

## REVIEW



# Recent Progress in High-Performance Conductive Hydrogels for Wearable Sensors

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**Abstract:** Hydrogel materials are regarded as an ideal platform for constructing next-generation high-performance, bio-integrated wearable sensors due to their unique softness, high water content, excellent biocompatibility, and tunable conductivity. This review aims to systematically summarize recent advances in the field, providing a comprehensive exploration of the entire technical pathway—from material design and performance regulation to device integration and application. We first analyze optimization strategies for key properties of hydrogels—such as conductivity, self-healing, adhesion, and environmental stability—through approaches like nanocomposite integration and dynamic cross-linking. And we further discuss advanced fabrication techniques, including the construction of hybrid physical–chemical cross-linking networks and 3D printing. In addition, we categorize five types of hydrogel sensors—resistive, capacitive, piezoelectric, triboelectric, and electrochemical—based on their sensing mechanisms, detailing their working principles and performance characteristics. Finally, this review outlines their specific applications in cutting-edge fields such as physiological monitoring, human–machine interaction, and implantable medical devices. Research findings demonstrate that hydrogel sensors can balance flexibility and sensing performance under moderate strain levels (100–200%) and controlled hydration conditions, whereas extreme deformation or prolonged exposure to dry environments often leads to signal drift or mechanical degradation enabling strain and pressure sensing with reported gauge factors typically ranging from ~2 to 100, detectable pressure ranges from a few Pa to hundreds of kPa, and operational lifetimes from hours to several weeks depending on encapsulation and environment.

**Keywords:** hydrogels, conductive hydrogels, wearable sensors, flexible electronics, human–machine interaction

## 1. Introduction

The Artificial Intelligence of Things is driving a paradigm shift in wearable technology, evolving from passive data logging to autonomous, closed-loop bio-interactive systems. Future wearables are envisioned not merely as accessories but as imperceptible, skin-integrated interfaces capable of seamless physiological monitoring and environmental perception. However, a grand challenge impedes this vision: the intrinsic mechanical and biological mismatch between rigid silicon-based electronics and soft, curvilinear biological tissues, which severely compromises long-term signal fidelity and user comfort.

In this context, conductive hydrogels emerge as the quintessential candidate for next-generation bioelectronics [1]. Possessing a unique water-rich, three-dimensional porous architecture analogous to the extracellular matrix, hydrogels offer distinct advantages over traditional materials. Their tunable viscoelasticity, ionic conductivity, and intrinsic self-healing capabilities allow them to bridge the biotic–abiotic interface effectively. This modulus-matching characteristic minimizes interfacial impedance and motion artifacts, creating unprecedented opportunities for high-fidelity, continuous biometric sensing without inducing immune responses.

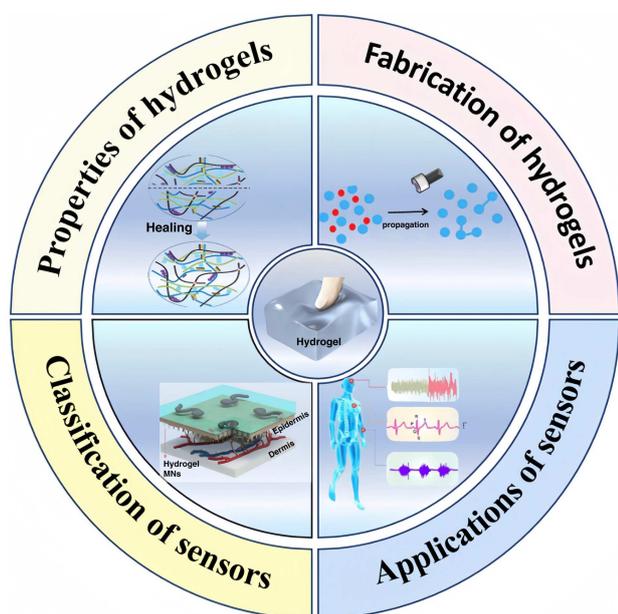
Despite their immense potential, translating laboratory prototypes into robust commercial products remains hindered by formidable technical bottlenecks. Conventional hydrogels are plagued by a notorious trade-off between mechanical robustness and electrical sensitivity, as well as susceptibility to environmental fluctuations. Specifically, inevitable dehydration and freezing in extreme conditions lead to the catastrophic loss of flexibility and conductivity. Furthermore, achieving stable bio-adhesion while maintaining dynamic reversibility remains a critical hurdle.

This review consolidates current research advancements in hydrogels, focusing on innovative materials engineering strategies—such as incorporating multifunctional nanomaterials and developing dynamic, reversible cross-linking networks—that “empower” hydrogels with enhanced properties critical for wearable sensors. These strategies facilitate the precise tuning of key characteristics, including conductivity, self-healing, adhesion, and frost resistance. Moreover, the application of additive manufacturing (3D printing) technology has enabled the realization of complex hydrogel sensor structures, allowing for personalized designs and system integration. This progress is pivotal for meeting the demanding requirements of intelligent wearable devices, enabling high performance, portability, and multifunctionality, and represents a critical step toward their practical application. Furthermore, we also classify and discuss hydrogel sensors based on various sensing mechanisms—piezoresistive, capacitive, and notably, self-powered piezoelectric and triboelectric modalities—highlighting

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their advantages in achieving high sensitivity, self-powering capabilities, and other intelligent features. By integrating frontier research with practical applications, we map advances in hydrogel sensors to transformative applications, including real-time biomonitoring, adaptive human-machine interfaces, and autonomous theranostic systems. It is hoped that those will not only serve as a comprehensive technical reference for future research but also inspire innovative ideas at the intersection of materials science, electronic engineering, and artificial intelligence (AI), accelerating the transition of hydrogel sensors from laboratory concepts to transformative, real-world wearable devices. The main content is illustrated in Figure 1 [2–4].

**Figure 1**  
The graphical abstract from hydrogel property optimization to wearable hydrogel sensors application description



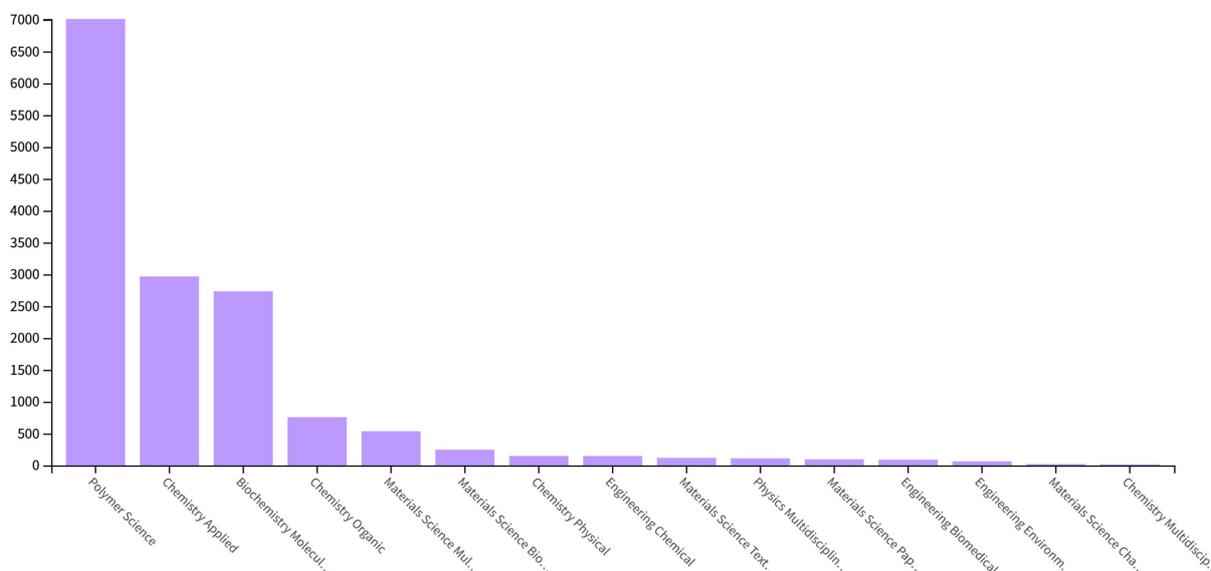
Recent years have witnessed extensive reviews on hydrogels and conductive hydrogels, hydrogel bioelectronics, self-powered generators (triboelectric/piezoelectric), and electrochemical wearable sensors. Yet, these reviews are frequently organized around either *material families* or *single transduction mechanisms*, which makes it difficult for readers to translate material-level choices into device-level performance and, ultimately, wearable-ready system decisions. In practice, wearable sensors require simultaneous optimization of multiple coupled metrics (e.g., sensitivity vs measurement range, conductivity vs mechanical robustness, adhesion vs removability, and environmental stability vs flexibility/dynamic responsiveness), but a unified, decision-oriented synthesis across mechanisms and engineering levels remains insufficient.

To address this gap, this review adopts a holistic engineering perspective along the entire technical pathway—from hydrogel property regulation and fabrication strategies to representative sensing mechanisms and application-driven integration—so that general design rationales and key trade-offs can be distilled and used for modality selection under realistic wearable constraints. We therefore discuss five representative routes (resistive, capacitive, piezoelectric, triboelectric, and electrochemical) within a comparable framework and further emphasize system-level considerations that often dominate real-world performance, such as dehydration/freezing tolerance, drift and durability, encapsulation, and scalable manufacturing toward integrated wearable and bio-integrated electronics.

## 2. Literature Search and Study Selection

To systematically review representative and cutting-edge research on high-performance conductive hydrogels for wearable sensors, we conducted a structured literature search. The databases and sources included Web of Science Core Collection, MDPI, ScienceDirect, and Nature, primarily covering the period from January 2024 to December 2025. The search combined hydrogel/material terms with keywords related to wearable sensing, such as (conductive hydrogel, ionic hydrogel, electronic hydrogel) AND (wearable sensor, electronic skin, strain sensor, pressure sensor, bioelectrode, bioelectronics) AND (self-healing, adhesion, antifreeze). The distribution of literature from the Web of Science search results is shown in Figure 2. In addition, we

**Figure 2**  
Distribution of Web of Science subject categories (analyze results)



screened references from key review articles and foundational papers to supplement potentially relevant studies not indexed in the databases.

Literature screening was carried out in two stages: title/abstract and keyword screening, followed by full-text content screening. The inclusion criteria were conductive hydrogels (ionic, electronic, or composite types) with clearly described compositions/network structures, applications targeting wearable or skin-integrated sensing or hydrogel bioelectronic interfaces, and reporting of at least one performance metric relevant to wearables (e.g., conductivity, stretchability, gauge factor/sensitivity, response time, durability, adhesion strength, or environmental tolerance). Any disagreements during the screening process were resolved through discussion and consensus.

Data extracted from the included studies encompassed hydrogel matrix and conductive phase, cross-linking strategies, key mechanical/electrical properties, device configurations, sensing modes, testing conditions (humidity/temperature/strain rate), and critical performance metrics. Given the heterogeneity in material systems, testing protocols, and reporting formats, this study did not perform a meta-analysis. Instead, we focused on a qualitative synthesis, emphasizing the extraction of universal design principles and performance trade-offs across different sensing mechanisms.

### 3. Properties of Hydrogels

Hydrogels, characterized by their unique three-dimensional hydrophilic networks and high water content, represent a class of soft matter that closely mimics the physical and chemical nature of biological tissues. This intrinsic similarity—particularly in terms of mechanical compliance and ionic richness—renders them the ideal candidate for bridging the gap between rigid electronics and soft human skin. However, conventional hydrogels generally suffer from inherent electrical insulation, mechanical fragility, and environmental instability (e.g., dehydration or freezing), which severely restrict their reliability in practical sensing scenarios [1, 5].

To overcome these barriers, recent advancements have transitioned from simple component mixing to rational molecular engineering and nano-microstructure design. By strategically introducing conductive fillers and optimizing cross-linking strategies, researchers have successfully endowed hydrogels with properties that were previously mutually exclusive. In this section, we provide a comprehensive overview of the critical properties essential for high-performance wearable sensors, including electrical conductivity, self-healing capability, adhesion, and environmental stability. Understanding and tailoring these physicochemical properties is pivotal for developing the next generation of skin-integrated electronics with superior sensitivity, durability, and user comfort.

#### 3.1. Electrical conductivity

Electrical conductivity, as one of the fundamental properties of conductive hydrogels, has consistently been a key parameter of focus for researchers. Enhancing conductivity is crucial for expanding the practical applications of hydrogels [6, 7]. Notably, current effective strategies can be primarily classified into two categories. The first involves incorporating electrolytes into the polymer network to increase ion concentration, which is typically employed to improve ionic conductivity. The second approach introduces conductive polymers or other conductive materials into hydrogels to enhance electronic conductivity. Consequently, based on their conductive mechanisms, hydrogels can be classified

into ionic conductive hydrogels and nonionic conductive hydrogels, which utilize ions or electrons as charge carriers, respectively (Figure 3(a, b)) [8, 9].

##### 3.1.1. Electronic-conductive hydrogels

Electronic-conductive hydrogels (ECHs) rely on the transport of electrons or holes through a percolated network formed by conductive fillers within the insulating polymer matrix. Common fillers include metal nanoparticles (e.g., Ag, Au), carbon nanomaterials (e.g., CNTs, graphene), and conductive polymers (Figure 3(c)) [10, 11]. However, a critical challenge in ECHs is the trade-off between conductivity and stretchability. High filler loading, required to reach the percolation threshold, often leads to aggregation and stress concentration, severely embrittling the hydrogel network. To circumvent this, recent research focuses on nano-microstructural design to balance these properties. Zhang et al. [12] engineered a hybrid network of cellulose nanofibers (CNF) and CNTs, where CNF assisted the dispersion of CNTs, maintaining stable conductivity under strain, simultaneously achieving durability exceeding 1000 cycles [13–15].

To address the potential toxicity and processing rigidity of inorganic fillers, intrinsically conductive polymers (ICPs) such as polyaniline (PANI), polypyrrole, and poly(3,4-ethylenedioxythiophene) (PEDOT) have gained prominence [7, 16, 17].

