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REVIEW

Multifunctional Hydrogel Interfaces: Reshaping the Future of Flexible Electronics

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ABSTRACT

Flexible electronics is undergoing a transition from single-function devices to intelligent systems capable of multimodal perception and closed-loop operation. Multifunctional hydrogels have emerged as a core platform for next-generation electronics, owing to their structural tailorability, biomimetic compatibility, dynamic responsiveness, and exceptional interfacial properties. This review outlines a cross-scale design pathway of hydrogel electronics spanning molecular strategies and microstructural architectures to macroscopic functionalities (mechano-electro-thermo-chemical responses) and system-level integration. We critically survey recent advancements in hydrogel-based applications, including wearable health monitoring, electronic skin, soft robotics, and self-powered devices, highlighting their unique advantages in high-fidelity signal acquisition, autonomous energy management, and long-term stability under complex conditions. Furthermore, we explore how AI-driven inverse design, digital twins, and in situ characterization are accelerating the shift from empirical to model-driven development of hydrogel electronics. A performance evaluation framework based on the "energy–signal coupling coefficient" is introduced, combining with green design principles promoting circular sustainability. Finally, we outline future challenges and opportunities to achieve extreme environmental adaptability and promote standardization and scalable manufacturing. Interdisciplinary integration and AI-assisted multimodal data analytics will ultimately advance hydrogel electronics from functional devices to fully intelligent bio-integrated systems.

1 | Introduction

Flexible electronics are rapidly evolving from single-function devices into integrated systems capable of multimodal perception, intelligent closed-loop control, and autonomous operation [1]. This paradigm shift necessitates materials that exhibit not only exceptional flexibility and conductivity but also the ability to synergistically interface with biological tissues, harvest energy,

Boya Song and Jing Zhang contributed equally to this work.

transduce multimodal signals, and enable adaptive feedback, serving as dynamic bridges between the physical and digital worlds [2]. Hydrogels have thus emerged as a foundational platform for next-generation bio-integrated electronics due to their high hydration, tissue-like softness, biocompatibility, tunable physicochemical properties, and highly designable structures. However, with the development of distributed sensing, in vivo longitudinal monitoring, and personalized interventions become standard practice, the benchmark for hydrogel sensors shifts from the peak sensitivity and response time parameters to sustained performance under real-world constraints including sweat, temperature–humidity swings, multi-axial strain, wireless telemetry, and limited power [3, 4]. Consequently, the co-design of energy harvesting and management with signal fidelity has emerged as the critical inflection point for translating multifunctional hydrogel-based electronics into clinical practice.

In recent years, significant progress has been achieved in molecular design, functional compositing, and device integration of hydrogel interfaces [5–7]. At the molecular level, rational deployment of covalent, dynamic covalent, and non-covalent bonding has enabled concurrent enhancement of mechanical robustness and intrinsic self-healing behavior [8–10]. Advancements in multi-network architectures have markedly improved strength and fatigue resistance, while the integration of conductive nanofillers such as conducting polymers, carbon nanomaterials, and MXenes, has significantly extended electrical functionality and application horizons [11, 12]. From a fabrication standpoint, advanced manufacturing techniques, including direct-ink-writing, lithographic micro-/nano-patterning, laser micromachining, and kirigami/serpentine layout engineering, have enabled precise cross-scale structural control spanning from 2D to 3D and from homogeneous to heterogeneous configurations, thereby furnishing a foundational platform for sophisticated functional integration [13–16]. Figure 1 depicts the evolution of multifunctional hydrogel interfaces in flexible electronics, showcasing their progression from initial material development, through the functional augmentation via multimodal sensing and energy harvesting, and ultimately to system-level intelligence integrating AI-driven feedback and biomedical applications.

Currently, most studies on hydrogel-based flexible electronics remain confined to individual functionalities or parallel categorization, lacking a comprehensive analysis of multimodal synergy and system-level integration. Particularly, the rapid evolution of artificial intelligence and Internet of Things (IoT) technologies has significantly heightened the demand for the next-generation flexible electronics [39]. Consequently, hydrogel interfaces have evolved from passive, single-function materials into dynamic, adaptive platforms capable of sensing, energy management, and decision-making. This paradigm shift underscores the urgent need for a comprehensive review that not only summarizes recent advancements but also redefines the fundamental requirements for hydrogel materials in intelligent systems. Emerging applications such as closed-loop biointerfaces, autonomous soft robotics, and environmentally responsive sensors, impose new criteria for supporting seamless integration with AI algorithms, facilitating real-time data processing, and sustaining energy-efficient operation [40–42]. Furthermore, the convergence of digital twins, machine learning-aided material design, and sustainable life-cycle management calls for hydrogel systems that are both functionally sophisticated and ecologically feasibility [43–45]. A systematic examination of these aspects is therefore imperative to bridge the gap between material-level innovation and system-level intelligence, steering the future development of hydrogel-based technologies toward truly adaptive, interconnected, and sustainable intelligent systems.

Unlike conventional reviews that focus primarily on material properties or device-level demonstrations, this article redefines hydrogel interfaces as cross-scale, multi-physical systems encompassing synthesis, processing, performance regulation, and multifunctional integration. We propose a comprehensive "material–device–system" framework centered on dual energy–signal coupling, which transforms hydrogels from passive substrates into active platforms capable of concurrent sensing, energy harvesting, communication, and decision-making. We systematically summarize recent progress in preparation strategies, advanced processing and micro/nanofabrication techniques, evolving performance requirements at biotic–abiotic interfaces, and standardized workflows for device assembly and validation. Representative multifunctional hydrogel devices are highlighted to illustrate how fundamental design principles can be translated into practical high-performance systems. In addition, we discuss paradigm-shifting approaches including AI-guided inverse design, digital twin modeling, and in situ characterization that are enabling the transition from empirical optimization toward predictive and system-level intelligence. This review expands the conceptual and technological scope of hydrogel-based flexible electronics and provides a roadmap for their evolution into intelligent, autonomous, and environmentally adaptive systems for next-generation applications.

2 | Hydrogel material fundamentals and design principles

Multifunctional hydrogel interfaces have rapidly emerged as a transformative class of materials in the development of flexible and implantable bioelectronic devices, owing to their unique capability to seamlessly integrate with biological tissues [46]. As illustrated in Figure 2, hydrogel-based devices have been demonstrated across a broad spectrum of cutting-edge applications including intraoperative monitoring, intraocular and auricular pressure sensing, post-stent surveillance, sleep analysis, swallowing detection, sweat metabolite profiling, cardiac rehabilitation, tactile perception, and electromyographic recording. Beyond conventional diagnostic and therapeutic functions, hydrogel interfaces are also increasingly enabling novel capabilities such as oral environment sensing and advanced human–machine interactions, further highlighting their versatility across both clinical and daily-life scenarios.

In these applications, multifunctional hydrogels function not only as passive supporting matrices, but also as active interfacial mediators that enable high-fidelity transduction of electrophysiological, biochemical, and mechanical signals. Their innate hydration, mechanical compliance, and dynamic chemical properties enable seamless and biocompatible integration with biological tissues. However, long-term wearability, signal fidelity, and environmental adaptability remain key challenges

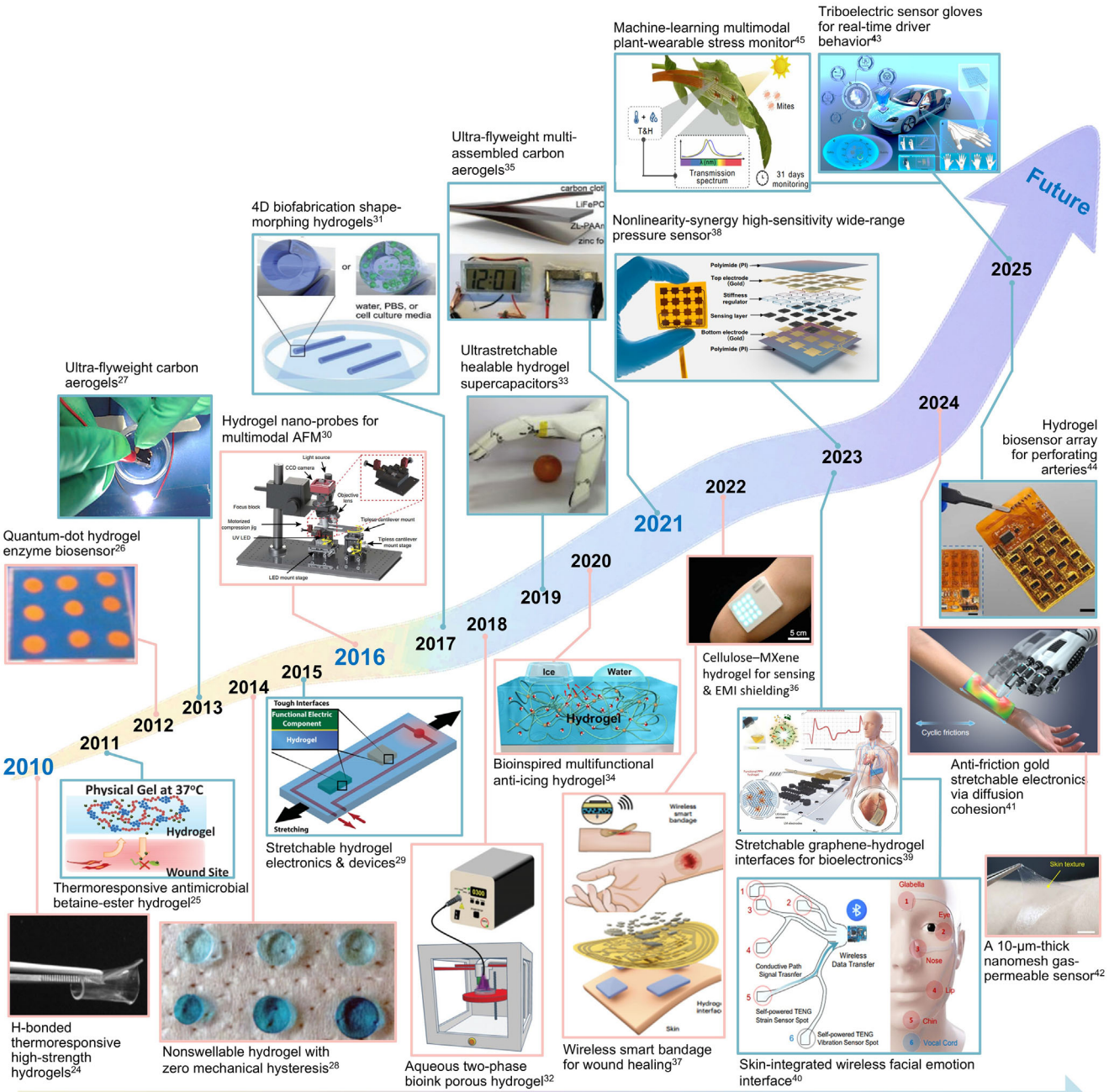


FIGURE 1 | Legend on next page.

that hinder the sustained clinical use of multifunctional hydrogel. Consequently, their successful integration into closed-loop systems will necessitate concerted interdisciplinary advances across materials science, electronics, and AI-driven optimization to keep pace with the evolving requirements of adaptive and self-monitoring medical technologies. The following sections focus on the design principles and synthesis strategies that drive hydrogel multifunctionality and adaptability, addressing key challenges such as mechanical performance, biocompatibility, and long-term stability. These innovations aim to enhance the clinical

FIGURE 1 | Evolution of hydrogel interface toward next generation intelligent flexible electronics. H-bonded thermoresponsive high-strength hydrogels. Reproduced With Permission [17]. Copyright 2010, Wiley-VCH; Thermoresponsive antimicrobial betaine-ester hydroge. Reproduced with permission [18]. Copyright 2011, Wiley-VCH; Quantum-dot hydrogel enzyme biosensor. Reproduced with permission [19]. Copyright 2012, Wiley-VCH; Ultra-flyweight carbon aerogels. Reproduced with permission [20]. Copyright 2013, Wiley-VCH; Nonswellable hydrogel with zero mechanical hysteresis. Reproduced with permission [21]. Copyright 2014, American Association for the Advancement of Science (AAAS); Stretchable hydrogel electronics & devices Reproduced with permission [22]. Copyright 2015, Wiley-VCH; Hydrogel nano-probes for multimodal AFM. Reproduced with permission [23]. Copyright 2016, Springer Nature; 4D biofabrication shape-morphing hydrogels. Reproduced with permission [24]. Copyright 2017, Wiley-VCH; Aqueous two-phase bioink porous hydrogel. Reproduced with permission [25]. Copyright 2018, Wiley-VCH; Ultrastretchable healable hydrogel supercapacitors. Reproduced with permission [26]. Copyright 2019, Elsevier Ltd; Bioinspired multifunctional anti-icing hydrogel. Reproduced with permission [27]. Copyright 2020, Cell Press; Ultra-flyweight multi-assembled carbon aerogels. Reproduced with permission [28]. Copyright 2021, Wiley-VCH; Cellulose–MXene hydrogel for sensing & EMI shielding. Reproduced with permission [29]. Copyright 2022, KeAi Communications Co., Ltd; Wireless smart bandage for wound healing. Reproduced with permission [30]. Copyright 2022, Springer Nature; Nonlinearity-synergy high-sensitivity wide-range pressure sensor. Reproduced with permission [31]. Copyright 2023 Springer Nature; Stretchable graphene-hydrogel interfaces for bioelectronics. Reproduced with permission [32]. Copyright 2023 Springer Nature; Skin-integrated wireless facial emotion interface. Reproduced with permission [33]. Copyright 2023 Springer Nature; Anti-friction gold stretchable electronics via diffusion cohesion. Reproduced with permission [34]. Copyright 2024 Springer Nature; A 10-μm-thick nanomesh gas-permeable sensor. Reproduced with permission [35]. Copyright 2024, American Association for the Advancement of Science (AAAS); Triboelectric sensor gloves for real-time driver behavior. Reproduced with permission [36]. Copyright 2025 Springer Nature; Hydrogel biosensor array for perforating arteries. Reproduced with permission [37]. Copyright 2025, American Association for the Advancement of Science (AAAS); Machine-learning multimodal plant-wearable stress monitor. Reproduced with permission [38]. Copyright 2025, American Association for the Advancement of Science (AAAS).

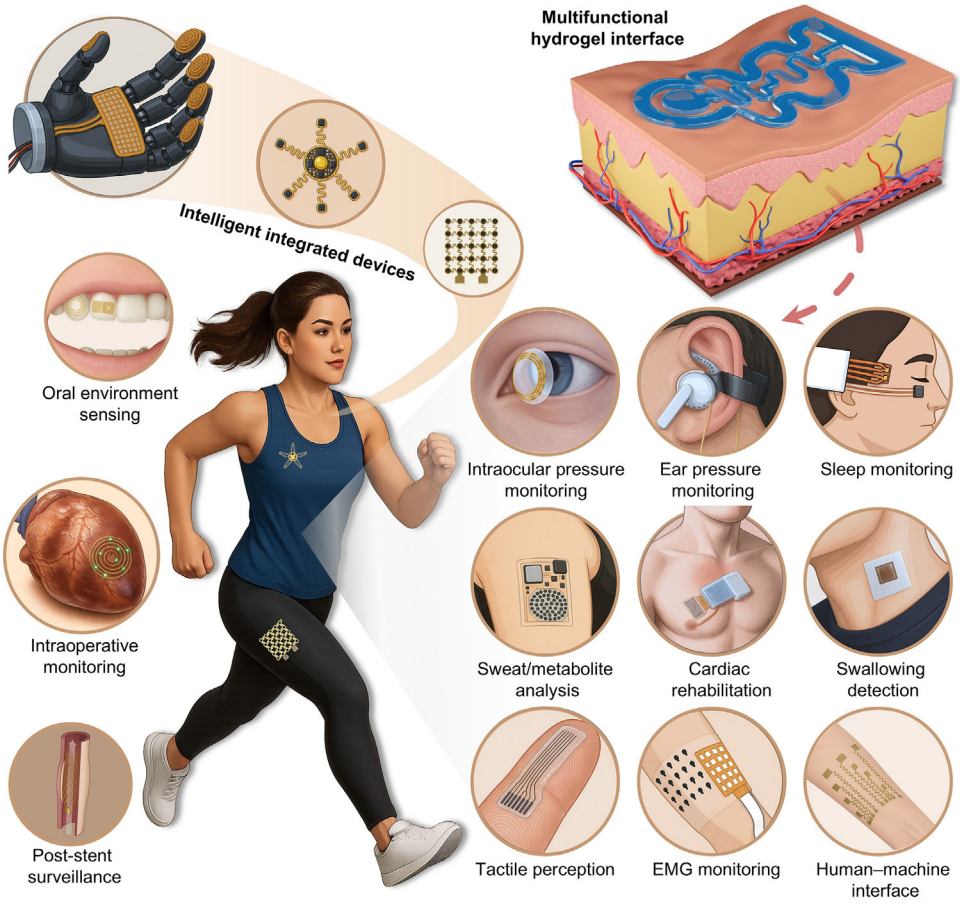


FIGURE 2 | Hydrogel interface for multimodal sensing, health monitoring and human–machine integration.

