Yin *et al. Soft Sci.* **2025**, *5*, 30
DOI: 10.20517/ss.2025.15

Soft Science

Perspective

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Tough gelatin-based biogels for wearable sensors

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How to cite this article: Yin, J. J.; Li, Y.; Sun, X.; Qin, Z. H. Tough gelatin-based biogels for wearable sensors. *Soft Sci.* **2025**, *5*, 30. https://dx.doi.org/10.20517/ss.2025.15

Received: 25 Apr 2025 **First Decision:** 27 May 2025 **Revised:** 11 Jun 2025 **Accepted:** 16 Jun 2025 **Published:** 27 Jun 2025

Academic Editor: YongAn Huang **Copy Editor:** Shu-Yuan Duan **Production Editor:** Shu-Yuan Duan

Abstract

Flexible wearable sensors that can intimately adhere to the human body for real-time monitoring of human activities and physiological signals have attracted great attention owing to their potential in personalized healthcare and human-machine interfaces. Gelatin-based biogels are promising materials in wearable sensors due to their good biocompatibility, biodegradability, and sustainability. However, conventional gelatin-based biogels are usually weak and brittle (tensile strength < 10 kPa and stretchability < 50%), and thus cannot be applied in flexible wearable devices. Therefore, further efforts are needed to engineer tough gelatin-based biogels that meet the demands of flexible wearable sensors. In this perspective, we summarize recent progress in designing tough gelatin-based biogels and their wide applications in wearable sensing devices, while highlighting potential future directions in this field.

Keywords: Gelatin, biogel, stretchability, mechanical toughness, wearable sensor

INTRODUCTION

Flexible wearable sensors have undergone rapid development in recent decades due to their significant role

in personalized healthcare, precision medicine, and human-machine interfaces[1-3]. However, different from inherently soft human tissues, conventional wearable devices are usually made with rigid materials, causing unstable integration with tissues and discomfort[4]. Polymer gels, composed of three-dimensional polymer networks and dispersion media, have brought many opportunities to wearable sensors by harnessing their mechanical flexibility, structural permeability, and tissue resemblance[5,6]. However, general synthetic polymer gels have poor biocompatibility and non-degradability, leading to severe immune response and a serious threat to environmental sustainability[7].

Biogels derived from natural biopolymers (such as alginate, cellulose, and gelatin) are gaining research interest as building blocks for wearable sensors, ascribing to their biocompatibility and environmental sustainability[8-11]. The biogels' biocompatibility enables long-term skin contact without irritation or inflammation, while their environmental degradability presents a sustainable approach to mitigating electronic waste. Gelatin, as a hydrolyzed product of natural collagen, is a type of protein composed of polypeptides. Collagen is generally water-insoluble, composed of three long helix-shaped chains of amino acids [Figure 1A][12]. Compared to collagen, gelatin is a mixture of single- and multi-chain polypeptides composed of amino acids such as alanine, proline, and 4-hydroxyproline with good water solubility. The unique advantages of gelatin, including excellent biocompatibility, non-immunogenicity, biodegradability, and multiple reactive sites for functionalization, make it a promising candidate for constructing soft biogels as wearable sensors [Figure 1B][13,14]. Although gelatin-based hydrogels can be formed by cooling a hot gelatin solution to convert partial gelatin chains from random coil structures to triple-helix configuration, the obtained hydrogel is fragile with low toughness, which makes it difficult to apply in flexible sensing devices[12,15].