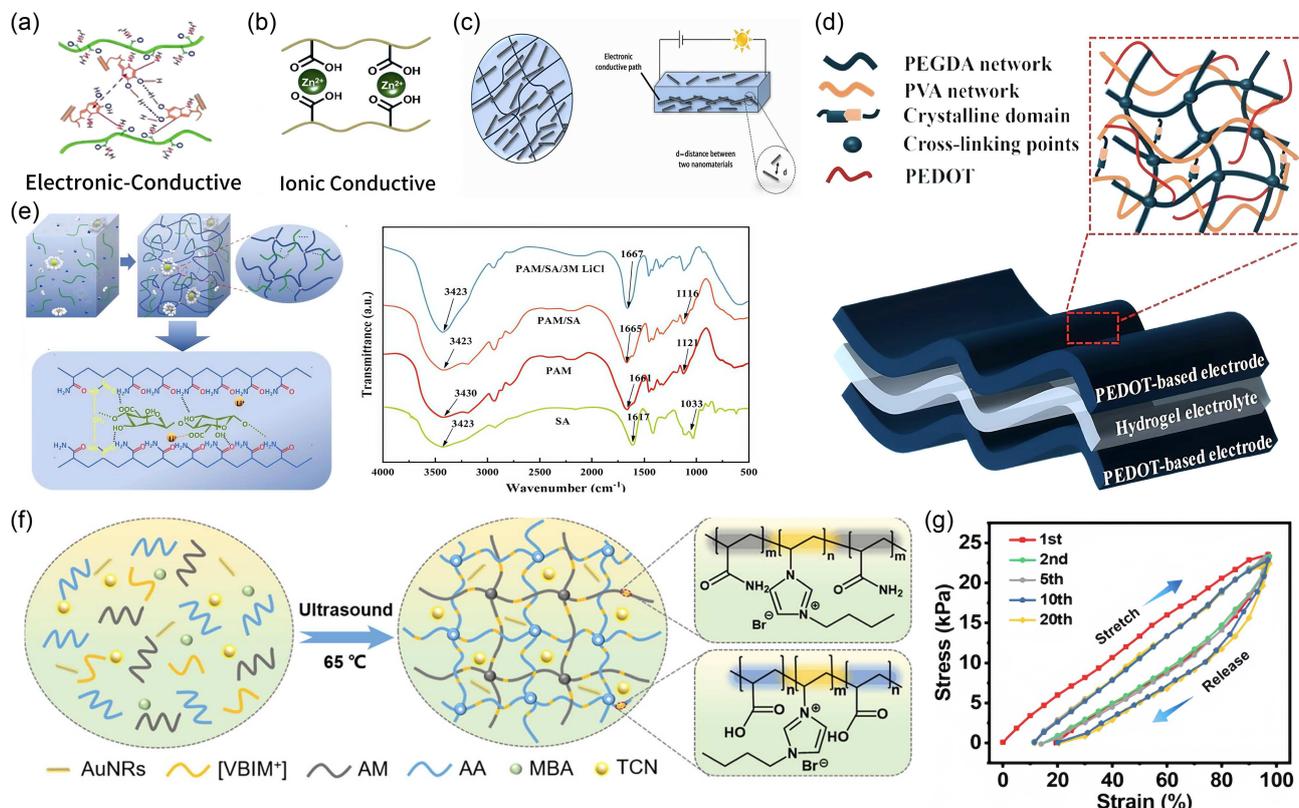
Unlike rigid carbon fillers, ICPs offer better molecular-level compatibility with hydrogel matrices. Conductive polymers not only enable the control of electrical conductivity through switching between two states (semiconducting and conductive, doped and dedoped) but also allow for the modulation and maintenance of mechanical properties through chemical modification, doping, and composite formation [18–20]. And some natural polymers, such as chitosan, are not only non-toxic and suitable for fabricating tough hydrogel-like materials but also present excellent properties in other aspects, such as wound healing and antibacterial activity [21–23]. This versatility facilitates the balanced integration of multiple functionalities. Among these, PEDOT:PSS is particularly distinguished for its high conductivity and water processability. For example, by interpenetrating semi-crystalline poly(vinyl alcohol) (PVA) chains with PEDOT networks, excellent electrochemical stability is achieved—combining a conductivity of 5 S/m with capacitance retention of 97.6% after 3000 charge–discharge cycles (Figure 3(d)) [24]. Furthermore, to tackle the brittleness of dry PEDOT:PSS, plasticizers like glycerol or ionic additives are often introduced to disrupt the rigid polystyrene sulfonate (PSS) domains. Reynolds et al. [25] utilized sodium trimetaphosphate to modulate the interactions within PEDOT:PSS, significantly reducing stiffness without sacrificing conductivity, achieving high-performance conductivity of  $(1.78 \pm 0.63) \times 10^7$  S/m and making it ideal for skin-conformal electronics [26]. Similarly, emerging assembly techniques, such as stretch-drying-induced alignment and salting-out-assisted aggregation, allow for the precise ordering of conductive polymer chains, yielding hydrogels with anisotropic conductivity and superior mechanical tunability (strength: 17.13–142.1 MPa; toughness: 50 MJ·m<sup>-3</sup>) [27, 28].

##### 3.1.2. Ionic conductive hydrogels

Unlike ECHs, ionic conductive hydrogels utilize mobile ions as charge carriers—a mechanism analogous to signal transmission in biological nerves, primarily achieved through electrolytes (Figure 3(e)) [29, 30]. This unique feature endows ionic conductive hydrogels with distinct advantages, including optical

Figure 3

Electrical conductivity of hydrogels: (a) charge transport mechanism in nonionic hydrogels, (b) ion migration mechanism in ionic conductive hydrogels, (c) conductive filler-based and nanomaterial-based hydrogel architectures, (d) structural configuration of PEDOT-PVA/PEGDA hydrogel, (e) electrolyte-based ionically conductive hydrogel system and its FT-IR spectra, (f) ionic liquid (IL)-enabled conductive hydrogel design, and (g) stretch-recovery cycles for the IL hydrogel at 100% tension strain



transparency, tunable adhesion, and superior impedance matching at the bioelectronic interface [29, 30].

Large amounts of water stored by hydrogels accommodate numerous free ions that act as charge carriers. Typically, ionic conductivity is enhanced by immersing hydrogels in ionic liquid (IL) solutions or by adding inorganic salts to increase the ion concentration within the hydrogel [27]. For example, directly immersing the hydrogel into an electrolyte salt solution, Ye et al. [31] improved both ionic conductivity and mechanical properties of a PVA/cellulose nanofibril hydrogel by inducing a gel-sol transition and immersing it in a salt solution, achieving a high stretchability of up to 660%. Another method is the solution replacement technique, which involves immersing a pre-formed hydrogel in an electrolyte solution. This allows for better control over ion concentration and solvent ratios. Zhang et al. [32] used this technique to enhance the conductivity and mechanical properties of an SA-gelatin hydrogel by immersing it in a  $\text{CaCl}_2$  solution, resulting in a uniform incorporation of calcium ions. Meanwhile, the challenge of ionic conductive hydrogels freezing at sub-zero temperatures needs to be solved, as this freezing leads to a significant loss of conductivity and stretchability. While increasing salt content in hydrogels enhances electrical conductivity and anti-freezing capability, excessively high salt concentrations can trigger a “salting-out” effect, adversely affecting the mechanical properties of the hydrogel. To overcome this, Wu et al. [33] applied a “salting-out” effect by adding potassium acetate to induce the salting-out of PVA. This strategy enhanced solubility while enabling it to reach a conductivity of 8.0 S/m at room temperature and remain 1.2 S/m at  $-60^\circ\text{C}$  [34, 35].

Beyond aqueous electrolytes, ILs have emerged as a powerful alternative due to their excellent properties (Figure 3(f, g)) [29, 30]. Composed entirely of ions, ILs possess negligible vapor pressure and wide electrochemical windows, fundamentally solving the dehydration issue [36]. While traditional imidazolium-based ILs may raise toxicity concerns, recent efforts have shifted toward bio-ionic liquids, such as choline-amino acid-based ILs. A multifunctional hydrogel synthesized by using biocompatible poly(ionic liquids), which exhibited stable conductivity, exceptional strain sensitivity, and long-term durability without the risk of solvent evaporation [37].

### 3.1.3. Summary

It is evident that the electrical conductivity of hydrogels largely depends on the type and content of the conductive materials incorporated. While increasing the content of traditional fillers can enhance conductivity, it often introduces multiple associated challenges. The introduction of ICPs for mechanical enhancement, along with the adoption of natural polymers and biocompatible ILs to mitigate toxicity, demonstrates the critical impact of selecting suitable novel conductive materials on improving conductivity. Furthermore, the rational combination of strategies based on new material approaches may present a promising research direction.

## 3.2. Self-healing ability

Wearable sensors are inevitably subjected to repetitive mechanical deformations (e.g., stretching, twisting) during long-term

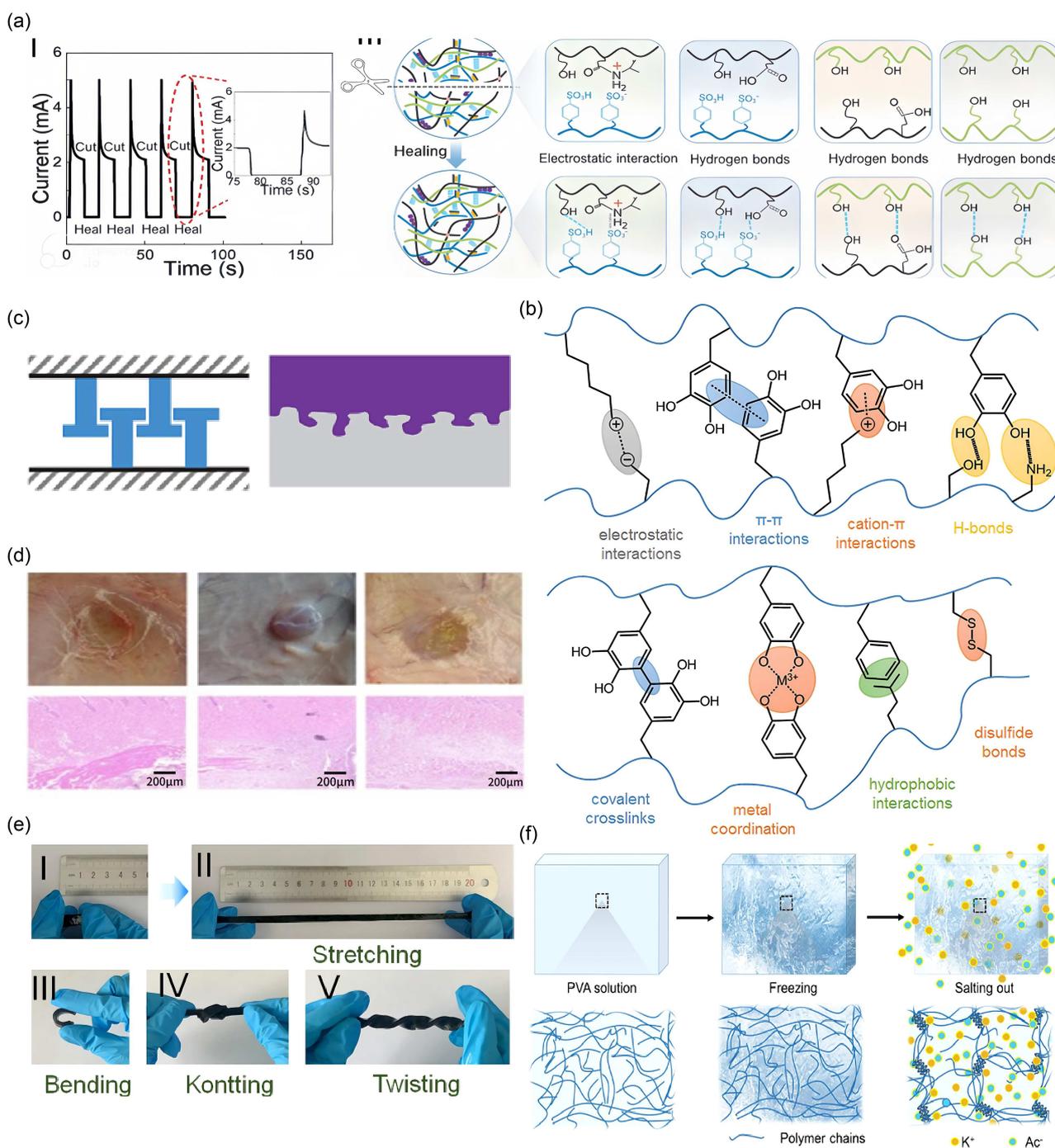
usage, leading to the accumulation of micro-cracks and eventual structural failure. More critically, for conductive hydrogels, these microscopic damages sever the electron transport pathways, causing irreversible signal drift or open-circuit failures. Therefore, imparting self-healing capability—the ability to autonomously restore both mechanical integrity and electrical functionality—is paramount for extending the service life and reliability of wearable electronics (Figure 4(a)) [2, 6].

Self-healing processes in hydrogels are primarily divided into two types: extrinsic and intrinsic. Intrinsic self-healing processes,

which are self-contained within the material, have garnered significant attention due to their simpler mechanisms and more robust repair capabilities compared to extrinsic processes that require external inputs. Fundamentally, intrinsic self-healing relies on the presence of dynamic reversible motifs within the polymer network. Unlike permanent covalent cross-links, these dynamic bonds can dissociate to dissipate energy under stress and re-associate to heal damage, driven by the system's tendency to minimize surface free energy and return to thermodynamic equilibrium [38]. These interactions are generally categorized into physical interactions

Figure 4

Key properties of hydrogels: (a) self-recovery behavior, (b) bonds and interactions governing adhesiveness, (c) mechanical interlocking mechanism, (d) biocompatibility evaluation in a murine model, (e) assessment of mechanical stability, and (f) anti-freezing ionic conductive hydrogel fabricated via a freeze-soaking method



(e.g., hydrogen bonding, electrostatic interactions, hydrophobic associations, host–guest interactions) and dynamic covalent bonds (e.g., Schiff base, disulfide bonds, boronate ester bonds).

Supramolecular interactions inspired by biological systems are widely adopted strategies. For instance, mimicking the adhesive proteins in marine mussels, a pH-responsive hydrogel based on metal–ligand coordination between DOPA (3,4-dihydroxyphenylalanine) and iron ions had been developed. The dynamic nature of the  $\text{Fe}^{3+}$ -catechol complex is susceptible to pH; a shift from acidic to alkaline conditions promotes the formation of higher-order coordination complexes (mono- to bis- to tris-complexes), thereby triggering a rapid sol–gel transition and efficient healing [39].

However, achieving self-healing in conductive hydrogels presents a unique challenge: the healing of the polymer matrix does not guarantee the reconnection of the conductive network. To address this, recent research focuses on the synergistic reconstruction of both the mechanical matrix and the electrical percolation network. The high mobility of PVA chains can facilitate the migration of conductive fillers (CNTs and carbon nanoparticles) to the fracture interface, and this dynamic network reconstruction achieves a healing efficiency of 89.82% and ensures that the re-establishment of mechanical contact is synchronously accompanied by the restoration of electrical conductivity, eliminating the need for external stimuli. Such autonomous electrical healing is critical for ensuring artifact-free signal acquisition in continuous health monitoring scenarios [40].

It can be observed that, on the one hand, the self-healing efficiency of hydrogel systems is closely coupled with environmental variations, limiting their universality in complex environments. On the other hand, even when environmental dependencies are overcome, inconsistencies in the restoration of various hydrogel properties persist. Future research should focus on developing environmentally robust dynamic bonding systems and designing conductive networks with topological adaptability, enabling real-time synchronization of mechanical and electrical recovery to advance self-healing sensing from laboratory demonstrations toward practical applications.

### 3.3. Bio-adhesive properties

The fidelity of physiological signal acquisition in wearable sensors is fundamentally governed by the quality of the bioelectronic interface. Unlike rigid electronics, the human epidermis is a curvilinear, dynamic, and moisture-secreting surface. Therefore, achieving robust conformal contact is critical not only to prevent device detachment but also to minimize interfacial impedance and suppress motion artifacts during physical activities [41].

Conventional adhesives (e.g., medical tapes) often fail in humid conditions or cause skin irritation upon removal. In contrast, hydrogels, with their tissue-like softness and abundant functional groups, offer a promising platform for intrinsic adhesion. Strategies to engineer high-performance adhesive hydrogels generally exploit two distinct mechanisms: chemical bonding and physical interactions, or a synergistic combination thereof (Figure 4(b)) [41].

