthcare sensors [38–40]. Therefore, biomaterials must not only possess fundamental properties such as biocompatibility, lightness, breathability, and responsiveness to external stimuli but also maintain their functionality and structural integrity in dynamic environments [38–43]. This necessity has driven extensive research focused on developing durable and stable biomaterials that can withstand daily wear and sustain their functionality over time [44–48].

This review explores the biomaterials utilized in wearable sensing devices, with a specific focus on strategies aimed at bolstering their durability and stability. In this pursuit, we examine three pivotal categories of biomaterials: hydrogels, fibers, and hybrid materials. Each of these categories possesses distinct attributes that render them well-suited for sustained utilization within health monitoring systems. Hydrogels, for instance, excel in their ability to retain moisture and conform to the skin, making them ideal for prolonged wear without causing discomfort. Fibers, on the other hand, offer versatility and strength, enabling the seamless integration of sensors into wearable garments or accessories. Hybrid materials, in this review, refer to those that combine colorimetric sensing capabilities with other functional properties, distinguished by their ability to change color when exposed to specific stimuli, provide a visual indication of physiological changes, facilitating real-time monitoring without the need for complex instrumentation. By exploring these three critical categories of biomaterials, we aim to elucidate their respective roles in advancing the field of wearable sensing technology. Furthermore, we delve into recent breakthroughs in biomaterial-based sensors and their application in skin and eye-mounted monitoring, as depicted in Fig. 1. Through this comprehensive analysis, we endeavor to establish a cohesive understanding of how hydrogels, fibers, and hybrid materials contribute to the development of innovative wearable sensing solutions. Finally, as we navigate through the landscape of material-based wearable sensors, we confront ongoing challenges and chart future research directions. By identifying areas for improvement and innovation, we strive to foster continuous progress in this dynamic field, ensuring that wearable sensing technology remains at the forefront of healthcare innovation.

2. Biomaterials

Biomaterials are materials, whether natural or synthetic, that are engineered specifically to interact with biological systems for medical purposes including diagnostics, therapy, or tissue regeneration [49]. In the realm of wearable health monitoring, biomaterials refer to those substances that are designed to interface directly with human tissues, such as skin and eyes, without causing adverse reactions [13]. However, ensuring long-term, dependable integration poses significant challenges, primarily due to issues like mechanical stability, environmental resilience, and permeability [38,39,50]. Durability is thus a critical focus, vital for achieving stable material integration, consistent data collection, and resistance to various conditions of wear. Durable biomaterials such as hydrogels, fibers, and hybrid materials must be capable of withstanding repeated mechanical stresses, such as stretching, bending, and compression, as well as exposure to environmental factors like drying, and temperature fluctuations, and permeability to gases and liquids, without compromising their structural or functional integrity [33, 51–53]. Proper permeability is particularly important for maintaining skin breathability and avoiding the accumulation of moisture, which can lead to discomfort, skin irritation, or compromised sensor performance [52]. Moreover, they must retain their biocompatibility and adhesive properties over prolonged periods of use, ensuring reliable performance in wearable sensors by preventing issues such as skin irritation or sensor detachment [54]. In response to these requirements, researchers are increasingly dedicated to developing biomaterials that maintain durable functionality for robust integration into wearable devices [44–48]. Among these, hydrogels, fibers, and hybrid materials are notable for their unique properties that contribute to maintaining the structural and functional integrity of wearable sensors. Hydrogels, in particular, have been enhanced with properties such as self-healing and anti-freezing capabilities to meet this requirement [55,56]. For example, self-healing hydrogels can autonomously repair damage from mechanical stress, ensuring that the sensor's functionality is preserved even in dynamic environments [55]. Anti-freezing hydrogels, on the other hand, maintain their flexibility and conductivity in low-temperature conditions, preventing the loss of performance due to freezing [56]. To enhance the durability of fiber-based wearable devices, additional treatments, such as protective coatings or encapsulation coatings can be applied to improve resistance against environmental factors to prevent sensor failure or circuit short. These enhancements help to preserve the fibers' mechanical properties and prevent degradation over time, thereby ensuring reliable performance in diverse conditions. Hybrid materials for colorimetric sensing are designed to deliver a visible and stable color change even under wear and tear, ensuring reliable long-term performance. Moreover, they are engineered for reusability, maintaining their colorimetric response over multiple uses without significant degradation, making them well-suited for extended wearable applications. These advancements make biomaterials such as hydrogels, fibers, and hybrid materials a particularly promising material for achieving the durability needed in next-generation wearable health monitoring systems. This chapter delves into the durable characteristics

Fig. 1. Schematic illustration of the human skin- and eye-interfaced health monitoring and biomaterials' representative durable properties (e.g., self-healing, washability, flexibility, permeability).

of these biomaterials and their crucial roles in enhancing wearable sensor technology.

2.1. Hydrogels

Hydrogels are increasingly recognized for their potential in wearable sensor applications, particularly due to their responsiveness to stimuli [48]. Their flexibility and biocompatibility make them highly suitable for integration with human skin, a crucial factor in wearable technologies [57]. Yet, their integration with human skin for long-term use remains challenging because of inherent durability issues [38,39]. Hydrogels are naturally prone to mechanical degradation over time, especially under continuous stress, which raises concerns about their long-term reliability in wearable devices [58]. Additionally, hydrogels can be sensitive to environmental factors including temperatures, leading to potential performance degradation in extreme conditions [59].

To overcome these challenges, research has focused on enhancing hydrogels with properties like self-healing and anti-freezing capabilities, which support robust and continuous integration into wearable devices, ensuring that hydrogels can maintain reliable sensor function and withstand various conditions of use [44,45]. These properties ensure that hydrogels can maintain reliable sensor function and withstand various conditions of use. Consequently, a variety of conductive hydrogels boasting these adaptive features have been developed, targeting long-term, reliable performance in health monitoring wearables across multiple scenarios [45–48]. Despite these advancements, there are still some limitations. The addition of self-healing and anti-freezing properties often involves complex fabrication processes, which can increase production costs and complicate the scalability of these materials for widespread use. Furthermore, these advanced properties can sometimes lead to trade-offs, such as reduced softness under certain conditions, which may limit the effectiveness of the hydrogels in specific applications [56]. This section explores recent advancements in hydrogel research, emphasizing their self-healing and anti-freezing qualities, which are critical for their application in wearable sensors. Self-healing hydrogels possess the remarkable ability to autonomously repair damage inflicted upon them, ensuring prolonged sensor functionality even in dynamic and demanding environments. This is particularly advantageous in applications where the sensor is subject to frequent mechanical stress, as it reduces the likelihood of sensor failure. On the other hand, anti-freezing hydrogels are engineered to withstand low temperatures without compromising their structural integrity or sensing capabilities. This makes them particularly valuable for wearable sensors that need to

function reliably in cold weather or in scenarios where temperature fluctuations are common. By integrating self-healing and anti-freezing properties into hydrogels, researchers aim to address the challenges associated with long-term wearability and performance of wearable sensors, ensuring reliable operation under diverse conditions.

2.1.1. *Self-healing hydrogel*

Since the early 2000s, self-healing hydrogels have marked a significant breakthrough in materials science due to their capacity to autonomously mend mechanical and electrical damage [60]. This capability is particularly crucial for long-lasting and durable applications in wearable sensors, as it improves device longevity and resilience by reducing the impact of environmental stress and mishandling. A notable instance of these materials in practice was in January 2020, when Apple's patent for integrating self-healing polymers into the display of a foldable iDevice was published by the US Patent and Trademark Office. This patent highlighted Apple's innovative strategy to boost the durability and toughness of its devices.

Fig. 2A (i) illustrates the mechanisms behind this capability: Self-healing hydrogels can be engineered through dynamic covalent interactions, such as borate ester bonds, or noncovalent interactions like hydrogen bonding, which allow for disruption and reformation [61]. Dynamic covalent bonding provides superior mechanical strength due to the relatively stronger bond strengths compared to noncovalent bonding. On the other hand, noncovalent bonding requires less time for self-healing compared to covalent bonding, allowing for quicker recovery after damage. For example, Liao et al., developed self-healing hydrogel based on polyvinyl alcohol (PVA), functionalized single-wall carbon nanotube (FSWCNT), and polydopamine (PDA) [62]. The dynamic nature of the hydrogels, driven by the dynamic supramolecular cross-linking among FSWCNT, PVA, and PDA, along with the dynamic complexing interaction between the hydroxyl groups of PVA and tetrafunctional borate ions, endows the hydrogels with rapid self-healing properties within 2 s, without requiring any external stimuli. As shown in Fig. 2A (ii), the hydrogels, when cut, can rapidly self-heal and demonstrate significant elongation, showcasing their practical utility [55]. The formulation of these hydrogels involves a dynamic interconnected network incorporating polyvinyl alcohol (PVA), borax, poly (3,4-ethylenedioxythiophene)/polystyrenesulfonate (PEDOT/PSS), and glycerol (GL). Glycerol's inclusion into the control group, which is composed of PVA, borax, and PEDOT/PSS, enhances the intermolecular interactions within the hydrogel matrix by moderating the balance between covalent (e.g., PVA-borax) and additional hydrogen bonds (e.g., glycerol-borax, glycerol-PVA, PEDOT/PSS-borax, and PEDOT/PSS-PVA) as shown in Fig. 2A (iii). This facilitates rapid self-healing in under 0.12 s, 5 times faster than the control group (4.2 s), which lacks glycerol in its composition. This rapid healing stems from the accelerated dynamics of non-covalent over covalent bonding. Additionally, the presence of

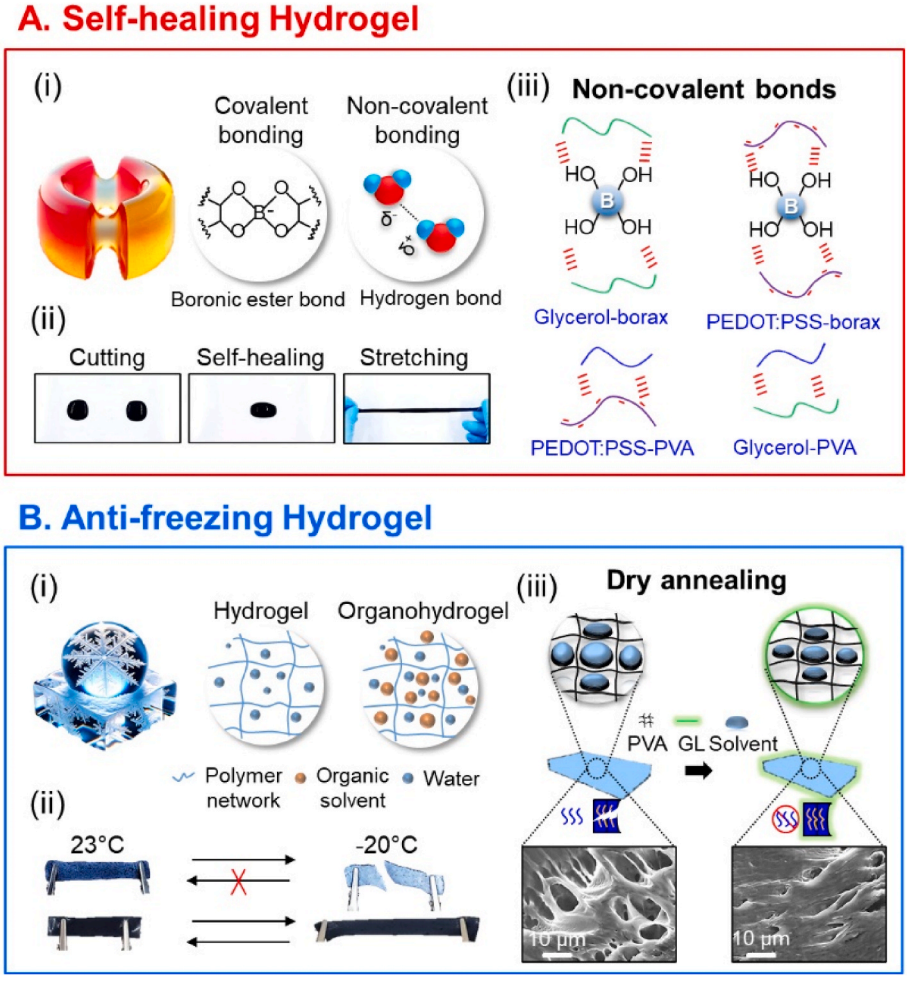

Fig. 2. Conductive hydrogels for wearable sensors. (A) Self-healing hydrogels. (i) Self-healing mechanisms. (ii) The elongation of self-healing hydrogel after healing process. (iii) Schematic diagrams showing the rapid self-healing mechanism of the hydrogel by multiple hydrogen bonding. Adapted with permission from Ref. [55] Copyright 2024 American Chemical Society. (B) Anti-freezing hydrogels. (i) Composition of hydrogel and organohydrogel. (ii) Photographs showing the organohydrogel and hydrogel after freezing while being stretched. (iii) Schematic representation and SEM images, showing the inner structure of organohydrogels before and after dry annealing. Adapted with permission from Ref. [56]. Copyright 2024 Wiley-VCH.

glycerol fosters abundant intermolecular interactions, resulting in ultralow electrical hysteresis (<0.64 %) under cyclic strains up to 500 %, crucial for consistent and interpretable data. Table 1 provides a comprehensive analysis including self-healing efficiency/time and electrical hysteresis of hydrogels. The hydrogel also exhibits notable properties including: (1) exceptional softness, with E < 5.2 kPa, four times lower than that of the control group; (2) high deformability, stretching beyond 10,000 %; (3) capability to reshape into various forms, such as the letters "P" and "S"; (4) complete gelation; (5) distinctive fingerprint patterns; (6) adaptability to conform to intricate contours, like the wrinkles on the wrist; (7) high ionic and electrical conductivity (>0.074 S m^{-1}); (8) self-adhesion to diverse surfaces, including plastic (0.35 ± 0.08 N cm^{-2}), glass (0.18 ± 0.002 N cm^{-2}), wood (0.17 ± 0.01 N cm^{-2}), and pigskin (0.10 ± 0.005 N cm^{-2}), exceeding that of the control group; (9) resistance to non-axial deformations like twisting, bending, and pressing; (10) rapid response and recovery times (238 ms); (11) no notable overshoot behavior and (12) durability (1000 cycles) at the strain level of 200 %. This remarkable combination of features makes hydrogel highly suitable for extended, reliable use in wearable sensing applications, including monitoring body movements and electrophysiological activities on the skin. Especially, the exceptional softness of the self-healing hydrogel, with a modulus below 5.2 kPa, closely matches that of human skin, enabling reliable and comfortable contact. Although these glycerol-enhanced hydrogels are promising for wearable sensors due to their rapid self-healing and enhanced intermolecular interactions, they still face challenges like susceptibility to freezing and dehydration, which may compromise their mechanical and electrical integrity under varying environmental conditions.