DESIGN OF TOUGH GELATIN-BASED BIOGELS FOR WEARABLE SENSING DEVICES

A general design principle for tough gels is the incorporation of energy dissipation domains within stretchable polymer networks[16]. The molecular engineering and structural engineering, including molecular interactions and topological network structures, are proposed to fabricate robust gels with high stretchability, mechanical strength, and toughness[17]. By manipulating functional groups at the molecular level, diverse crosslinking interactions (including permanent or reversible dynamic bonds) can be engineered within the polymer network for energy dissipation. Structural engineering mainly involves introducing high-order structure (such as phase separation and hierarchical structure) into polymer networks for strengthening and toughening gels. Various combinations of these effective strategies have been explored for designing tough gelatin-based biogels[18]. Considering the abundant functional groups in gelatin chains, an effective strategy for toughening gelatin biogels is to regulate non-covalent associations between gelatin chains[19-21]. For example, hydrophobic interaction domains and microphase separation regions were introduced into the gelatin network through the Hofmeister effect by soaking virgin gelatin gel in a $(NH_4)_2SO_4$ solution [Figure 1C][19]. The resulting gelatin hydrogels could withstand large tensile and compressive forces with tensile strength of ca. 3 MPa when stretching to 500% [Figure 1D]. Besides, introducing crosslinkers such as tannic acid (TA), sodium citrate, and sodium phytate to build non-covalent crosslinks with gelatin chains is another effective way to enhance the mechanical properties of gelatin-based biogels[22-25]. For example, Qin *et al.* reported a gelatin supramolecular organohydrogel by immersing a gelatin pre-hydrogel in citrate (Cit) water-glycerol mixture solution [Figure 1E][22]. Due to the formation of hydrophobic aggregation, ionic interactions between the $-NH_3^+$ of gelatin and Cit^{3-} anions, the organohydrogel exhibited high strength and toughness [Figure 1F]. Additionally, chemical modifications of gelatin chains with crosslinkable groups (such as methacrylate and dopamine) have also been investigated to create dually crosslinked structures for improving the mechanical properties of gelatin-based biogels[26,27]. With significant advances in mechanical enhancement, compared with the synthetic polymer gels (such as

