

Highly Conductive and High-tensile Methacrylated Hyaluronic Acid Hydrogels for Strain Sensing Applications

Meng Zhang and Xu-Dong Yu*

Hebei Provincial Key Laboratory of Photoelectric Control on Surface and Interface, and College of Science, Hebei University of Science and Technology, Shijiazhuang 050080, China

 Electronic Supplementary Information

Abstract Owing to their good biocompatibility, polysaccharide hydrogels have broad application prospects in the field of flexible strain sensors. However, there are still significant challenges in the preparation of polysaccharide hydrogels with good mechanical properties. MCA-LiCl hydrogels were prepared by introducing methacrylated hyaluronic acid (MeHA) into the polymer network in the presence of acrylic acid (AA), acryloyloxyethyltrimethyl ammonium chloride (CATAC), and metal ions. The polymer network not only has a chemically cross-linked network and a tough network structure, but also benefits from a variety of supramolecular interactions, such as hydrogen bonding and coordination covalent bonding, resulting in excellent mechanical properties, with an elongation at break of 1390%, a tensile strength of up to 1200 kPa, a toughness of 9.4546 MJ/m³, and adhesive properties towards various substrates. At the same time, the hydrogel has a high conductivity (5.33 mS/cm) and high strain-sensing sensitivity (Gauge factor=2.55). The flexible strain sensor assembled from the prepared MCA-LiCl hydrogel can be used to detect human movements, from micro-expressions (smiles, swallowing) to pulse signals and other physiological activities, as well as large-scale joint movements (wrists, elbows, knees, fingers, etc.), realizing the real-time monitoring of full-scale human movements. The prepared hydrogels have potential applications in wearable devices, electronic skin, and strain-sensor components.

Keywords Hyaluronic acid; High conductivity; Flexible strain sensor

Citation: Zhang, M.; Yu, X. D. Highly conductive and high-tensile methacrylated hyaluronic acid hydrogels for strain sensing applications. *Chinese J. Polym. Sci.* 2026, 44, 127–139.

INTRODUCTION

With the deepening integration of flexible electronics and modern information technologies, the demand for multifunctional flexible devices continues to grow across wearable technologies, smart sensing systems, and biomedical applications.^[1] In the context of the rapid development of materials science, hydrogels, as a new type of functional material with unique physical and chemical properties, have become an important research direction in the field of functional materials because of their outstanding characteristics such as excellent flexibility, highly adjustable mechanical properties, and good biocompatibility, especially in the frontier fields of biomedical engineering and flexible electronic devices. Hydrogels are polymer materials composed of three-dimensional (3D) crosslinked networks, and their structures are usually composed of hydrophilic polymers that can absorb and retain large amounts of water. The establishment of these three-dimensional networks can primarily be realized via two distinct approaches. First, a permanent network structure can be constructed by cross-linking stable covalent

bonds. Second, temporary crosslinks can be formed by means of dynamic and reversible noncovalent interactions. Among these, there are various forms of non-covalent interactions, including physical entanglement between polymer chains, hydrogen bonding between molecules, interactions between hydrophobic groups, supramolecular self-assembly, electrostatic interactions between charged groups, and metal coordination. This multi-layered cross-linking mechanism provides hydrogels with unique mechanical and response properties.^[2]

However, as an important branch of hydrogel materials, ionic conductive hydrogels realize charge transport through the directional migration of internal free ions, and their conductive mechanism is fundamentally different from that of traditional electronic conductors. Most importantly, the three-dimensional interpenetrating network structure of these materials not only provides excellent mechanical properties (such as high tensile and fatigue properties) but also provides a continuous three-dimensional channel for ion transport, which significantly improves their electrical conductivity. At present, ionic conductive hydrogels have been widely used in the design of flexible strain sensors and have shown good application prospects in the fields of human motion monitoring and human-computer interaction.^[3–5] However, polysaccharide-based hydrogels exhibit tremendous po-

* Corresponding author, E-mail: chemyxd@163.com

Received August 12, 2025; Accepted October 13, 2025; Published online December 22, 2025

tential for medical applications, such as wound dressings and drug delivery. However, their mechanical and ionic properties, particularly their low strength and brittleness, severely restrict their utilization in flexible strain sensors.

According to existing literature, the application of such materials in the sensing field is relatively limited. Ding^[6] *et al.* combined *in situ* polymerization and freeze-thaw cycle technology to synthesize PEDOT-based composite conductive hydrogels using poly(3,4-ethylenedioxythiophene) (PEDOT) as the monomer and ammonium persulfate (APS) as the initiator. Hyaluronic acid (HA) and polyvinyl alcohol (PVA) were introduced into the PEDOT polymerization process, and the porous structure of the PVA/HA/PEDOT (PHP) composite hydrogel was successfully prepared. The tensile strength of this hydrogel increased from 0.68 MPa to 0.74 MPa, and the maximum elongation at break was 416%. To create a non-covalent chitosan (CS)/hyaluronic acid (HA) hydrogel with strong toughness, a layered manufacturing strategy was proposed,^[7] which combines the use of standing layering (S-layering) and water-washing rearrangement (WW-rearrangement) to form a rigid honeycomb structure (block) of CS/HA hydrogel, which provides a regular structural basis for host-guest permeative cross-linking (HG-crosslinking) between the flexible adhesive and the block to form many flexible tangled domains, achieving 570 kJ/m³ toughness. Compared with physically cross-linked HA/CS hydrogels, BWA hydrogels achieve seven times greater toughness and two times higher Young's modulus. In tensile tests, BWA showed a significantly enhanced elongation (588%) while maintaining a tensile strength (0.1–0.2 MPa) comparable to that of conventional CS/HA hydrogels, which significantly exceeded the 200%–400% range characteristics of conventional CS/HA hydrogels. Ionic conductive hydrogels play an important role in flexible wearable devices owing to their excellent biocompatibility, mechanical adaptability, and transparency. Thus, Liu^[8] *et al.* showed that co-crosslinked *N*-isopropylacrylamide with acrylic acid (PNIPAM/PAA) was used to form the basic backbone. The final intertwined structure was formed by a network of iron(III) sodium salts of ethylenediaminetetraacetate and tannic acid (TA) cross-linked with the basic skeleton. Owing to the dynamic hydrogen bonding between the PNIPAM/PAA backbone and TA, a tensile rate of up to 2638% can be achieved with a tensile strength of less than 60 kPa. Although a variety of polysaccharide hydrogels have been developed, their mechanical properties are generally limited, and high-strain materials often lack strength, whereas high-stress materials are usually less malleable.