2.1.2. *Anti-freezing hydrogel*

Conductive hydrogels are highly valued in wearable sensing applications for their flexibility and responsiveness to stimuli [48]. However, their utility can be limited by inherent vulnerabilities, particularly their tendency to freeze at subzero temperatures, which can severely impair their mechanical and electrical integrity, potentially leading to failure. Moreover, their wider application is limited by insufficient mechanical properties, making them unsuitable for sports such as skiing, beach volleyball and exploration activities like space, freezing mountains and scorching deserts, which demand environmental tolerance, high mechanical properties and durability [31]. To address this, enhancing hydrogels with anti-freezing and improved mechanical properties has been pivotal, allowing the development of sensors that remain functional even in extremely cold environments. Fig. 2B (i) illustrates how the integration of organic solvents into hydrogels serves as an effective method to confer anti-freezing properties by tightly binding water molecules, preventing ice formation.

Fig. 2B (ii) highlights a conductive organohydrogel, notable for its resistance to freezing, unlike traditional hydrogels which solidify under similar conditions [56]. This organohydrogel consists of an intricate network of PEDOT:PSS, PVA, water, ethylene glycol, and glycerol while the hydrogel excludes the organic solvents. Remarkably, it maintains consistent mechanical and electrical properties, demonstrating exceptional adaptability across a range of environmental settings. This durability is chiefly due to the synergistic effects of ethylene glycol and glycerol. Ethylene glycol disrupts ice crystal formation by forming extensive hydrogen bonds with water, while glycerol aids in retaining moisture, thus providing a barrier against drying. Additionally, a dry annealing process optimizes the hydrogel's structure by enhancing molecular chain alignment, minimizing pores, and bolstering mechanical properties as shown in Fig. 2B (iii). Moreover, the reduced quantity of surface pores within the compact matrix enhances the hydrogel's resistance to dehydration. Dry annealing strengthens its network structure, thereby imparting anti-drying and robust mechanical characteristics due to its unique configuration of structural enhancements, strong interconnections, and dynamic component interactions. For instance, the treated organohydrogel exhibits a remarkable 147.2 % increase in toughness compared to its untreated counterpart, showcasing superior resilience to deformation and fracture under stress. Similarly, the mechanical modulus sees a significant fivefold enhancement post-dry annealing, indicating increased stiffness and rigidity. While the enhanced modulus may result in a mismatch with the mechanical properties of human skin, the primary focus of the dry annealing process is to enhance the hydrogel's durability and usability to a wider range of environmental conditions, as well as to mechanically demanding scenarios, thereby ensuring the sensor's long-term stability and functionality. Overall, the conductive organohydrogel demonstrates high toughness (30.4 MJ m^{-3}) and mechanical strength (2.6 MPa) across various environmental scenarios, including extreme temperatures (−20–70 °C), vacuum conditions (12 psi), and prolonged storage (one month under ambient conditions). Furthermore, a minor 3.7 % mass reduction post-dry annealing contrasts sharply with the significant 46.1 % decrease in mass observed in the untreated sample after 30 days of storage. This disparity underscores glycerol's crucial role in enhancing desiccation resistance, ensuring the hydrogel's longevity and effectiveness. Additionally, this improvement highlights the critical role of dry annealing in maintaining the hydrogel's structural integrity and functionality over time. Other notable features include exceptional electrical performance, such as perfect linearity (R^2 > 0.99) across high strain (>1000 %), rapid response/recovery times within 0.4 s without noticeable overshoot at 100 % strain, and remarkable resilience and cyclic stability even under extreme conditions. This impressive combination of attributes renders the organohydrogel highly suitable for sustained and robust use in wearable sensing applications, efficiently monitoring human movements across diverse environmental conditions. Table 2 provides a comprehensive analysis summarizing environmental tolerance and mechanical performance of hydrogels. With its advanced anti-freezing and robust mechanical properties, this organohydrogel is well-positioned as a promising, durable material for next-generation wearable sensors.

Table 1
Summary of the self-healing performance of hydrogels.

Hydrogel System	Components	Healing efficiency and time	Electrical hysteresis	Applications	Ref.
PVA-Borax	PEDOT:PSS, GL	100 %, 0.12 s	0.64 % @ 500 % (Strain)	Strain sensor, ECG, EMG, EOG	[55]
	FSWCNT, PDA	99 %, 2.0 s	–	Strain sensor	[62]
	CNT	98 %, 3.2 s	Negligible @ 100 % (Strain)	Strain sensor	[63]
	rGO, GL	95.7 %, 3s	–	Strain sensor, E-skim, ECG	[64]
	MFC	10 min	–	PH sensor	[65]
	PAA, EG	62.2 %, 10.0 s	–	Strain sensor	[66]
	CNTs or Graphene	8.6 s	–	Strain sensor	[67]
	PDA	0.25 s	–	Strain sensor	[68]
	Xylan	85.8 %, 30 s	–	–	[69]
	Ionic Liquid	6 min	–	Electrochromic strain sensor	[70]

2.2. *Fiber materials*

The realm of smart textiles marks a groundbreaking fusion of technology with traditional fabric, unlocking capabilities far beyond those of ordinary textiles [77–80]. These cutting-edge materials possess the remarkable ability to detect shifts in their environment, respond to mechanical, thermal, or chemical cues, and fundamentally alter our

Table 2
Summary of environmental tolerance and mechanical performance of hydrogels.

Materials	Low Temp. Stability	High Temp. Stability	Long-term Stability	Strength (MPa)	Toughness (MJ m^{-3})	Modulus (MPa)	Ref.
PVA/EG/PEDOT:PSS/GL	Yes			2.57	30.4	1.60	[56]
PVA/APTES-SiO_2	Yes	No		0.16	–	71	[71]
PVA/PSBMA	No			0.032	–	–	[72]
				0.066	–	–	
				0.153	–	–	
				0.228	–	–	
				0.596	0.596	–	
PVA	Yes	No		0.257	0.26	73	[73]
PVA/GL	Yes	No		0.576	1.3	0.072	
PVA/PAni	Yes	No		0.035	0.051	0.002	
PVA/GL/PAni	Yes	No		0.2	0.412	0.023	
Enhanced PVA	No			0.13	4.9	0.046	[74]
				2.1	14.8	1.119	
				3.9	13.4	3.088	
PVA/AgNPs	No			13.3	78.1	–	[75]
PVA/Epoxy	No			1.1	2.8	0.08	[76]

interaction with the world around us [81–83]. Among their myriad functions are thermoregulation [84–86], health monitoring [87], and energy harvesting [88–92]. Through the incorporation of conductive threads, wearable sensors, and miniature electronics, garments can now communicate, adapt to environmental conditions, and even gather data on the wearer's well-being [93–95]. Crafting smart textiles involves a meticulous selection of materials tailored to specific functionalities, drawing from a diverse and ever-expanding range of options such as metal-based [96], carbon-based [97,98], or conductive polymer-based [99,100] materials. Advanced fabrication techniques and design principles further enhance the development of these innovative textiles. We encounter two main categories of smart textiles: conductive materials-based and active materials-based. Conductive materials-based smart textiles predominantly utilize metals, carbon, or conductive polymers to impart electrical conductivity, enabling functionalities like sensing and communication. In contrast, active materials-based smart textiles leverage materials that undergo dynamic responses to external stimuli, enabling features such as light-emitting or power-generating properties. Each category offers unique advantages and challenges. Conductive materials-based textiles excel in electrical conductivity and data transmission, while active materials-based textiles shine in adaptability and responsiveness to environmental cues. Table 3 summarizes materials used for smart textile.

2.2.1. *Conductive materials-based smart textiles*

Fig. 3A (i) presents an innovative approach for fabricating metal-based interconnections on polyethylene terephthalate (PET) fabric [102]. The process involves metallizing the PET fabric using Polymer-assisted Metal Deposition (PAMD) treatment to convert it into conductive copper (Cu) fabric. The fabrication steps include dip-coating with photoresist, soft baking for solvent removal, double-sided ultraviolet (UV) exposure for comprehensive pattern coverage, pattern development, hard baking to enhance adhesion, wet etching for precise patterning, residue removal, and oven drying of the fabric. Subsequently, an epoxy-silver adhesive reinforces the connection between the electronic components' rigid metal pins and the Cu fabric. Distinguished from traditional semiconductor technology photolithography, the metallic fabric is coated with photoresist through dip-coating, not spin coating, enabling a conformal coating of materials on the three-dimensional (3D) textile network created by the interlaced warp and weft yarns. Moreover, this metallic patterning strategy enables a high air permeability of 79 mm s^{-1} and moisture permeability of 270 g m^{-2} per day.

Table 3
Summary of fiber materials used for smart textiles.

Materials for smart textiles	Applications	Ref.
Conductive materials	Conductive interconnection/ electrography	[96–100, 184]
Metals (for example: Ag, Cu, Ti, Au, Ni)		
Conducting polymers (for example: polythiophene, polyaniline, polypyrrole)		
Carbon materials (for example: CNT, graphene)		
Active materials	**Applications**	**Ref.**
Electrochemically active (for example: Ag_2O/Ag, MOFs)	Biochemical sensing/energy-storage textiles	[101]
Mechanically active (for example: PVDF, ZnO)	Mechanical sensing/Energy-harvesting textiles	[106]
Thermoactive (for example: PEDOT:PSS, Sb_2Te_3–Bi_2Te_3)	Thermal management/ Temperature-sensing textiles	[84–86]
Photoactive (for example: TiO_2, ZnS)	Photovoltaic/ electroluminescent textiles	[80,105, 192,242]

Fig. 3A (ii) illustrates a straightforward method for creating highly adhesive and dispersible carbon nanotube (CNT) ink [103]. Initially, γ-Aminopropyl triethoxy silane (APTES) serves as a cross-linking agent to modify cellulose. Additionally, APTES, acting as a weak base, obviates the need for an inorganic base or the typical Tris-HCl buffer and catalyzes the self-polymerization of dopamine (DA) into polydopamine (PDA). Alcohol adjusts the surface tension between the CNTs and water. The prepared cellulose solution facilitates the uniform distribution of CNTs via non-covalent bonds, resulting in CNT ink with strong adhesion to various substrates. This CNT ink is employed in the production of textile-based sensors via stencil printing, which equips the strain sensors to swiftly respond to external stimuli. Following this, the sensors are encapsulated with polydimethylsiloxane, which markedly broadens their operational strain range from less than 20 % to approximately 70 %.

In Fig. 3A (iii), a sensor composed of polyaniline-decorated poly(3,4-ethylenedioxythiophene)/poly(4-styrenesulfonate) (PANi/PEDOT/PSS) is developed by dip-coating PANi onto PEDOT/PSS cotton yarn [104]. The sensor's high conductivity (1343 S cm^{-1}) arises from the ethylene glycol (EG)-induced separation of PEDOT and PSS, leading to a swollen, highly conductive PEDOT layer alongside conductive PANi doped with PSS. These PANi/PEDOT/PSS sensors exhibit high selectivity towards ammonia gas and maintain excellent stability over extended periods in air. The sensing mechanism involves the protonation of PANi surfaces by ammonia and subsequent charge transfer at the interface of the PANi/PEDOT/PSS heterostructure. The sensor achieves an ammonia sensing response of 7.54 % at 50 ppm, coupled with an exceptionally low detection limit of just 5 ppm. Importantly, it maintains outstanding stability in both conductivity and sensing response across 1100 stretching/releasing cycles, and it demonstrates high durability after 10