Chemical adhesion relies on the formation of covalent bonds between the hydrogel matrix and the substrate. While static covalent bonds provide high bond strength, they are often irreversible. Consequently, recent research has pivoted toward dynamic covalent chemistries (e.g., Schiff base reactions, boronate ester bonds), which offer reversible and repeatable adhesion. For instance, a covalently cross-linked chitosan-acrylamide hydrogel that formed

robust bonds with diverse substrates, leveraging the abundant amine and hydroxyl groups for interfacial anchoring [42].

Physical adhesion, on the other hand, is driven by non-covalent interactions (e.g., hydrogen bonding, electrostatic interactions, van der Waals forces) and topological entanglement. The latter—often described as mechanical interlocking—is particularly effective on rough substrates (e.g., textiles or skin with micro-wrinkles), where polymer chains infiltrate surface micro-crevices and increase the effective contact area. From an engineering perspective, the resulting adhesion enhancement can be understood as a synergy between intrinsic interfacial bonding (intrinsic interfacial toughness) and bulk/network viscoelastic dissipation, both of which should be co-optimized and quantitatively reported when designing wearable hydrogel adhesives (Figure 4(c)) [41, 43–45].

However, a single adhesion mechanism often necessitates a trade-off between bond strength and removability. To address this, bio-inspired hybrid strategies have emerged as a standard. Drawing inspiration from the adhesive proteins of marine mussels, polydopamine (PDA) and catechol chemistry have been widely adopted to achieve “universal adhesion.” A tough double-network (DN) hydrogel integrating polyacrylic acid, chitosan, and PDA was constructed, and the adhesive strength of this hydrogel can reach  $(22.78 \pm 1.47)$  kPa. This design synergistically combines the strong wet adhesion of catechol groups (chemical) with the energy dissipation of the polymer network (physical) [46]. Similarly, Liu et al. [47] introduced nucleobases—fundamental units of DNA—into polyacrylamide networks. These hydrogels exhibited superior adhesion through a “lock-and-key” mechanism involving multiple hydrogen bonds and metal complexation. Such hybrid approaches not only ensure reliable fixation under extreme deformation but also enable on-demand detachment, solving the long-standing conflict between durability and biocompatibility in skin-integrated electronics.

Current hydrogel adhesion strategies have achieved significant progress through bio-inspired designs; however, both physical and chemical adhesion mechanisms exhibit inherent limitations. Drawing inspiration from the hybrid cross-linking approaches in hydrogels, integrating multiple adhesion mechanisms to construct dynamic adhesive interfaces that combine high stability with environmental responsiveness remains a promising and worthwhile direction for further exploration.

### 3.4. Biocompatibility and biodegradability

When wearable or implantable sensors come into contact with biological tissues—especially in scenarios involving implantation inside the body—they may trigger an immune rejection response from the host. This can disrupt the normal functioning of health monitoring devices and potentially lead to health issues such as inflammation or tissue damage. Therefore, ensuring excellent biocompatibility is crucial for preventing immune rejection and ensuring the proper operation of these devices.

Biocompatibility is generally defined as the ability of a biomaterial to perform its desired function in a medical therapy without causing undesirable local or systemic effects in the recipient. It should also generate the most beneficial clinical or performance outcomes. Essentially, it refers to a material’s ability to perform with an appropriate host response in a specific situation [48]. In the context of hydrogel development, improving biocompatibility often involves reducing the toxicity of the hydrogel and its components, preventing tissue and cell damage, and minimizing host rejection. Figure 4(d) [49] provides *in vivo* evidence that the hydrogel interface can mitigate host responses at

the tissue–device boundary, which is essential for stable long-term wearable/implantable signal acquisition.

Compared to synthetic polymers, natural biopolymers (e.g., alginate, gelatin, chitosan, cellulose, pectin) have emerged as the premier choice for constructing the hydrogel matrix in terms of biocompatibility. Unlike synthetic counterparts that may require toxic cross-linkers, these materials offer inherent bioactivity, hemocompatibility, and low immunogenicity. For instance, Markov et al. [50] engineered a pectin-based hydrogel that demonstrated exceptional blood compatibility, serving as an inert barrier to prevent post-operative adhesions without compromising red blood cell integrity. Similarly, Sun et al. [51] developed a skin-friendly sensor using a biocompatible matrix that effectively mitigated host rejection, ensuring safety during long-term monitoring.

Beyond mere safety, the field is rapidly evolving toward “Transient Electronics”—devices that can physically disappear after their service life. Biodegradability is becoming a critical dimension of biocompatibility, aiming to minimize electronic waste and obviate the need for secondary retrieval surgeries. Hydrogels based on cleavable linkages (e.g., ester bonds, disulfide bonds) or enzymatic degradation mechanisms allow sensors to decompose into harmless byproducts (CO<sub>2</sub>, water, inorganic salts). This paradigm shift transforms hydrogel sensors from passive “compatible” materials into active, eco-friendly systems that harmonize with both the human body and the environment.

Biocompatibility research has evolved from the stage of “safe coexistence” to “functional synergy.” While natural polymers provide a favorable biocompatibility foundation and more readily meet design requirements for degradability, they still face challenges in terms of precise functional regulation, stability of material properties, and scalability. Moving beyond their inherent functional limitations and achieving the precisely controllable, high-performance characteristics typical of synthetic polymers represents a critical direction for future breakthroughs.

### 3.5. Long-term stability and environmental tolerance

The transition of hydrogel sensors from laboratory prototypes to commercial products is contingent upon their stability under rigorous operating conditions. Unlike rigid electronics, hydrogels—as “wet” materials—face intrinsic thermodynamic instabilities, primarily including mechanical fatigue, dehydration, and freezing. Consequently, engineering hydrogels with robust environmental tolerance and long-term fatigue resistance—maintaining stable mechanical properties after undergoing processes such as folding and stretching—is pivotal for practical wearable applications (Figure 4(e)) [2, 28, 52].

Mechanical robustness serves as the physical foundation for reliable sensing. Conventional hydrogels often suffer from a strength-toughness trade-off due to their high water content and homogeneous networks. To overcome this, strategies focusing on energy dissipation mechanisms have been widely adopted. The construction of DN architectures, which incorporate sacrificial bonds (e.g., hydrogen bonds, ionic coordination) to dissipate energy during deformation, has proven effective in preventing catastrophic fracture [26, 28, 53, 54]. For instance, substantial research efforts have been devoted to utilize phase separation and sacrificial networks to fabricate tough hydrogels capable of withstanding high-stress cycles; through this, hydrogels can stretch up to 6564% of their original length [55, 56]. Meanwhile, the integration of liquid metal (LM) droplets into a Polyacrylamide (PAM) network, where the droplets acted as stress-transfer centers, resulted in a hydrogel that, despite a partial reduction in

tensile strain (2866%), attained a favorable electrical conductivity of 5.27 S/m [57].

Beyond bulk mechanical properties, dimensional stability becomes critical as sensors trend toward miniaturization. The high surface-to-volume ratio of micro-scale devices accelerates dehydration and structural collapse. Addressing this, Wang et al. [58] engineered an ultrathin (2.7 μm) hydrogel electrode reinforced by a polyurethane nanomesh. This bio-inspired exhibits relatively low stretchability (238%) yet provides exceptional mechanical support and moisture retention and also withstands ultra-low temperatures down to −90 °C, ensuring stable operation even at the micro-scale.

Environmental tolerance, particularly against extreme temperatures, is another major hurdle. Various methods, such as the freeze-soaking technique, have been applied to enhance the freeze-resistance performance of hydrogels (Figure 4(f)) [33]. At sub-zero temperatures, the crystallization of water molecules into ice lattices not only blocks ion transport channels—resulting in a loss of conductivity—but also mechanically damages the polymer network. The mainstream strategy to impart anti-freezing properties relies on the colligative properties of binary or ternary solvent systems. By introducing cryoprotectants (e.g., glycerol, ethylene glycol) or inorganic salts (e.g., LiCl, NaCl), the hydrogen bond network of water is disrupted, significantly depressing the freezing point [59, 60].

However, a critical challenge remains: the trade-off between anti-freezing performance and self-healing kinetics. To improve environmental tolerance while maintaining mechanical and self-healing properties, Jia et al. [61] introduced dynamic Schiff base bonds and LiCl into a hydrogel system. This salt not only prevented freezing but also plasticized the network, enabling rapid self-healing (within 5 min) even at −20 °C. Furthermore, to fundamentally eliminate the issues of evaporation and freezing, ILs have been employed as nonvolatile, wide-temperature solvents. IL-based ionogels could maintain stable sensing performance across a broad temperature window (−50 °C to 80 °C), while preserving the material’s mechanical properties (tensile strength: 554.37 kPa) and self-healing capability (self-healing efficiency = 92.51% ± 0.40% within 3 h) [62].

In conclusion, due to the inherent trade-off between mechanical stability and environmental tolerance, current strategies (such as introducing eutectic solvents or ILs), while effectively broadening the operational temperature range, often come at the cost of compromised mechanical or self-healing properties. Future breakthroughs may require moving beyond the simplistic “additive modification” paradigm and shifting toward the intrinsic design of hydrogel polymer networks. This approach would enable synergistic optimization of mechanical robustness and environmental adaptability at the molecular level, achieving a more balanced overall performance profile. The key data from the relevant studies in this section are summarized in Table 1.

## 4. Fabrication Strategies of Hydrogels: From Molecular Design to Structural Engineering

The macroscopic performance of conductive hydrogels is intrinsically dictated by their microstructure across multiple length scales. Therefore, fabrication is not merely a mixing process but a sophisticated structural engineering endeavor. Generally, fabrication strategies can be classified into two paradigms: (i) bottom-up network construction, which focuses on modulating molecular interactions (chemical or physical cross-linking) to determine

**Table 1**  
**Summary of reported properties of hydrogels**

Hydrogel system	Stretchability (Elongation at break / Strain)	Mechanical strength	Toughness/ Fracture-related properties	Electrical conductivity	Self-healing	Operating limit(s)	Ref.
Nanofiber hydrogels	335–471%	Tensile stress up to 0.79 MPa	Durability >1000 cycles	~0.4 S/m	Self-recovery without external stimulation	–	[12]
(PVA/PEGDA) hydrogel	1–498%	Young's modulus 15 kPa	Capacitance retention 97.6% after 3000 cycles	5 S/m	–	–	[24]
Trimetaphosphate-based PEDOT:PSS hydrogels	–	Stiffness (mean G' in Linear Viscoelastic Region (LVR)) 105~106 Pa	–	Conductivity (1.78 ± 0.63) × 10 <sup>7</sup> S/m	–	–	[26]
Super-robust hydrogels	~ 500%	17.13–142.1 MPa	Toughness 50 MJ/m <sup>3</sup>	Up to 30.1 S/m	–	–20 °C	[28]
Nanofibrils organohydrogel	Up to 660%	Ultimate tensile strength: 2.1 MPa	Up to toughness 5.25 MJ/m <sup>3</sup>	3.2 S/m	–	–70 °C	[31]
Ald-alginate–gelatin imine-hydrogel	~380%	Stress up to 35 kPa	–	1.5 S/m	Rapid self-healing (within 5 min)	–20 °C	[32]
Anti-freezing hydrogels	–	Tensile strength 8.2 MPa	Toughness 25.8 MJ/m <sup>3</sup>	8.0 S/m (RT); 1.2 S/m at –60 °C	–	–70 °C	[34]
Acid polyionic liquids hydrogels	Up to 900%	Tensile strength up to 0.42 MPa	Toughness up to ~ 1.9 MJ·m <sup>-3</sup>	0.409–0.798 S/m	Up to 87.1%	–	[37]
CNTs/C-NPs/PVA–borax hydrogel	611%	Tensile strength 0.026 MPa; Young's modulus 0.054 MPa	–	–	Healing efficiency 89.82%	–	[40]
Chitosan-based hydrogels	Exceeding 300%	–	Interfacial toughness up to 2413 J/m <sup>2</sup>	0.04 S/m	–	–	[42]
Tough double-network hydrogel	–	Fracture strength: up to (37.29 ± 1.15) kPa	–	–	–	–	[45]
Pectin-based hydrogel	Tested at 10–80%	Young's modulus up to 101 kPa	Enhanced elastic–plastic stability	–	–	–	[49]
Multifunctional hydrogel	322.5%	–	Interfacial toughness 30.9 J/m <sup>2</sup>	0.109 S/m	Recovery at 37 °C for 10 min	40 °C	[50]
Cross-linked network hydrogel	> 700%	Tensile fracture stress ~ 300 kPa	Toughness up to 1.6 MJ/m <sup>3</sup>	–	–	–	[55]
LM-doped hydrogel	2866.44%	Tensile stress 389.81 kPa	Toughness 5.75 MJ/m <sup>3</sup>	5.27 S/m	Tensile stress recovery up to ~100%	–	[56]

(Continued)

**Table 1**  
(Continued)

Hydrogel system	Stretchability (Elongation at break / Strain)	Mechanical strength	Toughness/ Fracture- related properties	Electrical conductivity	Self-healing	Operating limit(s)	Ref.
Nanomesh- reinforced composite hydrogel	238.4%	Flexural rigid- ity~ $1.0 \times 10^{-3}$ nN·m	Durability: 1000 cycles at 100% strain	0.1 S/m	–	–90 °C	[57]
Double- cross-linked organohydrogel	$1322 \pm 75\%$	Tensile strength up to $160 \pm 5$ kPa	Toughness up to $956 \pm 38$ kJ/m <sup>3</sup>	~2.4 S/m	–	~–30 °C	[58]
Dual dynamic cross-linking hydrogel	>800%	–	–	0.27 S/m	–	–20 °C	[60]
Ionically con- ductive organohydrogel	Up to 2119% $\pm 48\%$	Tensile strength up to 554.37 kPa	Toughness up to $4.25 \pm 0.13$ MJ·m <sup>-3</sup>	0.81 S/m	Healing effi- ciency $92.51 \pm 0.40\%$ (3 h)	–50 °C to 80 °C	[61]

intrinsic material properties, and (ii) top-down advanced manufacturing, such as 3D printing, which tailors the geometric configuration for specific device integration [63, 64].

#### 4.1. Gel network construction

The polymer network serves as the topological skeleton of the hydrogel, governing its mechanical modulus, fracture toughness, and environmental stability. The strategy employed to construct this network—whether through permanent fixation or reversible association—is the primary determinant of the sensor's durability and dynamic responsiveness. Based on this, network construction is categorized into chemical cross-linking and physical cross-linking (Figure 5(a, b)) [1, 64].

##### 4.1.1. Chemical cross-linking

Chemical cross-linking relies on covalent bonds to form the hydrogel's network, offering superior mechanical stability compared to non-covalent interactions. Unlike physical cross-linking, which is reversible, chemical cross-linking involves irreversible reactions such as free radical polymerization and reactions between complementary functional groups [65]. A distinctive feature of chemical cross-linking is the formation of an integrated network through reactions between cross-linkers and polymer chains, making the selection of cross-linking agents critically determinant of hydrogel performance [64, 66].