Natural polymers are a class of polymer materials produced by photosynthesis or biosynthesis processes in nature, or obtained through the extraction of natural products^[9]. These materials are ideal substrates for the construction of hydrogels owing to their unique biocompatibility, degradability, and eco-friendliness. At present, hydrogels based on natural polymers such as cellulose, chitosan, collagen, and hyaluronic acid not only exhibit excellent biodegradability and good histocompatibility but also have non-toxic properties. In addition, the mechanical properties of these hydrogels are highly matched to those of natural tissues, and the raw materials are renewable.^[10] These significant advantages give natural polymer-based hydrogels broad application prospects in the biomedical field, especially in tissue engineering, drug delivery, and wound dressings. Hyaluronic acid

(HA) is produced by repeatedly linking d-glucuronic acid and *N*-acetylglucosamine disaccharides. It is a naturally occurring linear polysaccharide that is widely distributed in connective tissue, skin, synovial fluid of joints, and vitreous humor of the eye.^[11] Chemical modification of hyaluronic acid mainly targets three key functional groups in its molecular structure: the carboxylic acid group of glucuronic acid, the primary and secondary hydroxyl groups, and the *N*-acetamino group. The chemical modification of these functional groups can significantly affect the physicochemical properties of HA-derived materials, including key performance indicators such as mechanical strength, chemical stability, and biological activity.^[12] Specifically, selective modification of these active sites ensures that the properties of hyaluronic acid-based materials can be precisely controlled. There are various chemical modification methods for hyaluronic acid, among which the orthodiol structure can be efficiently converted into an active aldehyde group by the selective oxidative cleavage of sodium periodate to generate aldehydized hyaluronic acid (HA-CHO).^[13] In addition, based on the functional groups such as active hydroxyl and carboxyl groups of hyaluronic acid, a variety of functionalization modification strategies have been developed. For example, phenylboronic acid-modified hyaluronic acid (HA-PBA) was prepared by introducing phenylboronic acid groups through an esterification reaction,^[14] which has glucose-responsive properties; or photocross-linked methacrylated hyaluronic acid (HAMA) can be synthesized by methacryloylation of hydroxyl groups, which imparts the material with photocuring capabilities.^[15] These modification methods have significantly expanded the range of applications for hyaluronic acid.

Based on the above understanding of polysaccharide hydrogels as well as flexible sensors, herein, we designed hydrogels by introducing methacrylated hyaluronic acid (MeHA) into the polymer network in the presence of acrylic acid (AA), acryloyloxyethyltrimethyl ammonium chloride (CATAC), and metal ions. In this system, CATAC not only dissolved methacrylated hyaluronic acid but also made the hydrogel have good electrical conductivity (5.33 mS/cm). In addition, coordination between Li⁺ and the polymer chain in the system further enhanced the mechanical properties of the hydrogel (breaking stress of 1200 kPa, breaking strain of 1390%, and toughness of 9.4546 MJ/m³). In particular, methacrylate-based hyaluronic acid-based hydrogels can be used to assemble strain sensors for monitoring human movements, from micro-expressions (smiles, swallowing) to physiological activities such as pulse signals, as well as large-scale joint movements (wrists, elbows, knees, fingers, etc.), enabling real-time monitoring of full-scale human movements and exhibiting good strain sensitivity (GF=2.55). The prepared hydrogels are expected to have potential applications in wearable devices, strain sensors, and other applications.

EXPERIMENTAL

Raw Materials

Hyaluronic acid (HA) ($M_w=1\times10^5\text{--}2\times10^5$), *N,N,N*-dimethylformamide (DMF), methacrylic anhydride (MAC), sodium chloride (NaCl), sodium hydroxide (NaOH), absolute ethanol (AR), acryloyloxyethyltrimethyl ammonium chloride (CATAC, 80 wt%),

lithium hydroxide (LiCl, 99 wt%), acrylic acid (AA, >99 wt%), and photoinitiator 2959 (2-hydroxy-4'-(2-hydroxyethoxy)phenyl)-2-methylpropan-1-one, 99 wt%). Dialysis tubing with a molecular retention of 7000 Da. Deionized water was used in this study and the reagents used were not further treated.

Preparation of Methacrylated Hyaluronic Acid

HA (1.8 g) was dissolved in 90 mL of deionized water. Then, 57.6 mL of DMF was then added and mixed thoroughly. The mixture was then placed in a condensation circulation system at 4 °C. Subsequently, 2.88 mL of methacrylic anhydride was slowly added dropwise over 30 min. The pH value was gradually adjusted to 8–9 using a 1 mol/L sodium hydroxide solution and stabilized without further changes. After 24 h, 0.9 g of sodium chloride was added for the demulsification reaction, which lasted for 1 h. Absolute ethanol (400 mL) was used as a precipitant to obtain a white flocculent precipitate. This precipitate was extracted by centrifugation and dialyzed in deionized water with a dialysis bag (molecular weight cutoff of 7000) for 3 days at room temperature. Finally, it was freeze-dried to obtain the double-bonded hyaluronic acid.