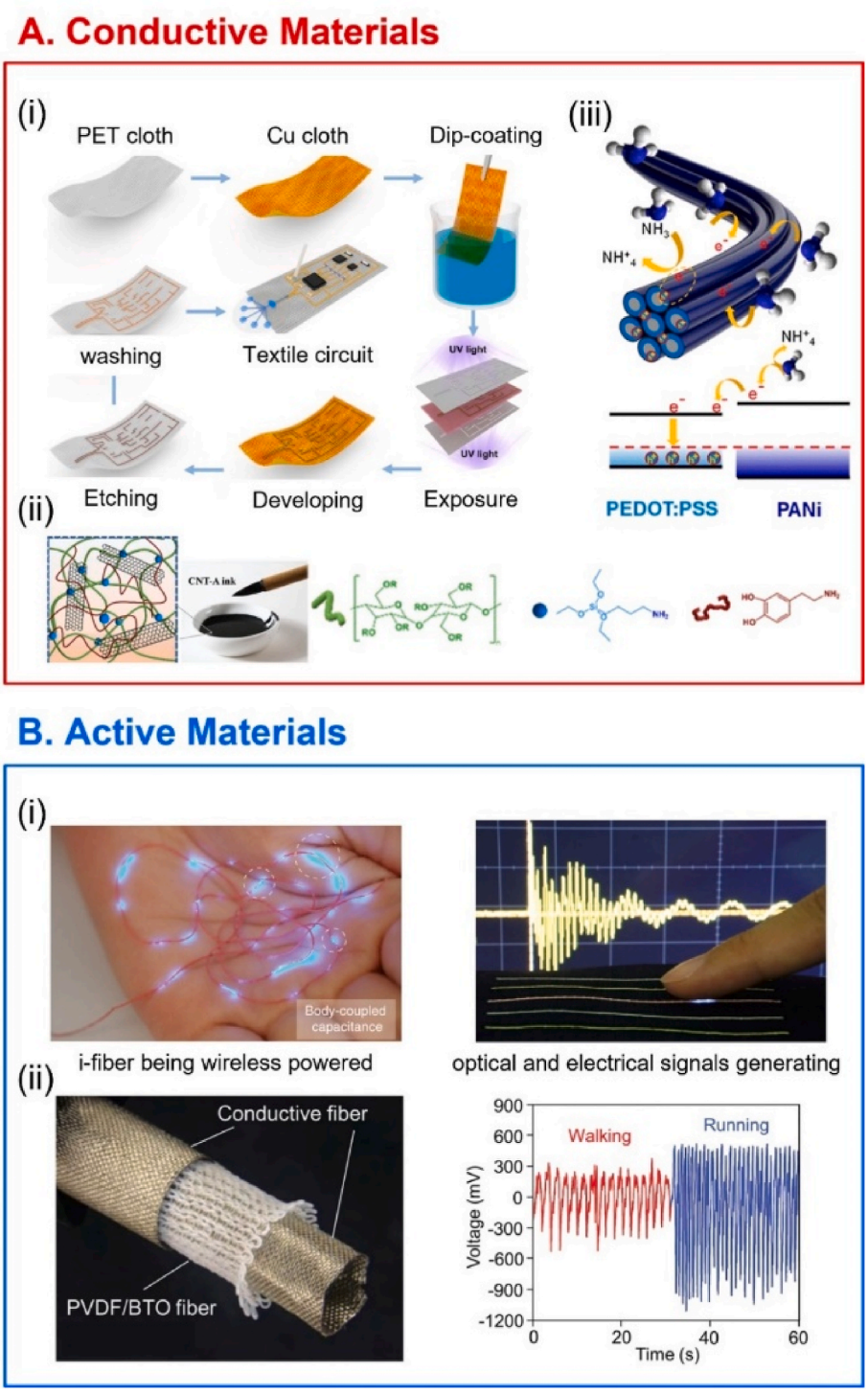

Fig. 3. Materials for smart textiles. (A) Conductive materials. (i) The fabrication process of Cu-based conductive interconnects and system assembly. Adapted with permission from Ref. [96]. Copyright 2023 American Association for the Advancement of Science. (ii) Schematic illustration displaying the molecular structure of the CNT-based dispersion. Adapted with permission from Ref. [103]. Copyright 2022 American Chemical Society. (iii) Diagram of the PANi/PEDOT/PSS yarn sensor. Adapted with permission from Ref. [104]. Copyright 2022 American Chemical Society. (B) Active materials (i) ZnS-based electroluminescent fiber being wirelessly powered. Adapted with permission from Ref. [105]. Copyright 2024 American Association for the Advancement of Science. (ii) PVDF-based textile energy generator. Adapted with permission from Ref. [106]. Copyright 2020 Wiley-VCH.

wash cycles.

2.2.2. *Active materials-based smart textiles*

Expanding beyond the discourse on conductive-based materials, it is imperative to widen our vista to encompass a broader spectrum of active materials integral to smart textiles. These materials extend beyond mere conductivity, incorporating diverse functional substances that dynamically respond to environmental stimuli. This paradigm shift enables the exploration of innovative applications and technologies, enhancing the interactive capabilities of smart textiles and heralding a new era of adaptive and responsive fabric-based solutions. Through the integration of these versatile active materials, smart textiles attain unprecedented

levels of functionality and versatility, catering to diverse needs in modern wearables and beyond. Fig. 3B (i) introduces a pioneering approach to fiber electronics that leverages the human body as an interactive medium to interface with ambient electromagnetic (EM) energy [105]. This body-coupled fiber electronics method facilitates the generation of bound charge pairs between the body and the electronic fiber, enabling oscillation between bound and radiation states for the wireless transmission of sensor data. Moreover, electric field-sensitive luminescent dielectrics embedded within the fiber antennas offer visual feedback, seamlessly integrating sensors and actuators into a singular fiber. This holistic design obviates the necessity for chips, batteries, and other rigid components, consolidating all electronic elements into a diminutive fiber structure, rendering it as flexible and lightweight as conventional textiles. Additionally, the double-network cross-linked resin effectively prevents dye fading and phosphor shedding, ensuring the textile retains its luminous brightness and demonstrates robust resistance to washing.

Fig. 3B (ii) introduces a method for crafting wearable energy generators and sensors utilizing nanostructured hybrid piezoelectric fibers and leveraging the vast diversity of textile structures [106]. By employing a mixture of barium titanate (BT) nanoparticles and poly (vinylidene fluoride) (PVDF) at a mass ratio of 1:10, highly efficient hybrid piezofibers have been developed. These fibers are woven into a wearable energy generator capable of producing a peak voltage output of 4 V and a power density of 87 μW cm^{-3}, which is 45 times greater than previously documented for piezoelectric textiles. This wearable generator can charge a 10 μF capacitor in just 20 s, significantly faster than earlier versions made from PVDF/BT and pure PVDF, by four and six times, respectively. Additionally, the knitted design of the energy harvester shows a sensitivity that is 6.3 times greater compared to previous piezofiber generators. A knee sleeve prototype featuring a PVDF/BT wearable device has been demonstrated for real-time precise health monitoring. The fabrication technique developed is scalable, facilitating the production of large quantities of smart textiles for industrial use.

Fiber materials are inherently flexible, making them ideal for designing wearable sensors and devices that need to conform to the natural movements of the human body. This flexibility allows fibers to bend, stretch, and twist in sync with the wearer's motions, ensuring consistent performance and comfort without compromising the functionality of embedded sensors. However, fiber materials can be sensitive to environmental conditions such as moisture and UV radiation, which can degrade their mechanical properties or interfere with sensor performance. Moisture penetration can cause fibers to swell, weaken, or even result in electrical short circuits in conductive applications, while prolonged UV exposure may lead to discoloration and reduced structural integrity. To address these challenges, protective measures like encapsulation and waterproof coatings are often employed, enhancing durability and allowing fibers to endure conditions such as regular washing. For example, smart textiles, like the jacket from Google's Project Jacquard with Levi's, are designed to withstand multiple wash cycles. Moreover, continuous advancements in material science and textile engineering are enhancing textiles' inherent abilities to self-clean, resist stains, and recover from mechanical wear, significantly improving their washability. These advancements enable smart textiles to maintain reliable performance in various environments, extending their use beyond casual wear to more demanding applications in medical and athletic apparel, where hygiene, durability, and consistent sensor functionality are essential.

2.3. Hybrid materials

Hybrid materials integrate selected organic and inorganic components into new materials that merge their best properties, creating synergistic combinations with unique characteristics and enhanced performance across a wide range of applications [107–110]. In the context of this work, hybrid materials are those that integrate colorimetric sensing capabilities with other material properties, enabling them to respond to specific analytes with a visible color change. These materials can be engineered by embedding colorimetric agents such as nanoparticles or enzyme catalysts into various matrices, thereby creating multifunctional sensing platforms, making them especially suited for wearable sensing applications. Due to the capability to produce visible results with minimal equipment for interpretation, colorimetric sensors have shown considerable promise in detecting a wide range of biomarkers, including pH levels, electrolytes, uric acid, glucose, and proteins [111]. However, despite their widespread popularity, the use of existing colorimetric sensing technologies in long-term wearable health monitoring is somewhat limited. This limitation primarily stems from challenges related to sensitivity, selectivity, and rigid designs that often lead to discomfort and constrained sensing ranges [112]. Recent advances in nanotechnology and material science have harnessed the potential of hybrid materials, [113–115]. This section not only highlights the broad potential of hybrid materials but also examines two primary types of materials used in wearable colorimetric sensor fabrication: nanoparticle-based and enzyme-catalyzed hybrid sensing materials. Nanoparticle-based hybrid materials utilize the unique optical properties of nanoparticles to induce color changes in response to specific analytes, enabling quick and direct observation of results [116]. Enzyme-catalyzed hybrid materials, meanwhile, exploit catalytic amplification to detect very low concentrations of target molecules [117]. While both material types offer distinct advantages, nanoparticle-based sensors are particularly noted for their efficiency and versatility [118], whereas enzyme-catalyzed sensors are prized for their high specificity and compatibility with biological environments [119].

To provide a clearer understanding of the materials and their applications, Table 4summarizes various nanoparticle-based and enzyme-catalyzed hybrid sensing materials along with their respective sensing targets and detection limits. This compilation demonstrates the breadth of research and innovation in the field, highlighting key materials and their effectiveness in detecting a wide range of analytes, including temperature, glucose, bacteria, proteins, and more. Such hybrid materials, which seamlessly integrate sensing capabilities with structural functionalities, represent a significant advancement in the development of wearable colorimetric sensors.

Table 4
Summary of hybrid material based colorimetric sensors for healthcare application.

Type	Materials	Sensing Targets	detection limit	Ref.
Nanoparticle based	AuNPs/PEG/Anti-spike antibody	COVID-19 antigen	1 ng/mL	[120]
	AuNPs/PNIPAM	Temperature	0.2 °C	[121]
	AuNPs/sodium hydroxide	Glucose	0.29 mM	[122]
	AuNPs/Antibody	Bacteria (S. aureus and Lactobacillus)	120 and 105 CFU/ml	[123]
	AuNPs/AgNPs	Protein/bacteria	0.5 μM	[124]
	AgNPs/Ca^{2+}	cysteine	0.1 μM	[125]
	AgNPs/Citrate	creatinine	66 nM	[126]
	CuNPs/curcumin	Na+	10 mM	[127]
Nanozyme based	PVA-AuNPs/TMB	Glucose	1 mM	[128]
	Ppy-MoS_2-Au/Ag/Pd/TMB	Oral Bacteria	10^7 CFU/mL	[129]
	DNA-templated Ag/Pt bimetallic nanoclusters/TMB	Thrombin	1 nM	[130]
	silica–gold nanocluster hybrid (MSN–AuNC)/TMB	HER2+ cancer cells	10 cells	[131]
	MOFs/PVP-GO_x	Glucose	5 μM	[132]
	Porphyrin–ZnS/TMB	Glucose	0.005 mM	[133]

2.3.1. Nanoparticle-based hybrid materials

Nanoparticle-based hybrid materials have garnered significant attention in colorimetric sensing recently, owing to their distinctive optical properties, biocompatibility, and responsiveness to external stimuli [134]. At the heart of their functionality lies surface plasmon resonance (SPR), where the oscillation of conduction electrons on the surface of metal nanoparticles in response to light induces striking color changes [135]. Precise control over the size, shape, and composition of these nanoparticles enables fine-tuning of their optical responses, rendering them highly effective for colorimetric sensing applications. In wearable sensors, these nanoparticles are typically integrated with antibodies or other biological components in a hybrid design, allowing them to respond to specific analytes through mechanisms such as aggregation or dispersion [136]. The interaction between the analyte and the nanoparticles results in a visible color change directly correlated with the concentration of the target substance. Among various nanoparticle types, gold nanoparticles (AuNPs) are particularly favored due to their strong localized surface plasmon resonance (LSPR), facilitating visible and easily interpretable color changes for precise biomarker detection [137]. The high surface area-to-volume ratio of AuNPs

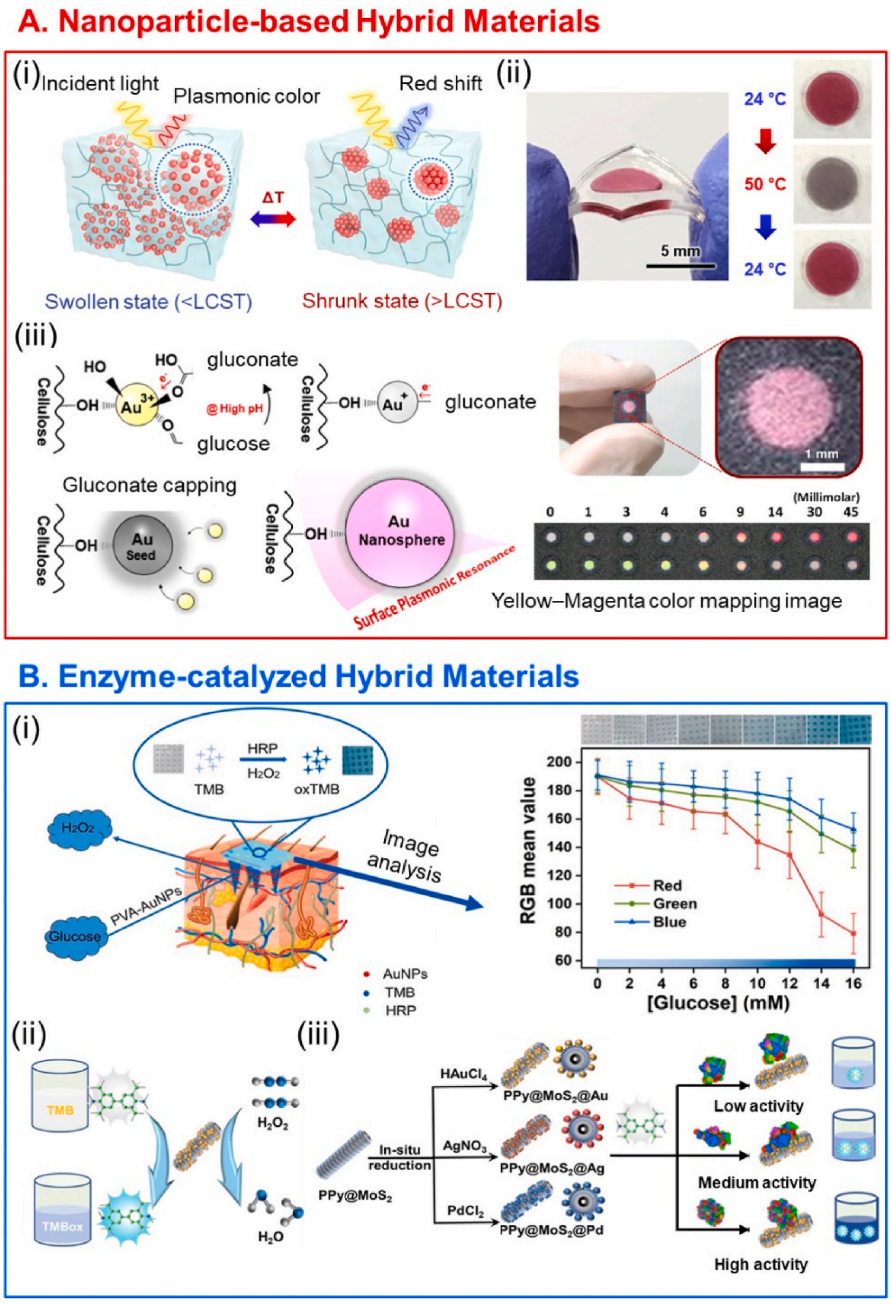

Fig. 4. Hybrid materials for wearable sensors. (A) Nanoparticle-based hybrid materials. (i) The working principle of the temperature-responsive plasmonic microgel-based colorimetric sensor. (ii) The smart colorimetric patches in response to temperature changes. Adapted with permission from Ref. [121]. Copyright 2018 Springer Nature. (iii) Schematic illustration of glucose sensing in ocular fluid through stepwise reactions and color changes at different glucose concentrations. Adapted with permission from Ref. [122]. Copyright 2020 MDPI. (B) Enzyme catalyzed hybrid materials. (i) Schematic diagram of the color-development principle of colorimetric MN patch and the detection of glucose solutions with different concentrations. Adapted with permission from Ref. [128]. Copyright 2024 Elsevier. (ii) Sensing mechanism of a nanozyme-based colorimetric sensor array for precise identification of proteins and oral bacteria. (iii) Synthetic process of metal-NP-supported nanozyme (MNN) and the modulation of the catalytic oxidation by different proteins. Adapted with permission from Ref. [129]. Copyright 2022 American Chemical Society. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