Phototriggered cross-linking is a key manufacturing lever for wearable hydrogel sensors because it enables spatially programmed curing and patterning, which underpins light-based fabrication such as digital light processing (DLP)/Stereo lithography (SLA) printing and on-demand *in situ* gelation. As an illustrative example of emerging “dual-role” photochemistry, Pt(IV)-complex-mediated photolysis can generate radicals to initiate polymerization while concurrently contributing to network formation (Figure 5(c)) [3]. Importantly, for wearable/bioelectronic translation, such chemistries should be assessed against application-specific constraints, rather than being viewed only as a synthetic novelty. In addition, PVA, as a hydrophilic material,

is often used in chemical cross-linking due to its processing ease, high mechanical strength, and lightweight properties. PVA can generate free radicals necessary for polymerization and is commonly used with cross-linkers like ethylene glycol dimethacrylate to create pH-sensitive hydrogels [67].

However, purely static chemical cross-linking, as a singular and static methodology, exhibits several limitations, including the lack of dynamic behavior, inability to self-heal, and relatively brittle mechanical properties. These drawbacks render it inadequate for meeting the stringent material performance demands of cutting-edge applications. To overcome these limitations, dynamic covalent chemical cross-linking, which employs reversible covalent bonds as cross-linking points, has become the mainstream approach within chemical cross-linking. However, they are often associated with drawbacks such as complex preparation procedures and slow response times. Consequently, increasing attention has shifted toward physically cross-linked hydrogels, which feature fast, reversible interactions and can be triggered under mild conditions [62].

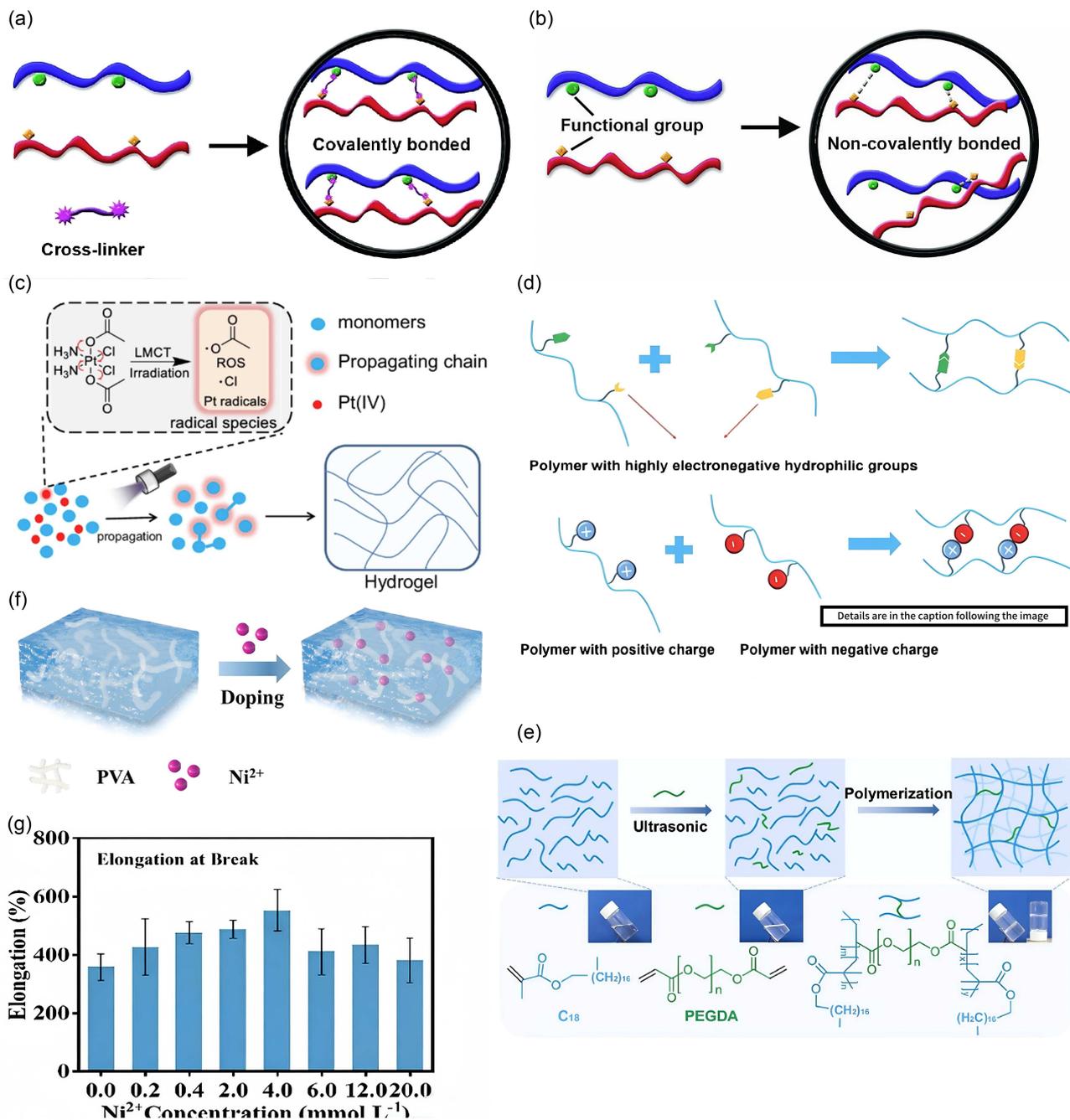
##### 4.1.2. Physical cross-linking

Unlike the rigid architecture of chemically cross-linked networks, Physically Cross-Linked Hydrogels rely on reversible non-covalent interactions—such as hydrogen bonding, electrostatic interactions, hydrophobic associations, and host-guest interactions—to form dynamic networks (Figure 5(d)) [68]. The defining feature of these hydrogels is their ability to dissipate mechanical energy through the rupture and reformation of physical bonds, a mechanism that endows them with viscoelasticity, self-healing capabilities, and high toughness. Based on their topological complexity, they are categorized into single-network (SN) and multi-network (MN) systems.

In SN systems, toughness is derived from the density and strength of reversible cross-links within a homogeneous network. Early strategies focused on singular interactions, such as electrostatic attraction. For instance, Rahmani et al. [69] utilized LM droplets coated with cellulose nanocrystals (CNCs) as multifunctional cross-linkers to initiate the polymerization of acrylic acid.

Figure 5

Physical and chemical cross-linking in hydrogels: (a) schematic illustration of the chemical cross-linking principle, (b) schematic representation of the physical cross-linking mechanism, (c) generic photoinitiated polymerization/cross-linking, (d) schematic of hydrogen bonding and electrostatic interactions as physical cross-linking strategies, (e) preparation process of an organogel via free radical polymerization, (f) fabrication of Ni<sup>2+</sup>-doped PVA hydrogel through immersion of PVA hydrogel in a nickel ion solution, and (g) elongation at break of PVA/Ni<sup>2+</sup> hydrogels with different Ni<sup>2+</sup> doping concentrations



The electrostatic interactions between the carboxyl groups and the LM surface created a robust network, achieving a remarkable stretchability of ~2000% (Figure 5(e)) [63]. Similarly, the strengthening mechanism of polyampholyte hydrogels was elucidated that the stochastic distribution of opposing charges leads to the formation of strong ionic bonds that serve as effective energy dissipation centers to achieve high toughness with a fracture energy of 4000 J·m<sup>-2</sup> and 100% self-recovery [70, 71]. To further elevate mechanical performance, beyond strategies that

integrate multiple interactions—including hydrogen bonding, electrostatic interactions, and covalent bonding, “Phase Separation Engineering” also has been widely adopted to introduce heterogeneous structures, such as crystalline domains, which act as high-functionality physical cross-links [68, 72]. Freeze-thaw cycling has become a common practice for the treatment of PVA [13]. Huo et al. [73] demonstrated that additives like carbon dots (CDs) can serve as nucleation agents and CDs could accelerate the crystallization of PVA, promoting energy dissipation through

crack deflection and bridging. In contrast to Rahmani et al. [69], this study focused on tensile strength and achieved a tensile strength as high as 156 MPa. The Ni<sup>2+</sup>-doped PVA hydrogel engineered by Wang et al. [74], where ice crystal formation induced the segregation of PVA chains into dense crystalline domains, balanced tensile strength with other properties. These domains, reinforced by Ni<sup>2+</sup>-coordination bonds, functioned as “physical anchors,” significantly enhancing the modulus, fracture strain (580%), and tensile strength (1.1 MPa) (Figure 5(f) and (g)) [74].

Despite the elegance of SN designs, they often face an insufficiency in balancing stiffness and stretchability. To break this limit, the construction of DN or MN architectures has become the gold standard. The core principle, pioneered by Gong et al. [75], involves the interpenetration of a rigid, tightly cross-linked first network (sacrificial network) and a soft, loosely cross-linked second network (matrix) [76]. Upon deformation, the sacrificial network fractures internally to dissipate massive amounts of energy, while the soft matrix maintains macroscopic integrity.

Recent advances have extended this concept to all-physical DN hydrogels to incorporate self-healing properties. Zhuang et al. [77] designed a fully physical DN system where dynamic borate ester bonds served as the sacrificial first network, and a hydrogen-bonded PVA/CNF network acted as the elastic matrix. The rupture of borate bonds provided energy dissipation, while the CNF reinforcement ensured structural stability. Crucially, the addition of NaCl not only facilitated the phase separation of conductive PEDOT:PSS but also provided mobile ions for conductivity.

Pushing the complexity further, Zhang et al. [78] constructed a triple-network architecture by integrating PVA, quaternized chitosan (QCS), and a polyacrylate copolymer. This design synergistically leveraged three distinct physical mechanisms: electrostatic attraction between QCS and polyacrylate, extensive hydrogen bonding, and chain entanglement. The resulting hydrogel exhibited a “1 + 1 + 1 > 3” synergistic effect, combining high stretchability (~2113%), robust adhesion, and superior sensitivity (electrical conductivity: 0.77 S/m), thereby meeting the multifaceted requirements of high-performance wearable sensors.

#### 4.1.3. Hybrid cross-linking

While chemical cross-linking provides structural permanence and elastic recovery, and physical cross-linking offers dynamic reversibility and energy dissipation, neither alone suffices for high-performance sensors. Consequently, hybrid physical–chemical cross-linking has emerged as the paradigm of choice (Figure 6(a)). [41] The design philosophy hinges on a “division of labor”: a sparse chemical network maintains the macroscopic shape (entropic elasticity), while a dense physical network sacrifices itself to dissipate energy under stress (viscoelasticity), thereby achieving a synergistic balance of stiffness, toughness, and fatigue resistance [79].

The most direct implementation is the hybrid DN strategy, combining a rigid chemical network with a ductile physical network. Zhang et al. [80] realized a conductive hybrid DN by interpenetrating a chemically cross-linked PAM network with a physically cross-linked PANI/CNT/PAA network. Here, the covalent bonds ensured structural stability, while the hydrogen bonds between PANI/CNTs and the polymer matrix acted as reversible “sacrificial fuses.” This synergy endowed the hydrogel with favorable mechanical properties—fracture strain ~659%, tensile strength ~200 kPa, and toughness ~691 kJ/m<sup>3</sup>—and endowed the

sensor with robust environmental stability (anti-freezing/drying) and high sensitivity, surpassing SN counterparts.

Incorporating nanofillers as “Multifunctional Cross-linking Hubs” represents another frontier. Unlike simple blending, these fillers (e.g., CNTs, CNCs, micelles) form strong interfacial interactions with polymer chains [81, 82]. Similarly, Pan et al. [83] utilized vesicular CNCs as physical anchors within a chemically cross-linked NIPAM-SA network. The CNCs, combined with hydrophobic associations and ionic coordination, created a hierarchical energy dissipation system, significantly toughening the hydrogel. Micelles can also serve as macro-cross-linkers; Zhou et al. [84] utilized Sodium Dodecyl Sulfate (SDS) micelles to physically anchor hydrophobically modified PAM chains, creating a transparent, highly stretchable hydrogel via a “micellar copolymerization” strategy [85].

Given the inadequate fatigue resistance of many DN and nanocomposite hydrogels, along with the structurally heterogeneous networks often resulting from free radical polymerization-based chemical cross-linking. To resolve this, topological cross-linking—often termed “slide-ring” technology—has been developed. Karino et al. [86] introduced polyrotaxanes where cross-linking points are figure-of-eight molecules threaded onto polymer chains. Under tension, these cross-links slide along the chains like pulleys (“pulley effect”), automatically equalizing chain tension and homogenizing stress distribution (Figure 6(b)) [86]. Building on this, Zhu et al. [87] integrated topological sliding entanglement into a hybrid DN system. The resulting hydrogel combined the permanence of chemical bonds with the infinite deformability of sliding cross-links, achieving exceptional fatigue resistance and fracture toughness (Figure 6(c)) [87]. These studies, while balancing multiple properties, have respectively achieved high electrical conductivity (13.9 S/m) and high fracture strain (>3000%).

#### 4.1.4. Summary

In summary, the fabrication of high-performance hydrogels has undergone a paradigm shift from simple polymerization to sophisticated network topology engineering. While traditional chemical cross-linking ensures structural permanence at the cost of brittleness, and physical cross-linking offers self-healing capabilities but limited stability, state-of-the-art hybrid strategies (e.g., DN, nanocomposite, slide-ring) have effectively reconciled the inherent stiffness-toughness trade-off. The field is now advancing toward “programmable networks,” where the spatiotemporal control over cross-linking density and dynamics promises to underpin the next generation of intelligent, adaptive wearable sensors. The key data from the relevant studies in this section are summarized in Table 2.