Preparation of Conductive Hydrogel MCA-LiCl

First, the prepared MeHA (1 wt%) was dispersed in deionized water at room temperature. After vigorous stirring for several hours, a uniform and transparent solution was obtained, which was set aside for later use. Subsequently, the acryloyloxyethyltrimonium chloride (CATAC) solution and acrylic acid (AA) solution were evenly mixed with MeHA (1 wt%) in a specific proportion. To endow the prepared hydrogel with a certain level of conductivity, different amounts of LiCl were added to adjust the conductivity. Subsequently, a small amount of 2959 photoinitiator was added to the mixed solution, which was then sonicated to ensure thorough mixing and complete crosslinking. The resulting solution was then transferred to a PTFE dumbbell-shaped mold, and the MCA-LiCl hydrogel was formed by exposure to an ultraviolet lamp for 1–2 min. Specific adjustment experiments are presented in Tables S1–S4 (in the electronic supplementary, ESI).

Characterization

Fourier transform infrared (FTIR) spectroscopy of the MCA-LiCl hydrogel was performed on a Thermo Fisher Scientific Nicolet 6700 spectrometer (USA) in ATR mode, with spectral measurements in the 600–3800 cm⁻¹ wavenumber range. The ¹H-NMR spectra of HA and MeHA were obtained using a Bruker AVANCE III spectrometer. Five hundred MHz spectrometer at 298 K. The optical transmittance of the gel was measured using a UV-Vis spectrophotometer (UV-Vis, HITACHI UH5300, Japan) in the wavelength range of 400–800 nm. Gel samples were prepared by pouring the prepolymer solution into a quartz cuvette with a thickness of 2 mm by *in situ* photopolymerization. Rheological measurements of MCA-LiCl hydrogels were performed using an Anton Paar rheometer (MCR302). The surface morphology of the MCA-LiCl hydrogel was examined using a Hitachi SU8600 cold-field scanning electron microscope (SEM).

RESULTS AND DISCUSSION

The Design of MCA-LiCl Hydrogel

To develop a conductive hydrogel that integrates excellent flexibility, high stretchability, good electrical conductivity, strong

adhesion, high sensitivity, high transparency, self-healing capability, and human motion strain-sensing functions, an MCA-LiCl hydrogel was successfully prepared using a simple one-pot method.

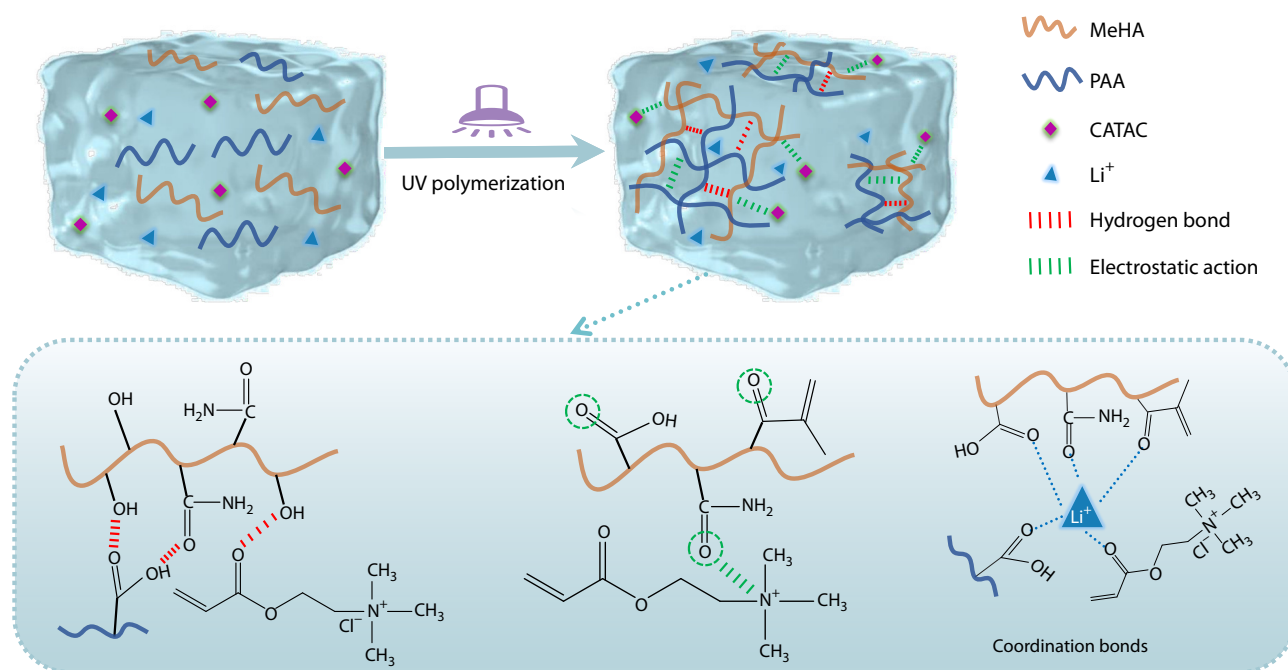
The specific preparation process is shown in Scheme 1. 1 wt% methacrylated hyaluronic acid (MeHA), a cationic monomer (CATAC), acrylic acid (AA), and lithium chloride (LiCl) were sequentially dissolved in an aqueous system. Under the action of photoinitiator 2959, ultraviolet light was used to initiate free-radical polymerization, resulting in the formation of the MCA-LiCl hydrogel.