promotes quick interactions with analytes, allows for quick interactions and visible color changes upon analyte binding. Additionally, AuNPs' ability to be easily functionalized with various molecular receptors enables direct and efficient signal transduction. For example, the aggregation of AuNPs, which typically exhibits a red color in solution, can shift to blue as they clump together in the presence of certain analytes. This change is often triggered by specific biochemical interactions or alterations in the electrostatic balance among the nanoparticles, serving as a robust indicator of analyte presence [138].

Recent years have seen significant advancements in the development of metal nanoparticle-based hybrid materials for colorimetric sensing. For example, Ray et al. demonstrated the synthesis and application of spike protein antibody-functionalized AuNPs for the rapid detection and inhibition of severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2) [120]. The sensor effectively identifies specific viral antigens or virus particles of the coronavirus disease that began in 2019 (COVID-19) through a simple color change from pink to blue within 5 min, triggered by nanoparticle aggregation upon binding with the virus. This aggregation notably enhances the Raman signal, allowing for detection thresholds as low as 4 pg per milliliter for the antigen and 18 virus particles per milliliter for the virus using surface-enhanced Raman spectroscopy (SERS). Furthermore, these antibody-conjugated AuNPs not only detect but also inhibit the virus, blocking its entry into cells by preventing the interaction between the virus's spike protein and the angiotensin-converting enzyme 2 (ACE2) receptor on human cells. This dual functionality highlights a promising strategy for both diagnostic and therapeutic applications in the fight against COVID-19.

Fig. 4A (i) depicts the design of stretchable and wearable colorimetric patches that utilize thermo-responsive plasmonic microgels embedded in a hydrogel film, designed to enable dynamic and precise temperature sensing [121]. By integrating AuNPs with poly (N-isopropylacrylamide) (PNIPAM) microgels, the research team crafted a flexible sensor capable of significant color shifts from red to grayish-violet in response to temperature changes, highlighted by an impressive extinction peak shift of 176 nm occurring within just 1 s. The sensor patch can be stretched up to 90 % in length, without disturbing the thermally shifted plasmonic colors. The sensor offers a high temperature resolution of 0.2 °C and functions effectively across a temperature range of 25–40 °C. This capability is due to the efficient plasmon coupling between the AuNPs, facilitated by the reversible, thermally induced structural transitions of the PNIPAM microgels, as shown in Fig. 4A (ii). With its rapid response and broad temperature sensing range, coupled with its ability to maintain conformal contact with the human body, this sensor is highly suitable for wearable applications.

Fig. 4A (iii) depicts the stepwise reactions of a colorimetric sensor employing in-situ synthesized gold nanoparticles on plasmonic paper for glucose detection in ocular fluids [122]. This sensor leverages the colorimetric changes induced by the interaction of glucose with gold nanoparticles, which are effectively stabilized by the paper matrix. This stabilization facilitates a distinct color shift from yellow to magenta as glucose concentrations vary, allowing the sensor to detect levels as low as 0.29 mM. This sensitivity falls well within the range necessary for effective monitoring in diabetic patients. These attributes establish it as a promising tool for non-invasive diabetes monitoring and diagnosis, with potential applications extending beyond ocular fluids to other biological matrices where glucose monitoring is essential.

Moreover, the versatility of nanoparticle-based materials transcends conventional metals such as gold and silver [139]. Recent progress has introduced an array of nanocomposites and novel nanoparticle formulations, augmenting both the sensitivity and selectivity of colorimetric sensors. These materials can be tailored to respond to a diverse spectrum of environmental and biological stimuli, thus expanding the potential utility of wearable colorimetric sensors across various domains, including medical diagnostics [131,140]. Despite the significant advantages of nanoparticle-based hybrid materials, most research on the surface plasmon resonance (SPR) of nanomaterials has concentrated on the synthesis of functional gold and silver nanostructures. However, the high costs of these noble metals have constrained their broader application. Fortunately, recent studies suggest that Copper Nanoparticles (CuNPs), which are more cost-effective, could serve as an alternative for conventional metals such as gold and silver [127,141–147]. However, the main disadvantage of CuNPs is their tendency to oxidize and form copper oxide, which limits their current applications primarily to the detection of heavy metals. Further research is needed to fully explore and expand the utility of copper nanoparticles and other alternatives in wearable colorimetric sensors.

2.3.2. *Enzyme-catalyzed hybrid materials*

Enzymes, highly efficient and specific substances produced by living cells, offer remarkable catalytic properties. However, challenges such as low stability and high production costs impede the widespread utilization of natural enzymes in biosensors [148]. Nanotechnology has paved the path for the development of innovative enzyme mimics with enzyme-like activities. Termed nanozymes, these nanomaterials exhibit inherent enzymatic characteristics, withstand high temperatures, and demonstrate notable bioselectivity, presenting significant potential for diverse biomedical applications [149]. The construction of colorimetric sensing systems using hybrid materials, which combine enzymes and chromogenic substrates, typically relies on a color transition induced by the catalysis of 3,3′,5,5′-tetramethyl biphenyl amine (TMB) through enzymatic or biomimetic enzyme activity. Incorporating TMB into mimetic nanozyme systems enables the creation of various colorimetric sensors with enhanced selectivity and sensitivity through the nanozyme-catalyzed oxidation of TMB [150].

Fig. 4B (i) unveils a groundbreaking microneedle-based colorimetric sensor designed for in-situ glucose detection, employing glucose oxidase-like gold nanoparticles (AuNPs) [128]. This sensor features a double-layered microneedle (MN) patch that ensures seamless and conformal contact with the body while rapidly absorbing interstitial fluid from the skin, achieving 60.2 mg within 10 min. Within the tip layer, the AuNPs catalyze the oxidation of glucose, generating hydrogen peroxide. This compound subsequently reacts with 3,3′,5,5′-tetramethylbenzidine (TMB) in the backing layer, facilitated by horseradish peroxidase, inducing a visible color change for the quantitative assessment of glucose levels across a concentration range from 0.5 mM to 100 mM. This study presents a highly sensitive, minimally invasive approach for real-time blood glucose monitoring.

Fig. 4B (ii) shows the mechanism of a nanozyme-based colorimetric sensor array for precise identification of proteins and oral bacteria [129]. This sensor employs in-situ reduced metal nanoparticles on MoS_2 on polypyrrole surfaces, which interact uniquely with various bio-analytes, offering a distinct fingerprint for each, as shown in Fig. 4B (iii). Utilizing a cross-reactive pattern recognition approach akin to the mammalian olfactory system, the system can discriminate between eleven proteins and five types of oral bacteria. It demonstrates high sensitivity and specificity, capable of identifying proteins and bacteria at nanomolar concentrations and distinguishing closely related hemoglobin from different species, thus showcasing its potential for real-world precise clinical diagnostics.

The integration of nanozyme-based hybrid materials into colorimetric biosensors to create a novel sensing platform holds immense promise for the rapid, sensitive, and convenient detection of various target molecules. This includes, but is not limited to, cancer markers, pathogens, dopamine, cholesterol, and glutathione [151–154]. The versatility of nanozymes render them invaluable in biomedicine, environmental remediation, and diagnostic applications. However, current research faces challenges such as achieving natural enzyme-like catalytic activity, preventing instability and aggregation, and addressing non-specific protein adsorption. Overcoming these issues through surface modifications and doping will be crucial to enhancing their catalytic performance and stability, particularly in complex biological samples.

2.4. Comparison of biomaterials

In this section, we summarize and compare the suitability of hydrogels, fibers, and hybrid materials in various scenarios, highlighting their distinct characteristics, advantages, and limitations.

Hydrogels are particularly well-suited for applications requiring prolonged skin contact due to their biocompatibility and flexibility [155]. Their ability to adhere to and move with the skin without causing discomfort makes them ideal for dynamic environments where flexibility is crucial. Additionally, hydrogels with self-healing properties can autonomously repair damage caused by mechanical stress, ensuring the long-term functionality of wearable devices even under rigorous conditions [55]. This makes hydrogels especially valuable in scenarios where the wearable device is subjected to frequent movement or mechanical strain, such as in continuous health monitoring or athletic performance tracking. Furthermore, the anti-freezing capabilities of some hydrogels enable their use in cold environments, maintaining sensor performance and flexibility in low temperatures [56]. However, the development of these advanced properties often requires complex fabrication processes such as adding chemicals and specific treatments, which can increase production costs and pose challenges for large-scale manufacturing.

Fiber materials are suitable for seamless integration into wearable technologies. Their inherent flexible structure makes them ideal for scenarios where the wearable device is frequently subjected to bending, stretching, and other dynamic movements. The breathable structure of textiles ensures that they can be worn comfortably over extended periods, making them suitable for continuous health monitoring. However, while the porous nature of textiles offers excellent breathability, it may also expose the material to environmental factors such as moisture and contaminants, potentially affecting sensor performance.

Hybrid materials are particularly advantageous for applications that require rapid and visible detection due to their sensitivity and direct signal transduction capabilities [156]. Their intrinsic property to change color in response to specific analytes makes them highly effective for real-time monitoring and diagnostics. This feature is invaluable in environments where timely responses are crucial, such as wearable healthcare applications. Moreover, hybrid materials that incorporate enzyme-catalyzed reactions can provide enhanced specificity and sensitivity, enabling the detection of low-concentration substances [117]. Additionally, some hybrid materials possess robust stability against environmental variables such as pH and temperature changes, making them suitable for use in diverse and challenging conditions. However, the development of highly specific and sensitive hybrid materials can require intricate synthesis processes, which may involve precise molecular imprinting or the incorporation of complex biochemical pathways. These processes can escalate production costs and present hurdles in scaling up for widespread commercial use.

3. Biomaterials integrated into wearable sensors

Biomaterials possess a diverse array of properties, including biocompatibility, lightweight nature, breathability, sensing capabilities, and responsiveness to stimuli, making them uniquely suitable for integration into wearable sensing applications. However, challenges such as integrity issues hinder their direct use in wearables. To overcome this, the integration of biomaterials into wearables emerges as crucial to ensure stable, reliable data collection, and resilience to various environmental conditions. Biomaterials like hydrogels, fibers, and hybrid materials offer distinct properties that render them well-suited for integration into wearable sensors, facilitating applications ranging from healthcare monitoring to fitness tracking. This section delves into the integration of biomaterials into wearables, shedding light on their diverse applications and contributions to the field of wearable technology.

3.1. Integration of conductive hydrogels into wearable sensors

Conductive hydrogels, with their unique properties, facilitate the integration of wearable sensors, enabling the measurement of external stimuli such as stress, strain, and bioelectric signals [15–19]. Their utilization in applications like strain sensors, pressure sensors and electrophysiological sensors underscores their versatility and significance in wearable technology [55,56,67]. In the context of strain and pressure sensors, conductive hydrogels offer exceptional flexibility and responsiveness to mechanical stress such as tensile stress and compressive stress, enabling accurate and reliable strain measurements [55,67]. Additionally, their ability to maintain conductivity under various environmental conditions addresses challenges such as dehydration, ensuring consistent performance over time [56,157–159]. On the other hand, in electrophysiological sensors, the integration of conductive hydrogels provides seamless interfaces between skin and electrodes, crucial for acquiring precise bioelectric signals [55]. Their biocompatibility and adaptability contribute to the stability and accuracy of electrophysiological measurements, offering valuable insights into users' health status [55]. Therefore, the integration of conductive hydrogels enhances the functionality and reliability of wearable sensors across diverse applications, underscoring their pivotal role in advancing wearable technology.

3.1.1. Strain sensors

Conductive hydrogels are highly valued for their flexibility and responsiveness to stimuli, showing great potential in the field of wearable strain sensors [48]. Specifically, they are widely used in strain sensors to measure the amount of deformation or displacement in electrical resistance change that occurs when a material is subjected to tensile stress. However, their practical application often faces significant challenges due to their reliance on water as a dispersion medium, making them vulnerable to dehydration [56,157–159]. For example, in arid climates or at temperatures above 37 °C, these hydrogels can rapidly dry out, severely limiting their practical utility. This susceptibility to dehydration can compromise both their mechanical and electrical properties, increasing the risk of material failure. To address this issue, innovative approaches such as elastomer encapsulation have been developed to reduce water evaporation and preserve structural integrity [68,160–163]. Moreover, recent advancements in material science and integration techniques have aimed to enhance the functionality of these hydrogels, paving the way for improved performance in wearable applications.