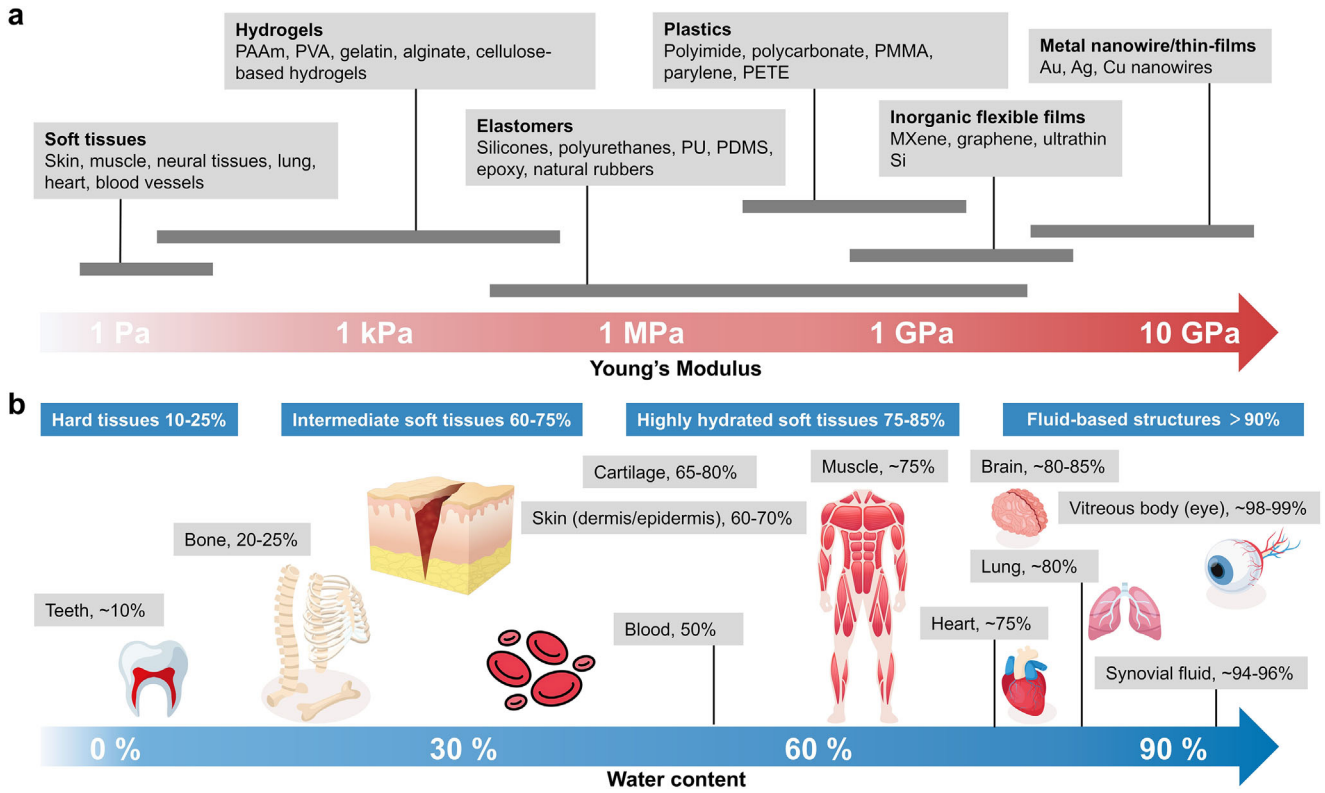


FIGURE 3 | Hydration and mechanical properties of soft tissues and conventional materials. (a) Young's modulus comparison from soft tissues (low Pa) to rigid materials like metals and inorganic films (high GPa), highlighting the inherent soft and flexible characteristics of hydrogel with tissue. (b) Hydration levels of various tissues ranging from 10% in bone to over 90% in fluid-based structures.

viability of hydrogel interfaces for flexible, self-monitoring, and closed-loop medical systems.

2.1 | Intrinsic Characteristics of Hydrogels

Hydrogels hold a unique position among soft electronic materials due to their intrinsic hydration, tunable viscoelasticity, and biochemical versatility, collectively enabling close conformity to the diverse mechanical and aqueous environments inherent in living tissues. Their cross-linked polymeric networks can retain hundreds of times their dry weight in water, thereby forming a highly hydrated microenvironment that mimics the extracellular matrix [47, 48]. This pronounced hydration not only imparts softness and mechanical compliance but also facilitates efficient ion transport, allowing seamless interfacing with the innate ionic signaling mechanisms of biological systems [49, 50].

As shown in Figure 3a,b, and Table 1, the mechanical diversity of native tissues spans from approximately 1–100 kPa in neural tissues to 1–30 GPa in bone. Similarly, hydration levels range from ~10% in teeth to over 95% in synovial fluid. Tissue hydration and mechanical properties are inherently connected to their biomechanical functions. Highly hydrated soft tissues such as synovial fluid possess low stiffness to maintain flexibility, while stiffer tissues like bone achieve structural integrity through reduced hydration. Hydrogels, synthesized from materials such as polyacrylamide, poly(vinyl alcohol), gelatin, alginate, and cellulose derivatives, are uniquely capable of spanning the wide biomechanical property range found in various human tissues [51]. By modulating factors such as crosslinking density and water content, their Young's modulus can be tuned from kPa to GPa, making them adaptable to soft tissues like skin, muscle, and brain [52, 53]. This extensive tunability enables hydrogels to effectively bridge the mechanical mismatch between rigid electronics and compliant biological substrates, offering unmatched compatibility with soft tissues. Although hydrogels offer versatile tissue-mimetic properties, their long-term performance in real-world applications depends critically on precise molecular design and tailored network architecture. In particular, ensuring mechanical stability and maintaining consistent functionality within dynamic living environments remain fundamental challenges. Therefore, a strategic focus on these design parameters is essential, which will be examined in detail throughout the following sections.

2.2 | Material Selection, Molecular Design, and Chemical Interaction for Advanced Hydrogels

2.2.1 | Material Selection for Hydrogels Construction

The design of hydrogels for flexible electronics begins with the selection of materials, where the polymeric matrix plays a central role in dictating the mechanical integrity, biological compatibility, and electronic performance of the hydrogel. As emphasized in earlier sections, hydrogels must balance the need for softness and biocompatibility with mechanical strength and long-term stability in dynamic biological environments.

TABLE 1 | Literature summary of advanced soft materials for biomedical applications.

Material	Representative Materials	Young's Modulus	Stretchability	Water Content	Damage Tolerance	Biocompatibility	Typical Applications	References
Soft tissues	Skin, muscle, blood vessels, neural tissues	1 Pa – 100 kPa	High, 100%–300%	60%–85%	Fragile but self-healing	Excellent (native tissues)	Biomimetic materials Tissue engineering	[54]
Hydrogels	PAAm, PVA, gelatin, alginate, cellulose-based hydrogels	10 Pa – 1 MPa	High, 50%–1000% (tunable)	70%–95%	Moderate	Good, close to soft tissues	Biosensors Drug delivery Soft robotics	[55]
Elastomers	PDMS, PU, natural rubber	10 kPa – 10 MPa	High, 100%–800%	<5%	Excellent, fatigue-resistant	Moderate	Wearable electronics Flexible devices	[56]
Ionogels	Polymer–ionic liquid networks	100 Pa – 10 MPa	Moderate, 50%–300%	10%–60%	Limited, often brittle	Good (depends on ionic liquid toxicity)	Flexible electrolytes Energy storage	[57]
Conductive polymers	PEDOT:PSS, PANI, PPy	100 kPa – 1 GPa	Moderate, 20%–200%	10%–40%	Limited, poor cycling durability	Moderate	Bioelectrodes Electronic skin	[58]
Liquid metal composites	EGaIn, Ga–In–Sn embedded in elastomers	Matrix-dominated, ~10 kPa – 1 MPa	High, 100%–600%	Matrix-dependent (~0%–10%)	Excellent, self-healing	Good (Ga-based alloys biocompatible)	Stretchable circuits Flexible electrodes	[59]
Plastics	PI, PC, PMMA, PET	1 MPa – 10 GPa	Low, <10%	<1%	Excellent, wear-resistant	Poor (non-biodegradable)	Substrates Encapsulation layers	[60]
Cellulose/natural polymer composites	Nanocellulose, chitosan films	10 MPa – 10 GPa	Moderate, 10%–50%	5%–20%	Excellent, strong and tear-resistant	Excellent (biodegradable, renewable)	Biodegradable sensors Green packaging	[61]
Inorganic flexible films	MXene, graphene, ultrathin Si	1 – 200 GPa	Low, <5%	0%	Poor, brittle and notch-sensitive	Moderate (requires surface coating)	Energy storage Flexible electronics	[62]
Metal nanowire/thin-film networks	Au, Ag, Cu nanowires	>1 GPa	Moderate, 10%–100%	0%	Excellent, stable conductivity	Moderate (surface treatment required)	Transparent electrodes Flexible interconnects	[63]

1. Natural Polymers such as gelatin, chitosan, alginate, cellulose derivatives, and hyaluronic acid offer inherent biocompatibility, biodegradability, and the ability to recognize biological cues, making them ideal for bio-integrated and transient electronic devices [64]. These materials mimic the extracellular matrix, providing desirable mechanical compliance and tissue affinity. However, batch-to-batch variability and limited mechanical strength are common issues that need to be addressed. Natural hydrogels often lack the consistency needed for scalable production and fail to offer sufficient mechanical robustness for long-term, high-performance applications in electronic devices. This variability poses a challenge, particularly in applications that require long-term stability and reliable performance.
2. Synthetic Polymers, on the other hand, such as polyacrylamide (PAM), poly(vinyl alcohol) (PVA), polyethylene glycol (PEG), and polyacrylic acid (PAA), offer well-defined molecular structures and precise control over mechanical properties and degradation rates [51]. These synthetic polymers can be designed to exhibit superior reliability and long-term stability, rendering them particularly suitable for high-performance applications that require consistent mechanical properties. Compared with natural hydrogels, synthetic hydrogels offer enhanced control over mechanical consistency, yet they often lack the inherent biocompatibility and biodegradability of their natural counterparts [65]. This leads to a fundamental trade-off in performance. Synthetic hydrogels typically offer superior mechanical performance and structural integrity, whereas their natural counterparts excel in biological mimicry, biocompatibility, and inherent degradability under physiological conditions.

For future closed-loop flexible electronics, a hybrid approach combining natural hydrogels for biological compatibility and synthetic hydrogels for mechanical strength and stability is crucial. Therefore, achieving an optimal balance between these competing properties requires a deliberate and application-specific design approach.

2.2.2 | Molecular Design for Hydrogels Functionality

The molecular design of hydrogels is crucial for further optimizing both natural and synthetic hydrogels to meet the specific demands of biomedical and flexible electronic applications. This balance is necessary to reconcile the biomechanical properties, biological integration, and electronic performance required in such systems. Strategies such as group modification, grafting copolymerization, and surface functionalization are employed to optimize hydrogel performances.

1. Group modification strategies aim to enhance key properties such as hydration, ionic conductivity, and biological adhesion. Hydrophilic groups (e.g., –OH, –COOH, $–NH_2$) are incorporated into the polymer backbone to improve bioadhesion and tissue interaction, making hydrogels suitable for medical sensors and biosensing applications. For instance, catechol or thiol groups significantly enhance hydrogel adhesion to biological tissues and metallic electrodes under wet conditions [66]. Meanwhile, anionic groups such as carboxylate and sulfonate substantially improve ionic conductivity by introducing mobile counter-ions into the hydrogel network and providing fixed negatively charged sites. These features collectively facilitate ion transport through the hydrated polymer matrix, leading to more efficient bioelectronic signal transduction [67, 68].
2. Grafting and copolymerization are key methods used to optimize the properties of hydrogels for applications in bioelectronics and biomedicine [69]. Specific grafting methods include free radical polymerization, click chemistry, and carbodiimide coupling, which enable the attachment of functional groups to the polymer backbone [70]. For example, polyethylene glycol (PEG) can be grafted via grafting-to or grafting-from methods, enhancing water retention and biological adhesion, making it ideal for wound dressings and drug delivery systems [71]. Chitosan grafting, using methods such as carbodiimide coupling, improves adhesion and provides antimicrobial properties, suitable for biomedical implants and wound healing [72]. In copolymerization, methods like radical copolymerization, ionic liquid-mediated polymerization, and click chemistry are used to combine different monomers. For example, PEG-based copolymers with acrylic acid (AA) or polyvinyl alcohol (PVA) are synthesized through free radical polymerization, resulting in hydrogels with tuned mechanical strength, swelling behavior, and water retention for flexible sensors and drug delivery [73]. Poly(lactic acid) (PLA)-PEG block copolymers synthesized via ring-opening polymerization offer biodegradability and ionic conductivity, making them suitable for implantable devices [74]. These techniques enable the precise engineering of hydrogels with enhanced biocompatibility, mechanical stability, and electronic performance, advancing their use in self-monitoring medical systems and wearable bioelectronics.
3. Surface functionalization enhances hydrogel performance by adding bioactive molecules such as growth factors, antimicrobial agents, or antibodies to improve bioactivity and tissue compatibility. Common methods include grafting, layer-by-layer assembly, and plasma treatment, with materials like PEG and PVA used as polymeric coatings to reduce immunogenicity and increase stability for long-term implantation [75]. Covalent functionalization via carbodiimide coupling or click chemistry allows attachment of bioactive molecules, enabling targeted drug delivery and biosensing. Although surface functionalization can substantially enhance biocompatibility and bioactivity, it is often constrained by surface-dependent properties that may not always align or integrate seamlessly with the bulk properties of the hydrogel, potentially limiting its overall performance in more dynamic or complex applications. For optimal performance in biomedical and electronic applications, integrating all three strategies is essential to achieve the right balance of mechanical stability, biological compatibility, and functional adaptability required for long-term and reliable use.

2.2.3 | Chemical Interaction for Hydrogels Optimization

Hydrogels represent a cornerstone of next-generation biointerface materials, whose performance boundaries are fundamentally

defined by the chemistry of their chemical interactions. The three molecular design strategies discussed earlier involve a wide array of chemical bonding interactions, which can be systematically categorized based on their nature and strength, encompassing covalent, dynamic covalent, and non-covalent bonds (Table 2). Both synthetic and natural hydrogel systems, rooted in their distinct chemical foundations, demonstrate divergent strategic preferences within this framework. This divergence ultimately guides them toward unique technological pathways and application scenarios, particularly in medical sensing, where the choice of interaction type plays a critical role in performance and adaptability.

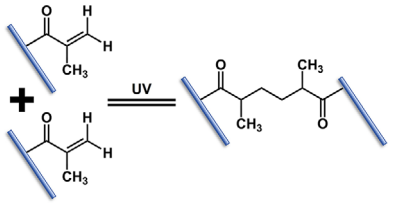
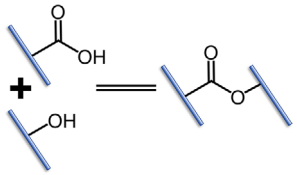
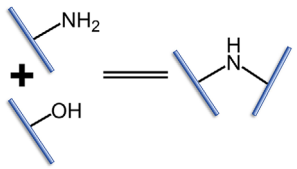
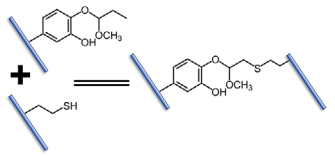
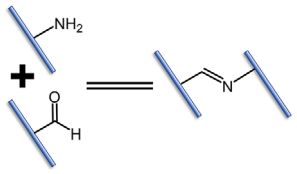
1. Covalent bonds such as C–C, C–O (ester, amide bonds), C–N (urea, carbamate bonds), and C–S (thioether bond), create permanent 3D networks that provide structural rigidity, dimensional stability, and controlled mass transport [76]. Synthetic hydrogels, employing precise reactions like thiol-ene click, azide-alkyne cycloaddition, Michael addition, or photo-crosslinking to tailor network pore size, mechanical modulus, and degradation timeline [77]. This capability meets the stringent requirements of implantable sensors for long-term structural integrity and consistent signal baseline stability. Natural hydrogels, typically achieve covalent crosslinking via reactions involving their native functional groups, such as amine, carboxyl, or hydroxyl moieties. Common strategies encompass Schiff base formation, genipin-mediated crosslinking, or enzymatic coupling catalyzed by transglutaminase [78]. While these methods proceed under mild conditions and better preserve bioactivity, the resulting natural hydrogels often lack the uniformity and mechanical strength of their synthetic counterparts. Consequently, they are more suited for constructing sensing interfaces designed for short-term use, biodegradation, or those requiring robust cellular integration and sustained release of bioactive factors.
2. Dynamic covalent bonds combine the strength of covalent linkages with conditional reversibility, serving as a key enabler for adaptive and self-healing sensing interfaces [79]. Representative bond types include disulfide bonds capable of reductive exchange, boronic ester bonds with reversible diol complexation, pH-responsive imine and oxime bonds, as well as thermally reversible Diels-Alder adducts. Synthetic hydrogels allow for the systematic design and incorporation of such dynamic chemistries [80]. For instance, disulfide bond exchange can impart self-repair capabilities to sensors operating in oxidative stress environments [81]. The glucose-sensitive complexation between phenylboronic acid and diols can be leveraged to construct closed-loop glucose monitoring interfaces. Furthermore, thermally reversible Diels-Alder reactions enable gentle device retrieval and recalibration [82]. This chemical programmability empowers synthetic systems to respond to specific chemical cues within complex physiological microenvironments. In contrast, natural hydrogels possess few inherent dynamic covalent structures that are amenable to precise manipulation. A notable exception includes the disulfide bonds found in proteins. However, their reorganization is often nonspecific and challenging to engineer as a core responsive mechanism.
3. Non-covalent interactions, encompassing ion coordination, hydrogen bonding, hydrophobic association, and van der Waals forces, are characterized by their reversibility, dynamism, and stimulus-responsiveness [83]. They form the physical basis for energy dissipation and microenvironmental communication within the material. Synthetic hydrogels can be engineered to incorporate high-strength, high-density non-covalent crosslinking points. Examples include the introduction of non-covalent ion coordination between Fe^{3+}, Zn^{2+}, or Al^{3+} ions and carboxyl/catechol groups to achieve exceptional toughness and conductivity [84, 85]. The design of units such as ureidopyrimidinone (UPy) quadruple hydrogen bonds or benzotriazole-based triple hydrogen bonds can significantly enhance a material's resistance to tearing and fatigue, which is critical for flexible sensors enduring dynamic physiological loads like heartbeat or muscle contraction. The core advantage of natural hydrogels lies in their rich repertoire of endogenous, biologically functional non-covalent interactions. Prominent examples are the "egg-box" structure formed between alginate and Ca^{2+} ions, the pH-dependent protonation and electrostatic networking of chitosan, and the thermally reversible gelation of collagen or gelatin driven by hydrogen bonding and hydrophobic stacking. These interactions are inherently biocompatible, enabling such hydrogels to achieve gentle yet intimate integration with host tissues. This makes them particularly suitable for epidermal sensing or short-term, absorbable monitoring applications.