## 4.2. Incorporation of additive manufacturing technologies

While the molecular engineering of hydrogel networks (as discussed in Section 3.1) determines intrinsic material properties, the macroscopic performance of a wearable sensor is equally governed by its geometric configuration. Traditional fabrication methods, such as mold casting, are limited to simple shapes and lack the precision required for microstructural modulation and personalized customization [88–90]. Consequently, Additive manufacturing (3D printing) has emerged as a transformative tool, bridging the gap between material synthesis and device

**Table 2**  
**Properties of hydrogels prepared by different fabrication methods**

Hydrogel system	Fracture strain	Tensile strength	Toughness/ fracture-related properties	Electrical conductivity	Self-healing	Ref.
Multifunctional conductive gel system	~2000%	170–180 kPa	~1.8 MJ/m <sup>3</sup>	3.8 S/m	Toughness recovery ~90% (8 h)	[68]
Polyvinyl alcohol hybrid hydrogel	150–1500%	0.1–2 MPa	Fracture work 0.1–7 MJ m <sup>-3</sup> ; tearing energy 1000–4000 J m <sup>-2</sup>	–	~30% (1 h, MPTC); ~99%(DMAEA-Q)	[69]
Conductive hybrid hydrogel	580%	1.1 MPa	Withstands 500 cycles at 50% strain	–	–	[73]
Carbon dots hydrogel	~200–300%	156 MPa	Toughness 225.2 MJ/m <sup>3</sup>	Up to 1.2 S/m	–	[74]
Cellulose nanofibers-based hydrogels	Up to 926%	36.82 kPa	Toughness: from 43 to 228.83 kJ/m <sup>3</sup>	–	Healing efficiency: 46.8% (1 h); 82.5% (12 h)	[77]
Quaternized chitosan-based hydrogel	~2113%	463 kPa	Toughness up to 6.03 MJ/m <sup>3</sup> ; 2.54 kJ/m <sup>3</sup> energy dissipation at 50% strain	0.77 S/m	~91% to 96% (3 h)	[78]
Polyaniline@ligno sul-fonate/CNT hydrogel	~659%	~200 kPa	~691 kJ/m <sup>3</sup>	0.05 S/m	–	[80]
Carboxymethyl chitosan hydrogel	749%	0.247 MPa	Toughness: 0.959 MJ/m <sup>3</sup> ; withstands 500 cycles at 100% strain	4.29 S/m	–	[82]
Tunicate cellulose nanocrystals hydrogel	626.9%	152.4 kPa	Withstands 10 cycles at 400% strain	13.9 S/m	–	[83]
Ultrastretchable ionic hydrogel	>3000%	>180 kPa	Toughness: 890 kJ/m <sup>3</sup> ; withstands 1500% strain	Up to 4.34 S/m	–	[85]
Entangled network hydrogels	350%–400%	~3 MPa	Fracture energy 8340 J/m <sup>2</sup> ; withstands 1000 cycles at 200% strain	–	Nearly 100% reversible	[87]

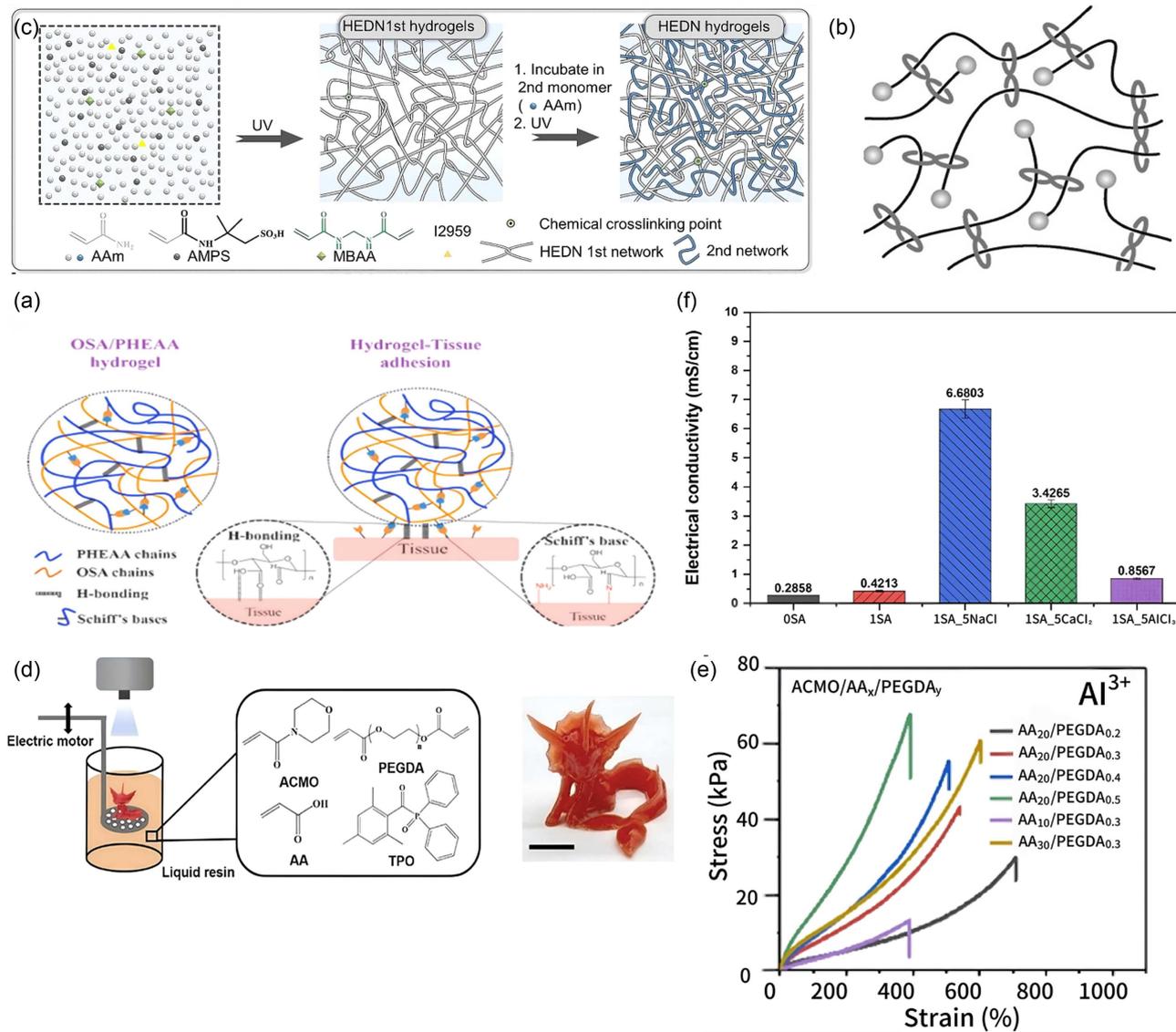
fabrication by enabling the programmable assembly of hydrogels into complex, hierarchical architectures.

Direct ink writing (DIW) is the most widely adopted technique for hydrogel sensors due to its material versatility. It relies on the extrusion of shear-thinning inks that flow under pressure and solidify upon deposition. This method is particularly potent for fabricating DN hydrogels with high viscosity. For instance, a shear-thinning ink was engineered by integrating alginate-functionalized graphene oxide into a PAM network. The resulting hydrogel could be printed into intricate lattices, maintaining structural fidelity while offering robust mechanical strength (Figure 6(d) and (e)) [89, 91–93]. Similarly, Antonia

et al. [94] utilized DIW to process DN particle organogels into customizable 3D constructs, demonstrating the feasibility of rapid prototyping for patient-specific skin patches. Beyond macroscopic shaping, aerosol jet printing (a high-resolution variant of DIW) enables the deposition of micro-scale conductive tracks, critical for miniaturized biosensing interfaces [88, 95].

For applications requiring micron-level resolution and complex internal porosity, light-based techniques such as DLP are superior. Unlike DIW, DLP forms structures by spatially controlled photopolymerization of liquid precursor resins. Tsupphayakorn-aek et al. [96] demonstrated the capability of SLA to fabricate electrolyte-conductive hydrogels into precise

**Figure 6**  
**Hybrid physicochemical hydrogels and 3D printing: (a) dual-network architecture of hydrogels, (b) structural configuration of a slide-ring hydrogel, (c) highly entangled double-network hydrogel incorporating sliding entanglement motifs, (d) efficient fabrication of three-dimensional hydrogel structures via 3D printing, (e) stress-strain curve of the 3D-printed hydrogel, and (f) the electrical conductivity of 3D-printed hydrogels**



geometries, overcoming the design constraints of conventional molding while maintaining excellent electrical conductivity (Figure 6(f)) [96]. The noncontact nature of DLP avoids shear stress, making it ideal for delicate network structures.

The frontier of hydrogel engineering lies in the convergence of advanced network topologies with digital fabrication. A prime example is the integration of “slide-ring” chemistry with DLP printing. Xiong et al. [92] synthesized a polymerizable pseudorotaxane precursor based on cyclodextrin and cholic acid. By formulating this into a photo-curable ink, they successfully printed complex, highly stretchable sensors via DLP. This approach synergistically combined the structural homogeneity of topological cross-linking with the geometric freedom of 3D printing, yielding sensors with high sensitivity and fatigue resistance that were previously unattainable [89, 91].

In essence, additive manufacturing transforms hydrogels from passive bulk materials into intelligent devices. It allows for

(i) personalization, matching the curvilinear topography of human skin; (ii) sensitivity enhancement, via printed micro-pyramids or porous lattices that amplify signal response; and (iii) functional integration, embedding interconnects and encapsulated electronics directly during the printing process. This trend signifies a paradigm shift toward fully integrated, customizable wearable electronics.

## 5. Classification of Hydrogel-Based Wearable Sensors

Hydrogel-based sensors have emerged as the premier interface for wearable electronics, bridging the gap between soft biological tissues and rigid data acquisition systems. While they can be categorized by material composition or structural design, a classification based on sensing mechanisms offers the most fundamental insight into their signal transduction pathways. This

mechanism-oriented perspective elucidates how external stimuli (strain, pressure, temperature, humidity) are converted into readable electrical signals (resistance, capacitance, potential), thereby providing the theoretical roadmap for optimizing sensitivity, linearity, and dynamic range [97]. Accordingly, this section systematically discusses hydrogel sensors based on their underlying transduction principles.

### 5.1. Resistive sensors

Resistive sensors, which function as chemiresistors or piezoresistors, represent the most widely investigated category due to their simple readout circuitry and effective signal response. The fundamental principle relies on the mechano-electrical transduction, where mechanical deformation modulates the sensor's electrical resistance [98]. This modulation arises from the synergistic coupling of two distinct mechanisms:

(i) Geometric Deformation: Governed by classical Ohm's law ( $R = \rho L/S$ ), tensile strain elongates the conductive path ( $L$ ) and reduces the cross-sectional area ( $S$ ), leading to a resistance increase.

(ii) Intrinsic Network Evolution: More critically for nanocomposite hydrogels, strain alters the internal connectivity of the percolation network. This includes the disconnection of conductive pathways, the variation of contact resistance between fillers, and the exponential change in tunneling resistance as inter-particle distances shift.

The gauge factor (GF)—the ratio of relative resistance changes to strain—serves as a key metric for sensitivity. Recent works have shown GF values exceeding 10 or even several tens, demonstrating improvements in sensor sensitivity (Figure 7(a)) [99]. The pursuit of high-performance strain sensors has shifted from relying on geometric changes (yielding low GF  $\sim 2$ ) to exploiting “Disconnect–Reconnect” and micro-cracking mechanisms for high sensitivity (GF  $> 10$ ) (Figure 7(b)) [99]. Under strain, the expanding hydrogel network disrupts the pathways of rigid conductive fillers. Two-dimensional fillers like MXenes/graphene achieve exceptional sensitivity through nanosheet sliding that drastically alters contact area and tunneling distance [99, 100]. In contrast, one-dimensional fillers such as AgNWs/CNTs maintain electrical percolation over larger strains due to their high aspect ratio, effectively balancing sensitivity with stretchability.

Recent advances leverage multi-scale structural engineering to optimize the sensitivity–stretchability–linearity trade-off. For high sensitivity, designs incorporating controlled crack propagation, such as an MXene/bacterial cellulose hydrogel, enable the detection of subtle strains (0.05%) with rapid response (130 ms) (Figure 7(c)) [101]. Conversely, wide-range stretchability is achieved through hierarchical networks, exemplified by a pure MXene-based hydrogel that maintains conductivity under extreme deformation via a dynamic percolation network. Distinct from tensile strain, piezoresistive pressure sensing utilizes pore compression in porous structures to increase conductive contacts and reduce resistance, as demonstrated in a lignocellulose/graphene composite (Figure 7(d)) [102].

Despite these sophisticated designs, intrinsic limitations persist due to the viscoelastic nature of hydrogels. Key challenges include significant hysteresis and signal drift from time-dependent polymer relaxation, nonlinear electrical output governed by the exponential nature of tunneling currents, and inadequate dynamic stability under high-frequency cyclic loading [103, 104]. Addressing these fundamental issues is critical for the development of reliable, high-performance wearable sensors.

### 5.2. Capacitive sensors

Unlike resistive sensors that rely on percolation networks, capacitive hydrogel sensors operate on the principle of charge storage and separation. They are particularly favored for their low power consumption, minimal hysteresis, and superior linearity. In the context of ionic hydrogels, sensing mechanisms typically fall into two distinct regimes:

(i) Dielectric Capacitive Sensing: Governed by the classic parallel-plate model ( $C = \epsilon_r \epsilon_0 A/d$ ), where external pressure compresses the hydrogel dielectric, reducing the distance ( $d$ ) and increasing capacitance. However, in soft ionic materials, the effective permittivity ( $\epsilon_r$ ) is not constant but strain-dependent due to ion concentration polarization.

(ii) Electric Double Layer (EDL) Sensing (Iontronics): This is the dominant mechanism for high-sensitivity tactile sensors. Here, the hydrogel acts as an ionic conductor rather than a simple dielectric. When an AC voltage is applied, mobile ions accumulate at the electrode/hydrogel interface, forming an EDL with nanometer-scale thickness ( $d_{EDL}$ ). The resulting interfacial capacitance is orders of magnitude higher than bulk capacitance ( $C_{EDL} \propto 1/d_{EDL}$ ), enabling the detection of ultra-low pressures and subtle physiological signals (Figure 7(e)) [105, 106].

To break the sensitivity limits of traditional elastomers, recent research focuses on three pillars: dielectric modulation, microstructural engineering, and interfacial iontronics.

(i) Dielectric Modulation for Strain Sensing: By tuning the polymer network, the dielectric properties can be customized. Kesava Rao et al. [107] synthesized a self-healing PDA-PAA-Cr<sup>3+</sup> hydrogel dielectric. The dynamic coordination bonds allowed for massive deformability (500% strain), yielding a GF of 160. This high sensitivity stems from the synergistic effect of geometric thinning and the strain-induced polarization of ionic clusters (Figure 7(f, g)) [107].

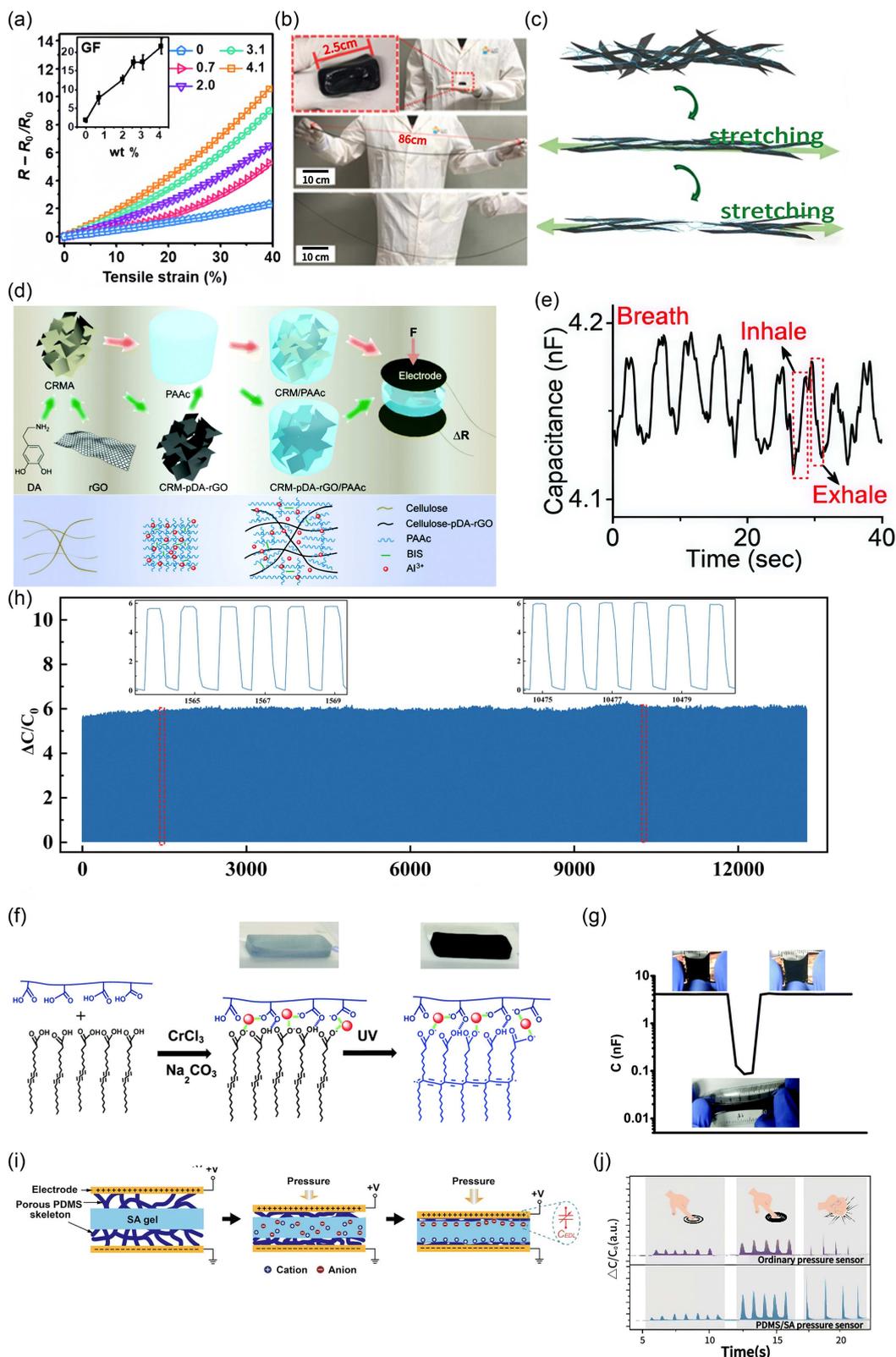
(ii) Microstructural Engineering for Pressure Sensing: Introducing micro-patterns increases the effective contact area and compressibility. Qiao et al. [108] engineered a capacitive sensor featuring an ionic hydrogel with a “lateral-bending” microstructure. Under pressure, the bending of micro-beams rapidly increased the effective dielectric constant and electrode contact area, achieving a high sensitivity of 2.79 kPa<sup>-1</sup> in the low-pressure regime ( $< 1$  kPa) with exceptional cycling stability ( $> 13,000$  cycles) (Figure 7(h)) [108].