Fig. 5A (i) illustrates the fabrication process of the hydrogel, created through a distinct microphase semi-separated network design [163]. This process involves merging PEDOT:PSS nanofibers with PVA and employing facile fabrication methods using 3D printing and successive freeze-thawing. Such methods lead to the physical crosslinking of PEDOT-rich semicrystalline and PVA crystalline domains within an amorphous matrix, solidifying the phase configuration and ensuring the formation of a well-distributed, stable microphase-separated hydrogel network. In this PEDOT:PSS-PVA hydrogel, the hydrophobic PEDOT-rich crystalline domain serves as the electrical phase, ensuring stable electrical performance for strain sensing. Simultaneously, hydrogen bonding-crosslinked PVA domain acts as the mechanical phase, enabling mechanical stretchability and robustness. The phase boundaries between these electrical and mechanical phases are merged and interlocked through various intermolecular interactions among PEDOT, PSS, and PVA chains. These interactions include electrostatic interactions between PSS/PVA and PEDOT, hydrogen bonding within the PVA chain, and chain entanglement among long PVA and/or PSS chains. This phase configuration effectively minimizes hysteresis (<1.5 %) under high strain (>300 %) without compromising electrical/mechanical properties, addressing interfacial detachment and slippage issues commonly observed in conventional hydrogel-based strain sensing materials. Additionally, other critical sensing properties

such as high linearity, mechanical cyclic stability, and inherent resilience against off-axis deformations like torsion and pressure have been successfully achieved. Fig. 5A (ii) demonstrates the encapsulation of the 3D printed hydrogel in silicone elastomers to prevent dehydration, ensure strong bonding between the hydrogel and encapsulating layers, and enhance durability. Silicone elastomers are chosen for their inherent flexibility and compatibility with hydrogels, ensuring the encapsulation layer minimally impacts the hydrogel strain sensor's high stretchability and ability to conform to complex, dynamic surfaces for applications requiring significant deformation [164]. This integration of the hydrogel with electronic skins can be utilized for identifying hand gestures, enabling a soft gripper for object recognition, and remote control of an industrial robot. Strain sensors based on metals and alloys typically exhibit limited stretchability and high hysteresis due to direct stress transfer from the polymeric substrate to the metals and alloys, leading to crack or delamination [165,166]. However, strain sensors based on hydrogel show high stretchability and ultralow-hysteresis due to its unique microphase semi-separated network design. This work offers promising conducting polymer hydrogels with enhanced sensing functionalities and technical platforms for stretchable electronic skins and intelligent robotic systems.

3.1.2. *Pressure sensors*

Conductive hydrogels show great promise in the development of pressure sensors, which measure the force exerted over a specific area by detecting compressive stress. These sensors respond to the application of pressure and convert this compressive stress into an electrical signal. Pressure sensors need to be robust, making them essential in healthcare applications. While elastomer encapsulation can slightly mitigate water evaporation in hydrogels, it remains ineffective in preventing freezing

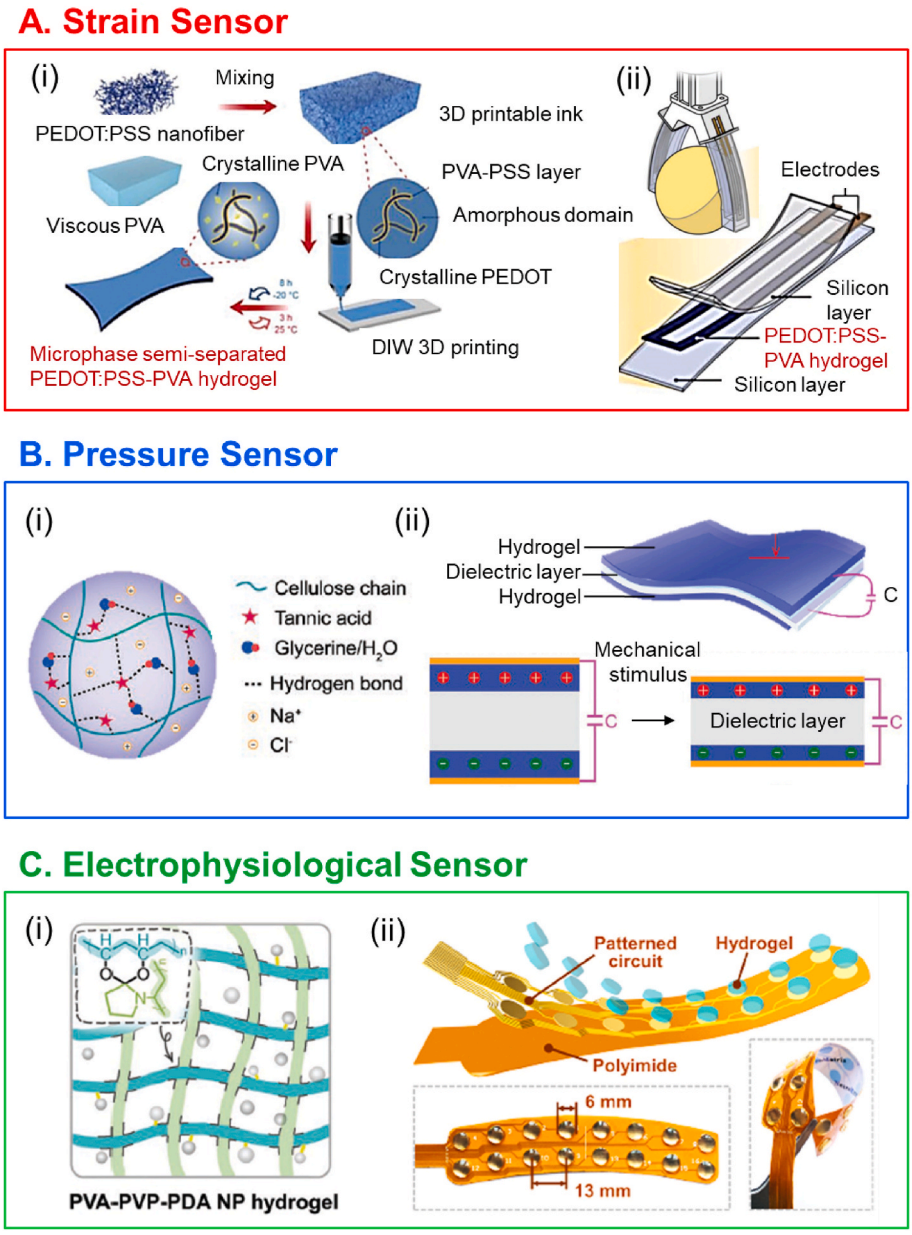

Fig. 5. Hydrogel integrated wearable sensors. (A) Strain sensor. (i) Fabrication process of the PEDOT/PSS-PVA conducting hydrogel. (ii) Illustration of PEDOT/PSS-PVA hydrogel strain sensor integrated into a soft robotic machine. Adapted with permission from Ref. [163]. Copyright 2022 Wiley-VCH. (B) Pressure sensor. (i) Schematic illustration of cellulose-based hydrogel. (ii) Schematic illustration of structural design and sensing mechanism of cellulose-based hydrogel sensor. Adapted with permission from Ref. [10]. Copyright 2022 Wiley-VCH. (C) Electrophysiological sensor. (i) Schematic illustration of PVA-PVP hydrogels with PDA NPs. (ii) Schematic illustration and photographs of the multichannel hydrogel electrode. Adapted with permission from Ref. [183]. Copyright 2023 Wiley-VCH.

under subzero temperatures, thus hindering their use in cold environments [160,167]. Therefore, there is a pressing need to develop conductive hydrogels that are intrinsically resistant to not only drying and high temperatures but also freezing. Recent advancements in material science have focused on enhancing the environmental stability of these hydrogels, enabling their reliable operation across a wide range of conditions and improving their utility in wearable pressure sensors [168–170].

Fig. 5B (i) shows a schematic illustration of a double cross-linked cellulose hydrogel (CH-GT) [10]. Cellulose was chosen as the hydrogel backbone material due to its bio-renewability, multi-hydroxy functionality, abundant availability, and biocompatibility. To enhance the toughness of traditional cellulose hydrogels, a double cross-linked cellulose hydrogel (CH-GT) was developed. This was achieved by using tannic acid (TA), glycerin, water, and NaCl to create an additional physical cross-linked network alongside the existing chemical cross-linked network. Due to the synergistic effects between chemical crosslinking and multi-hydrogen bonding within the hydrogel network, the synthetic CH-GT hydrogel exhibits excellent mechanical strength (2.61 MPa), 21.7-fold higher than that of chemical cross-linked cellulose hydrogel (CH) hydrogel. Additionally, the incorporation of glycerin and water imparts the CH-GT hydrogel with long-term stability at room temperature (2 weeks) and under extreme temperature conditions (−40–60 °C). Overall, the hydrogels exhibit a balanced performance in terms of mechanical strength, long-term, and temperature tolerance. Fig. 5B (ii) shows that the produced hydrogel sensor, with a dielectric layer (VHB 4905, 3 M) sandwiched between two hydrogel layers, senses mechanical stimuli based on the parallel-plate capacitance mode. When the capacitive sensor was compressed or stretched, the distance between the electrodes decreased or the effective overlap area increased, resulting in an increase in capacitance. From this principle, the mechanical vibration from chest/abdomen and pulse could be detected with independent signal output of capacitance. This integrated hydrogel sensor was successfully applied for real-time monitoring of obstructive sleep apnea syndrome (OSAS) with high reliability and accuracy. The multi-modal integrated hydrogel sensor developed in this study shows great potential for personal healthcare monitoring working under various environmental conditions.

3.1.3. Electrophysiological sensors

Conductive hydrogels, which mimic the mechanical properties of human skin, are increasingly valued for their versatile application in establishing seamless and conformal interfaces between skin and electrodes, facilitating the acquisition of electrophysiological signals [171–173]. These hydrogels must possess properties such as low interfacial impedance, self-adhesiveness, low modulus, flexibility, high transparency, and biocompatibility to ensure stable and high-performance sensing capabilities [174]. A vital application of wearable sensing utilizing hydrogel-based technology is the monitoring of users' electrophysiological status, providing crucial insights into their health by detecting abnormal vital signs associated with heart, muscle tissue, and brain-related diseases such as arrhythmia, myocardial infarction, neuromuscular diseases, and epileptic seizures [175–180]. Therefore, the integration of hydrogels into wearable electrophysiological sensors is essential, with recent advancements in material science and integration processes aimed at enhancing the functionality of these hydrogels and paving the way for improved performance in wearable applications.

One significant approach in this domain is the use of self-healing hydrogels, which possess the remarkable ability to autonomously repair both mechanical and electrical damage, making them highly suitable for long-term, durable use in electrophysiological sensing. Wang et al., developed a skin-friendly epidermal electronic made of a self-healing polyvinyl alcohol (PVA) hydrogel to stably monitor multiple physiological signals [181]. The abundant dynamic hydrogen bonds give the hydrogel rapid self-healing ability. Specifically, the hydrogen bonding between tetrafunctional borate ions and the –OH groups from PVA and HPC acts as reversible sacrificial bonds, which can break and reform dynamically at room temperature, enabling rapid self-healing in the hydrogel networks. This epidermal self-healing hydrogel represent a promising wearable platform for high-quality vital sign measurement. Another key strategy in this field is the use of permeable materials, which are vital for maintaining skin breathability and preventing moisture buildup, making them ideal for durable, long-term electrophysiological sensing. Zhang et al. developed an ultrathin gas-permeable hydrogel for long-term, continuous, high-precision electrophysiological monitoring, demonstrating effectiveness under daily life conditions for up to 8 days [182]. The gas permeability is attributed to the thinness geometry and porous nature of the hydrogel. Specifically, the ultrathin, reinforced structure is readily fabricated by immersing PU nanomeshes into a temperature-responsive phase transition hydrogel solution. This permeable hydrogel offers promising skin bioelectronics for long-term, continuous high-precision electrophysiological monitoring, marking a significant step toward noninvasive personalized healthcare.

Fig. 5C (i) shows the preparation process of the hydrogel, which is based on the biocompatible materials PVA, polyvinylpyrrolidone (PVP), and polydopamine nanoparticles (PDA NPs) [183]. PDA NPs were synthesized by treating PDA particles with alkaline H_2O_2, causing the decomposition of PDA into nanoparticles. These PDA NPs were then combined with PVA and PVP to form a hydrogel through a ketalization reaction involving the pyrrolidone and hydroxyl groups of PVP and PVA. Notably, both PVA and PVP chains exhibit excellent biocompatibility and flexibility, making them ideal candidates for producing low-modulus hydrogels suitable for direct contact with the skin. For this purpose, PVA and PVP with a moderate degree of polymerization were chosen to create hydrogels with suitable mechanical properties. The synthesis process facilitated the homogeneous dispersion of PDA NPs within the hydrogel, linking them to the PVA chains via esters. PDA NPs, derived from polydopamine peroxidation, incorporated into the hydrogel are used to enhance its transparency, self-adhesion, and reduce impedance. Overall, the hydrogels exhibit a balanced performance in terms of biocompatibility, high conductivity (<50 Ω), transparency (91.2 ± 2.9 %), low modulus (3.5–6.9 kPa), excellent resilience (94.9 ± 0.2 %) and tissue adhesion strength (39.7 ± 2.5 kPa), showcasing the most comprehensive performance among reported studies. Fig. 5C (ii) shows that the hydrogel is integrated into the flexible printed circuit (FPC) for wearable electrophysiological sensing applications. The hydrogels were then integrated onto the surface of the FPC based on a polyimide substrate with 16 circle-shaped contacts and placed in direct contact with the skin to extract the EEG signals. This resulted in a portable and multichannel hydrogel electrode exhibiting excellent flexibility. Subsequently, an EEG signal analysis system was integrated with the hydrogel electrode for real-time signal transmission, amplification, quantification, and analysis. This technology holds promise not only as a portable device for evaluating sustained attention in both scientific research and daily activities but also as a tool for multifunctional diagnoses and health assessments in medical applications.