As a quick summary, this chapter have elucidated the integral roles of material selection, molecular design, and intermolecular interactions in engineering hydrogels for medical sensing. Material selection establishes the foundational paradigm: synthetic polymers enable top-down, tunable architectures with controlled mechanics and electronic function, whereas natural biopolymers offer innate biocompatibility and support bottom-up integration with biological tissues. Molecular design, affords precise tailoring of hydrogel properties. Strategic modifications enhance hydration, ionic conductivity, bioadhesion, and tissue compatibility, thereby aligning molecular-scale features with macroscopic sensor performance. The operational efficacy of these systems is governed by the interplay of covalent, dynamic covalent, and non-covalent interactions. Covalent cross-linking ensures structural integrity, while reversible bonds and supramolecular forces enable adaptive, bioaffinitive interfaces capable of sustained physiological engagement. Together, these three elements form a coherent design framework that harmonizes biocompatibility, mechanical resilience, and electronic functionality. This integrated approach thus enables hydrogel-based sensors to evolve from passive monitors into adaptive systems capable of bidirectional communication with living tissues, which represents a pivotal advancement for next-generation medical sensing.

3 | Network Design, Interfacial Coupling, and Micro/Nanoscale Fabrication for Advanced Hydrogel Interfaces

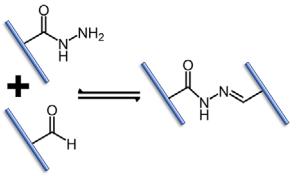
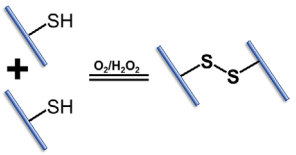
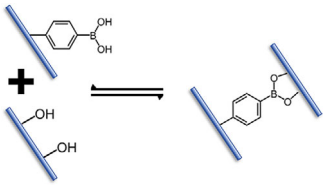
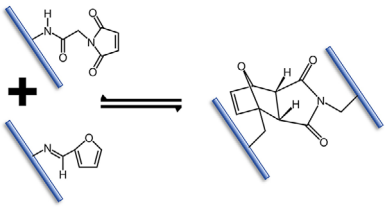
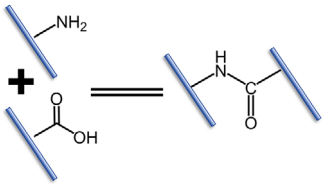
The advancement of hydrogel-based flexible electronics requires a multidimensional design approach. It is not enough to simply optimize material selection and molecular design. Achieving multifunctional systems necessitates a cohesive integration across molecular-level properties, interfacial characteristics, and

TABLE 2 | Overview of covalent bonds, dynamic covalent bonds, and non-covalent bonds in hydrogel systems.

Category	Specific Bonds	Methods	Molecular Diagram	Applications and Functions	Natural	Synthetic	Ref.
Covalent Bonds	**Single Bonds** C–C, C–O, C–N, C–S	Radical polymerization, esterification, amination, or thiolation		1. Enhancing hydrogel structural stability 2. Improving mechanical strength 3. Ensuring long-term stability in biomedical applications	Gelatin Chitosan Alginate Seaweed Sugar	Polyacrylamide (PAM) Polyvinyl Alcohol (PVA) Polyethylene Glycol (PEG) Polyacrylic Acid (PAA)	[86]
	Ester Bond C–O	Esterification reaction		1. Enhancing chemical stability 2. Increasing biocompatibility 3. Suitable for drug delivery and temporary biological interfaces	Alginate Gelatin Natural Rubber	Polyamide Polyether Polyester	[87]
	Amine Bond C–N	Amine reactions, nucleophilic substitution		1. Enhancing hydrophilicity. 2. Increasing bioactivity 3. Supporting cell adhesion and interactions with biological tissues	Chitosan Gelatin Alginate	Polydimethylsiloxane (PDMS) Polyurethane (PU)	[88]
	Thioether Bond C–S	Thiolation reaction		1. Antioxidant properties 2. Enhancing stability for long-term sensors and implants	Lignin Seaweed Sugar	Polyvinyl Chloride (PVC) Polystyrene (PS)	[89]
	Amine and Aldehyde Reaction C = N (Schiff base)	Schiff base formation reaction		1. Facilitating cross-linking under mild conditions 2. Ideal for short-term or biodegradable medical sensors	Gelatin Chitosan Agar	Polyacrylamide (PAA) Polyvinyl Alcohol (PVA)	[90]

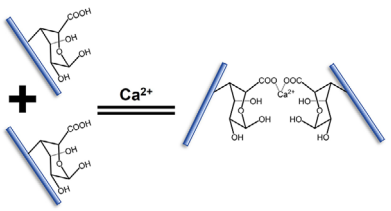
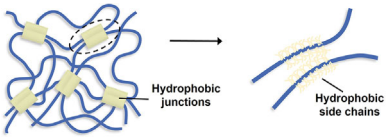
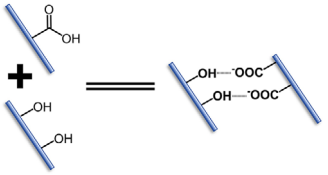
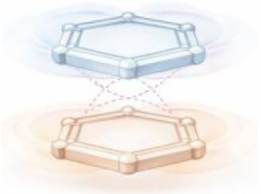
(Continues)

TABLE 2 | (Continued)

Category	Specific Bonds	Methods	Molecular Diagram	Applications and Functions	Natural	Synthetic	Ref.
Dynamic Covalent Bonds	**Disulfide Bond** –S–S	Redox reactions, oxidation of thiols		1. Self-healing 2. Restoring structural integrity 3. Suitable for wearable electronics.	Keratin Collagen	Polydisulfide Polyamide	[91]
	Boron Ester Bond Phenylboronic acid reacting with alcohol groups	Esterification or click chemistry reaction		1. Reversible crosslinking 2. Responsive to pH changes 3. Enhancing smart sensors and drug delivery systems	Glucosamine Pectin	Polyacrylamide (PAA) Polyvinyl Alcohol (PVA)	[92]
	Aldehyde and Amine Reaction –C = N (Schiff base)	Schiff base formation, condensation reaction		1. Self-healing 2. Enabling responsiveness in hydrogels	Chitosan Gelatin	Polyacrylamide (PAA) Polyurethane (PU)	[93]
	Diels-Alder Reaction Diene reacting with dienophile	Diels-Alder reaction, cycloaddition reaction		1. Reversible, temperature-sensitive crosslinking 2. Tunable material properties	Collagen Gelatin	Polystyrene (PS), Polyether Sulfone (PES)	[94]
	Reversible Amide Bond N–C = O	Amide bond formation and hydrolysis reaction		1. pH-sensitive crosslinking 2. Useful in dynamic drug delivery and biosensing applications	Chitosan Gelatin Collagen	Polyvinyl Alcohol (PVA), Polyacrylamide (PAA)	[95]

(Continues)

TABLE 2 | (Continued)

Category	Specific Bonds	Methods	Molecular Diagram	Applications and Functions	Natural	Synthetic	Ref.
Non-Covalent Bonds	**Hydrogen Bond** Hydrogen bonding between Polyvinyl Alcohol and water	Hydrogen bonding interactions		1. Improving hydrophilicity and biocompatibility 2. Facilitating cell interaction and tissue engineering	Gelatin Alginate Collagen	Polyvinyl Alcohol (PVA) Polyacrylic Acid (PAA)	[96]
	Hydrophobic Interaction Non-polar group interactions	Van der Waals interaction, hydrophobic interactions		1. Enhancing mechanical strength 2. Crucial for biomedical applications	Alginate Natural Rubber	Polystyrene (PS) Polyvinyl Alcohol (PVA)	[97]
	Van Der Waals Force Short-range interactions between non-polar groups	London dispersion forces		1. Stability and flexibility 2. Reducing dehydration and enhancing adaptability	Gelatin Seaweed Sugar	Polyacrylic Acid (PAA) Polyvinyl Alcohol (PVA)	[98]
	Coordination Bond Metal ions (e.g., Cu^{2+}, Zn^{2+}) coordinating with hydrogel surfaces	Metal coordination, chelation reaction		1. Enhancing metal-ion responsiveness 2. Increasing sensitivity to environmental stimuli in sensors and drug delivery systems	Alginate Gelatin Chitosan	Polyacrylic Acid (PAA) Polyurethane (PU)	[99]
	π-π Interaction Interactions between aromatic ring structures	π-π stacking interactions, aromatic interactions		1. Strengthening mechanical properties 2. Enabling multifunctional applications in flexible electronics	Agarose Gelatin	Polystyrene (PS) Polyvinyl Chloride (PVC)	[100]

device-level fabrication techniques. This section reviews how each of these factors contributes to creating next-generation hydrogels for biomedical devices and flexible electronics.

1. At the molecular level, rational cross-linking networks are essential to balance softness with mechanical toughness, and dynamic responsiveness with operational stability.
2. At the interfacial level, tailored adhesion mechanisms are critical for achieving efficient coupling between hydrogels and either biological tissues or electronic substrates under physiologically dynamic conditions.
3. At the device level, advanced manufacturing techniques are indispensable for transforming hydrogel materials into precise, scalable, and application-specific architectures.

By highlighting these critical "processing–structure–property" relationships, this section not only establishes a conceptual bridge between material fundamentals and device applications but also lays the logical groundwork for subsequent discussions on multifunctional integration and system-level intelligence.

3.1 | Network Architecture for Tunable Hydrogel Interface Functions

The structural integrity and multifunctionality of hydrogel-based electronic interfaces are fundamentally governed by their underlying cross-linking networks [101, 102]. These distinct network architectures not only dictate the bulk mechanical properties but also critically determine interfacial robustness and long-term operational stability when integrated into flexible electronic systems (Figure 4a).

1. Single-network hydrogels represent the simplest architectural, relying on homogeneous covalent or physical crosslinks. Although straightforward to fabricate, these systems often exhibit limited fracture toughness and are prone to rapid crack propagation [103]. Nevertheless, they serve as essential baseline models for elucidating fundamental structure–property relationships in hydrogel-based bioelectronics.
2. Double-network hydrogels overcome inherent brittleness by integrating two interpenetrating networks with contrasting mechanical properties, namely a rigid, energy-dissipating skeleton embedded within a soft, stretchable matrix. The synergistic combination of networks enhances both toughness and, in optimized designs, resistance to fatigue crack growth. Fatigue performance can be further improved through reversible energy dissipation, hierarchical structuring, and nanomaterial reinforcement. These advances position double-network hydrogels as promising materials for dynamic, load-bearing interfaces such as electronic skin and articular sensors, though their practical fatigue behavior remains critically governed by network architecture, crosslinking chemistry, and specific loading conditions [104, 105].
3. Multiple-network hydrogels incorporate three or more independent networks, substantially enhancing mechanical cohesion and resilience [106]. The hierarchical cross-link distribution effectively disperses stress and mitigates localized failure, ensuring long-term structural integrity under cyclic mechanical loading [107]. These architectures are particularly promising for implantable bioelectronic devices to withstand repetitive stresses. However, the complexity of these systems introduces challenges in scalability and cost for commercial production, presenting a trade-off between enhanced performance and practical feasibility.
4. Topological networks, composed of physically interpenetrating polymer chains without relying on covalent crosslinks, enable reversible chain slippage under deformation. This "sliding-chain" mechanism effectively dissipates energy and prevents catastrophic failure, endowing the material with self-healing capabilities and mechanical adaptability [108]. These features make topological networks ideal for interfacing with dynamically active tissues like the heart or lungs.
5. Nanofiller-reinforced networks incorporate conductive or reinforcing nanomaterials (e.g., carbon nanotubes, nanoclay, MXenes) as multifunctional crosslinking sites [109]. These nanostructured additives significantly enhance the elastic modulus, toughness, and electrical (or ionic) conductivity of the hydrogel, thereby improving stability and performance at the bioelectronic interface. While they offer enhanced structural stability and performance, incorporating nanofillers can result in increased complexity and cost, and may also introduce toxicity concerns when used in implantable devices.
6. Dynamically crosslinked networks utilize reversible interactions including hydrogen bonding, ionic coordination, and host–guest complexation to form structures capable of stimulus-responsive behavior, self-repair, and tunable mechanical properties [80]. These networks maintain interfacial adhesion and structural integrity even under repeated mechanical or chemical stress, ensuring reliability in biosensing and therapeutic applications.
7. Crystalline domain-reinforced networks incorporate semicrystalline polymer segments into an amorphous matrix, serving as robust physical anchors [110]. These crystalline regions effectively resist creep and stress relaxation, significantly improving long-term dimensional stability. Such networks are particularly suitable for hydrogel devices operating under variable mechanical loads or requiring long-term implantation.
8. Nanoconfined networks represent an advanced design paradigm where polymer chains are spatially restricted within nanoscale domains (e.g., micelles, nanocages, layered structures) [111]. This confinement effectively inhibits chain slippage and crack propagation, yielding exceptional mechanical robustness. Simultaneously, the nanoconfined architecture allows precise regulation of ion transport pathways, offering great potential for multifunctional hydrogels that require both mechanical durability and controlled ionic conductivity.

Collectively, these crosslinking strategies significantly expand the design landscape of multifunctional hydrogels. Through rational design of energy dissipation pathways, stress distribution

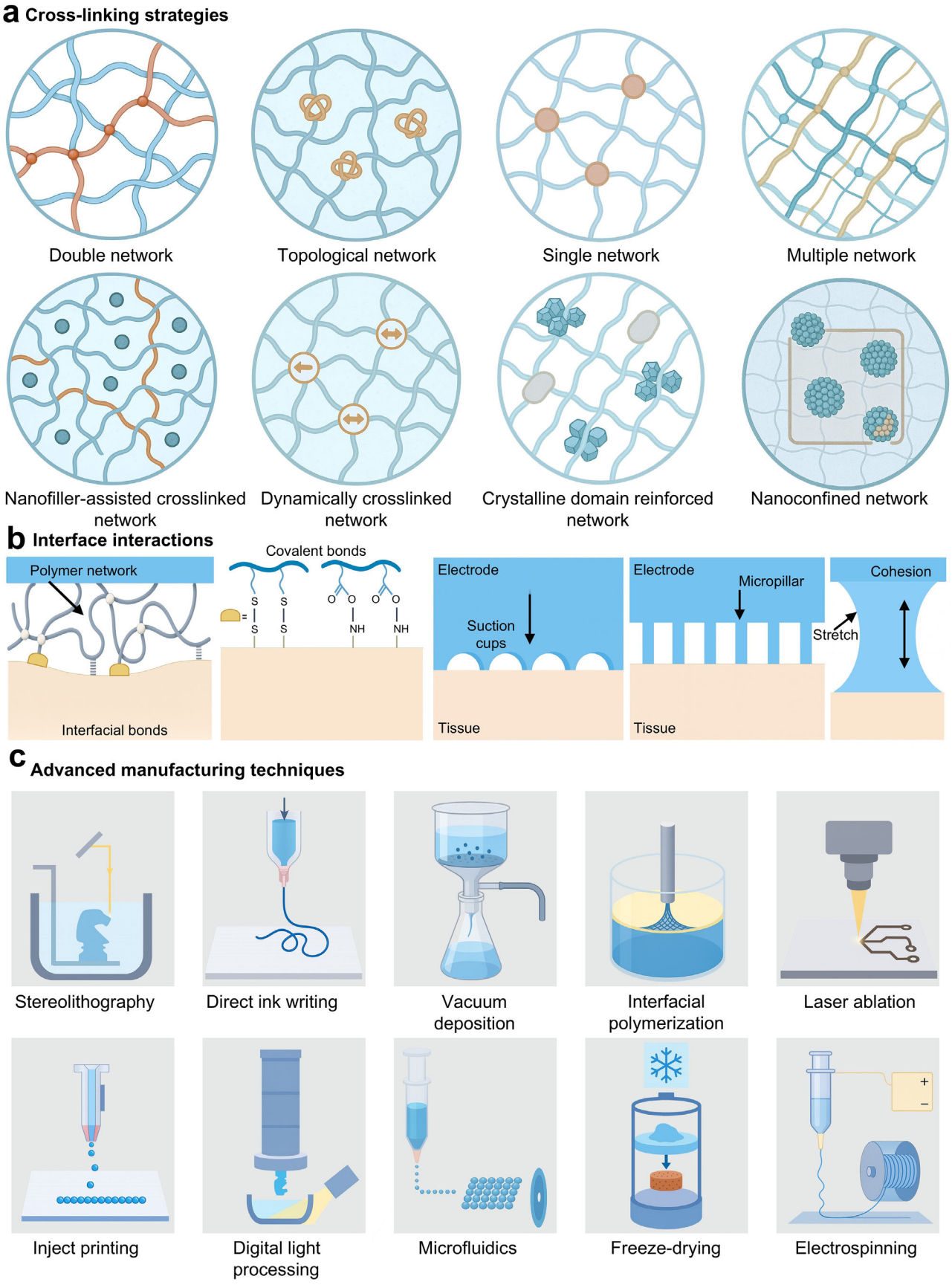


FIGURE 4 | Engineering strategies for multifunctional hydrogel interfaces. (a) Network architectures for functional adaptability. (b) Interfacial bonding mechanisms to ensure stable coupling between hydrogels and bioelectronic systems. (c) Advanced manufacturing techniques for hydrogel-based electronics.

mechanisms, and interfacial stabilization modes, researchers can fabricate hydrogel networks that exhibit high cohesion, superior mechanical toughness, and stable operation in flexible electronic and bio-integrated environments. This progress provides a solid foundation for the development of next-generation high-performance soft electronics.