The introduction of methacrylated hyaluronic acid (MeHA) is primarily motivated by the inherent limitations of native hyaluronic acid (HA), which lacks active groups for efficient, controllable covalent crosslinking. By chemically grafting methacrylic anhydride onto the HA backbone, these C=C bonds were activated and underwent copolymerization with other monomers containing C=C bonds. The cationic monomers CATAC and acrylic acid (AA) formed a robust and stable covalently crosslinked network. If unmodified HA was used directly, it would be difficult to form a stable three-dimensional network structure *via* UV-induced free-radical polymerization (Fig. S1 in ESI). Together with AA, CATC, and LiCl, abundant supramolecular interactions among them, including hydrogen bonding, electrostatic interactions and hydrophobic/hydrophilic^[16] interactions could be supplied, endowing the hydrogel with intriguing mechanical properties.^[17] The free ions provided by LiCl formed continuous ion-conducting pathways within the hydrogel network, enabling a stable electrical resistance response. These combined properties make the MCA-LiCl hydrogel highly promising for application in wearable sensors, flexible electronics, and human motion monitoring.

In summary, the hydrogel formed a unified single-network structure based on a covalently crosslinked skeleton, integrating a high density of dynamic physical crosslinks, including hydrogen bonds, ionic coordination, and hydrophobic/hydrophilic interactions. Through this hybrid crosslinking design—"one covalent skeleton, multiple dynamic interactions"—the different types of interactions operate synergistically to collectively endow the hydrogel with the following properties: (1) high mechanical strength and toughness with dynamic supramolecular interactions;^[18] (2) excellent adhesion: abundant polar groups (*e.g.*, carboxyl groups) enable strong adhesion to various material surfaces *via* hydrogen bonding; (3) self-healing ability: dynamic hydrogen and coordination bonds can reversibly recombine after rupture, allowing the material to self-heal rapidly; (4) good electrical conductivity: the free ions provided by LiCl formed continuous ion-conducting pathways within the hydrogel network, enabling a stable electrical resistance response. The combination of these properties makes the MCA-LiCl hydrogel highly promising for application in wearable sensors, flexible electronics, and human motion monitoring.

Basic Characterization of MCA-LiCl Hydrogel

FTIR spectroscopy confirmed the formation mechanism of the MCA-LiCl hydrogel. Fig 1(a) shows the FTIR spectra of MeHA, CATAC, AA, and MCA-LiCl hydrogels. The bands at 1728 cm⁻¹ in MeHA, 1725 cm⁻¹ in CATAC, 1732 cm⁻¹ in AA, and 1720 cm⁻¹ in



Scheme 1 Schematic diagram of the synthesis of MCA-LiCl hydrogels.

MeHA, CATAc, AA, and MCA-LiCl, respectively, corresponded to the C=O stretching vibrations of MeHA, CATAc, AA, and MCA-LiCl, respectively. After the formation of the MCA-LiCl hydrogel, the peaks show a slight redshift, moving towards lower wavenumbers, indicating the formation of hydrogen bonding interactions. The band at 1635 cm^{-1} in MeHA is the C=C stretching vibration peak. CATAc also shows a stretching vibration of its double bond (C=C) at 1635 cm^{-1} , but this band is weaker owing to molecular symmetry. The band at 1480 cm^{-1} in CATAc was attributed to the asymmetric bending vibration of the methyl group ($-\text{CH}_3$), which is characteristic of quaternary ammonium salts. The band at 960 cm^{-1} in CATAc corresponds to the C—N stretching vibration of $\text{N}^+(\text{CH}_3)_3$. The weak band at 3100 cm^{-1} in AA corresponds to the C—H stretching vibration associated with the double bond. The sharp band observed at 3390 cm^{-1} for the MCA-LiCl hydrogel was due to O—H stretching vibrations. As shown in Fig. S2 (in ESI), the $\delta=5.6\text{--}6.2$ ppm interval in the $^1\text{H-NMR}$ spectrum shows a set of chemically shifted symmetrical doublets (with a 1:1 peak-area ratio), which is in good agreement with the typical range of chemical shifts of olefin protons in methacrylates. The characteristic signal can be unequivocally attributed to the methacryloyl double-bond peak that was successfully introduced through esterification of the hyaluronic acid (HA) molecular chain. This clearly demonstrated that the polymerizable double-bond functional group was successfully grafted onto the HA backbone via esterification.

To directly confirm the coordination interaction between Li^+ and the carboxyl group ($-\text{COOH}$), X-ray photoelectron spectroscopy (XPS) analysis was performed. As depicted in Fig. S3 (in ESI), the O 1s spectrum of MCA hydrogel can be deconvoluted into two distinct peaks corresponding to the C—O and C=O bonds, with binding energies of 532.2 and 531.1 eV, respectively. Upon coordination with Li^+ in the MCA-LiCl hydrogel, the strong electron-withdrawing induc-

tion effect of Li^+ causes an overall shift of the O 1s peak towards higher binding energies.^[19] Specifically, the binding energies of the C—O and C=O bonds shift to 532.6 and 531.2 eV, respectively. Furthermore, a new peak emerges at 533.4 eV, which is attributed to oxygen atoms coordinated with Li^+ (O—Li). These results confirm the coordination interaction between Li^+ and the carboxyl groups, which constitutes an effective and dynamic cross-linking mechanism in this hydrogel system.