3.2. Integration of fibers into wearable sensors

Regarding smart textiles fabrication, yarn and fabric are fundamental elements that play distinct yet complementary roles [184–186]. Yarn, as the basic building block, undergoes processes like coating to imbue it with desired properties such as conductivity or responsiveness to stimuli, ensuring these characteristics are inherently integrated into the textile's structure [187–190]. On the other hand, fabric, formed by interlacing yarns through weaving or knitting, serves as the canvas for these functional enhancements, with post-construction coating allowing for the addition of tailored smart properties [191]. While yarn coating ensures that functionalities are intrinsic to individual fibers, fabric coating offers versatility by treating the entire textile surface, enabling

the incorporation of multiple functionalities for diverse applications in smart textiles. The inherent structure of textiles naturally provides excellent permeability, making them highly suitable for wearable applications. The interlaced fibers in woven or knitted textiles create a porous network that allows for the free passage of air and moisture. Together, these methods synergize to produce garments that seamlessly merge technology with fabric, offering consumers not only comfort and aesthetics but also dynamic interactive capabilities and advanced functionalities. In the following sub-sections, examples of yarn coating of active materials and fabric coating of conductive materials are presented.

3.2.1. *Yarn coating*

Fig. 6A illustrates a detailed yarn coating process, specifically focusing on the preparation of luminescent warp fibers crucial for fabricating electroluminescent (EL) units in smart textiles [192]. This process entails coating commercially available zinc sulfide (ZnS) phosphor onto silver-plated conductive yarn using a simple, scalable solution-based method, ensuring continuous production of luminescent warp fibers. Initially, the conductive yarn undergoes uniform dip-coating in a ZnS phosphor slurry, facilitated by a specially designed scraping micro-pinhole apparatus to achieve consistent and smooth coating. This apparatus, pivotal for controlling the thickness of the ZnS phosphor layer, allows adjustments through different micro-pinhole diameters, with the optimal phosphor layer thickness determined to be approximately 70 μm for this application. Subsequently, the coated luminescent warp undergoes drying, followed by meticulous evaluation of its uniformity to ensure consistent light emission capabilities along its length. The yarn is then integrated into a larger fabric structure using an industrial rapier loom, where it interacts with conductive weft fibers to form the EL units. This interlacing of luminescent and conductive fibers at the weft-warp contact points enables the creation of a display textile suitable for large-scale production and practical application in smart textiles. The process highlights the significance of uniform luminescent coating to achieve reliable and stable light emission across the textile.

3.2.2. *Fabric coating*

Fig. 6B demonstrates a sophisticated dual-regime spray technique employed in the production of custom-designed electronic textiles [193]. This innovative method utilizes a programmable spray system capable of achieving high spatial resolution and deep penetration of conducting nanoparticles into fabrics without necessitating shadow masks or vacuum equipment. The dual-regime spray technique integrates a three-axis computer numerical control (CNC) gantry system, facilitating precise control over the spray's mass loading and depth of penetration. The system employs two separate airflows: a low-speed flow for carrying atomized droplets and a high-speed flow to concentrate and penetrate the droplets into the fabric. The process initiates with the atomization of a solution containing silver nitrate, which is then transported to the fabric through controlled airflows. The nanoparticles are embedded into the fabric via a narrow and focused jet, ensuring uniform coverage and deep penetration. Subsequent layers of copper, Ecoflex, and gold are applied, each serving specific functions such as completing conduction paths, providing electrical insulation, and enhancing biocompatibility. Moreover, the application of an Ecoflex overcoat is crucial in preserving the functionality of smart textiles through multiple wash cycles, protecting embedded electronics from water and detergent. This enhancement extends the life and effectiveness of smart garments, making them more practical and durable for everyday use.

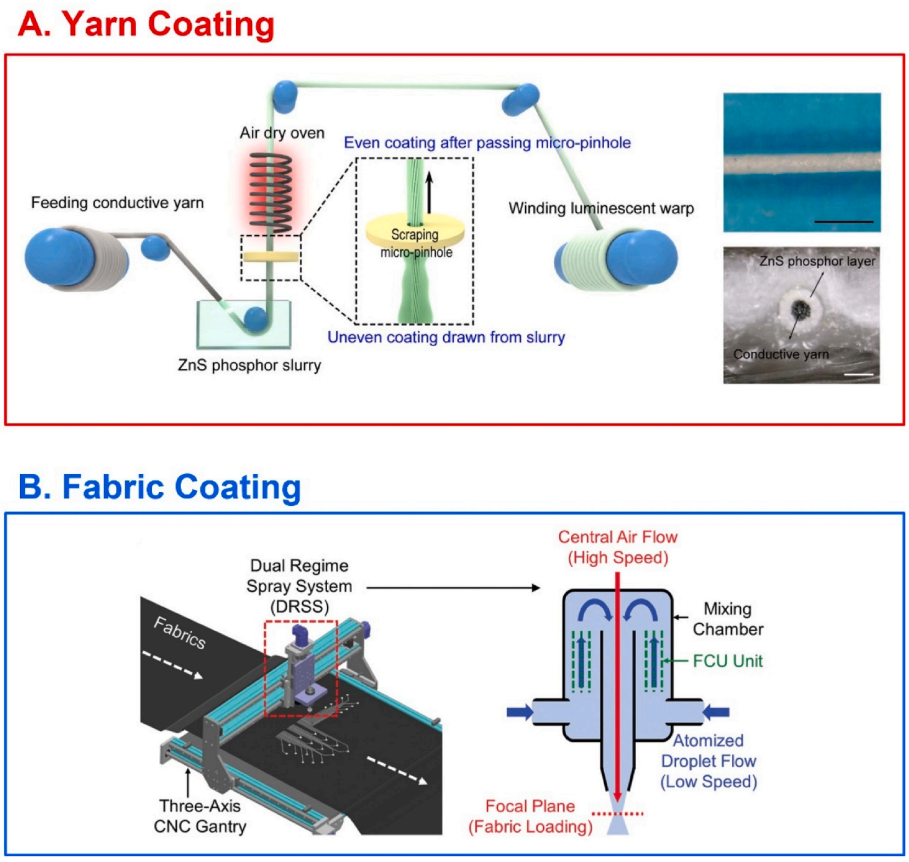

Fig. 6. Coating for smart textiles. (A) Schematic illustration of continuous fabrication of luminescent warp. Adapted with permission from Ref. [191]. Copyright 2021 Springer Nature. (B) Schematic illustration of the dual-regime spray process using a three-axis CNC gantry. Adapted with permission from Ref. [192]. Copyright 2022 Wiley-VCH.

3.3. Integration of hybrid materials into wearable colorimetric sensors

In the realm of wearable technology, the integration of hybrid materials into colorimetric sensors opens avenues for real-time health monitoring and environmental analysis, capitalizing on visible color changes for immediate insights [194,195]. As we explore the integration of these materials into wearable sensors, it's essential to recognize the diverse applications they offer. Microfluidic-based colorimetric sensors, for instance, offer non-invasive monitoring of physiological parameters, providing invaluable data directly from bodily fluids [196,197]. On the other hand, textile-integrated colorimetric sensors represent a seamless and practical approach to health diagnostics, easily blending into everyday clothing for continuous monitoring while providing great permeability [198]. By delving into these subsections, we can gain a comprehensive understanding of how wearable colorimetric sensors are revolutionizing the field of personal healthcare and performance tracking.

3.3.1. Microfluidic-based colorimetric sensors

Wearable colorimetric microfluidic sensors have gained considerable traction for their capability to provide non-invasive, real-time monitoring of various physiological parameters directly from bodily fluids like sweat, saliva, and tears [199–202]. Leveraging microfluidic technology and colorimetric analysis, these sensors offer advantages such as minimal sample requirements, integration into flexible platforms, and the capacity for multiplexed analysis, offering insights into health status during daily activities or athletic performance [199,202]. Particularly adept at capturing and analyzing sweat, these devices employ soft, skin-conformable materials to continuously monitor metabolites, electrolytes, and other essential health indicators with precision and minimal user discomfort [203]. Recent advancements focus on enhancing functionality and user-friendliness through innovations in material science, microfabrication techniques, and integration with phone analysis apps for precise color readings [204].

A cost-effective approach in this domain is paper-based microfluidic sensors, ideal for point-of-care testing and analytical devices [205]. Combining the simplicity and affordability of paper substrates with the specificity and ease of interpretation of colorimetric detection, these sensors offer significant potential [206]. The incorporation of enzymes with paper-based analytical devices significantly improves analytical performance while exhibiting excellent chemical and storage stability. In the work conducted by M. Ariza-Avidad and colleagues, a novel three-dimensional microfluidic paper-based analytical device (3D μPAD) is developed utilizing organic-inorganic hybrid nanoflower technology for glucose detection [207]. This innovative approach incorporates a bi-enzymatic system, specifically glucose oxidase and horseradish peroxidase, embedded within nanoflowers on cellulose paper. By integrating microreactor zones and colorimetric probes within a structured paper device, this system combines the precision of fluid management with the efficiency of enzymatic reactions, harnessing paper-based microfluidics' inherent properties like capillary action and minimal sample needs for accessible and efficient biochemical assays. In an innovative study by Cheng et al., a dual-signal readout paper-based wearable biosensor featuring a 3D origami structure is developed to enhance multiplexed analyte detection in human sweat [208]. Integrating colorimetric and electrochemical sensing zones within a single paper-based device allows simultaneous detection of various sweat biomarkers such as glucose, lactate, uric acid, magnesium ions, cortisol, and pH levels. By employing hydrophilic and hydrophobic treatments on filter paper, effective microfluidic pathways are established, ensuring optimal timing and interaction within the sensing regions. Additionally, Vaquer et al. present a novel chrono-sampling approach for wearable paper-based analytical devices, utilizing dissolvable polymer valves to control sequential sweat flow to multiple sensors [209]. This technique enables time-resolved measurement of biomarkers, significantly improving the temporal resolution of colorimetric sensors in wearable formats.

Fig. 7A illustrates a pioneering wearable microfluidic patch equipped with finger-actuated pumps and valves, which enhances the capabilities for on-demand, longitudinal, and multi-analyte sweat sensing [210]. This innovative device enables users to precisely control the timing of sample analysis in a wearable format, marking a significant advancement toward practical "lab-on-skin" technology. Its design features soft, conformal materials integrated with microfluidic architectures that collect sweat and allow for selective testing through simple manual activation. Demonstrating robust functionality in a two-day human pilot study, the device confirms its ability to perform accurate and reliable sweat analysis. This technology heralds a promising future for non-invasive health monitoring devices, combining flexibility, user-friendliness, and detailed physiological monitoring. Moreover, the use of colorimetric analysis in these microfluidic sensors provides a straightforward and continuous method for biomarker data acquisition, with the potential for enhanced accuracy through image processing [200,211]. This approach shows great promise for wearable body fluid analysis across multiple parameters [212].

3.3.2. Textile-integrated colorimetric sensors

Textile-integrated colorimetric sensors are presenting themselves as an inventive and pragmatic solution for wearable health monitoring, seamlessly integrating into everyday clothing to offer real-time, non-invasive health diagnostics [213–215]. These sensors leverage textiles embedded with nanoparticles or chemically reactive dyes that alter color based on shifts in the wearer's physiological conditions. Their primary application resides in health monitoring, enabling the tracking of metabolic and physiological changes such as pH levels, the presence of metabolites like lactate, and electrolyte levels [216].

Depositing nanoparticles in textiles has been a promising strategy to achieve multifunctional materials for colorimetric sensing. In the study presented by Ji-eun Lee and colleagues, a novel approach for on-fabric, one-touch colorimetric detection using aptamer-conjugated gold nanoparticles is developed [217]. This method leverages the specific binding properties of aptamers with thrombin to induce a visible color change from red to blue on bandages, signaling the presence of the target molecule. This simple yet effective diagnostic tool operates without the need for complex equipment, showcasing potential for rapid, on-site testing of various biomarkers.

Fig. 7B illustrates a wearable colorimetric sweat pH sensor made from curcumin-infused thermoplastic polyurethane, designed to monitor cystic fibrosis by detecting changes in sweat pH [218]. The sensor changes color from yellow to red as pH levels increase, a critical indicator of the disease's progression. This innovative design incorporates electro-spun fibers to enhance sensitivity and reduce response time, allowing for seamless, adhesive-free integration into everyday clothing with excellent permeability. The sensor's functionality and durability are further demonstrated by its ability to withstand multiple wash cycles without losing sensitivity. The sensor's functionality and durability are further demonstrated by its ability to withstand multiple wash cycles without losing sensitivity. The study highlights the sensor's suitability for continuous health monitoring among vulnerable groups such as infants and the elderly, emphasizing its user-friendliness, biocompatibility, and resilience.