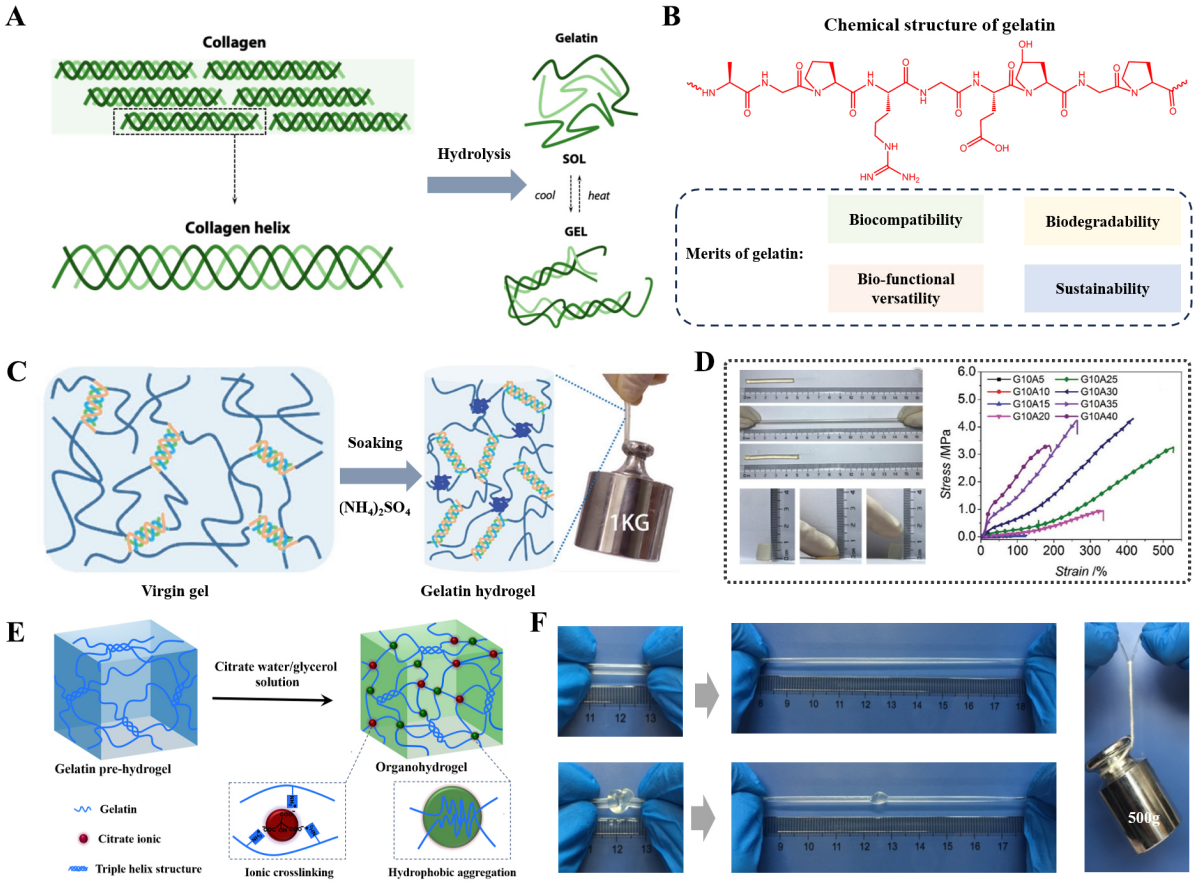

Figure 1. (A) Schematic diagram of processing collagen to gelatin and the mechanism of sol-gel transition of gelatin hydrogel upon heating and cooling[12]. Reprinted with permission. Copyright 2019, Multidisciplinary Digital Publishing Institute; (B) Chemical structure and properties of gelatin[13]. Reprinted with permission. Copyright 2021, Wiley-VCH; (C) Fabrication; and (D) photos and tensile stress-strain curves of high-strength gelatin-ammonium sulfate hydrogels via the Hofmeister effect[19]. Reprinted with permission. Copyright 2018, WILEY-VCH; (E) Schematic of formation, structure, and non-covalent interactions in gelatin organohydrogels; and (F) Photos showing the high mechanical performances of gelatin organohydrogels[22]. Reprinted with permission. Copyright 2019, ACS Publications.

polyacrylamide-based and polyvinyl alcohol-based gels), gelatin-based biogels possess the unique advantages of biocompatibility, biodegradability, mechanical tunability, sustainability and multifunctionality [Figure 2], showcasing superior balance of these properties, which make them an outstanding material for wearable sensor applications.

For applications in wearable sensors, tough gelatin biogels are typically employed in two key ways: as soft, stretchable substrates for flexible electronic devices, and as active sensing materials that respond to external stimuli. The performance of representative tough gelatin-based biogels and their application in wearable sensors are summarized in Table 1. The appealing features of mechanical robustness, good biocompatibility, and biodegradability make tough gelatin biogels a promising substrate for wearable devices on which functional sensing elements are integrated[28-30]. The fabrication of such flexible electronics is achieved by integrating individual sensing units via patterned conductive circuits embedded within or printed onto tough biogels. For example, a versatile gelatin biogel with self-adhesive, stretchable, self-healing, and fully degradable properties was fabricated by combining gelatin with sugars as extensibility promoters and water/glycerol as dispersion medium. The elastic modulus of this biogel can be widely regulated by modifying gelatin content. By utilizing rapid healing ability, biogels with different elastic moduli were assembled into

Table 1. Performance comparison and their application in wearable sensors of representative gelatin-based tough biogels