It is noteworthy that, as shown in Fig. 1(b), the MCA-LiCl hydrogel membrane exhibited excellent transparency, and the ultraviolet spectrophotometer measured the transmittance of the MCA-LiCl hydrogel to characterize its optical performance, achieving a transmittance of 86% in the visible-light region, significantly broadening its application scope. Fig. 1(c) shows the MCA-LiCl hydrogel placed under different temperature conditions for 35 days to test the environmental stability of the hydrogel. It can be seen that at -25°C , the moisture content of MCA-LiCl hydrogel changes very little; at 25 and 50°C , the weight change rate stabilizes at around 20 days, and the final moisture content is also within a relatively acceptable range of deviation. The rheological properties of the MCA-LiCl hydrogel were studied through rheological testing. Fig. 1(d) shows the strain sweep test results of the hydrogel. It can be observed from the figure that the G' of the hydrogel remains higher than G'' as the strain increases, indicating that the hydrogel has good resistance to mechanical damage. An oscillation frequency sweep test was conducted, as shown in Fig. 1(e). Throughout the entire frequency range, G' is significantly higher than G'' , and it increases with frequency, suggesting that the MCA-LiCl hydrogel maintains a stable state. The microstructure of the prepared MCA-LiCl hydrogel was further characterized using scanning electron microscopy. As seen in Fig. 1(f), the prepared hydrogel exhibited a three-di-

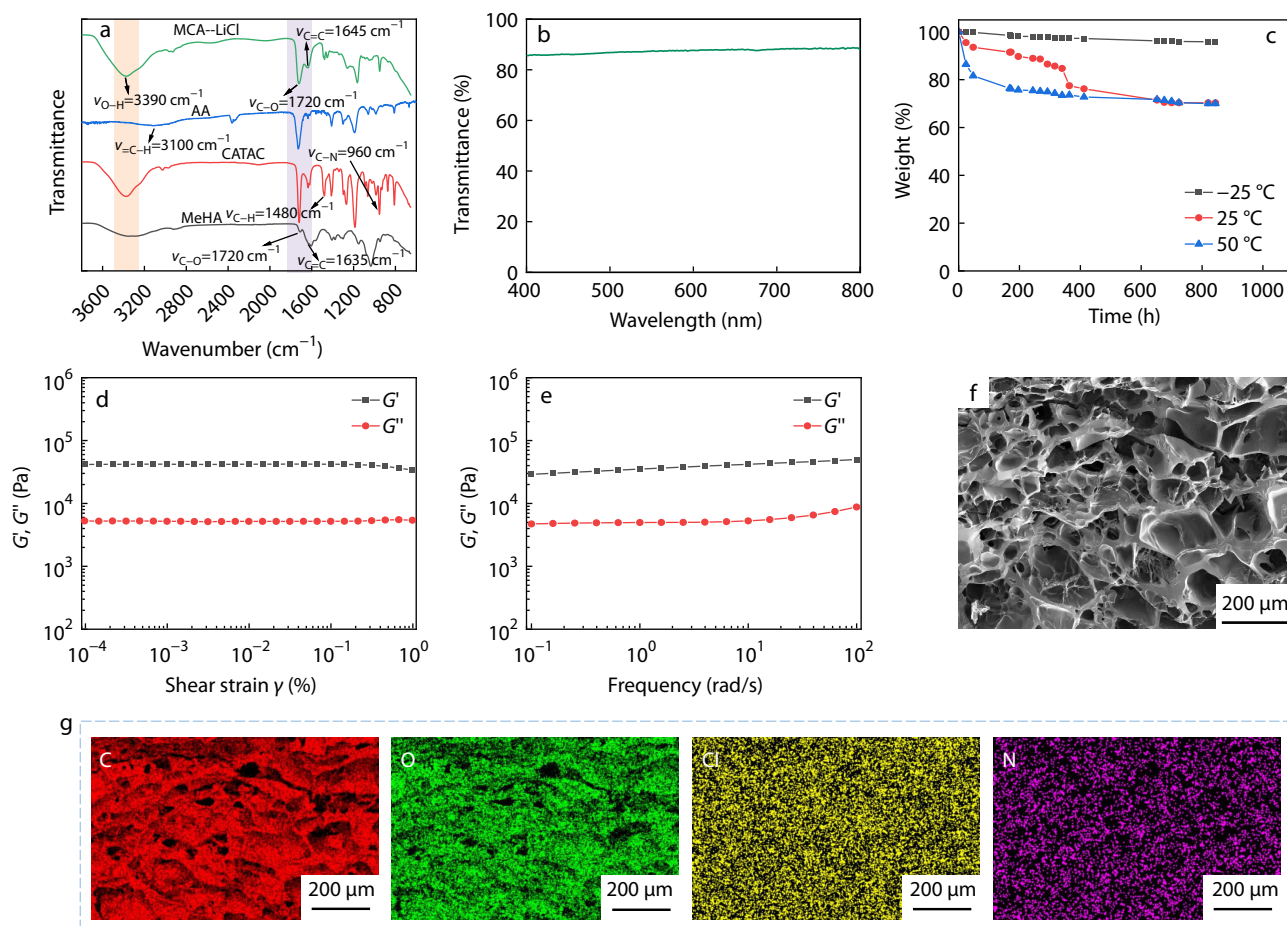


Fig. 1 (a) FTIR spectra of MCA-LiCl hydrogels; (b) Transparency of MCA-LiCl hydrogels; (c) Environmental stability of MCA-LiCl hydrogels; (d) G' and G'' of MCA-LiCl hydrogels in oscillatory strain scanning assays; (e) G' and G'' of MCA-LiCl hydrogels in oscillating frequency sweep assays; (f) SEM image of MCA-LiCl hydrogel; (g) EDS elemental diagram of MCA-LiCl hydrogel.

mensional porous network structure with a surface presenting intricate curved surfaces. The porous structure served as a transport channel that facilitated ion transfer, thereby further enhancing the conductivity of the hydrogel. The multiple hydrogen bonds within the network contributed to energy dissipation, significantly improving the flexibility and stretchability of the hydrogel. Furthermore, EDS spectrometry showed that C, N, O, and Cl were evenly distributed in the MCA-LiCl hydrogel, demonstrating the uniform distribution of components in the gel.