Fig. 7C presents a novel thread/fabric-based microfluidic wearable device, specifically designed for the colorimetric analysis of sweat biomarkers [219]. This sensor integrates embroidered threads on hydrophobic fabric to create complex microfluidic channels and reservoirs, utilizing the natural wicking properties of the thread to guide sweat to various sensing zones. These zones are chemically treated to respond to changes in sweat composition, facilitating real-time detection of pH, chloride, and glucose levels. The resulting color changes in response to these biomarkers can be quantified using a smartphone, offering a streamlined monitoring solution that combines accuracy and ease of use. Detection limits are established at 10 mM for chloride concentration, a

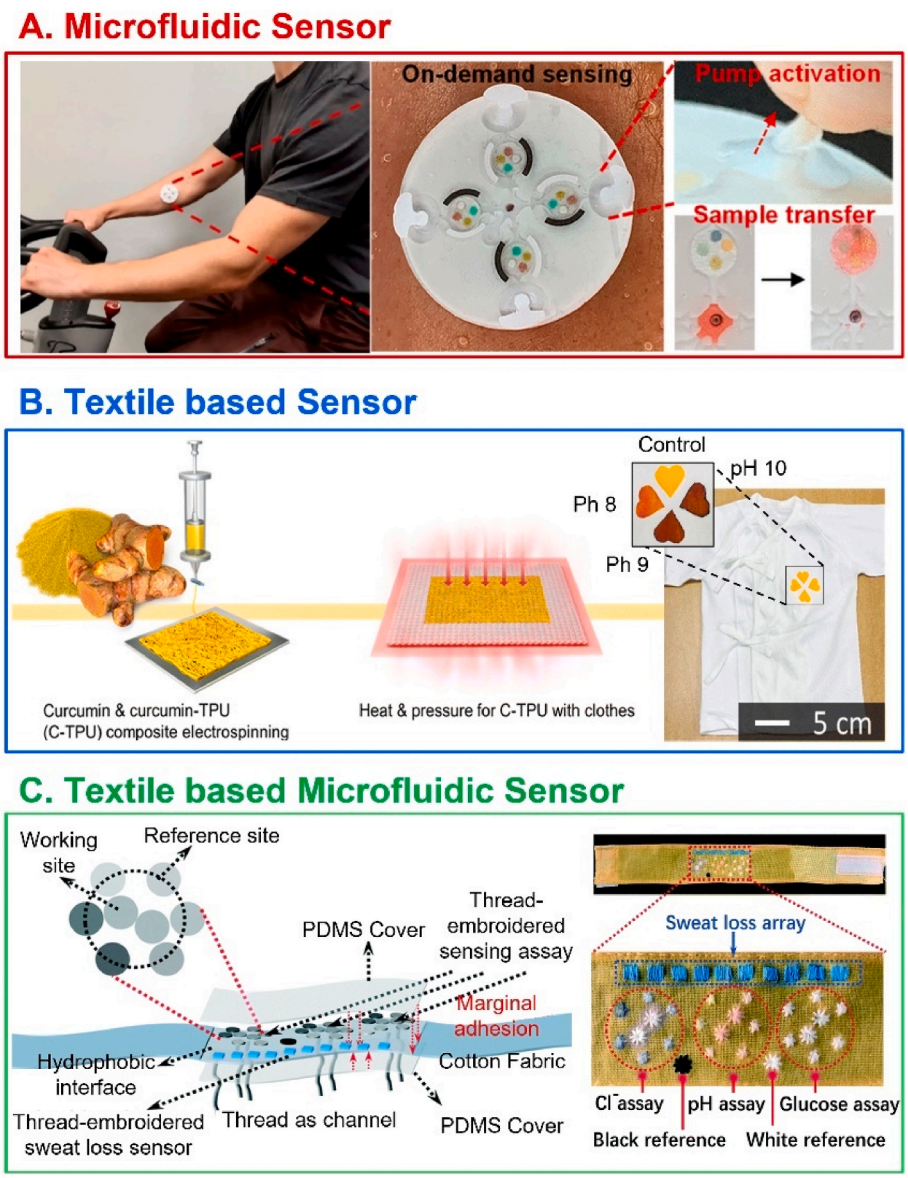

Fig. 7. Wearable devices for colorimetric sensors. (A) Photograph of the microfluidic patch device mounted on the forearm of the candidate with close-up images showing device design and activation of the pump by pulling the tab. Adapted with permission from Ref. [209]. Copyright 2022 American Chemical Society. (B) The scheme shows the electrospinning process of curcumin composite, attachment on fabric by heat and pressing, the C-TPU based colorimetric sensor attached to clothing, showing color change under different sweat pH. Adapted with permission from Ref. [217]. Copyright 2023 Royal Society of Chemistry. (C) Schematic illustration of the detection area in the band as a microfluidic device with digital images of the thread-embroidered patterns showing detection sites in the band. Adapted with permission from Ref. [218]. Copyright 2021 American Chemical Society. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

pH range of 4.0–9.0, and 10 μM for glucose concentration. The study highlights the potential of integrating textile-based sensors with microfluidic systems, providing notable benefits such as flexibility, scalability, customization, lightweight design, and robust mechanical strength.

Furthermore, integrating colorimetric sensors into textiles offers promising opportunities for large-scale production. Matzeu et al. introduce an innovative approach for large-scale patterning of reactive surfaces suitable for wearable and environmentally deployable sensors using screen printing, utilizing the capabilities of silk-based biomaterial inks [220]. These biomaterial-based inks are used to print high-resolution patterns on textiles that are responsive to biochemical stimuli, enabling the colorimetric detection of various analytes, including pH changes. The printed textile sensors can discern pH variations with high precision, detecting changes as minute as 0.1 pH units. These fabrics maintain their functionality and reversibility even after undergoing mechanical durability tests, such as abrasion and dry cleaning, showcasing the robustness of the ink formulations and their practical utility for everyday wear. This technique marks a significant advancement in wearable sensor technology, as it integrates the screen printing of biomaterial-based colorimetric inks that preserve stability and functionality, thus enhancing the practical application of wearable colorimetric sensors in health monitoring.

4. Advancements in health monitoring

Early disease detection is crucial for enhancing patient outcomes and mitigating healthcare expenses [93]. Relying solely on clinic or hospital visits based on observable symptoms often proves insufficient or delayed in detecting numerous diseases [221]. Therefore, there exists a pressing

need for body-interfaced health monitoring systems that provide accurate, on-demand evaluation of health conditions and symptoms in a noninvasive and comfortable manner. To meet this imperative, wearable sensors are integrated with biomaterials, capitalizing on their multifunctionality [222–224]. Recent advancements in biomaterials, including hydrogels, fibers, and hybrid materials, have significantly advanced the development of wearable sensors for health monitoring. This section will delve into the latest innovations in health monitoring, with a particular focus on cutting-edge eye- and skin-mountable sensors facilitated by these functional materials.

4.1. Hydrogel sensor

Hydrogels, crafted from biologic and synthetic hydrophilic polymers, create a network adept at absorbing aqueous fluids, rendering them ideal for ophthalmic applications due to their high hydrophilicity [225]. Given the eye's wealth of physiological data, it serves as an optimal setting for noninvasive disease monitoring via smart contact lenses [21, 226–230]. In this context, hydrogels are pivotal in the development of these lenses for effective disease monitoring in the eye [231–233]. Thus, hydrogels are integrated into contact lenses to monitor glucose levels in tears, aiming to establish a correlation with blood glucose levels. Despite rapid advancements in this field, a significant challenge remains: the inconsistent correlation between glucose concentrations in tears and blood [234–236]. This inconsistency likely stems from the slow diffusion of glucose into tears and the variable sensitivity of the sensors, leading to inaccuracies in glucose level readings [237].

Fig. 8A(i) presents a smart contact lens, integrated with hyaluronate-modified gold-platinum (Au–Pt) bimetallic nanocatalysts immobilized within nanoporous hydrogels, positioned in direct contact with the eye [238]. This configuration offers a durable and sustainable solution for continuous glucose monitoring (CGM) over the long term. The lens

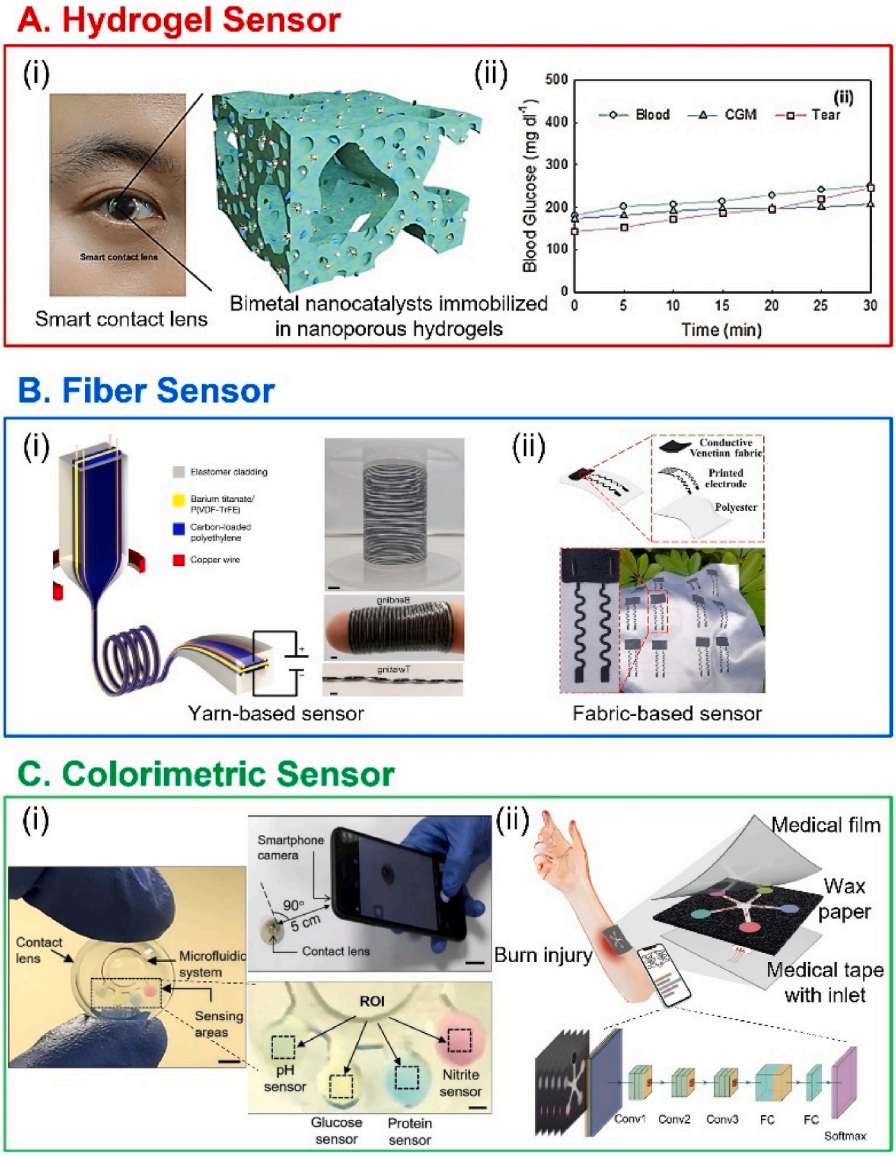

Fig. 8. Skin- or Eye-interfaced health monitoring sensors. (A) Hydrogel sensor. (i) Schematic illustration of smart contact lens for diabetes monitoring. (ii) Continuous glucose monitoring with a glucometer, a commercial CGM and hydrogel-based smart contact lens. Adapted with permission from Ref. [172]. Copyright 2022 Wiley-VCH. (B) Fiber sensor. (i) Yarn-based acoustic sensor. Adapted with permission from Ref. [238]. Copyright 2022 Springer Nature. (ii) Fabric-based pressure sensor. Adapted with permission from Ref. [243]. Copyright 2022 Elsevier B.V. (C) Colorimetric sensor. (i) Microfluidic contact lenses for the colorimetric sensing of tear metabolites. Adapted with permission from Ref. [246]. Copyright 2020 Elsevier. (ii) Illustration of the PETAL sensor for holistic wound assessment by leveraging deep learning algorithm. Adapted with permission from Ref. [252]. Copyright 2023 American Association for the Advancement of Science.

facilitates efficient glucose detection via a redox reaction catalyzed by glucose oxidase within the hydrogel, boasting high sensitivity, rapid response time, and minimal hysteresis. The embedded nanopores in the hydrogels enable reversible and continuous monitoring of glucose concentrations, supported by rapid diffusion of reaction species and swift swelling rates. Post-optimization, the biosensor demonstrated a glucose sensitivity of 180.18 μA cm^2 mM^{-1}, a response time of 3.6 s, and a detection limit of 0.01 mg dl^{-1}. Clinical accuracy assessments using the Pearson correlation coefficient and Clarke error grid analysis showed notable results. The smart contact lens achieved a high correlation value ($\rho = 0.82$), with 94.9 % of the data falling within the clinically acceptable A and B regions of the Clarke error grid, although some data appeared in the D and C regions, indicating underestimated values. The decrease in muscle movement around the eyes due to anesthesia may have contributed to reduced tear circulation and resultant inaccuracies. Further comparative evaluations against a glucometer and a commercial CGM were conducted. Tear glucose levels measured by the smart contact lens were converted to blood glucose levels using a correlation equation. Fig. 8A (ii) illustrates a strong correlation between glucose levels measured in tears by the smart contact lens and those measured in blood by a glucometer. The correlation between blood glucose levels measured by a glucometer and tear glucose levels measured by the smart contact lens ($\rho = 0.95$) demonstrated a strong association, comparable to the correlation observed between a glucometer and a commercial CGM ($\rho = 0.96$). This underscores the potential of contact lens sensors for blood glucose monitoring, positioning the smart contact lens as an excellent alternative to both glucometers and commercial CGM systems, given its remarkable clinical accuracy.

4.2. Fiber sensor

Smart textiles represent a transformative area within wearable technology, where sophisticated materials and advanced fabrication techniques merge to enhance human interaction with their environment [239–242]. In exploring the diversity of smart textiles, it becomes evident that they primarily manifest in two distinct forms: fiber-based sensors and fabric-based sensors. Each type offers unique capabilities and applications, leveraging the intrinsic properties of fibers and fabrics respectively, to serve as conduits for data collection and environmental interaction. This delineation not only highlights the versatility of smart textiles but also underscores their potential in a wide range of applications from healthcare to performance monitoring.

Fig. 8B (i) illustrates a thermally drawn composite piezoelectric fiber that converts audible frequency pressure waves into mechanical vibrations and subsequently into electrical signals [243]. Embedded within the fabric, this fiber enables the textile to function as a highly sensitive audible microphone while retaining traditional textile properties such as washability and flexibility. The fiber is encased in an elastomeric cladding that boosts sensitivity by concentrating mechanical stress onto a piezocomposite layer with a high piezoelectric charge coefficient. Although the piezoelectric fiber constitutes less than 0.1 % of the fabric's volume, it facilitates the creation of large areas of fabric with microphone capabilities. This innovative textile has diverse applications, from determining the direction of sound in a woven shirt to auscultating cardiac sounds, demonstrating its broad potential in sound detection and communication.