3.2 | Interfacial Coupling Mechanism Between Tissue and Hydrogel Interface

The performance and long-term durability of hydrogel-based flexible electronics are ultimately realized at the interface between biological tissues and device substrates. Effective adhesion arises from the synergy between molecular-scale interactions and microstructural design, determining interfacial toughness, cohesion, and operational stability under dynamic physiological conditions.

3.2.1 | Interfacial Coupling Forces

At the molecular scale, hydrogel–substrate adhesion must be tailored to meet the specific demands of different substrates in biomedical and electronic applications (Figure 4b). Rigid substrates, such as electronic devices, require permanent adhesion, which is achieved through covalent bonds (e.g., thiol-ene reactions, carbodiimide chemistry) to ensure long-term fixation. For soft, dynamic tissues, reversible adhesion is essential to accommodate tissue movement without causing damage. In these cases, non-covalent interactions, such as hydrogen bonding, ionic complexation (Fe^{3+} or Ca^{2+}), and van der Waals forces, provide adaptive bonding that supports dynamic conformal contact. For specialized interfaces, such as those with functionalized substrates (e.g., metallic electrodes or biomolecular coatings), supramolecular interactions like host–guest complexes and metal–ligand coordination introduce dynamic stability, allowing for repeated attachment-detachment cycles without compromising interfacial integrity [112]. This makes them ideal for smart bioelectronic devices requiring stimuli-responsive behavior.

Covalent anchoring through various strategies, such as amide coupling (e.g., carbodiimide-mediated coupling), thiol-ene click chemistry (e.g., thiol-ene photopolymerization), Michael addition reactions, and alkyne-azide click chemistry, results in strong, nearly irreversible fixation, with adhesion energies typically ranging from 100 to 500 mJ m^{-2} [113]. The thermodynamic work of adhesion is expressed as:

$$W_{ad} = \gamma_1 + \gamma_2 + \gamma_{12} \tag{1}$$

where γ_1 and γ_2 are the surface energies of the hydrogel and substrate, and γ_{12} is their interfacial energy. These covalent bonds provide long-term stability in implantable devices or biosensors (blood glucose monitoring sensors, neural interfaces, and cardiac pacemakers) where permanent adhesion is necessary for consistent performance [114]. In contrast, hydrogen bonding and ionic interactions provide moderate but reversible adhesion (10–100 mJ m^{-2}), enabling stress relaxation and conformal contact under cyclic deformation. Van der Waals forces, although weak, provide a universal adhesive baseline scaling inversely with interfacial separation:

$$\mathrm{F} \propto \mathrm{A}/6\pi\mathrm{D}^3 \tag{2}$$

where A is the Hamaker constant and D the distance. These forces, though weaker than covalent or ionic bonds, are essential for ensuring basic adhesion and interface stability in bioelectronics (flexible wearable sensors and soft neural electrodes), where lightweight designs are critical for comfort and long-term wearability [46]. Finally, supramolecular motifs such as host–guest complexes or metal–ligand coordination introduce dynamic and reversible stability, allowing repeated attachment–detachment cycles without loss of performance. These repeat adhesion properties are particularly useful in implantable bioelectronics that must endure cyclic loading while maintaining functional performance over extended periods [115].

3.2.2 | Microstructural Adhesion Strategies

At the structural level, bioinspired architectures dramatically enhance adhesion and robustness. Suction-cup–like structures generate negative pressure differentials, with the adhesion force approximated by

$$F = \Delta\mathrm{P} \cdot \mathrm{A} \tag{3}$$

where ΔP is the pressure difference and A the effective contact area. Such structures are particularly effective on wet interfaces. Micropillar arrays, inspired by gecko setae or octopus suckers, maximize contact area and shear resistance [116]. Their detachment energy approximates the Kendall peel model,

$$G = \frac{P^2}{2Eb} \tag{4}$$

where P is the peel force, E the elastic modulus, and b the film thickness. Interpenetrating network adhesion, in which polymer chains diffuse across the interface and form entangled domains, further strengthens cohesion [117]. The adhesion strength scales with chain density (ρ) and interdiffusion depth (d), following

$$\sigma_{adh} \propto \rho \cdot d^{1/2} \tag{5}$$

Additiolly, gradient and multiscale interfaces mitigate modulus mismatch between soft hydrogels and rigid electrodes, alleviating stress concentration and preventing delamination under repeated strain. The integration of these interfacial coupling strategies decisively influences the overall performance of hydrogel-based devices across various flexible electronic applications [118]. This advanced capability establishes a foundation for closed-loop therapeutic systems and real-time physiological monitoring platforms. Essentially, strategic interface engineering transforms hydrogels from passive mechanical buffers into active, intelligent mediators facilitating seamless energy and information exchange [119]. By harnessing multi-scale adhesion mechanisms, hydrogel-based bioelectronics achieve unprecedented integration with biological systems, ensuring mechanical conformity and functional reliability under demanding conditions of daily use or long-term implantation.

With material systems meticulously design and interfaces engineering, the ultimate challenge transitions to system-level

assembly and rigorous application validation. Hydrogels must be reliably incorporated into multilayer device architectures that harmonize soft, hydrated networks with flexible conductors, functional substrates, and protective encapsulation layers for advanced flexible electronics. A critical challenge remains the reconciliation of the innate compliance of hydrogels with the mechanical and operational stability required by conventional electronics, especially when considering self-healing and hybrid systems that integrate conductive nanomaterials like MXene to ensure long-term functionality and biocompatibility. This balance of compliance with electrochemical stability, achieved through multifunctional and hybrid material strategies (such as microstructural adhesion hydrogels, graphene oxide reinforcement, Diels–Alder self-healing mechanisms, catechol-based bioadhesive chemistry, and biodegradable cellulose-based materials), is central to enhancing the integration of hydrogels in flexible electronics and ensuring their reliable, long-term performance in future real-world applications [120, 121].

3.3 | Advanced Manufacturing Technology for Hydrogel Interface Optimization

The emergence of hydrogel-based electronics depends on the integration of network design, interfacial coupling, and advanced fabrication techniques. While molecular crosslinking establishes the mechanical properties and biocompatibility of hydrogels, advanced fabrication enables precise scaling, enhancing device-level performance. These fabrication methods ensure structural integrity and ionic conductivity, bridging the gap between lab-scale prototypes and functional devices. Meanwhile, advanced fabrication improves the interfaces stability by creating dynamic, stable adhesion, ensuring long-term reliability and flexibility under cyclic deformation, essential for applications in implantable systems and wearable electronics. Moreover, in contrast to conventional molding or casting methods, next-generation fabrication strategies offer precise control over material architecture, local composition, and integrated functionality, thereby effectively bridging the gap between laboratory-scale prototypes and practical, high-performance devices (Figure 4c).

Additive manufacturing techniques (e.g., stereolithography, digital light processing, direct ink writing) provide exceptional micron-scale precision and extensive design freedom [122]. These methods facilitate the fabrication of biomimetic geometries, hierarchically porous structures, and patient-specific architectures that conform seamlessly to intricate biological surfaces. For example, 3D printing of conductive hydrogels has been used for wearable sensors that monitor motion and body temperature while maintaining flexibility and high conductivity [123]. Importantly, the digital nature of these approaches allows rapid prototyping and straightforward customization, significantly accelerating the translation of hydrogel-based devices into personalized medical applications [124].

Microfluidic and interfacial assembly techniques represent another promising frontier, enabling the precise encapsulation of bioactive molecules, conductive nanomaterials, or even living cells within hydrogel matrices [125]. For instance, hydrogel microfluidic chips have been developed to mimic vascular or neural networks, which are essential for the fabrication of implantable bioelectronics that integrate sensing, actuation, and therapeutic delivery functions [126]. By manipulating flow and polymerization dynamics in confined microscale channels, microfluidics produces compartmentalized, multi-lumen hydrogel architectures mimicking vascular or neural networks. Such designs are particularly valuable for constructing implantable bioelectronic interfaces capable of integrating sensing, actuation, and therapeutic delivery functions [127].

Nanomanufacturing and laser-based processing methods further push the boundaries of spatial resolution and functional integration [128]. Techniques like femtosecond laser ablation and laser-induced forward transfer enable localized modification of hydrogel conductivity, topography, and chemical functionality, creating reconfigurable circuits and micropatterned sensor arrays [129]. These methods have enabled the integration of electronic components with hydrogel substrates without compromising the inherent softness or biocompatibility of hydrogels [130, 131].

Electrospinning and freeze-drying considerably expand the morphological diversity of hydrogel constructs. Electrospun hydrogel fibers exhibit high surface-area-to-volume ratios and provide mechanical reinforcement, while freeze-dried scaffolds produce highly interconnected porous networks that enhance ion transport, water retention, and drug-loading capacity. When functionalized with conductive fillers, these architectures produce lightweight, breathable, multifunctional hydrogel composites suitable for wearable and implantable applications [132]. For example, electrospun hydrogel scaffolds have been successfully used in wearable bioelectronics such as biosensing platforms for real-time health monitoring [133].

Hybrid integration strategies merge complementary manufacturing paradigms, such as 3D printing and electrospinning, to produce multilayered hydrogel-based devices for soft robotics and wearable bioelectronics. For instance, 3D-printed hydrogel soft actuators are combined with electrospinning to achieve high conductivity and mechanical properties suited for fine motor control in soft robotics [134]. Similarly, microfluidics coupled with laser patterning creates hydrogel devices with intricate fluidic channels, as seen in organ-on-a-chip applications, where laser patterning builds complex microfluidic networks mimicking vascular systems for drug testing. These hybrid methodologies enable adaptive, multifunctional systems capable of dynamic interactions with biological environments, exemplified by 3D-printed microfluidic hydrogel biosensors for real-time health monitoring [135]. Such convergent methodologies represent the transition from single-function hydrogels to multifunctional, intelligent systems capable of dynamic and adaptive interactions with biological environments [136].

The transformative potential of these advanced manufacturing techniques lies in their ability to generate complex structures and encode multifunctionality directly into hydrogel-based devices. By providing precise spatial control over composition, stiffness, conductivity, and porosity, these approaches facilitate fabrication of adaptive, biocompatible, and multifunctional hydrogel electronics [137]. Ultimately, advanced manufacturing is revolutionizing the field, transforming hydrogel-based electronics from conceptually promising materials into practical, scalable, and application-ready devices for

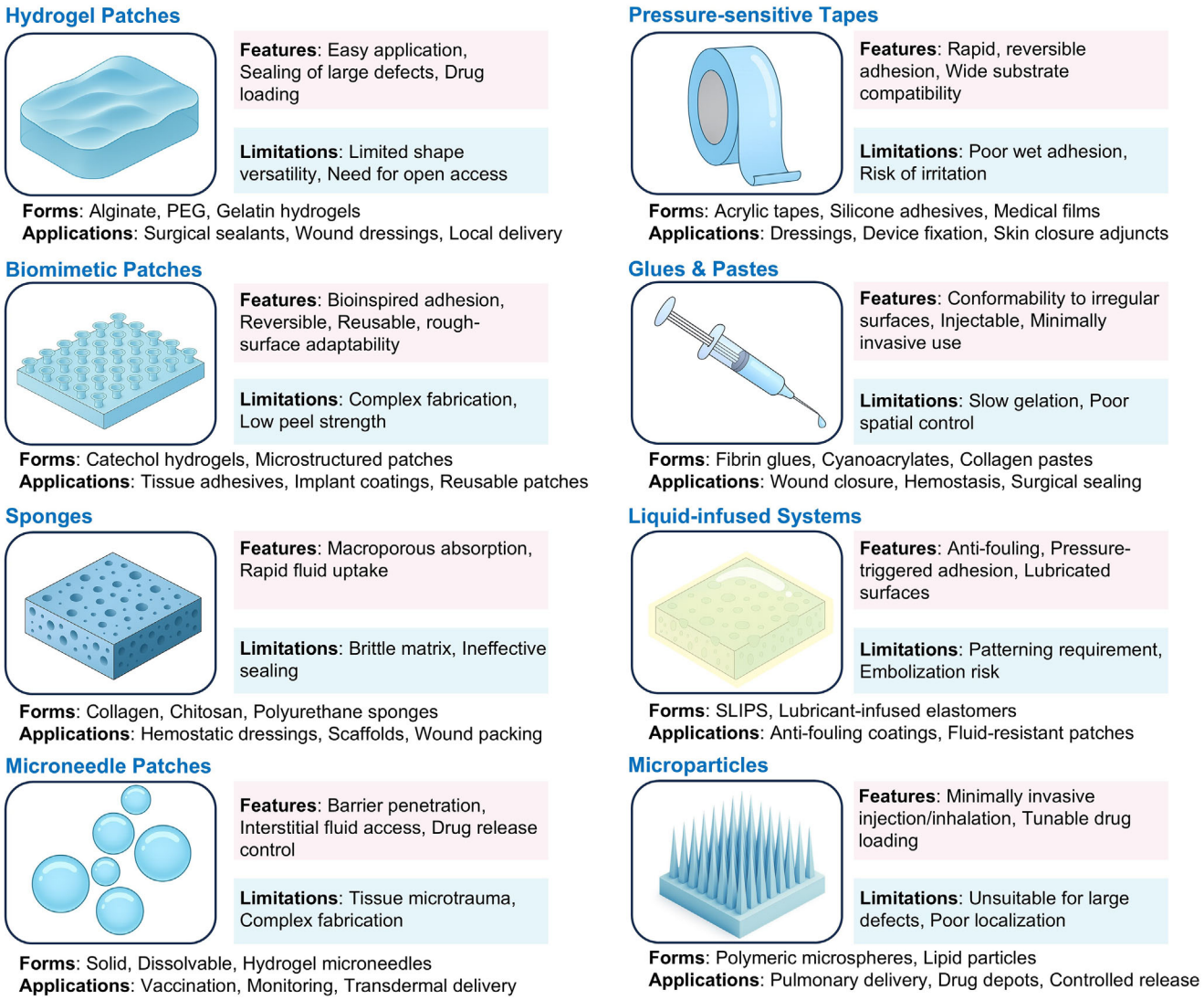


FIGURE 5 | Features and limitations of various interface platforms for sealing, sensing, and therapeutic delivery.

next-generation healthcare, robotics, and human–machine interfaces.

4 | Multifunctionality Regulation and Advanced Characterization of Hydrogel Electronics

4.1 | Hydrogel Versus Conventional Interface Materials

Benefiting from advanced crosslinking strategies and processing techniques, hydrogels are clearly distinguished from conventional interface materials such as pressure-sensitive adhesives and medical tapes (Figure 5) [138, 139]. Although emerging biomimetic patches and microneedle systems show promise for tissue integration and controlled release, hydrogels further distinguish themselves through intrinsic softness, extreme flexibility, and dual functionality as adhesives and sensing elements [140]. These attributes enable hydrogels to conform intimately to biological surfaces, ensuring superior comfort and extended wearability for long-term applications. Moreover, alternative material systems (e.g., sponges, liquid-infused surfaces, microparticulate composites) are often constrained by inherent limitations like mechanical brittleness, complex patterning requirements, or limited functional adaptability. In contrast, hydrogels exhibit inherent self-healing and finely tunable mechanical behavior, allowing recovery from deformation and maintained functional integrity across diverse, dynamic physiological conditions [141, 142]. As multifunctional and environmentally responsive materials, hydrogels are poised to redefine flexible bioelectronics by offering an integrated platform that merges electrical functionality with biological interfaces [143]. This positions them at the forefront of health monitoring technologies, smart wearable systems, and sustainable electronic devices.

4.2 | Application-Driven Performance Requirements

The rapid evolution of flexible electronics demands interface materials that withstand dynamic deformations, maintain reliable signal coupling, and ensure long-term biological safety.