Mechanical Properties of MCA-LiCl Hydrogels

To ensure the durability of wearable sensors, hydrogels must possess sufficiently strong mechanical properties such as ductility and toughness. To optimize the mechanical performance of the MCA-LiCl hydrogels, we employed a controlled variable method to determine the optimal ratio of MCA-LiCl hydrogels that achieved the best combined performance in terms of ductility and toughness (Tables S1–S4 in ESI). As shown in Figs. 2(a) and 2(b), we adjusted the amount of MeHA while maintaining the CATAc:AA ratio constant. At low MeHA contents, the network was primarily governed by electrostatic crosslinking between CATAc and AA. A high density of dynamic ionic bonds enables energy dissipation *via* reversible bond breaking and reformation, resulting in high toughness and extensibility, but low

elastic modulus and fracture strength. The tensile performance of the MCA hydrogel was the best when the amount of MeHA was 600 μL (49.5 wt%), the tensile performance of the MCA hydrogel is at its best. Fig. 2(c) shows the toughness and elastic modulus corresponding to varying amounts of MeHA. As shown in Figs. S4(a) and S4(b) (in ESI), we adjusted the CATAc content while maintaining the MeHA:AA ratio constant. With a lower CATAc content, insufficient charge pairing reduces the electrostatic crosslink points, resulting in a loose network and weak mechanical properties. With a higher CATAc content, excess charge causes like-charge repulsion, damaging structural integrity and leading to lower tensile strength. It was found that when the CATAc volume was 400 μL , the stress-strain curve showed significantly better tensile strength and elongation. Fig. S3(c) (in ESI) shows the toughness and tensile strength corresponding to the varying amounts of CATAc. Similarly, as shown in Figs. S3(d) and S3(e) (in ESI), the AA content was adjusted while maintaining a constant MeHA:CATAc ratio. The stress-strain curve indicated that the tensile strength and elongation of the hydrogel were 672% and 1180%, respectively. Therefore, Fig. S3(f) (in ESI) shows the toughness and tensile strength corresponding to the final adjusted ratio, reaching 4.0444 MJ/m^3 and 0.07653 MPa, respectively.

In addition, the introduction of ionic salts is a more direct method for improving the conductivity of hydrogels. There-