(iii) Interfacial Design for Biomimetic Perception: Advanced interfaces can emulate biological functions. For wearable health monitoring and human–machine interaction (HMI), Huang et al. [109] developed a hybrid PDMS/alginate hydrogel sensor that emulates the skin's “Touch-Pain” perception. The design is specifically tailored for on-skin electronics: the PDMS matrix offers durability and flexibility for dynamic body surfaces, while the alginate hydrogel enables high-fidelity signal acquisition. By optimizing the electrode/hydrogel interface, they achieved a context-aware dual-mode response—linear for subtle physiological signals (e.g., pulse) and nonlinear for excessive mechanical stress. This allows a single wearable device to cover a 400 kPa dynamic range, from gentle touch to painful impact, mirroring the protective logic of biological nociceptors and enhancing situational awareness for wearables (Figure 7(i, j)) [109].

Notwithstanding their potential, capacitive hydrogel sensors face specific challenges: their dielectric constant is highly susceptible to water content, leading to baseline drift due to dehydration; their high impedance makes them prone to

Figure 7

(a) Electrical response of M-hydrogel to tensile strain. (b) Photographs demonstrating the stretchability of M-hydrogel. (c) Schematic illustrations of the sensing mechanism of the rBC/MXene sensor. (d) Fabrication process and piezoresistive sensor assembly schematics of the elastic composite lignocellulose/polyanion hydrogels. (e) Capacitive response when the subject did breathing actions. (f) Synthesis of the polydiacetylene (PDA)–polyacrylic acid (PAA)–Cr<sup>3+</sup> strain sensor. (g) An example of a capacitive strain sensing experiment utilizing the PDA–PAA–Cr<sup>3+</sup> hydrogel. The recorded capacitance decreases significantly upon stretching, reverting to its initial value upon strain release. (h) Variation of capacitance under a long-term cyclic loading mode. (i) The sensing mechanism of the hybrid porous PDMS/SA pressure sensor. (j) Comparison of the relative capacitance variations of the traditional pressure sensor and the hybrid porous PDMS/SA pressure sensor under gentle touch, heavy pressing with a finger, as well as striking with the palm



electromagnetic interference; and a fundamental trade-off exists between high sensitivity and a broad dynamic range, often resulting in signal saturation at high pressures. Future progress hinges on developing advanced encapsulation, alongside graded microstructures or hybrid dielectrics, to simultaneously achieve ultra-low detection limits and wide operational ranges.

### 5.3. Piezoelectric sensors

Unlike resistive and capacitive sensors that require external power sources, piezoelectric sensors are inherently self-powered, converting ambient mechanical energy directly into electrical signals. In the realm of hydrogels, this energy conversion manifests through two distinct physical mechanisms: classical piezoelectricity (based on bound charge polarization) and the emerging piezoionic effect (based on mobile ion migration) [110].

Traditional piezoelectricity arises from the displacement of charge centers in non-centrosymmetric domains. Since hydrogels themselves are typically amorphous and non-piezoelectric, the primary strategy involves embedding piezoelectric nanofillers (e.g., BaTiO<sub>3</sub> nanoparticles, Polyvinylidene Fluoride (PVDF) nanofibers) into the hydrogel matrix. When deformed, the embedded fillers generate a polarization potential. The conductive hydrogel matrix then acts as a compliant electrode, facilitating charge collection. The challenge lies in the mechanical mismatch between rigid ceramic fillers and the soft matrix, which often dampens stress transfer. Du et al. [111] emphasized the importance of tailoring polymer–filler interactions and achieving high  $\beta$ -phase content in PVDF for maximizing output in hydrogel-based devices (Figure 8(a)) [111]. Wang et al. [112] addressed this by developing a PVA/PVDF composite hydrogel where the interface was tailored for efficient stress transmission. This device not only sensed motion but also generated sufficient electric fields to accelerate diabetic wound healing via electrostimulation. Furthermore, Vinikoor et al. [113] introduced an injectable collagen hydrogel loaded with Poly-L-Lactic Acid (PLLA) nanofibers. Ultrasound activation of these piezoelectric fibers provided wireless stimulation for cartilage regeneration, showcasing the potential for minimally invasive bioelectronics.

A paradigm shift in soft sensors is the utilization of the piezoionic effect, which does not rely on crystalline asymmetry but on the differential mobility of ions. In a hydrated polyelectrolyte network, mechanical stress induces a pressure gradient or fluid flow. If the cations and anions have different mobilities (e.g., small mobile counter-ions vs large, tethered polymer chains), a net charge separation occurs, generating a transient voltage or current. This mechanism closely resembles the signal transduction in biological mechanoreceptors. Dobashi et al. [110] demonstrated that simple saline-doped hydrogels could produce ionic currents with time constants tunable from milliseconds to seconds. By adjusting the hydrogel thickness and cross-linking density, they successfully mimicked the firing patterns of both fast-adapting and slow-adapting mechanoreceptors (Figure 8(b)) [110]. This offers a route to “soft-material-only” sensors that are chemically identical to biological tissues.

The frontier of research lies in coupling these two effects. Xu et al. [113] designed a sandwich structure (PVDF/Nafion/PVDF) that integrates electronic polarization with ionic migration. The synergy between the high voltage output of PVDF and the high charge density of the ionic interface significantly enhanced the effective piezoelectric coefficient and pressure sensitivity.

Despite the promise of self-powered sensing, several bottlenecks persist:

(i) **Signal Deconvolution:** In hybrid systems, distinguishing between the fast electronic response (piezoelectric) and the slow ionic relaxation (piezoionic) is complex.

(ii) **Rigorous frequency-domain analysis and selective blocking layers** are required to decouple these signals. **Impedance Mismatch:** High-performance piezo-materials are typically high-impedance dielectrics, while hydrogels are ionic conductors. Optimizing the interfacial charge transfer efficiency without short-circuiting the piezoelectric domains remains a material engineering challenge.

(iii) **Long-Term Stability:** The piezoionic effect is fundamentally driven by solvent flow. Dehydration or ion leaching will drastically alter the sensor’s baseline and sensitivity. Future designs must incorporate ion-locking chemistries and robust encapsulation (as detailed in Section 3.5) to ensure stable operation in open-air environments.

### 5.4. Triboelectric sensors

Hydrogel-based triboelectric sensors (H-TENGs) operate by coupling contact electrification and electrostatic induction. When two materials with different positions in the triboelectric series come into contact and then separate, charge transfer occurs at the interface, generating equal and opposite surface charges. In the context of wearable electronics, hydrogel-based TENGs (H-TENGs) have emerged as a superior alternative to traditional metal-electrode TENGs. By utilizing hydrogels as transparent, stretchable ionic current collectors, H-TENGs resolve the long-standing mechanical mismatch between rigid electronics and soft biological tissues, enabling fully compliant self-powered sensing systems. In H-TENG configurations, the hydrogel plays a dual role. It serves both as the triboelectric layer, offering excellent flexibility, deformability, and tunable surface chemistry, and as an ionic conductor, facilitating fast charge transport and enhancing signal stability. This dual functionality allows self-powered sensing, mechanical adaptability, and biocompatibility, which are essential for wearable and biomedical applications [114, 115].

Recent advances in H-TENGs have been primarily focused on four main areas:

(1) **Material Innovation:** To improve conductivity and mechanical robustness, researchers have developed conductive or ionically doped hydrogels by incorporating salts, conductive polymers, or carbon-based nanomaterials. These modifications reduce internal resistance and improve output stability during repeated mechanical deformation [116, 117].

(2) **Structural Design:** Innovations in micro/nano-patterning, surface modification, and hydrophobic–hydrophilic regulation have been adopted to increase charge density and retention. These modifications help to enhance the overall performance of the triboelectric sensor by improving charge generation and retention [116, 118]. Moreover, multilayer, woven, and textile-integrated H-TENG architectures have been developed to improve the device’s conformability, enabling real-time monitoring of motion, tactile stimuli, and physiological signals [116, 118].

(3) **Self-healing and Environmental Robustness:** Self-healing properties and environmental durability have been significant areas of research. Jin et al. [115] demonstrated that hydrogel materials can address key technical bottlenecks in traditional

triboelectric sensors, particularly in terms of flexibility, biocompatibility, and environmental robustness. For example, Figure 8(c) [115] shows that if the hydrogel is cut into two pieces, it can self-heal within 10 min. This makes hydrogel-based sensors resistant to environmental degradation and more reliable in real-world applications.

(4) Multifunctional Integration: Several studies have focused on integrating triboelectric sensors into functional systems for real-time monitoring of various physiological and mechanical stimuli. For instance, Lin et al. [119] developed a stretchable and adhesive triboelectric sensor integrated into a textile substrate for musculoskeletal monitoring, highlighting its potential for rehabilitation and human-machine interfaces. The process of contact electrification and electrostatic induction is shown in Figure 8(d) [119], which explains the physical principles of TENG operations [120].

To propel H-TENGs from laboratory prototypes to commercial wearables, future efforts must converge on a “triple optimization” strategy [116]:

(1) Interfacial Engineering: Developing ultrathin, hydrophobic, yet highly charged dielectric coatings to maximize electrostatic induction while preventing hydrogel dehydration.

(2) Architecture Modularity: Designing modular, self-healable units that can be assembled into large-area sensor arrays (e.g., electronic skin).

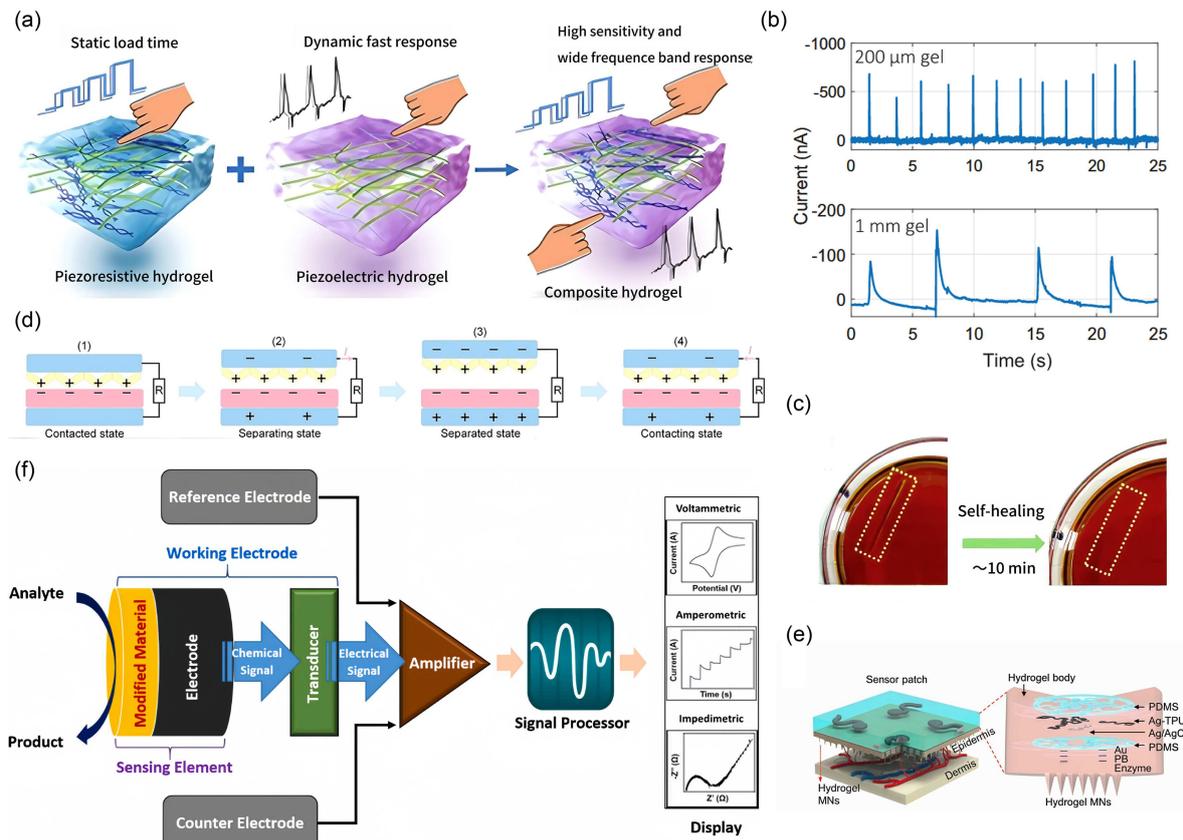
(3) Intelligent Signal Processing: Integrating machine learning (ML) algorithms to decouple the multimodal signals (e.g., pressure vs humidity) often convolved in TENG outputs. In summary, H-TENGs represent the convergence of iontronics and energy harvesting, offering a self-powered, skin-conformal solution that is indispensable for the next generation of sustainable wearable electronics [121].