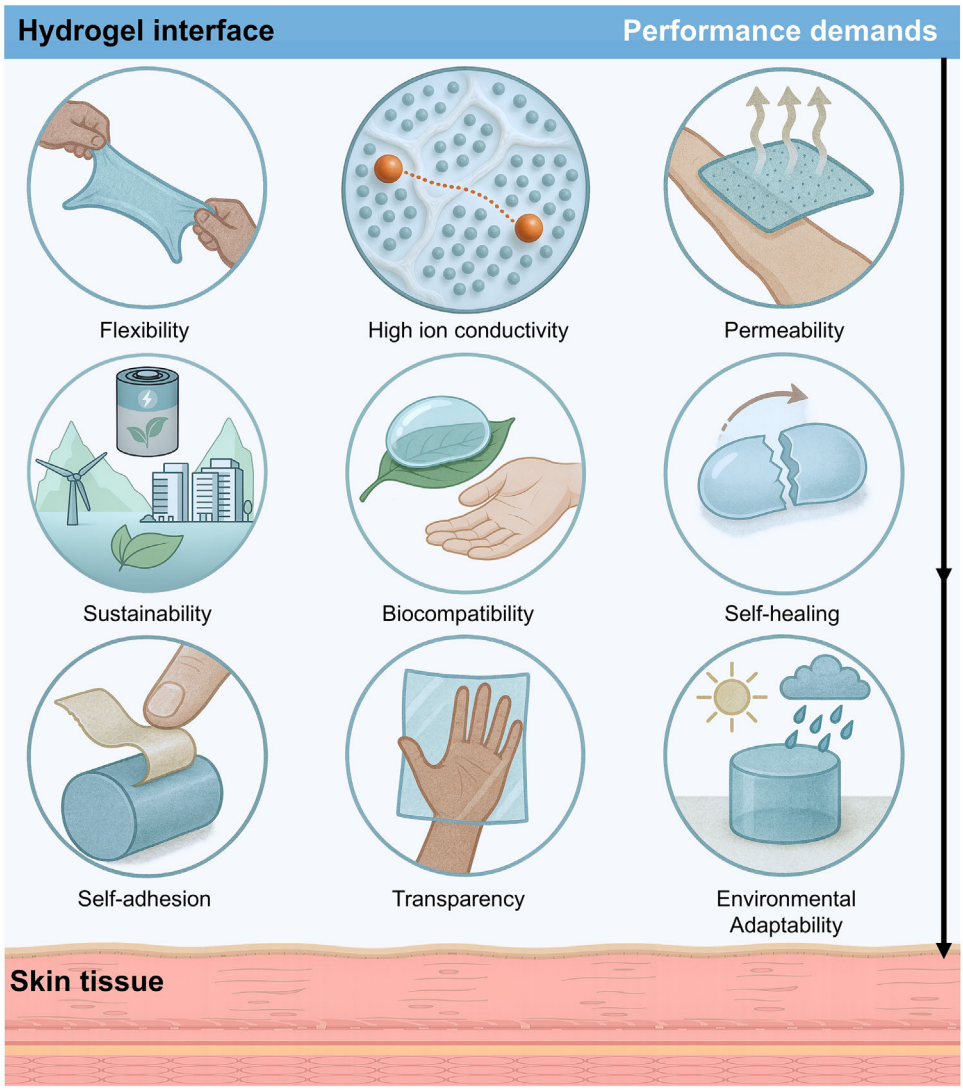


FIGURE 6 | Key performance demands of multifunctional hydrogel interfaces for skin-integrated electronics.

Conventional adhesives, tapes, and polymer films often fail to meet these stringent requirements due to brittleness, poor breathability, or limited biocompatibility [132]. In contrast, hydrogels offer a distinctive solution by integrating softness, ionic conduction, and adaptive responsiveness into a single material platform (Figure 6). Their potential can be systematically understood by linking performance construction mechanisms, validation strategies, and application-specific advantages, thereby forming a coherent chain from material design to system-level deployment [144].

Mechanical compliance and conformability form the foundation for hydrogel applications, enabling intimate and continuous contact with complex, dynamic biological surfaces. Benefiting from their tunable crosslinking density, hydrated polymer networks can achieve modulus values comparable to those of skin or muscle [145]. In applications such as epidermal patches, joint motion sensors, and soft robotics, these attributes ensures both stability and comfort addressing the common limitations of traditional materials. High ionic conductivity is essential for reliable bioelectronic signal transmission [146]. The aqueous network within hydrogels facilitates efficient ion transport, which can be further enhanced by incorporating salts, zwitterionic polymers, or nanofillers such as MXene. Electrochemical impedance spectroscopy and electrophysiological tests confirm that hydrogels maintain low impedance and high signal fidelity even under sweating or motion, outperforming dry electrodes and pressure-sensitive films in demanding applications such as continuous ECG or EMG monitoring [46]. Permeability enhances hydrogel wearability by enabling selective exchange of gases and moisture, maintaining skin homeostasis and reducing irritation. By preventing moisture buildup and balancing hydration, hydrogels reduce skin maceration and inflammation, especially in applications like wound dressings and wearable sensors [146]. Their soft, porous structure improves conformity, reduces friction, and enhances user comfort [147]. Additionally, permeability ensures stable ionic conductivity and biocompatibility under physiological conditions (e.g., sweat, humidity), maintaining long-term functionality in wearable electronics [148].

Sustainability and biocompatibility are inherent strengths of many hydrogel systems, addressing both environmental and clinical imperatives. Hydrogels derived from renewable polymers such as cellulose and gelatin can degrade naturally

under physiological conditions, although the degradation rate varies depending on crosslinking density, polymer type, and the local enzymatic activity. For example, cellulose-based hydrogels typically degrade over weeks to months in wound dressings under physiological conditions, with enzymatic degradation by cellulases accelerating the process [149]. In contrast, gelatin-based hydrogels degrade much more quickly, typically within 7–14 days, due to their susceptibility to collagenase activity. Their biodegradation has been confirmed through enzymatic assays and life-cycle assessments, highlighting their value in disposable electronics and biodegradable implants [150]. Meanwhile, as validated by cytotoxicity and implantation studies, their high-water content and biomimetic chemistry minimize immune rejection. These properties align well with applications in disposable electronics and biodegradable implants, where environmental and clinical requirements converge.

Practical deployment in complex environments also demands resilience and usability. Self-healing properties, introduced via dynamic hydrogen bonds or reversible covalent chemistry, enable hydrogels to recover structural and functional integrity after mechanical rupture. Rheological recovery and fatigue threshold tests confirm this durability, which is vital for wearable sensors and electronic skins subjected to repeated bending [151]. In parallel, self-adhesion, achieved by incorporating catechol or cationic groups, allows hydrogels to attach securely to wet and moving tissues without auxiliary glues. Adhesion strength tests under dynamic conditions validate this feature, making hydrogels ideal for emergency ECG electrodes or wound-sealing patches where rapid and reliable deployment is crucial. Additional functionalities such as transparency and environmental adaptability broaden the operational landscape of hydrogels. Optical transmittance studies confirm their suitability for integration with imaging and phototherapy systems, enabling applications ranging from corneal patches to light-triggered drug release. Simultaneously, hydrogels reinforced with nanofillers or zwitterionic polymers retain stability under fluctuating humidity and temperature, as demonstrated by accelerated aging and thermal cycling tests [152]. These attributes provide unique advantages in outdoor health monitoring, rescue medicine, and extreme climate operation, where material reliability directly determines device survival.

Taken together, these interconnected properties form a coherent framework that advances hydrogel-based technologies. Mechanical compliance enables conformal integration with biological surfaces, while high ionic conductivity ensures faithful signal transmission across dynamic interfaces. Permeability contributes to extended wear comfort, and inherent sustainability combined with biocompatibility supports safe integration and environmentally benign operation. Self-healing mechanisms and spontaneous adhesion enhance durability and ease of use, whereas transparency and environmental adaptability open avenues for multifunctional applications. By addressing persistent challenges such as motion artifacts, skin irritation, mechanical failure, and environmental fluctuations, hydrogels provide integrated solutions beyond the capabilities of conventional interfacial materials. Thus, hydrogels should be regarded not as passive alternatives to adhesives or films, but as intelligent, multifunctional, and sustainable interfaces. Their unique combination of tunable properties, validated performance, and application-specific adaptability establishes them as a transformative material platform, offering robust support for next-generation health monitoring, implantable bioelectronics, and adaptive wearable systems.

4.3 | AI and Advanced Characterization for Performance Optimization

Performance optimization presents a critical, multifaceted challenge in developing hydrogel-based flexible electronics, requiring careful reconciliation of competing demands like softness versus robustness, hydration versus environmental stability, and high sensitivity versus long-term durability within an integrated platform [153, 154]. Traditional trial-and-error approaches often fall short in addressing this complex multi-parameter design space, which spans from molecular chemistry and network architecture to device-level functionality [155]. The convergence of artificial intelligence (AI), digital twin technology, and advanced in situ characterization now enables a transformative paradigm shift, advancing hydrogel electronics from experience-based exploration to model-driven precision engineering (Figure 7).

Artificial intelligence introduces a fundamentally new and efficient pathway for targeted performance enhancement. By leveraging large and multi-dimensional datasets interconnecting polymer chemistry, cross-linking strategies, and functional device metrics, machine learning algorithms, such as convolutional neural networks (CNNs) and recurrent neural networks (RNNs), can uncover hidden structure-property-function relationships inaccessible through intuition or conventional methods [156]. Inverse design frameworks enable researchers to set performance targets (e.g., ionic conductivity, tissue adhesion) and use generative models like generative adversarial networks (GANs) and variational autoencoders (VAEs) to propose new hydrogel formulations that meet these criteria, accelerating discovery for bioelectronic applications [157]. Reinforcement learning further optimizes hydrogel design by iteratively adjusting parameters, as demonstrated by deep reinforcement learning (DRL) in iontophoretic drug delivery systems [158]. For example, Yang et al. leveraged generative AI (including GPT-based and diffusion-based models) to design polymer electrolytes with improved ionic conductivity. Their work demonstrates how generative models can propose novel candidate materials that achieve significantly higher conductivity than those in the training set [159].

Digital twin technology extends data-driven performance optimization across the operational lifetime of hydrogel electronic devices by creating a dynamic virtual replica of the physical system that continuously integrates real-time sensor data with multi-physics simulations. A digital twin captures the evolving state of its physical counterpart including hydration levels, mechanical fatigue, ion transport, and other critical parameters, and enables predictive assessment of performance and failure modes, guiding proactive design modification rather than reactive troubleshooting [160]. Although direct digital twin studies of hydrogel bioelectronics are still emerging, analogous work in sensor-enabled digital twin systems shows how continuous data streams from flexible devices can be leveraged to simulate performance degradation (e.g., moisture loss or mechanical delamination), predict device aging, and recommend design adjustments before failure occurs. For instance, sensor-driven

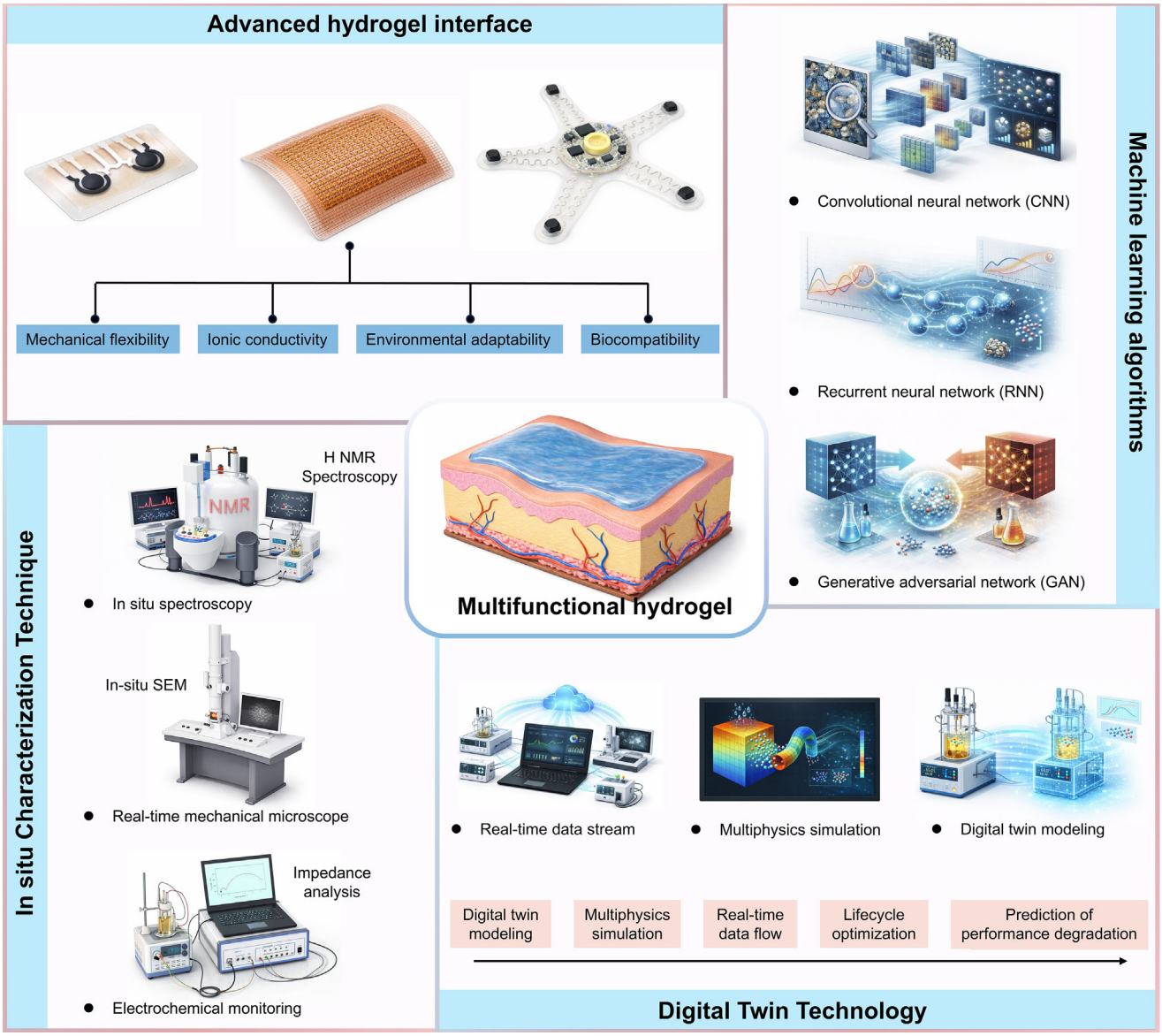


FIGURE 7 | Integration of multifunctional hydrogels with machine learning algorithms, digital twin technology, and in situ characterization techniques for advanced hydrogel interface optimization.

digital twin frameworks have been proposed to integrate wearable health device measurements with digital twin models for personalized healthcare delivery, demonstrating how real-time biosignal data can be fused with AI and simulation to anticipate changes and optimize device behavior [161]. Moreover, digital twin platforms facilitate context-specific adaptation and user customization by incorporating individual physiological and environmental data into device predictions, ensuring consistent performance across diverse conditions and users. This capability aligns with the broader vision of precision medicine digital twins, which are already being explored to model metabolic health, disease progression, and treatment responses at the individual level [162].

In situ characterization techniques complete the optimization loop by delivering real-time, high-resolution insight into hydrogel behavior under actual operating conditions. These methods capture dynamic processes such as molecular rearrangements, network deformation, and evolving ion-transport pathways that static tests cannot resolve. Techniques such as operando spectroscopy, live mechanical microscopy, and continuous electrochemical monitoring probe materials as they function, directly correlating structural changes with device performance. For example, in situ monitoring of hydrogel dehydration has revealed internal structural reorganization and changes in network cohesion, guiding material optimization [163]. This empirical feedback validates predictions from AI and digital twin models while revealing early indicators of degradation or impending failure. By correlating microstructural dynamics with macroscopic device performance, in situ analysis identifies critical governing parameters that dictate functional durability, thereby guiding targeted optimization in material composition, processing routes, and device integration strategies.

Collectively, AI, digital twins, and in situ characterization establish a closed-loop iterative framework for continuous

performance enhancement. AI proposes novel material designs with tailored properties; digital twins simulate their behavior and predict longevity within complex real-world environments; and in situ tools provide empirical verification and mechanistic understanding. Through this iterative cycle of computational design and experimental validation, hydrogel-based electronics development accelerates markedly, ultimately yielding systems that harmonize high signal fidelity, extended operational lifetime, and scalable manufacturability. It is foreseeable that the deep integration of these advanced technologies will accelerate the transition of hydrogel devices from laboratory prototypes to reliable, intelligent, and commercially viable electronic platforms.

5 | Hydrogel Interface in Close-Loop Flexible Electronics

The versatility of hydrogel interface has enabled applications across the entire body, from neural and oral diagnostics to cardiac monitoring, epidermal electronics, and self-powered skins, forming the basis of body-wide closed-loop healthcare systems [164]. To realize their full potential, however, these interfaces must be understood through a systematic framework: (i) sensing systems that demonstrate distributed physiological sensing; (ii) sensing mechanisms that couple mechanical, electrical, and biochemical stimuli to measurable outputs; (iii) performance benchmarks for close-loop electronics; and (iv) translational challenges including degradation, immune responses, and mechanical mismatch.

5.1 | Mechanistic Pathways and Application Frameworks

The advancement of hydrogel-based bioelectronics can be organized into four interrelated dimensions: multimodal transduction, structural engineering, material design, and intelligent data analytics.

Multimodal transduction underpins the functional versatility of hydrogel interfaces. Mechanical stimuli are commonly transduced via piezoelectric, piezoresistive, and triboelectric mechanisms, while thermoelectric and electrochemical modalities enable temperature sensing and biochemical monitoring (for example metabolites and glucose). Complementary optoelectronic and magnetic-response modalities extend detection to optical and magnetic signatures, thereby broadening applications from physiological surveillance to motion tracking and environmental sensing (Figure 8a).