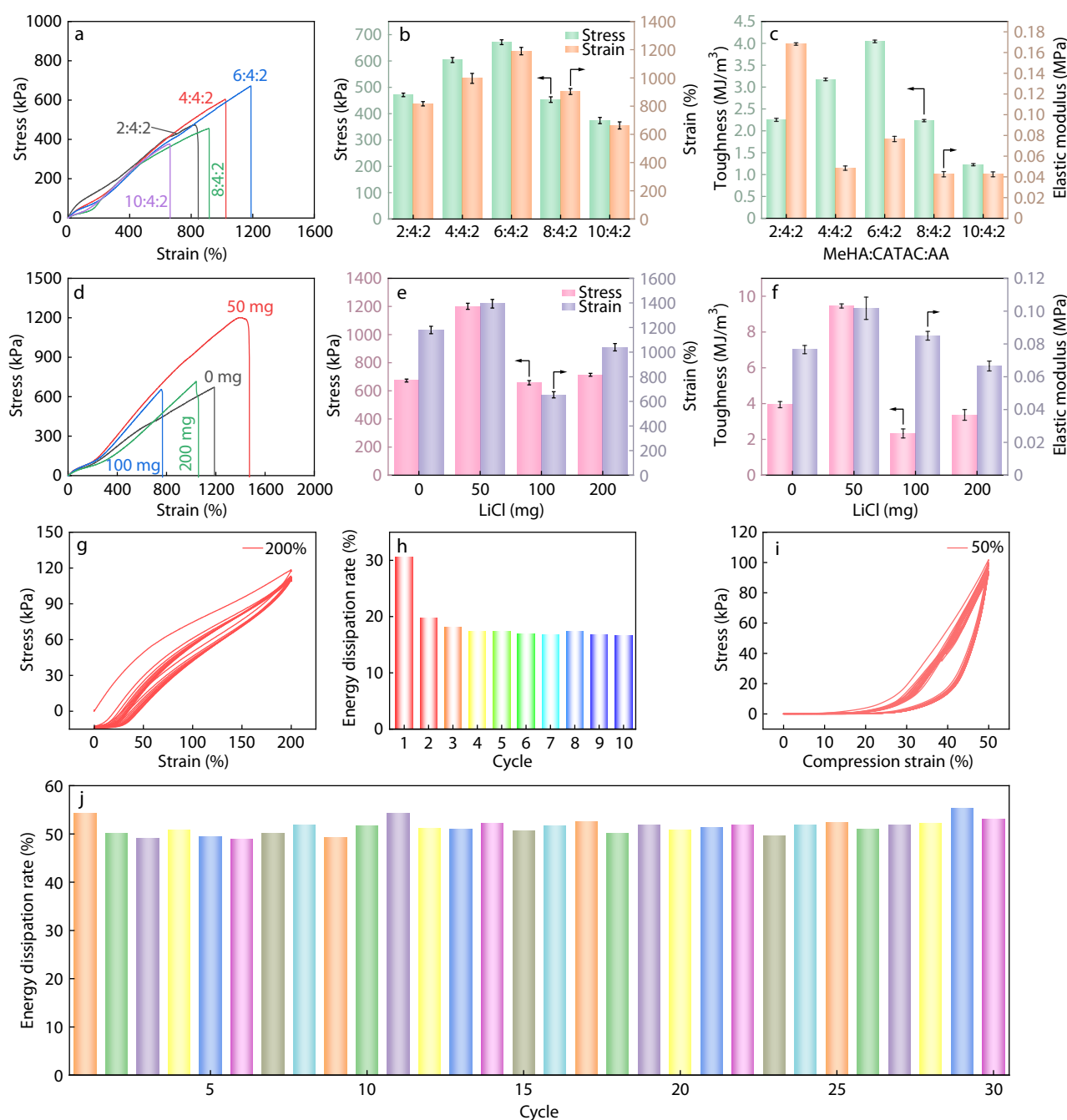


Fig. 2 Mechanical properties of MCA-LiCl hydrogels. (a, b) Tensile stress-strain curves and histograms of MCA-LiCl hydrogels with different 1% MeHA contents; (c) Toughness and elastic modulus of MCA-LiCl hydrogels with different 1% MeHA contents; (d, e) Tensile stress-strain curves and histograms of MCA-LiCl hydrogels with different LiCl contents; (f) Toughness and elastic modulus of MCA-LiCl hydrogels with different LiCl contents; (g) 10 tensile loading-unloading cycles of hydrogel at 200% elasticity and (h) corresponding dissipation energy; (i) Compressive loading-unloading cycle curves of hydrogels under 50% elastic strain and (j) corresponding dissipation energy.

fore, we introduced the conductive ion LiCl. The addition of ions also affected the mechanical properties of the hydrogel, as shown in Figs. 2(d) and 2(e). Stretching tests were conducted on the samples with different LiCl contents using a universal tensile testing machine. The hydrogel exhibited optimal mechanical properties when the LiCl content was 50 mg (1200 kPa, 1390%). Fig. 2(f) shows the toughness and Young's modulus of the MCA-LiCl hydrogel, which reached 9.4546 MJ/m³ and 0.1017 MPa, respectively. Figs. S4(a) and S4(b) (in

ESI), and Fig. 1(g) show the continuous stretch-retract curves for the MCA-LiCl hydrogel at 50%, 100%, and 200% strain for 10 cycles, respectively, as well as the energy dissipation corresponding to 200% stretch-retract cycles repeated 10 times (Fig. 1h). It can be seen that from the second cycle to the tenth cycle, the energy dissipation remains at a stable level. Only in the first stretch-retract cycle is a larger amount of energy dissipated, which is due to the irreversible crosslink points in the MCA-LiCl hydrogel being broken and not being

reformed, resulting in significant energy dissipation. In the other nine stretch-retract cycles, the reversible dynamic crosslink points acted as 'sacrificial bonds' for reconstruction, leading to less energy dissipation.^[20,21] Fig. S4(c) (in ESI) shows a composite diagram of the first cycle under stretching and unloading of the MCA-LiCl hydrogel at 50%, 100%, and 200% cycles. Compression-unloading cyclic tests typically characterize the fatigue resistance of materials. Fig. 2(i) shows the compression-unloading curve of the MCA-LiCl hydrogel under 50% strain over 30 continuous cycles, whereas Fig. 2(j) shows the corresponding energy dissipation graph.

Conductive Properties of MCA-LiCl Hydrogel

The conductivity of a hydrogel can be realized through ionic or electronic pathways. To further enhance the conductivity of the hydrogel in flexible electronic devices, LiCl was added to the hydrogel and its impact on the system was examined. As shown in Figs. 3(a) and 3(b), the MCA-LiCl hydrogel acts as a conductor in the circuit to light an LED, and it can be observed that the LED light changes when the strain of the hydrogel increases. This is due to the lengthening of the ionic transport pathways, which increases the ionic transport resistance, consequently leading to

a decrease in LED brightness, indicating that this hydrogel possesses good strain sensitivity. This means that the hydrogel can convert the applied mechanical deformation into a detectable resistance signal, which is widely applied in strain sensors. As depicted in Figs. 3(c) and 3(d), the highest conductivity was achieved by varying the amount of LiCl. Specifically, the conductivity increases sharply with the initial addition of LiCl, reaching a maximum value of 5.33 mS/cm at 50 mg of LiCl. This enhancement is primarily attributed to the significant increase in the number of charge carriers (Li^+ and Cl^-), which dominates under low-ionic-strength conditions. However, when the LiCl content exceeded this optimal value, the conductivity decreased markedly, reaching a local minimum of approximately 100 mg of LiCl. This behavior can be explained by the following factors: enhanced ion association leading to a reduction in effective carrier concentration^[22] and a notable increase in the local viscosity of the aqueous phase within the hydrogel due to excessive salt, which impedes ion mobility.^[23] In addition, in the MCA-LiCl hydrogel system studied here, high salt concentrations may screen the electrostatic repulsion between polymer chains, causing partial contraction of the network and thereby reduc-