### 5.5. Electrochemical sensors

Hydrogel-based electrochemical sensors leverage the intrinsic properties of hydrogels—such as their high water content, tunable network structure, and ionic mobility—to convert biochemical or ionic interactions into measurable electrical signals. In these sensors, the hydrogel typically acts as an interface or immobilization matrix for the electrode, facilitating analyte recognition and signal transduction. Significant advancements have been made in enhancing the sensitivity, flexibility, wearability, and miniaturization of these sensors. Hydrogel-based electrochemical sensors can be classified into three main categories based on their electrochemical transduction mechanisms: amperometric (current-type), potentiometric (voltage-type), and impedimetric (impedance-type) sensors. The classification is determined by how the hydrogel-electrode system responds: by generating a current at a fixed potential, modulating an equilibrium potential at zero current, or altering the impedance spectrum under small

Figure 8

(a) Enhancing strain-sensing properties of the conductive hydrogel by introducing PVDF-TrFE. (b) Piezoionic transient response under repeated step compressions onto a 200- $\mu\text{m}$ -thick (top) and a 1-mm-thick (bottom) pAAm gel swollen with 1.5 M NaCl, exhibiting decay times of  $\sim 50$  ms and  $\sim 5$  s, respectively, emulating rapid- and slow-adapting mechanoreceptors. (c) The hydrogel was able to heal itself within 10 min after being cut if the two cut parts were placed together. (d) Schematic illustration of the working mechanism of the MB-TENG in a contact–separation mode. (e) Schematics and images of the sensor patch applied to the skin. (f) Schematic diagram of an electrochemical sensor



alternating current (AC) excitation. This section will explore each of these sensor types in detail.

5.5.1. Amperometric sensors

Amperometric sensors measure the current generated by the oxidation or reduction of electroactive species at a constant potential. They are the gold standard for metabolic monitoring (e.g., glucose sensing). The primary challenge in wearable amperometry is maintaining the stability of biocatalysts (enzymes) on flexible substrates. Hydrogels provide a biocompatible 3D immobilization matrix that prevents enzyme leaching and denaturation.

Fu et al. [122] highlighted that hybrid hydrogels doped with conductive nanomaterials (e.g., AuNPs, CNTs) create “wired” enzyme systems, significantly enhancing electron transfer kinetics. This architecture not only amplifies the signal for trace analytes like H<sub>2</sub>O<sub>2</sub> but also lowers the overpotential, minimizing interference from other electroactive species. Furthermore, Zhang et al. [123] demonstrated a hydrogel-modified interface for detecting antibiotic-resistant bacteria, where the hydrated network facilitated rapid reagent diffusion, achieving a limit of detection superior to conventional 2D film sensors.

5.5.2. Potentiometric sensors

Potentiometric hydrogel sensors function by measuring the potential difference between a working electrode and a reference electrode under near-zero current conditions, in response to changes such as ion concentration, pH, or selective binding events. In these systems, the hydrogels may embed ion-selective membranes or responsive ionic groups, enabling selective potential shifts. For example, a study by Harun-Or Rashid et al. [4] detailed wearable sweat sensors using hydrogel-integrated electrodes for pH and ionic detection, showing that the hydrogel structure enhanced conformal skin contact and improved signal drift performance. Figure 8(e) [4] shows schematics and images of the sensor patch applied to the skin. Moreover, Bauer et al. [124] reviewed how deposition of hydrogels and polymer membranes influences potentiometric sensor stability and selectivity. These studies elucidate hydrogels’ multifunctional roles in stabilizing ionic environments, enhancing selective binding, and maintaining seamless compatibility with soft substrates—consistent with the potentiometric mechanism of potential modulation without major current flow.

5.5.3. Impedimetric sensors

Impedimetric hydrogel sensors measure changes in the impedance at the electrode–hydrogel interface, often under AC excitation. Changes may result from analyte binding, network swelling/shrinkage, or ion mobility shifts within the hydrogel. For instance, Singh et al. [125] presented a review of recent trends in electrochemical sensing, emphasizing impedance techniques and

highlighting hydrogel-based platforms for cell-substrate impedance monitoring and wearable interfaces. Figure 8(f) [125] is a schematic diagram of the electrochemical sensor structure, clearly showing the core components of the sensor and the signal processing flow, and includes modules related to impedance technology. Additionally, de Oliveira et al. [126] used bbv, a hydrogel-based electrochemical cell to detect trace pesticides in matrices, using impedance changes induced by hydrogel–analyte interactions. These studies reveal the core working mechanism of such sensors: the hydrogel modulates the impedance pathway, altering charge transfer resistance or double-layer capacitance when analytes or deformation occur.

In summary, hydrogel-based electrochemical sensors uniquely leverage the synergy between biological compatibility and versatile electrochemical functionality: amperometric modes for monitoring metabolic flux, potentiometric modes for analyzing electrolyte balance, and impedimetric modes for label-free detection of proteins and pathogens. Future research is decisively pivoting toward multiplexed sensor arrays—integrated single patches with distinct hydrogel zones capable of simultaneous monitoring of metabolites, electrolytes, and physical signals to provide a holistic view of human health.

5.6. Summary

This section has systematically classified hydrogel-based wearable sensors into five distinct categories based on their signal transduction mechanisms. This classification is fundamental to understanding the structure–function relationships that dictate sensor performance. A comparative analysis reveals distinct operational niches for each mechanism: physical transducers (resistive and capacitive) excel in motion and tactile sensing with simple circuitry but require external power, self-powered energy harvesters (piezoelectric and triboelectric) address the energy bottleneck but face challenges in high impedance and signal conditioning, and biochemical interfaces (electrochemical) provide unique molecular specificity for biofluid analysis.

Despite their diversity, all hydrogel sensors face a common set of “grand challenges” rooted in their material nature: environmental stability, viscoelastic hysteresis, and signal crosstalk. Looking forward, the field is evolving from single-mechanism optimization toward system-level integration. Future research should focus on developing “Lab-on-Skin” platforms for multimodal fusion, integrating ML for intelligent signal processing, and creating closed-loop systems where sensing triggers active feedback. By mastering these mechanisms and addressing interfacial stability, hydrogel electronics are poised to transition from laboratory prototypes to robust, clinical-grade wearable systems. The key characteristics of the different sensors are summarized in Table 3.

Table 3  
Classification of hydrogel sensors with key characteristics

Sensor type	Sensing mechanism	Advantages	Disadvantages	Application scenarios
Resistive Sensors	Mechanical deformation induces changes in electrical resistance through geometric deformation and the evolution of conductive percolation networks	Simple readout circuitry; high sensitivity (high gauge factor); wide strain detection range	Pronounced hysteresis; signal drift due to viscoelasticity; nonlinear response; limited high-frequency stability	Strain sensing, motion monitoring, pressure sensing, human–machine interfaces

(Continued)

**Table 3**  
(Continued)

Sensor type	Sensing mechanism	Advantages	Disadvantages	Application scenarios
Capacitive Sensors	External stimuli modulate capacitance via dielectric thickness variation or electric double layer (EDL) formation at the electrode–hydrogel interface	Low power consumption; minimal hysteresis; high linearity; ultra-low pressure detection	Sensitive to dehydration; prone to electromagnetic interference; trade-off between sensitivity and dynamic range	Tactile sensing, pressure mapping, physiological signal monitoring, electronic skin
Piezoelectric Sensors	Mechanical stress generates electrical signals through embedded piezoelectric fillers or ion migration (piezoionic effect)	Self-powered operation; fast response; biomimetic signal transduction	Signal deconvolution complexity; impedance mismatch; performance instability due to dehydration or ion leakage	Self-powered motion sensing, biomedical stimulation, and mechanoreceptor-inspired sensors
Triboelectric Sensors (H-TENGs)	Contact electrification and electrostatic induction convert mechanical motion into electrical output using hydrogel-based ionic electrodes	Self-powered; high flexibility and stretchability; good biocompatibility; self-healing capability	Signal affected by humidity and environmental conditions; complex signal processing; charge decay over time	Motion sensing, tactile sensing, wearable energy harvesting, rehabilitation monitoring
Electrochemical Sensors – Amperometric Sensors	Oxidation or reduction of electroactive species generates a current at a fixed potential	High sensitivity; suitable for metabolic monitoring; efficient enzyme immobilization	Enzyme stability issues; potential biofouling; need for calibration	Glucose sensing, metabolite detection, biochemical monitoring
Electrochemical Sensors – Potentiometric Sensors	Changes in ion concentration or selective binding modulate the equilibrium potential under near-zero current	Low power consumption; good selectivity; stable baseline potential	Susceptible to reference electrode drift; limited multiplexing capability	Sweat analysis, pH sensing, electrolyte monitoring
Electrochemical Sensors – Impedimetric Sensors	Analyte interaction or hydrogel deformation alters interfacial impedance under AC excitation	Label-free detection; high sensitivity to interfacial changes; suitable for miniaturization	Complex signal interpretation; sensitive to environmental noise	Biosensing, pathogen detection, wearable health monitoring

## 6. Applications of Wearable Sensors Based on Hydrogels

The transition from rigid silicon-based electronics to soft, bio-integrated systems represents a paradigm shift in modern healthcare and robotics. In this landscape, conductive hydrogels have emerged not merely as “promising materials” but as the ultimate interfacial bridge between abiotic devices and biological tissues. Unlike conventional elastomers (e.g., PDMS, PU) that suffer from mechanical and impedance mismatches with the skin, hydrogels offer intrinsic tissue-like compliance, high water content, and ionic conductivity. These biomimetic attributes enable

conformal skin contact, minimized motion artifacts, and seamless ion–electron signal transduction, which are critical for high-fidelity sensing [127–129].

While pure hydrogels often lack the electrical robustness required for advanced electronics, the convergence of polymer chemistry and nanoengineering has overcome these hurdles. By integrating functional nanomaterials—such as MXenes, graphene, CNTs, and conductive polymers—researchers have engineered hybrid hydrogel composites that combine toughness with metallic-level conductivity [130, 27, 131, 132]. This material evolution has propelled hydrogel sensors beyond simple strain gauges into the realm of multimodal intelligence, capable of simultaneously

tracking physiological signals, biochemical markers, and environmental stimuli [133–135]. Despite persisting challenges regarding dehydration and long-term stability, the trajectory of hydrogel electronics is undeniably moving toward clinical-grade diagnostics and closed-loop therapeutics.

Following a systematic exploration of hydrogel sensors based on various sensing mechanisms, this chapter will focus on their extensive practical applications. To clearly and systematically demonstrate how hydrogel sensors span multiple critical fields from health monitoring to intelligent machines, we have categorized the application domains into four main directions based on their core functions, interaction targets, and technical emphases: (1) wearable physiological monitoring; (2) environmental and chemical sensing; (3) soft robotics, human–machine interfaces, and artificial skin; and (4) implantable and in vivo biomedical devices. This classification framework aims to comprehensively outline the diverse application landscape of hydrogel sensors, ranging from in vitro to in vivo, from physical to chemical signals, and from sensing to interaction.

### 6.1. Wearable physiological monitoring

The cornerstone of personalized healthcare lies in the continuous, noninvasive acquisition of physiological data. Conductive hydrogels have revolutionized this field by serving as soft, ionic–electronic interfaces that match the mechanical impedance of human skin. Unlike rigid metal electrodes that suffer from high contact impedance and motion artifacts, hydrogels ensure conformal contact and maintain a stable electrode–skin interface, which is critical for clinical-grade monitoring of both bio-potentials (ECG, EMG, EEG) and mechano-acoustic signals (pulse, respiration) [127–129].

Conductive and functionalized hydrogels offer a soft, hydrated, and mechanically compliant interface, closely matching the mechanical properties of skin and soft tissues. This enables low-impedance, conformal electrical contact, making them ideal for long-term physiological monitoring. In recent years, hydrogel electrodes and hydrogel-based strain/pressure sensors have enhanced the fidelity and comfort of on-body monitoring devices for electrocardiography (ECG), electroencephalography (EEG), electromyography (EMG), pulse, and respiration by addressing the trade-offs between conductivity, stretchability, and adhesion.

For example, MXene-reinforced organohydrogels have demonstrated highly conductive percolating networks of two-dimensional nanosheets embedded in resilient polymer matrices. These hydrogels enable simultaneous detection of low-amplitude pulse signals and large joint movements, while also providing antifreeze behavior and self-adhesion to the skin [127]. Gong et al. [128] developed an MXene hydrogel constrained by nanochannels, featuring neuron-like conductive pathways. This hydrogel showed high sensitivity, excellent mechanical properties, and reliable strain/pressure sensing capabilities for wearable devices, capable of directly detecting human motion and pulse signals. Figure 9(a) and (b) [128] shows its quick response time in detecting swallowing movements and pulse signals, demonstrating its high sensitivity and stability in monitoring weak physiological signals.

Semidry, double-layer hydrogel electrodes, combining a conductive layer and an adhesive substrate, have been shown to record EEG signals comparable to wet electrodes for extended periods ( $\approx 12$  h). These electrodes address motion artifacts and

long-duration stability issues in ambulatory neurophysiological monitoring [128]. Moreover, Zheng et al. [27] developed a low-cost hydrogel electrode with high electrical conductivity, mechanical stretchability, and biocompatibility, enabling multifunctional sensing of ECG and EMG signals for long-term monitoring. Figure 9(c) [27] shows the ECG waveforms under different movement states (resting, walking, swinging arms, squatting), demonstrating that the hydrogel electrodes can still function stably in dynamic environments.

Research combining PEDOT:PSS with photocross-linked or PVA hydrogel matrices has resulted in stretchable, low-impedance electrodes that improve ECG/EEG signal quality and offer scalable fabrication for wearable sensors [136, 137]. Additionally, injectable and self-cross-linking hydrogel electrodes with long-term stability have been developed, addressing the challenge of minimally invasive placement and stable recording for on-tissue sensing applications. Hsieh et al. [138] reported an injectable self-cross-linking hydrogel electrode with enhanced long-term EEG stability and biocompatible placement. Figure 9(d) shows the electrical impedance curves of the on-skin skin-electrode interface over 5 consecutive days. Three pre-formed AIRTodes were applied to the subject's forearm, and impedance measurements were performed once daily. The impedance values were recorded at 500, 100, 10, and 5 Hz. Figure 9(e) shows the impedance stability of AIRTode on the skin over five consecutive days and its long-term wearing stability without skin irritation.

Current advancements are shifting from simple signal acquisition to integrated diagnostic systems. Recent reviews emphasize the need for hydrogels that are not only conductive but also environmentally robust (anti-drying via glycerol/ionic liquids) and intelligently integrated with wireless transmission modules for real-time health analytics [117, 129, 139]. This transition marks the evolution of hydrogel sensors from benchtop prototypes to field-ready, clinical-grade wearable platforms.

### 6.2. Environmental and chemical sensing

Beyond physical signal monitoring, the next frontier in wearable technology is the noninvasive analysis of biochemical markers in biofluids (sweat, interstitial fluid). Hydrogels are uniquely positioned as the ideal “Bio-Fluidic Interface” for these applications, serving a dual purpose: (1) acting as a super-absorbent pump to efficiently harvest biofluids via capillary force and (2) providing a biocompatible reservoir that immobilizes bioreceptors (enzymes, antibodies) while preserving their catalytic activity.

A critical bottleneck in sweat sensing is the reliable collection of fluids, especially during sedentary states with low perspiration rates. Granular and microstructured hydrogels have emerged as a solution. Lorestani et al. [136] engineered a granular hydrogel-microfluidic platform that leverages the high interstitial permeability of packed microgels. This system enables rapid sweat uptake and efficient transport to the sensor surface, facilitating real-time monitoring of lactate dynamics even during low-intensity activities (Figure 9(f)) [136].