Structural engineering imparts the mechanical adaptability required for reliable operation in dynamic biological environments. Architectures such as anisotropic lattices, porous and gradient networks, kirigami-derived patterns, and laminated constructs enhance compliance, strain sensitivity and multifunctionality, while also facilitating fluid and gas transport (Figure 8b). These design strategies are crucial for seamless integration with tissues subject to continuous deformation—including respiratory, articulatory and cardiac motions.

Material design must reconcile biocompatibility, controlled degradability and environmental tolerance. Hydrogels intended for prolonged contact or implantation should degrade in a predictable fashion without harmful residues, preserve breathability and wearer comfort, and, where feasible, employ sustainable constituents to mitigate environmental impact (Figure 8c). Such considerations are fundamental to both clinical translation and the development of green wearable technologies.

Advanced data processing algorithms play a pivotal role in interpreting complex, multimodal sensor outputs (Figure 8d). Deep learning techniques, such as convolutional neural networks (CNNs), recurrent neural networks (RNNs), and graph neural networks (GNNs), are employed to extract meaningful patterns from raw data, enabling real-time analysis and personalized feedback. These algorithms enhance the ability to interpret physiological signals, facilitating applications like emotion recognition, medical diagnostics, gait analysis, and motion detection. By integrating multimodal data from various sensors, hydrogel interfaces transform from simple sensing platforms into intelligent systems, capable of adaptive health monitoring and dynamic response based on real-time inputs (Figure 8f). The confluence of these four dimensions positions hydrogel bioelectronics as a promising platform for next-generation wearable and implantable systems.

5.2 | In-Depth Analysis of Sensing Mechanisms

Hydrogel-based bioelectronic utilize a range of sensing mechanisms, each suited to particular applications due to their unique characteristics. These systems operate on various physical principles to support multimodal sensing, real-time data collection, and intelligent feedback (Figure 9).

1) Capacitive sensing in hydrogel interfaces detects changes in capacitance resulting from variations in the electric field between electrodes, which are induced by alterations in the dielectric properties of the medium or changes in inter-electrode distance. The fundamental capacitance C is described by the Equation (6):

$$C = \frac{\varepsilon A}{d} \tag{6}$$

where ε is the dielectric constant, A is the area of the electrode, and d is the distance between the electrodes. The sensing response arises because deformation or interaction with a stimulus (e.g., strain, touch, proximity) alters the dielectric environment or electrode spacing, leading to a measurable change in capacitance. High ionic conductivity and a stable dielectric response are the basic prerequisites for hydrogel capacitive sensors. The hydrogel must permit efficient polarization under an alternating field and maintain contact with electrodes under mechanical deformation without significant signal drift. In practical wearable applications, these capacitive hydrogel sensors can detect subtle physiological signals (e.g., skin deformation, pulse wave) and interactive touch/proximity, owing to their low power consumption, high sensitivity to electrostatic perturbations, and mechanical compliance [165, 166]. The integration of hydrogels with ionic conductors (e.g., PVA/salts or nanofillers) ensures that even small dielectric changes translate into measurable capacitance shifts without invasive interfaces, making them suitable for

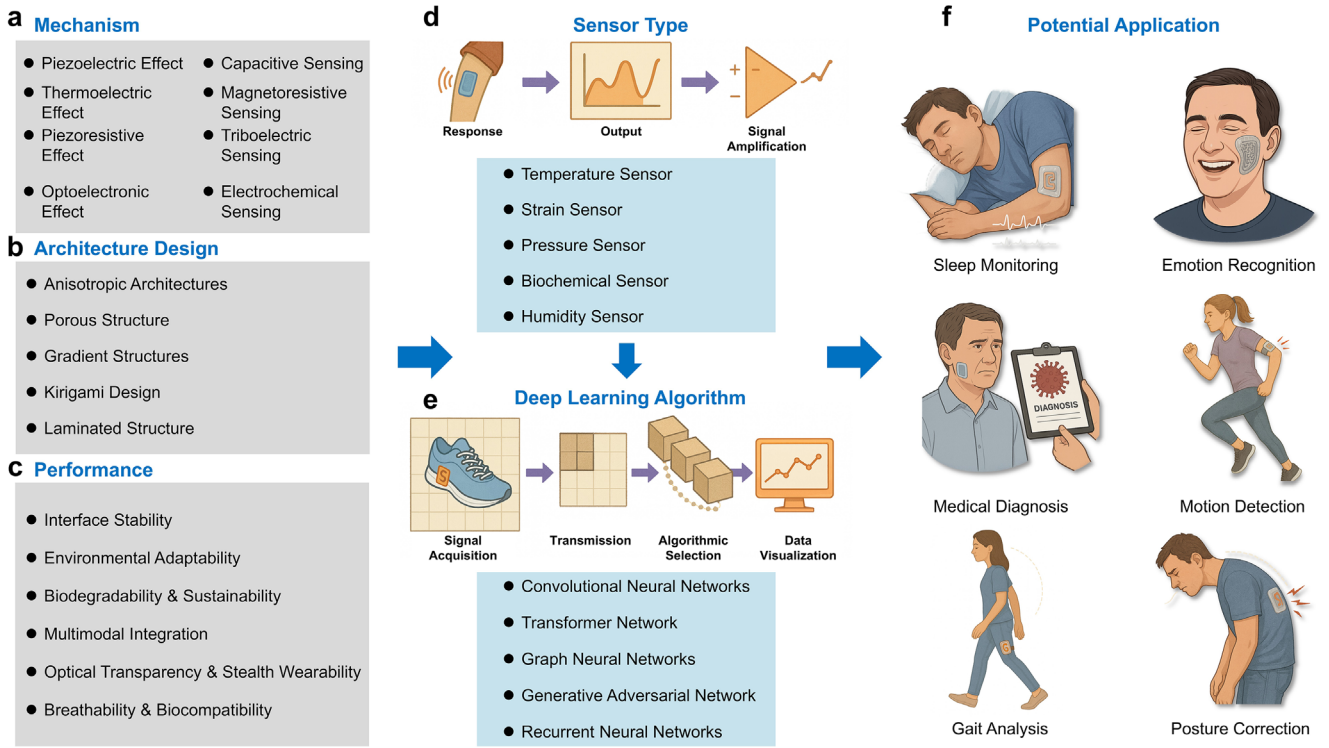


FIGURE 8 | Integrated framework of hydrogel-based intelligent sensing systems. (a) Representative sensing mechanisms for hydrogel-based electronics. (b) Structural design strategies enabling flexibility and adaptability. (c) Performance requirements ensuring stability, sustainability, and biocompatibility. (d) Sensor types targeting physical and biochemical signals. (e) Integration with deep learning algorithms for intelligent data processing. (f) Potential applications in health monitoring, diagnosis, and human–machine interaction.

human–machine interaction, interactive displays, and long-term personal health monitoring [167]. Water retention strategies (e.g., adding glycerol/LiCl) have also been used to minimize dehydration and sustain performance in real conditions [168].

2) Piezoelectric sensing relies on the piezoelectric effect, where mechanical stress induces electrical charge generation. The relationship between charge Q and applied stress σ is given by:

$$Q = d \cdot \sigma \tag{7}$$

where d is the piezoelectric coefficient. Piezoelectric sensors are highly sensitive to dynamic mechanical deformations such as force, pressure, and vibration. The key prerequisite for piezoelectric sensing is the hydrogel's ability to exhibit a significant piezoelectric effect upon mechanical deformation, often achieved through materials such as ZnO nanorods, graphene oxide, polyvinylidene fluoride (PVDF), and biomaterials embedded with piezoelectric nanoparticles. These materials can convert mechanical energy into electrical signals, enabling self-powered operation without the need for external batteries. A notable example is the use of PVDF/PVA hydrogels for posture correction and dynamic force sensing in healthcare and sports applications, where real-time feedback is essential for maintaining optimal posture or detecting early signs of injury [169, 170]. Thus, they have found broad application in motion detection, gait analysis, and posture correction, delivering highly responsive and real-time feedback.

3) Piezoresistive sensing relies on resistance changes in response to mechanical strain. The relationship between the change in resistance ΔR and strain ϵ is described by:

$$\Delta R = R_0 \cdot \left(\frac{\Delta L}{L_0}\right) \cdot \left(\frac{1}{1 + \alpha \cdot \varepsilon}\right) \tag{8}$$

where R_0 is the initial resistance, ΔL is the change in length due to strain, L_0 is the original length, and α is the piezoresistive coefficient. Piezoresistive sensors are highly sensitive to mechanical deformations, enabling them to detect small changes in strain in real-time. This makes them ideal for applications such as pressure sensing, force measurement, and continuous body movement monitoring. The key prerequisite for these sensors is the high sensitivity of the material's resistance to mechanical strain. Alginate-based hydrogels, for example, can undergo reversible deformation under mechanical stress while maintaining their ionic properties [171]. These materials have been successfully used in wearable sensors for force and pressure measurement, where continuous monitoring is required for applications like health tracking and motion detection [172, 173]. Moreover, their affordability and ease of fabrication also render them an appealing option for use in consumer electronics and smart textiles.

4) Triboelectric sensing is based on the contact electrification effect, where friction between materials generates electrical charge. The voltage V output follows:

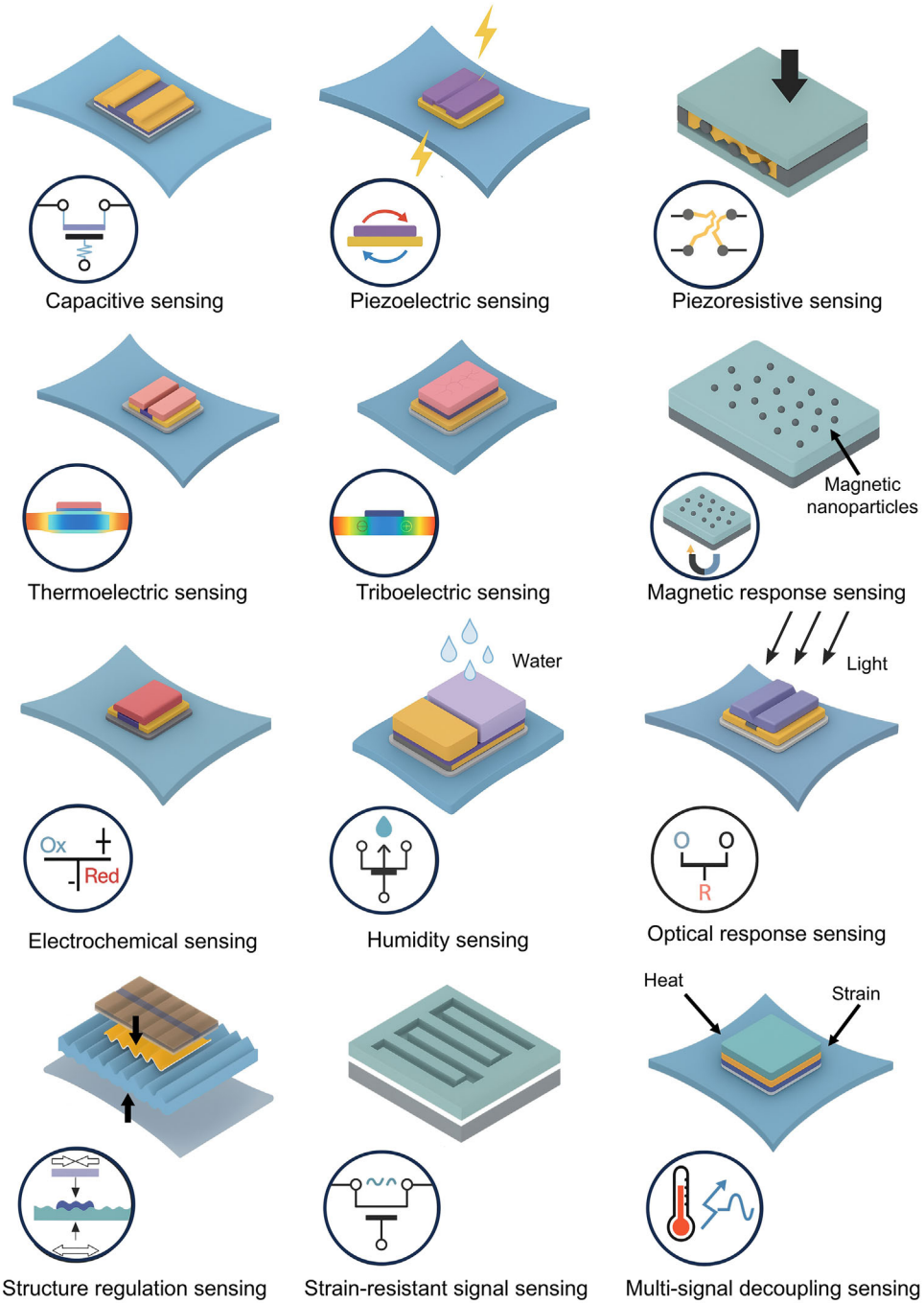


FIGURE 9 | The various mechanisms of hydrogel interfaces for sensing application.

$$V = \frac{Q}{C} = \frac{\eta \cdot A \cdot \Delta X}{\varepsilon} \tag{9}$$

where η is the triboelectric coefficient, A is the contact area, ΔX is the relative displacement, and ε is the permittivity of the material. Triboelectric sensors inherently transduce mechanical motion into electrical signals without external power sources, making them self-powered and highly suitable for autonomous wearable devices and energy harvesting systems. Their operation relies on repeated contact-separation or sliding cycles, which induce charge transfer and potential differences between triboelectric layers. The prerequisite for high performance is a combination of materials with large differences in triboelectric polarity and robust mechanical compliance to sustain repeated deformation without mechanical fatigue. For example, a hydrogel-based sandwich-structured TENG using PVA/carboxymethyl cellulose (CMC) hydrogel doped with ionic polyelectrolytes exhibited an open-circuit voltage up to ~330 V and a short-circuit current of ~44 μA, enabling high-sensitivity multifunctional sensing and mechanical energy harvesting for wearable applications [174]. Similarly, conductive hydrogels doped with boric acid and KCl have been used to fabricate flexible TENGs that combine high stretchability (>1500%), rapid response, and sensitive strain/pressure sensing, demonstrating the feasibility of triboelectric hydrogel sensors in human–machine interaction, motion tracking, and health monitoring [175].

5) Thermoelectric sensing leverages the Seebeck effect, where a temperature gradient across a material generates an electrical voltage, described by the Equation (10):

$$V = S \cdot \Delta T \tag{10}$$

where S is the Seebeck coefficient and ΔT is the temperature difference. Thermoelectric sensors are particularly well-suited for temperature monitoring and thermoregulatory applications, where their high real-time sensitivity to temperature changes enables non-invasive health monitoring and environmental sensing. These sensors are ideal for applications such as fever detection and smart textiles that regulate body temperature by continuously sensing the user's skin temperature. The Seebeck coefficient plays a critical role in determining the sensor's sensitivity, with materials exhibiting a high Seebeck coefficient providing greater voltage output for a given temperature difference. Hydrogels doped with conductive nanoparticles, such as graphene oxide or carbon nanotubes (CNTs), have been developed to improve the performance of thermoelectric sensors. For instance, graphene oxide (GO)-doped hydrogel thermoelectric sensors have been shown to achieve high thermoelectric performance, demonstrating high Seebeck coefficients and fast response times suitable for wearable body temperature monitoring [176]. These materials have been successfully used in smart clothing to monitor fever or hypothermia in real-time [177].

6) Electrochemical sensing detects ions or molecules through redox reactions at the electrode surface. The potential difference E follows the Nernst Equation (11):

$$E = E_0 + \frac{RT}{nF} ln \left(\frac{[C]_{out}}{[C]_{in}} \right) \tag{11}$$

where E_0 is the standard electrode potential, and $[C]_{\text{out}}$ and $[C]_{\text{in}}$ are the ion concentrations. Electrochemical sensors translate biochemical redox processes into measurable electrical signals typically current (amperometry), potential (potentiometry), or impedance (impedimetry), which enables high specificity and sensitivity for target ions or biomolecules. This makes them especially suitable for biosensing applications such as glucose monitoring, lactate detection, and sweat metabolite analysis in real time. A key requirement is the immobilization of selective recognition elements (e.g., enzymes like glucose oxidase or lactate oxidase, aptamers, or non-enzymatic catalytic surfaces) on a conductive interface that provides efficient electron transfer and stable chemical interaction with the analyte. Hydrogels are particularly advantageous as the sensing matrix because their hydrated, porous structure facilitates efficient analyte diffusion, bioreceptor immobilization, and mechanical conformability to skin or tissue, while maintaining intimate contact with the biofluid. For example, the hydrogel-integrated wearable sweat glucose sensor developed using a Prussian blue-doped PEDOT (poly(3,4-ethylenedioxythiophene)) composite electrode incorporated into a hydrogel patch. This device rapidly absorbs natural perspiration and enables continuous, non-invasive electrochemical detection of glucose during routine activities, demonstrating robust electrocatalytic activity and stable long-term performance for clinical-grade glucose management [178, 179].