Fig. 8B (ii) depicts a fabric-based pressure sensor, where the piezoresistive fabric is crafted by electrostatically depositing a polyurethane/carbon nanotube composite onto Venetian fabric [244]. The interdigital electrodes are fabricated through the screen printing of elastic carbon ink onto polyester fabric. These pressure sensors exhibit high sensitivity, rapid response and recovery times, outstanding cycling stability, and exceptional durability against mechanical bending and chemical washing. Integrated seamlessly into various garments, these sensors enable wireless, real-time monitoring of diverse physiological activities, including joint movements, muscle contractions, and object handling. This efficient and innovative method paves the way for significant advancements in smart garments and wearable technology.

4.3. Colorimetric sensor

Wearable colorimetric sensors have demonstrated significant potential in monitoring physiological parameters crucial for ocular and dermatological health. These sensors are especially beneficial for continuous monitoring, offering a non-invasive alternative to traditional methods that may be invasive or cumbersome [245,246]. In the realm of ocular health, wearable colorimetric sensors can be incorporated into contact lenses to track tear fluid biomarkers. This capability provides essential insights into conditions such as dry eye syndrome and diabetes, and can even potentially detect viral infections through specific biomarkers in tear fluid. Fig. 8C (i) illustrates a microfluidic contact lens engineered for in-situ sensing of pH, glucose, proteins, and nitrite ions in tear fluid [247]. Microchannels were etched into commercial contact lenses using CO_2 laser ablation, with biosensors embedded at strategic locations for rapid and sensitive detection of each analyte. The sensors respond within 15 s, achieving sensitivities of 12.23 nm per pH unit, 1.4 nm per mmol L^{-1} of glucose, 0.49 nm per g L^{-1} of proteins, and 0.03 nm per μmol L^{-1} of nitrites. Utilizing a similar laser patterning technique, Yetisen et al. developed an innovative microfluidic scleral lens sensor for the non-invasive, multiplexed measurement of electrolyte concentrations in the tear film, designed to diagnose dry eye syndrome in point-of-care settings [248]. The scleral lens integrates fluorescent probes to quantitatively assess pH and various ions (Na^+, K^+, Ca^{2+}, Mg^{2+}, and Zn^{2+}) within physiological ranges, enabling continuous monitoring of tear composition and potentially revolutionizing the management of dry eye disease. Additionally, Li et al. introduced a power-free smart contact lens for non-invasive glucose monitoring [249]. This lens features multiple electrochromic Prussian Blue (PB) electrodes that change color in response to glucose concentration variations in tear fluid. This device allows for the real-time visualization of glucose levels ranging from normal (0.16–0.5 mM) to abnormally high (0.9 mM) without the need for external power sources or complex electronics. Demonstrating consistent performance with minimal standard deviations of 0.0462 and 0.025 during repeated use and over several days, respectively, this lens underscores its reliability and potential for daily health monitoring. These innovations in colorimetric sensor-integrated contact lenses represent a transformative advancement in wearable technology, combining the subtlety and convenience of contact lenses with the precise biochemical monitoring capabilities of colorimetric sensors.

For skin monitoring, colorimetric sensors are often embedded into wearable patches or integrated into textiles that maintain direct contact with the skin. These sensors can detect variations in sweat composition or identify specific biomarkers excreted through the skin [250]. Wang et al. have developed a flexible, self-healing, and adhesive wearable hydrogel patch designed for on-demand sweat colorimetric detection [251]. Created using the solvent displacement method, this patch can be directly applied to the skin for in situ sweat sampling and analysis. It demonstrates excellent reliability and stability in measuring pH (4–9), glucose (0–2 mM), Cl^- (0–100 mM), and Ca^{2+} (0–16 mM) in human sweat and integrates seamlessly with smartphones, providing a practical approach for continuous sweat monitoring. The colorimetric hydrogel remains securely intact without separation or shedding during stretching, bending, and twisting, demonstrating the excellent mechanical flexibility of the wearable hydrogel patch. Building on the success of colorimetric sensors in sweat analysis, there is a natural extension towards their application in wound monitoring [252]. Fig. 8C (ii) introduces the paper-like battery-free in situ AI-enabled multiplexed (PETAL) sensor for holistic wound assessment by leveraging deep learning algorithms [253]. This innovative device uses colorimetric sensors embedded in a wax-printed paper panel to detect five critical biomarkers—temperature, pH, trimethylamine, uric acid, and

moisture—key to the wound healing process. Tested on rat models with burn wounds, the PETAL sensor achieves up to 97 % accuracy in distinguishing between healing and non-healing wounds. This technology not only provides a flexible and non-invasive method for tracking wound progression but also enhances wound care management by facilitating early detection and intervention, showing significant potential for clinical applications and home healthcare.

4.4. Biomaterials and sensor designs for wearables

The development of wearable sensors for health monitoring has led to innovations that cater specifically to the unique environments of the skin and eye. Both skin- and eye-mountable sensors demand tailored biomaterials that ensure durability, functionality, comfort, and biocompatibility. This section provides a summary of the biomaterials such as hydrogels, fibers, and hybrid materials, and their sensor designs, focusing on the specific requirements for skin-mountable and eye-mountable sensors.

Skin is a dynamic organ that stretches, contracts, and moves constantly. It undergoes constant motion and is exposed to diverse environmental conditions, such as varying temperature, and mechanical stress. Sensors for the skin must be flexible and durable without any malfunction due to skin movement and external environments.

Hydrogels are highly valued in skin-mountable sensors for their conductivity and flexibility, which can be further enhanced through capacitance-based mechanisms [10] and encapsulation techniques [163]. One common design employs capacitance, where the sensor consists of parallel conductive plates separated by a dielectric material, functioning as a pressure sensor. These sensors detect pressure by measuring the change in capacitance that occurs when the distance between the plates varies due to external forces. As the skin moves, the subtle changes in capacitance allow for the monitoring of physiological signals like breathing, enabling real-time, precise measurements [10]. Another design strategy to enhance durability under harsh conditions involves using encapsulation layers, where the hydrogel is encapsulated in a sandwich structure [163]. In this design, the hydrogel serves as the sensing layer, while two outer layers provide protective encapsulation. These protective layers shield the hydrogel, safeguarding the sensor from mechanical stress and dehydration, thus ensuring long-term stability and performance even in challenging environments [55,163].

Fiber materials in skin-mounted devices are often embedded into breathable fabrics or within stretchable substrates to provide a stable yet flexible platform for sensor integration, without compromising the intrinsic properties of textiles. The loop structure, for example knitted fabric, ensures close contact with the skin, allowing the fibers to move naturally with the body. Conductive and active coatings, such as metallic or polymeric layers, are applied to the fibers, enabling them to capture physiological signals, such as electrical activity or temperature changes, with high sensitivity. To enhance durability, protective coatings or encapsulation techniques are often used to safeguard the fibers against environmental factors like sweat, UV exposure, and moisture, ensuring consistent performance in various conditions.

Hybrid materials for colorimetric sensing are increasingly favored in skin-mountable sensors for their ability to provide visually interpretable data and ease of integration with wearable technologies. One prominent application involves microfluidic devices, where the colorimetric regents are embedded within a network of microchannels that guide fluids to specific sensing zones. These sensors are designed to detect biomarkers in sweat or interstitial fluid, with the color change occurring as the fluid interacts with the sensor, allowing for real-time monitoring of physiological conditions such as electrolyte balance or glucose levels. Another innovative approach integrates hybrid materials into fabrics, where the sensing components are woven or printed directly onto the textile. This design enables the continuous monitoring of skin conditions by observing the color changes that occur in response to the presence of specific analytes. The fabric-integrated sensors are designed to be lightweight, flexible, and comfortable for prolonged wear, making them ideal for daily health monitoring. Both microfluidic patches and fabric-integrated colorimetric sensors offer a non-invasive, user-friendly means of tracking health metrics, providing valuable insights through simple visual cues.

The eye is one of the most complex organs in the human body, holding vital physiological information such as intraocular pressure, glucose, pH, and electrolytes [248,254]. The eye presents a smooth, curved, and sensitive surface, making it essential for ocular sensors to conform to this curvature while maintaining optical clarity. Additionally, the eye's sensitivity necessitates the use of materials that are both biocompatible and permeable.

For eye-mountable sensors, hydrogels are widely used due to their biocompatibility and permeability, which ensure the safety and comfort of smart contact lenses. Moreover, optimal transfer technology is required to transfer function devices from rigid substrates to spherical surfaces of contact lens. In this regard, Zhu et al. developed a method for producing hydrogel-based smart contact lenses for wireless intraocular pressure monitoring using a conformal stacking technique [232]. The poly(2-hydroxyethyl methacrylate) (pHEMA) hydrogel enables permeability and biocompatibility, ensuring the safety and comfort of smart contact lenses. The conformal stacking technique solves the swelling of the hydrogel and the spherical integration of the pyramid-microstructured dielectric elastomer. The resulting device, which features a highly sensitive spherical pyramid-microstructured capacitive pressure sensor combined with a hydrogel substrate, successfully monitors intraocular pressure in an in vitro porcine eye model.

Hybrid materials-based colorimetric contact lens sensors present a compelling solution for non-invasive, real-time monitoring of crucial analytes like glucose, pH, and electrolytes directly from tear fluid [233,247]. These sensors are particularly valuable for detecting biomarkers associated with conditions such as diabetes, dry eye syndrome, and glaucoma, offering continuous health monitoring without the discomfort of invasive procedures. The fabrication of these sensors leverages advanced techniques to enhance their functionality and user comfort. One key approach involves the functionalization of contact lens surfaces with responsive materials, such as enzymes or pH-sensitive dyes, that undergo visible color changes upon interaction with specific analytes. Additionally, laser-engraved microfluidic channels are integrated into the lens, directing tear fluid to designated sensing zones embedded with these reactive chemicals, ensuring precise and efficient analyte detection. These design innovations not only improve the sensors' sensitivity and specificity but also maintain the flexibility, comfort, and durability of the lenses, making them suitable for extended wear. Furthermore, the hybrid materials used in these sensors provide the necessary biocompatibility and mechanical robustness, ensuring that the lenses are both safe and effective for long-term use in daily life.

This section highlights the critical role of tailored biomaterials, such as hydrogels, fibers, and hybrid materials, in the design of wearable sensors for skin and eye applications, ensuring they meet the specific demands of functionality in their respective applications.

5. Conclusion

Biomaterials have significantly propelled the advancement of wearable health monitoring by offering tailored and non-invasive healthcare solutions. The effectiveness of wearable sensors critically depends on the integration of sophisticated biomaterials, essential for ensuring seamless operation and compatibility with the human body. The durability and stability of these materials are crucial in maintaining the reliability of wearable technologies, fostering smooth integration with human tissues and consistent performance over time. By exploring key biomaterial categories such as hydrogels, fibers, and hybrid materials, researchers have identified unique traits that make them particularly suited for use in durable and stable wearable health monitoring systems. Recent advancements in biomaterial-based sensors hold

promising prospects for early detection, preventive interventions, and personalized healthcare, providing real-time monitoring capabilities and invaluable insights into users' physical well-being, which are vital for effective disease prevention and management.

Despite these benefits, challenges remain in effectively integrating these materials with skin and eye tissues, often exacerbated by environmental factors. These issues limit the practical application of wearable technologies in health monitoring and impede the industrialization of biomaterial-based sensors. Overcoming these challenges requires the development of biomaterials that feature durable and stable functionality, including self-healing properties, resilience to harsh conditions, washability, and flexibility, ensuring continuous operation in various environments without significant degradation.

The integration of these robust biomaterials into wearable sensors, equipped with wireless data processing, also ensures stable, reliable remote data collection under diverse environmental conditions. This integration enhances home-based telehealth services, enabling sustained use in various settings and reducing the frequency of hospital or clinic visits, thus improving the management of therapies and treatments for patients, doctors, and caregivers worldwide.

However, further challenges include the need for improvements in the scalability and cost-effectiveness of biomaterial-based sensors [255]. Achieving scalability involves developing manufacturing processes that can efficiently produce sensors in large volumes without compromising quality or increasing costs. Enhancing cost-effectiveness requires optimizing materials and production techniques to reduce manufacturing expenses while maintaining sensor performance and reliability. Addressing these challenges will necessitate interdisciplinary collaboration to innovate and refine sensor designs and production methods, ultimately advancing the accessibility and affordability of wearable health monitoring technology.

Moreover, there are significant opportunities for future enhancements in wearable health monitoring systems that could substantially improve clinical outcomes and healthcare delivery. The application of advanced algorithms, apps, artificial intelligence (AI), and machine learning could revolutionize clinical decision support and caregiver assistance. These technologies are capable of analyzing vast amounts of data from wearable sensors in real-time, providing predictive insights and personalized recommendations, which are crucial for proactive health management. Additionally, the simplification and batch fabrication of biomaterials synthesis and integration techniques could lead to high-volume production of low-cost devices, dramatically increasing the accessibility of advanced health monitoring technologies, especially in under-resourced areas or rural regions. This could facilitate widespread adoption and lead to improved health outcomes across diverse populations.

Given the significant impact of these technological and procedural advancements and their clinical relevance, ongoing research in the field of human skin and eye-interfaced health monitoring systems is essential. Continual innovation is necessary not only to enhance the functionality and efficiency of these devices but also to ensure their ethical use in healthcare settings. Such research will drive further innovations, refine existing technologies, and promote the widespread clinical adoption of wearable health monitoring systems, setting the stage for a more informed, empowered, and health-conscious global population.

CRediT authorship contribution statement

Seokkyoon Hong: Writing – review & editing, Writing – original draft, Visualization, Investigation, Formal analysis. **Tianhao Yu:** Writing – review & editing, Writing – original draft, Visualization, Investigation, Formal analysis. **Ziheng Wang:** Writing – review & editing, Writing – original draft, Visualization, Investigation, Formal analysis. **Chi Hwan Lee:** Writing – review & editing, Writing – original draft, Visualization, Supervision, Investigation, Funding acquisition, Formal analysis.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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