By integrating conductive polymers or ion-selective membranes, hydrogels transform into highly sensitive electrochemical probes. Li et al. [140] developed a PANI-PA-PVA hydrogel sensor where the protonation state of the PANI network responds rapidly to pH changes. This device provided ultra-sensitive, real-time mapping of skin pH, which is a vital indicator of skin

barrier function and metabolic status (Figure 9(g)) [140]. To achieve autonomous operation, recent innovations have coupled hydrogel biochemical sensors with self-powered modules. For instance, hydrogel-based biofuel cells (BFCs) can harvest energy from sweat lactate to power the sensor, or triboelectric nanogenerators (TENGs) can serve as active power sources, creating a self-sustained sensing system [116, 131].

Despite these strides, the path to commercialization faces hurdles: (1) Calibration: Variations in sweat rate and evaporation can alter analyte concentration, necessitating integrated flow-rate sensors for correction. (2) Bio-stability: Preventing the leaching of enzymes and maintaining their activity over weeks remains a challenge. (3) Anti-fouling: While hydrogels are inherently resistant to protein adsorption due to their hydration layer, further surface engineering (e.g., zwitterionic coatings) is needed to ensure long-term reliability in complex biofluids [131, 136, 141].

### 6.3. Soft robotics, human–machine interfaces, and artificial skin

Hydrogel sensors are evolving into key components for HMI and artificial skin (e-skin), endowing machines with human-like somatosensory perception. Their mechanical compliance, adhesion, and adaptable electrical properties allow for accurate strain, pressure, and shear mapping on curved, deforming surfaces. Multi-element hydrogel sensor arrays have been developed to distinguish normal and shear forces, enabling robots to perform “dexterous manipulation” of fragile objects while maintaining conformal contact.

The integration of hydrogel sensors with ML algorithms has ushered in a new era of “Smart HMI.” Hydrogel strain sensors embedded in smart gloves capture the subtle kinematic features of finger movements for data-driven control. For example, Luo et al. [139] developed a transparent, stretchable H-TENG based on MXene/PVA hydrogel. By feeding the triboelectric signals into a neural network, they achieved real-time, high-accuracy gesture recognition for the teleoperation of a robotic hand (Figure 9 (h–k)) [139]. Beyond physical touch, Zhou et al. [142] demonstrated a deep-learning-assisted system capable of noncontact gesture recognition via electrostatic coupling, allowing for intuitive control of virtual interfaces (Figure 9(l)) [143].

In soft robotics, the focus is shifting toward closed-loop control. MXene- or carbon-nanomaterial-reinforced hydrogel tactile layers applied to soft grippers provide real-time feedback on slip detection and contact pressure. When integrated with flexible readout electronics and ML classifiers, these systems enable the robot to dynamically adjust its grip strength based on sensory input, enhancing the durability and precision of robotic operations [117]. This synergy of material innovation and system engineering is unlocking applications in metaverse interactions, prosthetic feedback, and autonomous robotics.

### 6.4. Implantable and in vivo biomedical devices

The ultimate frontier for conductive hydrogels lies in implantable bioelectronics, where the material must bridge the stark mechanical and biological divide between rigid circuitry and soft living tissue. Traditional metal or silicon probes often trigger chronic inflammatory responses and glial scarring, leading to signal degradation over time. Conductive hydrogels, with their tissue-mimetic modulus and high water content, offer a transformative solution by acting as “stealthy” neural interfaces. When

employed as coatings on electrodes (e.g., PEDOT:PSS-modified probes), hydrogels significantly increase volumetric capacitance and reduce interfacial impedance. This enhancement improves the charge injection capacity for stimulation and the signal-to-noise ratio for recording, enabling stable, long-term monitoring of cardiac or cerebral activity with minimal foreign body response [133, 134, 138]. For instance, Li et al. [135] highlighted that PEDOT:PSS-based interfaces on flexible ECoG grids can maintain stable impedance even under severe mechanical distortions (twisting, folding), ensuring reliable brain–computer interfacing (Figure 9(m) and (n)) [135].

Beyond surface coatings, the development of injectable and self-healing hydrogels is revolutionizing minimally invasive surgery. These materials can be delivered via syringe into deep tissues or complex cavities, where they spontaneously cross-link to form conformal conductive pathways. Hsieh et al. [138] demonstrated this concept with “AIRTrode,” an injectable PEDOT:PSS-based hydrogel. While primarily demonstrated for scalp EEG, its shear-thinning and self-adhesive nature exemplifies the potential for creating robust bio-interfaces in hard-to-access anatomical sites without invasive implantation surgery. Furthermore, hydrogels are enabling the next generation of closed-loop theranostics—systems that simultaneously sense pathology and deliver therapy. A prime example is the multifunctional hydrogel device developed by Qu et al. [143], which integrates real-time seizure detection with electrically triggered antiepileptic drug release (Figure 9(o)) [133, 139, 143]. This “detect-and-treat” capability in rodent models underscores the potential of hydrogels to function not just as sensors, but as active, intelligent guardians of health. Despite these breakthroughs, the translation to clinical practice requires rigorous validation regarding long-term biodegradability, sterilization compatibility, and the stability of bio-interfaces in the complex in vivo environment.

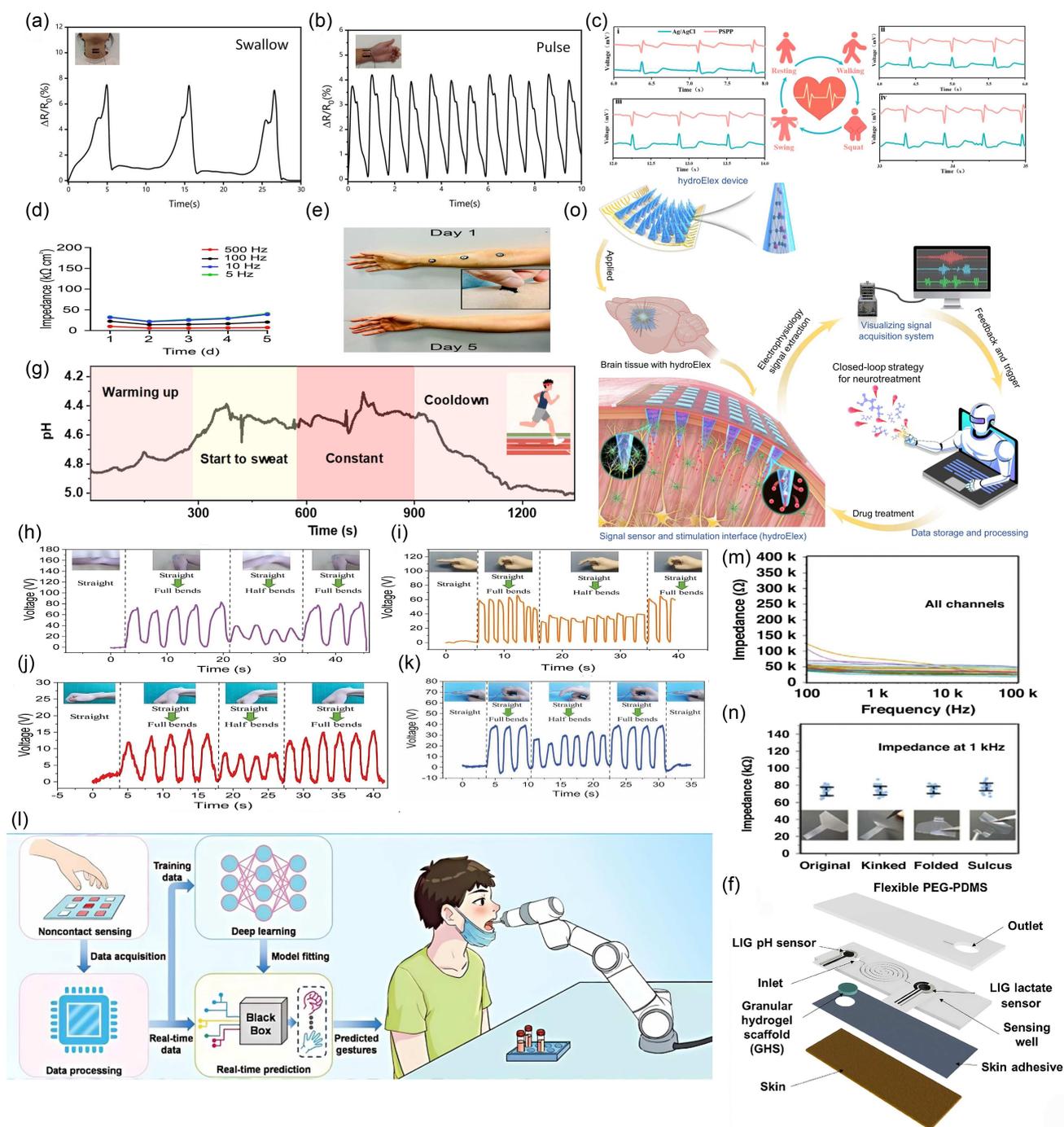
### 6.5. Summary

This chapter has provided a systematic overview of the burgeoning field of hydrogel-based wearable sensors, illustrating a trajectory from basic material synthesis to sophisticated system integration. By leveraging the synergistic properties of conductive fillers (MXenes, CNTs, PEDOT:PSS) and hydrated polymer networks, researchers have successfully engineered sensors that resolve the long-standing conflict between electrical performance and mechanical compliance. In physiological monitoring, we have witnessed a shift toward clinical-grade bio-potential recording and artifact-free motion tracking; in chemical sensing, hydrogels have evolved into “microfluidic labs” for real-time metabolite analysis; and in the realm of robotics and implants, they are endowing machines with somatic perception and enabling seamless neural interfaces.

However, the path toward widespread commercialization is not without hurdles. Critical challenges remain in ensuring environmental stability (preventing dehydration/freezing), minimizing viscoelastic hysteresis for high-frequency sensing, and achieving standardized, scalable manufacturing. Future research must pivot from optimizing individual metrics to holistic system-level engineering—integrating hydrogel sensors with wireless communication, energy harvesting, and ML algorithms. As these material and engineering challenges are addressed, conductive hydrogels are poised to become the foundational building blocks of the next generation of “Invisible Electronics,” seamlessly merging the digital world with the biological self.

Figure 9

Resistance changes of the PcAT MNC sensor during monitoring different physiological signals, including (a) swallow and (b) pulse, and (c) ECG waveforms under resting, walking, arm swinging, and squatting states: (i) ECG waveform in the resting state, (ii) ECG waveform in the walking state, (iii) ECG waveform in the arm swinging state, and (iv) ECG waveform in the squatting state. (d) The electrical impedance curves of the on-skin skin-electrode interface over 5 consecutive days. Three pre-formed AIRTodes were applied to the subject's forearm, and impedance measurements were performed once daily. The impedance values were recorded at 500, 100, 10, and 5 Hz. (e) The placement of the electrodes on the forearm on day 1 (top), the process of removing the electrodes on day 5 (center), and the appearance of the forearm after the removal of the electrodes on day 5 (bottom). (f) Exploded view of the patch that integrates electrochemical lactate and pH sensors with a microfluidic platform based on hydrophilic PEG-PDMS, as well as the Granular hydrogel scaffold (GHS) for sweat collection. (g) Real-time monitoring of sweat pH during physical activity, confirming stable, responsive, and noninvasive sensing performance under dynamic conditions. Voltage signals of the MH-TENG in response to continuous bends of (h) elbow, (i) finger, and (j) wrist. (k) Voltage signals of the MH-TENG in response to finger bends. (l) Working flow of the deep-learning-assisted noncontact gesture recognition system for controlling a robot to collect throat swabs, (m) Electrochemical impedance spectroscopy (EIS) of the 30-channel ECoG grid. (n) Mechanical stability test of the ECoG grid, showing stable EIS values across 30 channels in four different states: original, kinked, folded, and sulcus. (o) Schematic diagram of a closed-loop bioelectronic system for adaptive antiepileptic treatment



## 7. Conclusion and Perspectives

In this review, we have provided a holistic roadmap of the recent advancements in high-performance conductive hydrogels for wearable sensors, dissecting the strategies for tuning key properties and exploring the diverse sensing modalities and advanced fabrication techniques that transform these soft materials into functional devices. Collectively, this comprehensive analysis underscores the unique value of hydrogels not merely as substitutes for traditional conductors but as the ideal “bio-abiotic interface” capable of bridging the mechanical and biological gap between living tissues and rigid instrumentation.

However, the path toward widespread commercialization is obstructed by critical bottlenecks that demand interdisciplinary solutions. At the material level, intrinsic trade-offs persist, most notably the “conductivity-toughness” paradox and the conflict between high sensitivity and broad linear range. At the device level, environmental fortitude remains a primary concern; challenges such as dehydration, freezing, signal drift due to ion migration, and biofouling significantly hamper long-term reliability in real-world scenarios. Moreover, the current focus on performance often overlooks sustainability, necessitating urgent research into biodegradable and eco-friendly hydrogels. Finally, at the system level, a substantial “Lab-to-Fab” gap exists. The complexity of integrating heterogeneous components (e.g., combining rigid chips with soft gels) and the lack of scalable, low-cost manufacturing processes hinder the transition from benchtop prototypes to industrial-grade products.

Looking forward, the evolution of hydrogel-based wearable sensors will be defined by a shift from “single-function components” to “intelligent, integrated systems.” Future research priorities should focus on three strategic pillars:

- 1) All-in-one multimodal perception: Developing fusion sensors that decouple and simultaneously detect physical (strain, pressure, temperature) and biochemical (glucose, lactate, cortisol) signals within a single hydrogel matrix. This will enable a comprehensive “digital twin” of human physiology.
- 2) AI-enabled autonomy: Integrating hydrogel sensors with energy harvesting modules (e.g., TENGs, BFCs) and edge-computing algorithms. This moves beyond passive data collection to achieve self-powered, closed-loop systems capable of real-time signal interpretation and therapeutic feedback without external tethers.
- 3) Advanced bio-fabrication: Innovating manufacturing processes, such as 4D printing and heterogeneous integration, to create devices with spatially resolved functionality and dynamic deformability. Through these concerted efforts, conductive hydrogels are poised to transcend their current limitations, advancing toward application-oriented systems that require standardized testing protocols, improved encapsulation strategies, and reliable integration with rigid electronic components.

Through interdisciplinary collaboration and innovation, future development of hydrogel-based wearable sensors will likely depend not on further increasing single performance metrics but on optimizing multi-parameter trade-offs among sensitivity, durability, environmental tolerance, and manufacturability. Hydrogel sensors are expected to ultimately transform from a promising experimental platform into a technological driving the development of personalized healthcare, soft robotics, and intelligent human-machine interfaces.

## Materials and Methods

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## Ethical Statement

This study does not contain any studies with human or animal subjects performed by any of the authors.

## Conflicts of Interest

The authors declare that they have no conflicts of interest to this work.

## Data Availability Statement

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

## Author Contribution Statement

**Jianrui Zhao:** Methodology, Validation, Investigation, Resources, Data curation, Writing – original draft, Writing – review & editing, Visualization. **Qingwang Liu:** Investigation, Resources, Data curation, Writing – original draft, Writing – review & editing. **Xinzu Yue:** Conceptualization, Methodology, Software, Resources. **Weibao Wang:** Resources. **Weiwei Zhang:** Resources. **Xinning Liu:** Resources. **Guoshen Yang:** Conceptualization, Methodology, Validation, Formal analysis, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition.

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