7) Humidity sensing capitalizes on the hygroscopic swelling of hydrogels, where changes in moisture content cause the material to swell or shrink, which in turn affects its electrical resistance. The relationship between resistance and humidity H is given by:

$$R(H) = R_0 \cdot e^{\alpha H} \tag{12}$$

where $R(H)$ is the resistance at relative humidity H, and R_0 is the resistance at 0% humidity. Hydrogels with high water retention and swelling properties are ideal for humidity sensing, as they can undergo significant dimensional changes in response to even small variations in moisture levels, which directly affect their conductivity. An example is the development of wearable sensors for real-time sweat tracking based on PVA-PEG hydrogels, where the sensor's resistance changes with the amount of sweat evaporating from the body. These sensors provide critical insights into hydration status, making them invaluable for athletes and medical diagnostics to monitor and prevent dehydration during physical exertion [180].

8) Optical response sensing in hydrogels involves the measurement of changes in light absorption or reflectivity in response to external stimuli such as strain or temperature. This can be described by the Beer-Lambert law:

$$A = \varepsilon \cdot c \cdot l \tag{13}$$

where A is the absorbance, ε is the molar absorptivity, and l is the path length. Optical sensing enables non-contact monitoring, making it ideal for real-time biomolecular and temperature sensing, especially in situations where invasive sensors are impractical. The high sensitivity of optical sensors to material property changes (e.g., swelling from strain or temperature variations) facilitates non-invasive health diagnostics and environmental monitoring. For instance, graphene oxide (GO) or silver nanowire (AgNW)-integrated hydrogel sensors have been successfully applied to monitor temperature fluctuations and strain-induced optical changes. These materials demonstrate a significant shift in absorbance under mechanical deformation, making them suitable for wearable devices that track body temperature and biomolecular changes continuously [181, 182].

9) Magnetic sensing relies on magnetic fields interacting with magnetic nanoparticles embedded in the hydrogel. External fields induce particle rearrangement, altering magnetic permeability or inductive response. The induced voltage V is approximated by:

$$\upsilon = \mu \cdot H \cdot m \tag{14}$$

where μ is the magnetic permeability of the material, H is the magnetic field strength, m is the magnetic moment. Magnetic sensors enable remote detection without requiring direct contact. Their high sensitivity to subtle magnetic fluctuations allows precise monitoring of motion or positional changes, which is particularly valuable for biomarker tracking and prosthetic

applications. Importantly, magnetic sensing is non-invasive and compatible with both wearable and implantable devices, offering high spatial resolution for capturing localized physiological signals. For instance, the development of magnetic sensing hydrogel systems used for prosthetic limb control, where the sensor detects position or motion by sensing the magnetic fields generated by embedded magnetic nanoparticles in the prosthesis [183–185].

10) Strain-resistant sensing enables hydrogels to endure repetitive deformation and high strain while maintaining functional stability. This is accomplished through various design strategies such as serpentine and kirigami patterns that distribute stress and facilitate elastic recovery without cracking. Adaptive networks that respond to external stimuli, such as pH and temperature, ensure dynamic stability, and 3D mesh architectures provide structural integrity under extreme deformation. Together, these approaches enable hydrogels to recover after deformation, maintaining accurate sensing capabilities under real-world conditions, making them ideal for wearable sensors and biomedical applications [186].

11) Multi-signal sensing enables hydrogels to detect mechanical, electrical, chemical, and thermal signals simultaneously, enhancing their versatility in various applications. This is achieved by incorporating hybrid hydrogels that combine piezoelectric and capacitive materials for dual-mode sensing of mechanical and electrostatic signals. Multi-layered structures, with each layer responsive to different stimuli like pressure, temperature, or chemicals, allow for multi-dimensional detection, while modular designs integrate regions tailored to specific signals, further expanding the hydrogel's multi-parameter sensing capabilities [187].

Hydrogel interfaces leverage diverse sensing mechanisms, such as capacitive, piezoelectric, and triboelectric, to address the growing demands of healthcare, wearables, and environmental monitoring. However, there are inherent trade-offs between sensitivity, signal interference, and miniaturization. Higher sensitivity enables the detection of subtle signals but also amplifies interference, particularly in miniaturized systems where space limitations exacerbate the issue. Optimizing performance requires a careful balance between sensitivity and interference, while also considering the impact of miniaturization on overall functionality. Additionally, these sensors are challenged by environmental factors such as drying, freezing, temperature fluctuations, and chemical degradation, which can impair their mechanical properties, ionic conductivity, and dimensional stability [188, 189]. To address these challenges, material innovations like encapsulation, self-healing polymers, and chemical-resistant coatings are crucial for enhancing the long-term durability of hydrogel interfaces [190, 191]. The next step is integrating these optimized hydrogel platforms with machine learning algorithms, enabling adaptive, real-time feedback. This integration promises personalized monitoring systems, improving precision, biocompatibility, and resilience, and positioning hydrogel interfaces as a core technology for next-generation bioelectronics and sustainable healthcare applications.

5.3 | Body-Wide Closed-Loop Medical Monitoring

Hydrogel interfaces have emerged as transformative platforms for achieving full-body, closed-loop medical monitoring. Unlike rigid conventional sensors, hydrogels can seamlessly conform to the skin, mucosa, or even internal tissues, enabling long-term, non-invasive monitoring of physiological signals while maintaining comfort and stability. The integration of hydrogel electronics into various body regions establishes a distributed sensing network, bridging multiple organ systems and paving the way for personalized healthcare and closed-loop therapeutics (Figure 10).

One of the most prominent applications is in neurological and cognitive monitoring, where hydrogel-based EEG electrodes and magnetoelastic fatigue sensors achieve high-fidelity signal acquisition without the discomfort of traditional metal contacts. Their hydration-mediated ionic conductivity allows for stable neural interfacing, enabling real-time brain–computer interaction, sleep monitoring, and emotion recognition, while reducing motion artifacts. Similarly, ear-integrated sensors leverage the hydrogel's conformability to monitor intraocular and auricular pressure, lactate levels, or even auditory-driven neural activity, highlighting their capacity to integrate into non-traditional anatomical niches.

In the oral cavity, hydrogel-based nanosensors enable highly sensitive quantification of salivary metabolites and pathogens, providing a non-invasive diagnostic avenue for systemic conditions including diabetes and infectious diseases [195, 201]. On the skin surface, epidermal electronic membranes composed of conductive hydrogels facilitate continuous electrocardiogram (ECG) and respiratory monitoring, while electronic tattoo sensors capture muscle activity via electromyography (EMG) [199, 203]. These devices achieve seamless and conformal integration with biological tissues to ensure long-term biocompatibility and support real-time wireless data transmission, thereby enhancing the efficacy of integrated digital health systems [192].

Furthermore, hydrogels are playing an increasingly central role in cardiovascular monitoring and rehabilitation. Integrated smart vascular stents incorporating hydrogels enable real-time surveillance of blood flow and restenosis, thereby reducing the risk of potentially fatal complications. Similarly, implantable or skin-adherent hydrogel-based sensors allow for continuous neural activity monitoring and precise neuromodulation, supporting closed-loop therapeutic interventions for chronic neurological disorders such as Parkinson's disease and epilepsy [193]. Meanwhile, wearable deep-tissue ultrasound patches employing hydrogel-coupled layers enhance acoustic transmission, enabling continuous imaging of cardiac, breast, and abdominal tissues in dynamic settings [196–198]. Together, these advances signify a paradigm shift in medical diagnostics from intermittent clinical assessments toward continuous, patient-centered monitoring.

Hydrogel interfaces also demonstrate significant potential for tracking metabolic and biochemical biomarkers in sweat and interstitial fluid [194]. For instance, hydrogel-integrated sweat-sensing wristbands allow accurate quantification of electrolytes and metabolites. Biofuel cell-based hydrogel "skin" can directly harness biochemical energy from bodily fluids to self-power

FIGURE 10 | Legend on next page.

monitoring devices, highlighting the dual functionality of hydrogels in sensing and energy harvesting [202]. This capacity for operation independent of external power sources is further exemplified by triboelectric generators integrated with hydrogel films, which convert mechanical motion from the chest or limbs into electrical energy, thereby advancing self-sustained biosensing platforms. The exceptional flexibility, transparency, ionic conductivity, and environmental sustainability of hydrogels make them ideal materials for human–machine interfaces. For example, adhesive robotic skins and smart gloves embedded with hydrogel sensors enable precise motion capture, tactile feedback, and bidirectional communication, effectively bridging biological and robotic systems [200]. These developments not only expand hydrogel applications in healthcare but also promote their potential in soft robotics, rehabilitation engineering, and advanced prosthetics, underscoring a promising future for improved medical treatment and daily living.

5.4 | Reliability Standards and Clinical Translation Barriers

Despite rapid advances in hydrogel design, practical deployment in biomedical and electronic applications remains hindered by multiple failure modes at the hydrogel–tissue interface. These challenges arise from the interplay of material degradation, biological responses, and mechanical or anatomical constraints, each of which undermines long-term stability and functional reliability. Material degradation is a primary concern, as hydrogel breakdown releases small molecular by-products that compromise interface stability and may trigger local irritation or inflammatory cascades (Figure 11a). Over time, these degradation processes weaken adhesion and shorten device lifespan, particularly for biodegradable systems intended for transient electronics. Bacterial colonization further accelerates interface failure (Figure 11b). Hydrogel surfaces are prone to microbial adhesion, and biofilm formation not only disrupts interfacial bonding but also elevates infection risks. Microbial colonization remains a major cause of device failure. Without antifouling strategies, biofilms can form rapidly, leading to detachment and rejection. Host immune responses pose a similar risk (Figure 11c). Fibrosis and immune cell infiltration often encapsulate hydrogel interfaces in dense scar tissue, which reduces signal fidelity and limits biochemical exchange. Such reactions compromise integration with host tissue and undermine long-term stability. Mechanical mismatch adds further complexity (Figure 11d). Rigid adhesives tend to delaminate under continuous motion, while overly soft gels lack structural integrity. These observations underscore the critical importance of developing hydrogel adhesives with modulus-matched architectures that minimize stress concentrations at the interface, while incorporating fatigue-resistant networks capable of sustaining repeated deformation and dynamic loading over prolonged implantation.

In addition to these intrinsic issues, the deployment of hydrogels is further constrained by anatomical barriers in irregular or defective tissue sites, where poor conformability hinders uniform coverage (Figure 11e). Even after successful application, hydration-induced swelling stresses can enlarge hydrogels, compress surrounding tissues, and generate interfacial strain that compromises adhesion stability (Figure 11f). Fluid interference presents another critical issue about blood, lymph, and interstitial fluids that often invades the interface, disrupting bonding and diluting adhesive components (Figure 11g). This is particularly problematic in wet or hemorrhaging environments, where maintaining strong adhesion is essential. Furthermore, motion instability caused by continuous physiological dynamics such as cardiac contraction, respiratory movement, or joint flexion subjects the interface to cyclic shear and tensile stresses, progressively weakening hydrogel adhesion (Figure 11h). Without adaptive or self-healing properties, motion-induced instability constitutes a major cause of interfacial failure. Collectively, these eight challenges highlight the multifactorial complexity of sustaining stable hydrogel–tissue interfaces. Addressing them will require systemic strategies, including antifouling and biofilm-resistant coatings, immunomodulatory chemistries, modulus-matched and swelling-resistant architectures, fluid-tolerant adhesives, and motion-adaptive retention designs. Such integrated approaches are essential for advancing hydrogel interfaces from experimental prototypes to reliable long-term bioelectronic platforms.

5.5 | Systemic Energy-Signal Coupling Performance Evaluation

The integration of multimodal sensing, self-sustained power management, and closed-loop feedback in modern devices has made the traditional evaluation of isolated parameters increasingly inadequate. For instance, performance metrics such as energy conversion efficiency or signal quality have proven insufficient, as they fail to account for the interactions between energy management and signal processing. To bridge this gap, a preliminary concept known as the energy-signal coupling coefficient (ESCC) is defined. This dimensionless parameter provides an

FIGURE 10 | Scheme of hydrogel-based bioelectronics for physiological monitoring, therapeutic delivery, and human–machine interaction. Magnetoelastic fatigue sensor. Reproduced with permission [192]. Copyright 2025, Springer Nature; Ear-eeg sleep patch. Reproduced with permission [193]. Copyright 2019, Springer Nature; Ear-eeg lactate duo. Reproduced with permission [194]. Copyright 2023, Springer Nature; Wireless orthodontic iontronic senso. Reproduced with permission [195]. Copyright 2025, American Association for the Advancement of Science (AAAS); Wearable deep-tissue ultrasound. Reproduced with permission [196]. Copyright 2024, Springer Nature; Breast ultrasound patch. Reproduced with permission [197]. Copyright 2023, American Association for the Advancement of Science (AAAS); Single-transducer echopatch. Reproduced with permission [198]. Copyright 2024, Springer Nature; Non-contact skin-flux patch. Reproduced with permission [199]. Copyright 2025, National Academy of Sciences; Adhesive robotic skin. Reproduced with permission [200]. Copyright 2025, American Association for the Advancement of Science (AAAS); Diabetes cardio-metabolic monitoring. Reproduced with permission [201]. Copyright 2025, Springer Nature; Biofuel electronic skin. Reproduced with permission [202]. Copyright 2020, American Association for the Advancement of Science (AAAS); EMG tendon patch. Reproduced with permission [203]. Copyright 2023, Springer Nature.

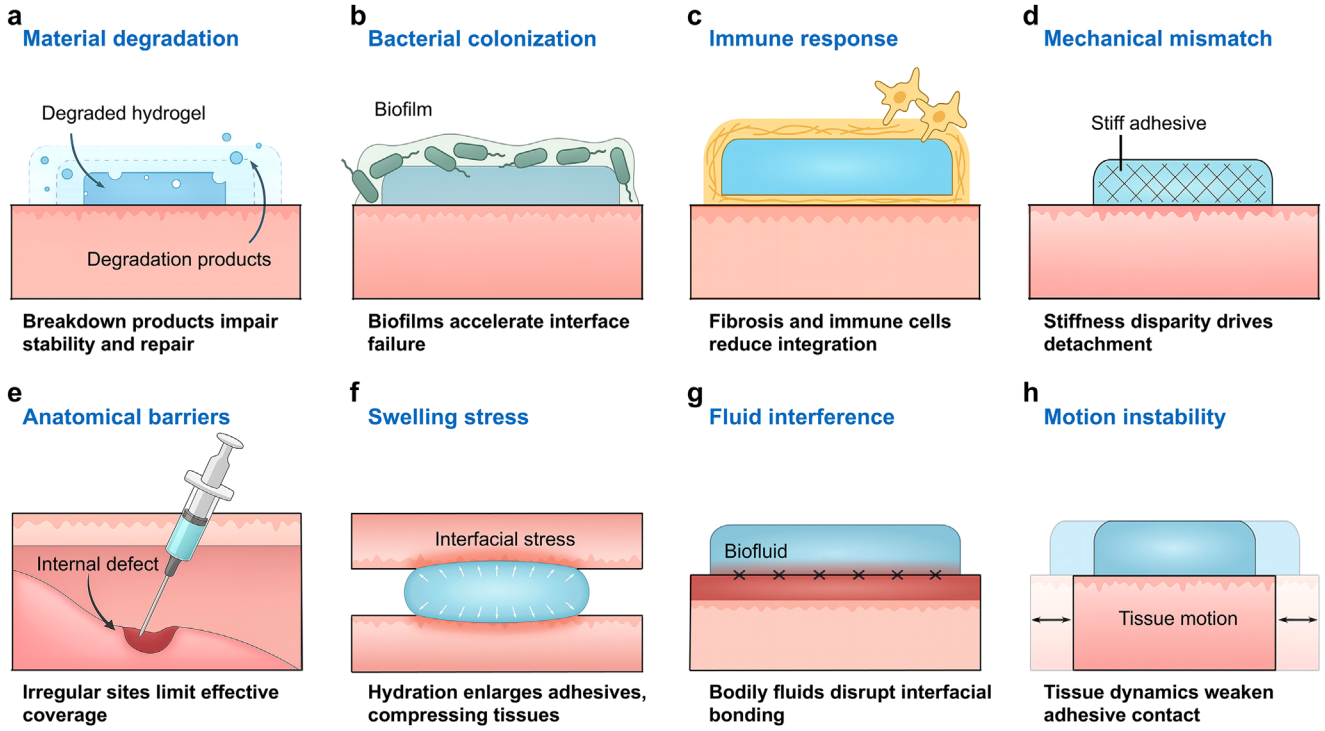


FIGURE 11 | Key challenges to the stability of hydrogel–tissue interfaces. (a) Material degradation impairs long-term adhesion. (b) Bacterial colonization and biofilm formation accelerating interface failure. (c) Immune responses inducing fibrosis and encapsulation that hinder interfacial coupling. (d) Mechanical mismatch induces stress concentrations and promotes detachment. (e) Anatomical barriers restrict conformal coverage at irregular or confined sites. (f) Swelling stress from hydration enlarges adhesives and compresses tissues. (g) Fluid interference disrupts interfacial bonding in wet or bleeding environments. (h) Motion instability from cyclic tissue dynamics progressively weakens adhesion.

integrated perspective on how hydrogels convert, store, and transmit energy, while simultaneously regulating signal acquisition and transduction under dynamic operating conditions. The ESCC framework establishes a unified benchmark that links energy flow and signal fidelity, offering a systematic approach to optimizing system design through the evaluation of the interdependence between energy and signal processes.