Composition	Mechanical property		Conductivity	Function	Admirable performance	Application	Ref.
	Strength	stretchability					
Gelatin/citric acid/sugar/glycerol	10-140 kPa	180%-325%	/	Substrate	Self-healing; Adhesion; Recycling;	Integrated multimodal e-skin	[28]
Gelatin/glycerol	/	Flexible	/	Substrate	Recycling;	Capacitive touch sensors	[29]
Gelatin/alginate/glycerol	~ 2.0 MPa	540%	/	Substrate	Self-healing; Recycling;	Integrated multifunctional soft electronics	[38]
Gelatin/alginate/glycerol/metal cations	0.64-1.88 MPa	45-150%	0.17-6.1 mS m^{-1}	Active sensing material	/	Thermal, humidity, strain sensors; Photodetector	[34]
Gelatin/betaine-$[Fe(CN)_6]^{3-/4-}$	0.44 MPa	247%	4.04 S m^{-1}	Active sensing material	Self-healing	Thermal sensor array	[35]
Gelatin/citric acid/NaCl	~ 0.8 MPa	~ 460%	~ 155 ohms at 100 Hz	Active sensing material	Temperature-controlled phase transition; Adhesion	On-skin bioelectrode for EEG recording	[36]
Gelatin/sodium pyrrolidone carboxylic acid	~ 0.18 MPa	256%	25 mS cm^{-1}	Active sensing material	Adhesion; Temperature-controlled phase transition; Self-healing	Bio-interface for recording electrophysiology signals	[38]
Gelatin/MXene/tannic acid/glycerol	1.81 MPa	330%	~ 80 mS cm^{-1}	Active sensing material	Adhesion	Thermal, strain sensors	[43]
Gelatin/PEDOT:PSS/glycerol-choline chloride	~ 1-3 MPa	200-375%	0.29-0.91 S/m	Active sensing material	Adhesion	Bioelectrode for ECG and EMG signal detection	[45]

"/" indicates "not available" in the references. The symbol "~" represents the estimated value in their articles. EEG: Electroencephalogram; ECG: electrocardiogram; EMG: electromyography; PEDOT: Poly(3,4-ethylenedioxythiophene); PSS: poly(styrenesulfonate).

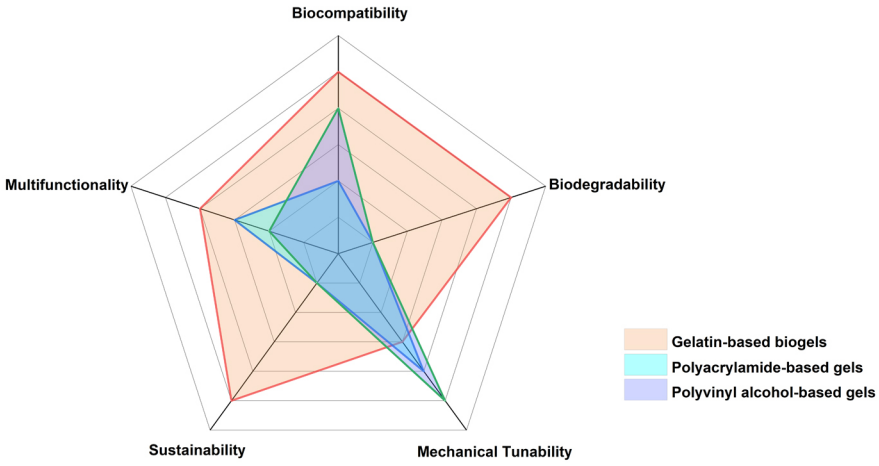

Figure 2. Radar chart comparing performance metrics of gelatin-based biogels with typical synthetic polymer gels.

graded modulus gels as tailored and sophisticated substrates for stretchable electronics [Figure 3A][28]. By integrating temperature, humidity, and strain sensors with a reusable flexible printed circuit board (PCB) on the biogel substrate, a stretchable and biodegradable on-skin electronic device was successfully fabricated, demonstrating potential as sustainable wearable electronics for real-time physiological monitoring

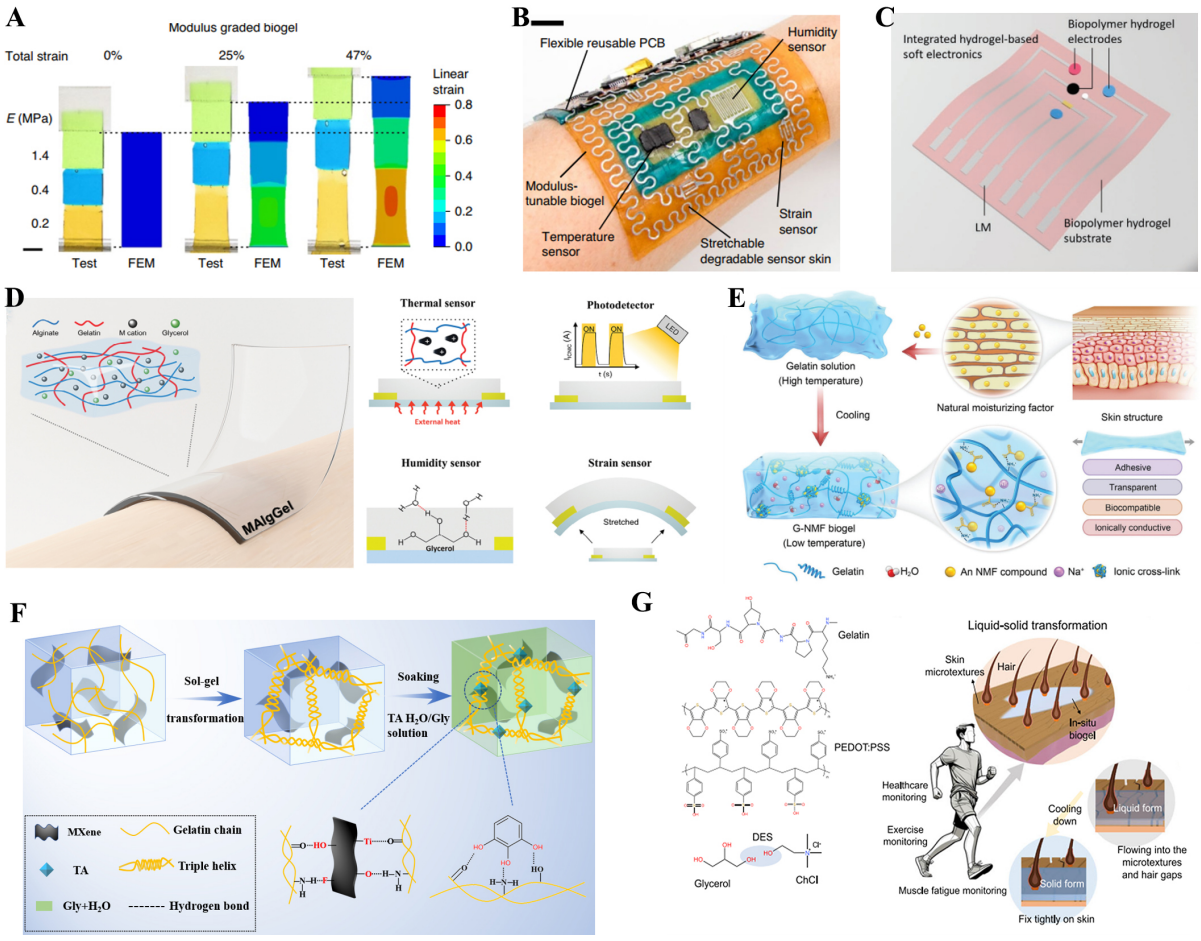

Figure 3. (A) Assembly of graded modulus gel and tensile test of a modulus-graded gel compared with its finite element modeling simulation[28]. Reprinted with permission. Copyright 2020, Springer Nature; (B) Construction of soft e-skin on biogel substrate attached to a human arm[28]. Reprinted with permission. Reprinted with permission. Copyright 2020, Springer Nature; (C) Illustration of integrated soft electronics with gelatin-alginate as biogel substrate[30]. Reprinted with permission. Reprinted with permission. Copyright 2022, WILEY-VCH; (D) Illustration of multiresponsive metal-crosslinked alginate/gelatin organohydrogel (MAlgGel) on skin for thermal, photo, humidity, and strain sensing applications[34]. Reprinted with permission. Copyright 2022, WILEY-VCH; (E) Schematic illustration of the ionic biogel fabrication process and its versatility[38]. Reprinted with permission. Copyright 2024, WILEY-VCH; (F) Schematic illustration of the preparation of the MCG organohydrogel[43]. Reprinted with permission. Copyright 2022, Elsevier; (G) Chemical composition, liquid-to-solid transformation, and application of *in situ* biogel[45]. Reprinted with permission. Copyright 2025, Springer Nature. PEDOT: poly(3,4-ethylenedioxythiophene); PSS: poly(styrenensulfonate); DES: deep eutectic solvent; MCG: MXene-composited gelatin.

[Figure 3B]. In another example, a transparent, robust, and recyclable biogel was fabricated via a sol-gel transition of a gelatin and alginate mixture dissolved in a water/glycerol solvent. Using this biogel as the substrate, a modulable and sustainable e-skin was developed by integrating sensing elements and patterned liquid metal as a soft conductor [Figure 3C][30].