The ESCC is a comprehensive metric that captures the correlation between energy input, storage, dissipation, and the fidelity of signal acquisition and transduction. Unlike conventional single-property metrics, the ESCC integrates mechanical, electrical, and biochemical responses, thus providing a holistic assessment of multifunctional system performance. While this concept remains in its early stages, its potential to bridge gaps in traditional performance evaluations is clear. For example, a hydrogel electrode with high ionic conductivity but poor mechanical resilience would suffer from signal distortion under cyclic strain, yielding a low ESCC. In contrast, systems capable of efficient energy harvesting (via piezoelectric or triboelectric mechanisms) while maintaining stable signal fidelity during dynamic operation would achieve higher ESCC values. By mapping device architectures and material compositions against the ESCC, it is possible to systematically identify trade-offs and guide multifunctional optimization.

The ESCC framework consists of six key stages that are interconnected and essential for ensuring high-performance operation (Figure 12). These stages include: (1) energy input (E_{input}), which focuses on converting external energy into usable electrical energy; (2) energy storage (E_{storage}), the efficiency with which harvested energy is stored for later use; (3) signal response (S_{response}), which determines how the system reacts to external stimuli and generates appropriate electrical signals; (4) signal transduction ($S_{\text{transduction}}$), ensuring signal fidelity and minimal noise during transmission; (5) energy output ($E_{\text{dissipation}}$), which accounts for the energy loss occurring during the overall closed-loop operation, including losses during energy conversion, transmission, and signal processing. This stage is crucial for ensuring sustained operation and minimizing system inefficiency. (6) lastly, personalized functional flex electronics, which integrates all prior stages into a modular, adaptable system tailored to specific user needs. These stages are mutually dependent, and the performance of one directly impacts the others, emphasizing the importance of optimizing the entire system rather than isolated components. The ESCC is calculated using the following formula:

$$ESCC = \frac{S_{response} \cdot S_{transduction} \cdot E_{storage}}{E_{input} \cdot E_{dissipation}} \tag{15}$$

The optimization of ESCC offers substantial benefits for flexible electronics design. The interdependencies between energy input, storage, signal response, and output require a holistic approach to optimize system performance. For example, by enhancing energy harvesting efficiency (e.g., through piezoelectric or triboelectric mechanisms), improving signal fidelity (via advanced material choices and interface design), optimizing energy storage capacity (using high-performance capacitors or batteries), and minimizing

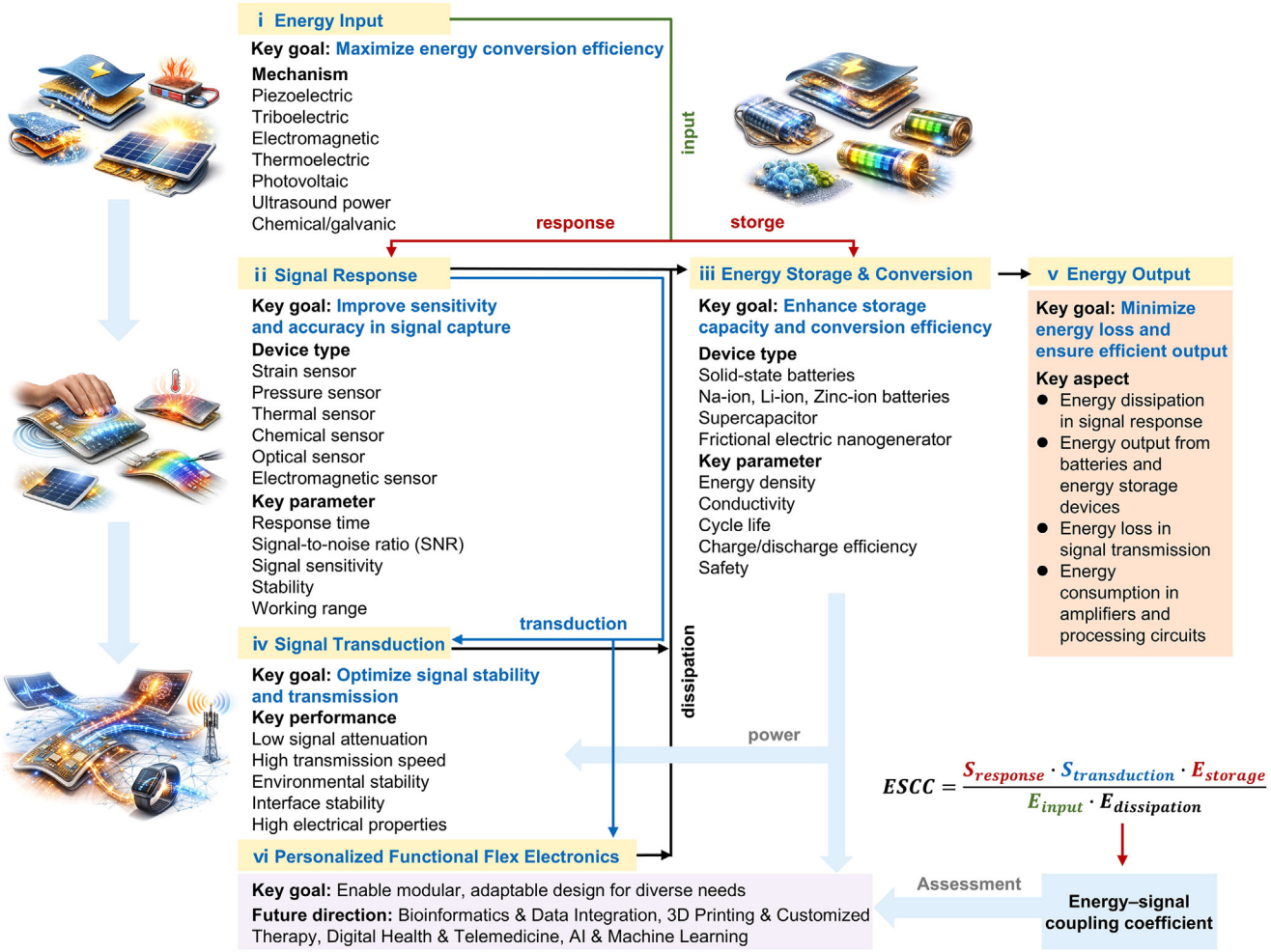


FIGURE 12 | Energy-signal coupling coefficient for close-loop flexible electronics applications.

energy dissipation (through efficient energy conversion and transmission paths), the ESCC can be maximized.

Recent research underscores how hydrogel-based flexible electronics align with the ESCC framework by optimizing both energy management and signal processing in integrated systems. Liu et al. developed mechanically compliant, impedance-matched hydrogel bioelectronics that interface with peripheral nerves at low voltage and high fidelity (Figure 13a). This demonstrates how optimizing both mechanical and electrical properties enhances signal transduction and energy efficiency in implantable systems, a key aspect of ESCC's signal response and signal transduction stages. Yu et al. introduced a breathable haptic textile that integrates flexible sensing and comfort features for extended wear. This multimodal textile platform ensures stable signal fidelity in dynamic environments, exemplifying ESCC's focus on maintaining signal integrity in real-world conditions (Figure 13b).

In the field of wearable healthcare electronics, Gao et al. developed a wearable echomyography system with a single transducer for deep-tissue muscle monitoring. This system combines high-quality signal acquisition with wireless processing and machine learning, aligning perfectly with ESCC's signal response and signal transduction metrics by enabling real-time interpretation of physiological signals (Figure 13c). Chen et al. developed a deep learning-enhanced wearable sensor system that integrates inertial measurement units (IMUs) and electromyography (EMG) to maintain signal fidelity despite motion noise (Figure 13d). This system aligns with the ESCC framework, as it optimizes both energy management (through efficient signal processing) and signal fidelity in dynamic environments, crucial for closed-loop wearable health monitors. By balancing signal response and energy efficiency, it exemplifies how ESCC can guide the design of adaptive, high-performance bioelectronic systems. Tian et al. advanced closed-loop wearable systems with a wearable obstructive sleep apnea management device (Figure 13e). The system integrates flexible piezoelectric monitoring with soft magnetoelastic actuation and real-time machine learning feedback, demonstrating how energy input, signal response, and feedback integration are key to adaptive healthcare devices, in line with ESCC's goal of dynamic system optimization.

From the energy storage perspective, Fu et al. designed paintable biogel electrolytes with dynamic imine chemistry for on-body Zn-ion batteries (Figure 13f). These batteries exhibit ultrafast self-healing and stable cycling under deformation, ensuring efficient energy storage and signal support for wearable systems, thus optimizing ESCC performance in dynamic, real-world applications. By optimizing the ESCC across these six stages, we achieve a system that balances energy efficiency with signal fidelity,

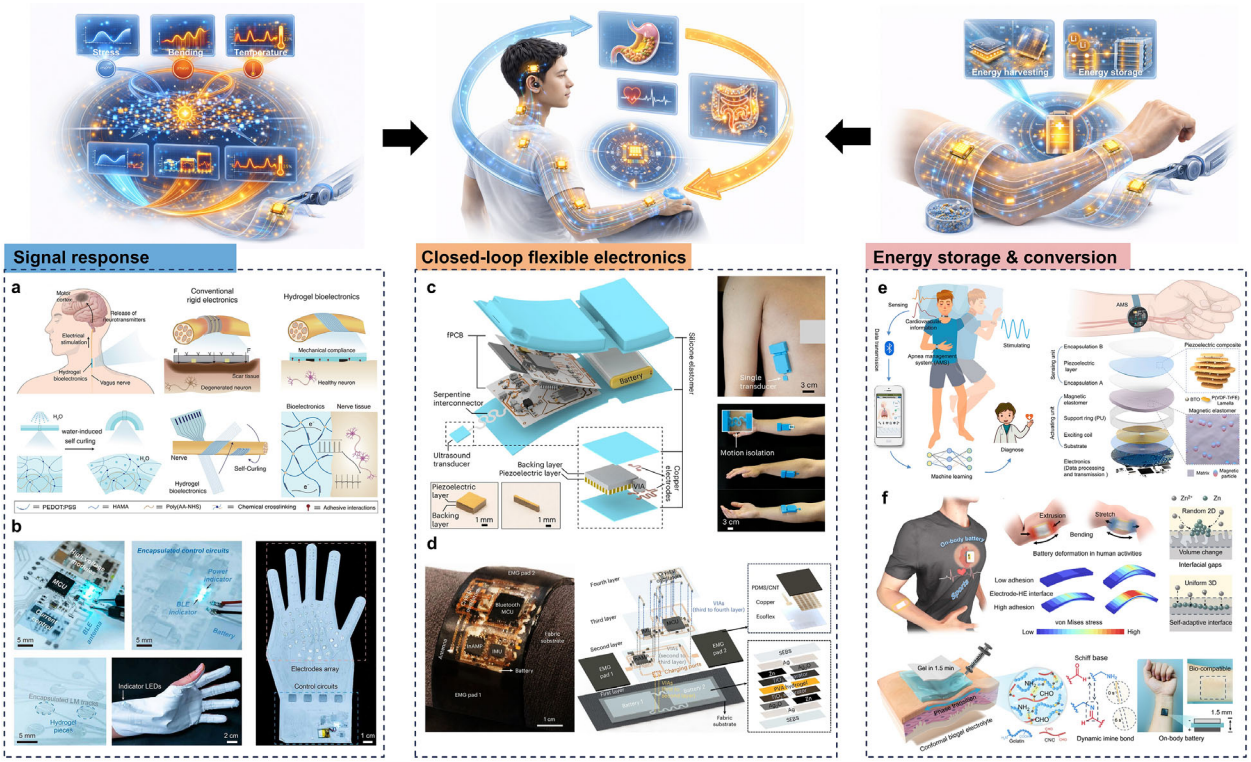


FIGURE 13 | Advanced hydrogel flexible electronics. (a) Hydrogel-based sensors for real-time monitoring of biological signals, ensuring high signal fidelity in wearable health devices. Reproduced with permission [204]. Copyright 2025, Wiley-VCH. (b) Integration of hydrogel electrodes in a wearable hand sensor, combining multimodal sensing and feedback mechanisms for adaptive health monitoring. Reproduced with permission [205]. Copyright 2024, AAAS (American Association for the Advancement of Science). (c) Piezoelectric and triboelectric hydrogel systems for energy harvesting, powering self-sustained wearable medical devices. Reproduced with permission [198]. Copyright 2024, Springer Nature. (d) Wearable bioelectronics with flexible hydrogel sensors for continuous physiological monitoring, optimizing both energy efficiency and signal fidelity. Reproduced with permission [206]. Copyright 2025, Springer Nature. (e) Eco-friendly hydrogel systems combining biosensing and energy storage, advancing sustainable wearable technologies. Reproduced with permission [207]. Copyright 2025, Cell Press. (f) Personalized wearable device with hydrogel-based electronics that adapt to individual physiological data for real-time health monitoring. Reproduced with permission [14]. Copyright 2025, American Chemical Society.

enhancing both performance and sustainability. This optimization is essential for closed-loop flexible electronics, which must operate in real-time, dynamic environments, such as wearable health monitors and bioelectronics.

6 | Perspectives and Outlook

The integration of multifunctional hydrogels with flexible electronics has established a transformative platform for next-generation bio-interfaces, effectively bridging biological systems and intelligent devices. From fundamental advances in chemical modifications, nanocomposite reinforcement, and hierarchical structural engineering, to the demonstration of multimodal sensing, energy harvesting, and interactive biomedical applications, hydrogel interfaces have rapidly evolved from material prototypes into versatile enablers of personalized healthcare and human–machine symbiosis. Yet, as the field transitions toward system-level intelligence and sustainable deployment, its trajectory is being shaped by a set of converging frontiers (Figure 14).

1. AI-driven inverse design and digital twins are already being actively implemented in the development of hydrogel electronics, transforming material discovery and device optimization. By harnessing large-scale data sets, generative design algorithms, and Bayesian-optimized learning techniques, hydrogels can be designed with a high level of predictive accuracy, optimizing properties across multiple dimensions, such as chemistry, network architecture, and functional performance. The integration of these tools with real-time digital twin platforms enables dynamic, real-time simulations of material behavior, offering the ability to continuously adapt and refine hydrogel designs based on operational feedback. This approach will drive autonomous lifecycle management, predictive maintenance, and performance optimization, enabling materials that respond intelligently to changing conditions in real-world applications.
2. Cross-scale operando and in situ characterization represents another critical frontier. By establishing quantitative causal chains linking external stimuli, structural reconfiguration, and functional output, multi-modal spectroscopies and simulations can provide predictive system health indicators. This shift from descriptive observation to mechanistic mapping will underpin reliability engineering, allowing hydrogel electronics to transition from laboratory prototypes to clinically and industrially deployable technologies.
3. Sustainability and circularity will become indispensable design principles. Future hydrogel electronics must embed life-cycle assessment (LCA) into R&D pipelines, prioritizing

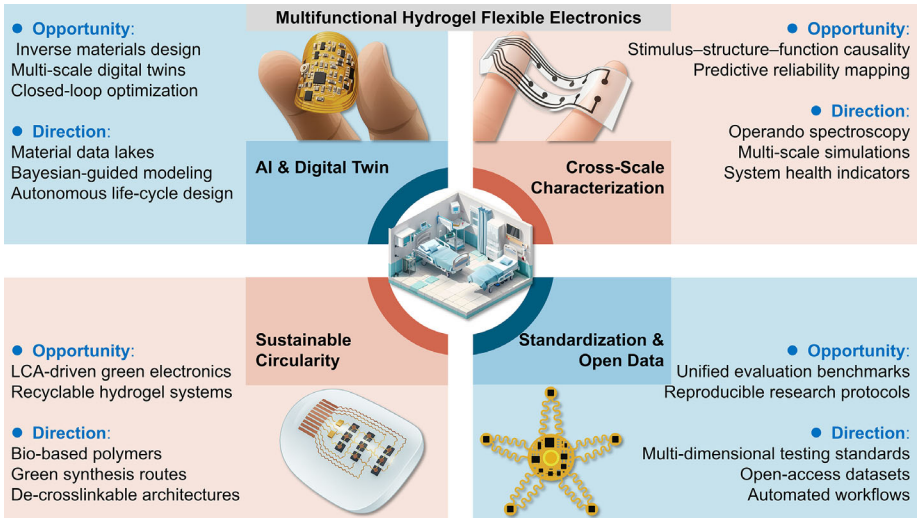


FIGURE 14 | Future opportunities and directions for multifunctional hydrogel flexible electronics.

bio-based polymers, green synthesis routes, and degradable or de-crosslinkable architectures. At the system level, recyclable device designs and self-consistent energy strategies will enable closed-loop circular electronics aligned with global carbon neutrality and green transition imperatives.

4. Standardization and open data infrastructures are equally vital to ensure reproducibility, interoperability, and accelerated translation. Unified benchmarks across sensitivity, durability, biocompatibility, and sustainability, together with open-access datasets and automated workflows, will reduce fragmentation, facilitate cross-disciplinary collaboration, and harmonize research outcomes with regulatory and clinical frameworks.

Despite these opportunities, key challenges remain, including reconciling conflicting material properties (softness vs. robustness, hydration vs. stability), extending device lifetimes in extreme environments, ensuring manufacturing consistency, and integrating AI with multi-omics and multimodal sensing to achieve autonomous system-level intelligence. Addressing these grand challenges requires dismantling disciplinary silos and fostering full-stack co-development across materials science, computational modeling, electronics engineering, and biomedicine.

Looking ahead, multifunctional hydrogel electronics will not merely serve as flexible substrates or transient prototypes. Instead, they will emerge as indispensable technological foundations for personalized healthcare, environmental intelligence, and a sustainable digital society. By coupling adaptive material design, cross-scale characterization, sustainable engineering, and standardized evaluation, the field is poised to transition from exploratory research toward a mature ecosystem that empowers both scientific discovery and societal impact.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The authors have nothing to report.

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