Aside from the use as flexible substrates for wearable devices, conductive compositions such as conductive fillers and mobile ions have been integrated into gelatin matrix to prepare conductive tough biogel as active sensing materials. In biological systems, ion conduction is ubiquitous and extremely important for tactile perception. Inspired by this, ionically conductive gelatin biogels, a class of conductive biogels containing mobile ions within their network to enable electrical signal conduction via ion migration, have been developed by introducing various types of mobile ions[31]. Early studies developed ionically conductive

gelatin-based biogels with excellent stretchability and environmental stability by constructing non-covalent interactions and incorporating hygroscopic agents such as glycerol, and ionic liquids[32-35]. These biogels have been effectively used as epidermal sensors to monitor strain, pressure, temperature, and humidity. Tordi *et al.*[34] reported gelatin-based organohydrogel by incorporating metal cations-crosslinked alginate into a stretchable gelatin network in water/glycerol binary solvent. The introduction of various metal cations could modulate ionic conductivity and mechanical properties, while glycerol enhanced long-term stability. The resulting organohydrogels exhibited high ionic conductivity and stretchability, with sensitivity to temperature, visible light, humidity, and strain, enabling the monitoring of environmental and physiological parameters [Figure 3D]. Lately, based on the reversibility of non-covalent crosslinking, tough, self-adhesive, and ionic-conductive gelatin-based biogels with the temperature-controlled reversible fluid-gel transition have been designed for electrophysiological monitoring [36-38]. For instance, Lan *et al.* developed ionic biogels by introducing sodium pyrrolidone carboxylic acid (PCA-Na) to gelatin hydrogel [Figure 3E][38]. This biogel exhibited good stretchability, high ionic conductivity and water retention ability. Besides, the temperature-controlled reversible sol-gel transition, enabling *in situ* gelation on various surfaces with strong adhesion, making it an effective interface for high-quality electrophysiological signal recording.

Though ion-conductive biogels possess advantages of optical transparency and bionic ion transport, some intrinsic drawbacks also exist, such as low conductivity and low sensitivity. Electronically conductive gelatin biogels that enable electrical conduction primarily through electron transport mechanisms have been widely investigated by incorporating conductive fillers into gelatin gel matrices. Different conductive materials, such as metal-based materials, carbon nanomaterials, and conducting polymers, are used to impart biogels with the required electrical conductivity[39-41]. To achieve high electrical conductivity, sufficient content of conductive fillers must be incorporated to establish continuous percolation pathways. However, the hydrophobic conductive fillers, including carbon nanotubes and graphene, usually have poor dispersion in the gel network, making it difficult to form an interconnected conductive network. Recently, MXene, a two-dimensional (2D) transition metal carbide or carbonitride with metallic conductivity, excellent hydrophilicity, and abundant functional groups, has emerged as a promising conductive filler for fabricating electronically conductive biogels[42]. For example, Wang *et al.* developed a conductive MXene-composited gelatin (MCG) organohydrogel by soaking a preformed MCG hydrogel in a TA water/glycerol solution [Figure 3F][43]. Gelatin facilitated the dispersion of MXene nanosheets through non-covalent interactions with their surface functional groups. Introduction of TA further crosslinked the network via supramolecular interactions among MXene, TA, and gelatin. The resulting MCG organohydrogel was applicable as a degradable, multifunctional wearable sensor. Compared to metallic or carbon-based conductive fillers, conducting polymers offer better compatibility with gelatin chains and can form interpenetrating conductive networks within the gelatin matrix[44,45]. For example, a biogel, composed of gelatin, poly(3,4-ethylenedioxythiophene) (PEDOT): poly(styrenesulfonate) (PSS), and deep eutectic solvents, was prepared based on the concept of liquid-to-solid transformation [Figure 3G][45]. Gelatin and PEDOT: PSS were combined by non-covalent interactions to a semi-interpenetrating network, endowing the biogel with superior tensile strength (~ 1-3 MPa) and skin-like modulus (~ 0.3-1.1 MPa). The *in-situ* biogel showed strong adhesion on skin and the dual conductive mechanism led to high conductivity. This biogel was greatly contented as epidermal electronics for monitoring exercise-related information during human activities.

CHALLENGE AND PERSPECTIVES

Despite significant advancements in the design of tough and functional gelatin-based biogels for wearable applications, several critical challenges remain to be addressed.

First, although various strategies have been employed to enhance the mechanical properties of gelatin-based biogels, their performance still lags behind that of synthetic polymer gels, particularly in terms of fracture toughness. It remains a great challenge to construct a gelatin-based biogel with the comprehensive performances of high strength, toughness, and fracture-resistance capability due to the insufficient crosslinking structure. Owing to the tunable configuration of gelatin chains and the presence of multifunctional groups, recent strategies for strengthening and toughening synthetic polymer hydrogels, such as highly entangled chain structures and solvent-induced toughening, may provide effective approaches to further enhance the mechanical properties of gelatin hydrogels. Enhanced mechanical performance will broaden the application scope of biogel-based wearable sensors in demanding mechanical environments.

Second, despite the introduction of conductive fillers and mobile ions into gelatin matrix endowing the biogel with conductivity, the conductivity of these tough gelatin-based biogels is lower than 1 s/m, which is insufficient for applications as active sensing materials, particularly in bioelectrodes. The low conductivity primarily arises from the intrinsically insulating nature of gelatin, which constitutes the bulk of the biogel and impedes the formation of continuous conductive pathways. To overcome this limitation, innovative strategies such as constructing dual-phase structures-comprising a conductive component-rich phase embedded within the gelatin matrix-may enable the high conductivity (> 1 s/cm) in gelatin-based biogels, realizing a material revolution in wearable bioelectronics.

Additionally, enhancing the multifunctionality of gelatin biogels, such as incorporating antibacterial or drug-releasing capabilities, could broaden their scope of application in health monitoring and therapeutic systems. By combining advanced material design with bioinspired functionality, these functionalized biogels will hold great promise for the development of next-generation flexible wearable bioelectronics that integrate sensing and therapeutic functions.

DECLARATIONS

Authors' contributions

Wrote the original draft: Yin, J. J.; Li, Y.
Supervised, reviewed, and revised the manuscript: Sun, X.; Qin, Z. H.

Availability of data and materials

Not applicable.

Financial support and sponsorship

The authors would like to acknowledge the support of the National Natural Science Foundation of China under Grant No. 22102139.

Conflicts of interest

Yin J. and Li Y. are affiliated with Xi'an Rare Metal Materials Institute Co., Ltd., while the other authors have declared that they have no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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