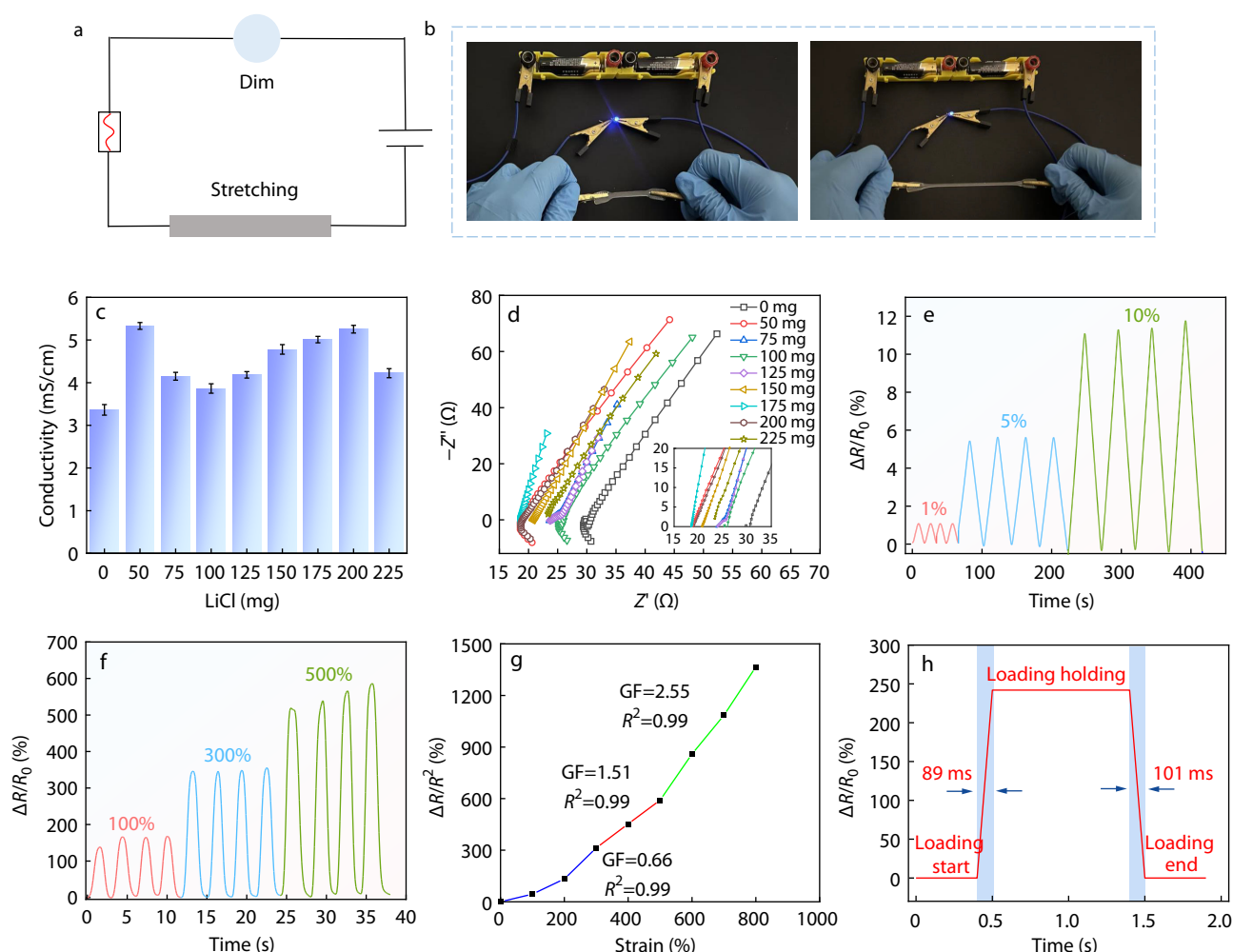


Fig. 3 Electrical properties of MCA-LiCl hydrogels. (a, b) Changes in the brightness of small bulbs under different hydrogel lengths; (c, d) Conductivity of MCA-LiCl hydrogels with different LiCl contents; (e) Cyclic tensile response in a small tensile deformation range of 1%–10% and (f) 100%–500% in a large tensile deformation range; (g) Sensitivity (GF) of the relative resistance of MCA-LiCl hydrogels under tensile strain (100%–800%); (h) Response and recovery time of MCA-LiCl hydrogels under bending strain.

ing effective pathways for ion transport.^[24] Notably, upon further increasing the LiCl content to 200 mg, the conductivity ceased to decline and instead rebounded. We speculate that under near-saturation salt conditions, although ion association and high viscosity persist, the absolute number of free ions increases markedly and emerges as the dominant factor regulating the conductive behavior. To accurately capture this complex trend, we conducted experimental validation at finer concentration intervals (see Figs. 3c and 3d). The results unequivocally confirm that conductivity exhibits a distinct "V-shaped" trend with increasing LiCl concentration, underscoring the complexity and uniqueness of the ion transport mechanism in this hydrogel matrix. In response to the reviewers' questions, the conductivity decreases when the LiCl content is 225 mg due to the imbalance of ion association effects at high salt concentrations, leading to negative migration phenomena.^[25] As shown in Fig. S6 (in ESI), when 200 mg of LiCl was added, the solution became saturated. Undissolved solids were clearly observed at 225 mg, indicating that a supersaturated state was reached. To confirm this, porosity tests were conducted. Porosity is an important parameter that reflects the performance of a hydrogel. A high porosity indicates that the hydrogel material has a porous structure with large pores, providing ample space for cell growth.^[26] The formula for calculating the porosity change of the MCA-LiCl hydrogel was $\text{porosity (\%)} = (w_2 - w_1) / (\rho \times V)$.^[27] First, record the mass w_1 of the freeze-dried hydrogel, then immerse it in anhydrous ethanol. After it was fully absorbed and saturated, its mass w_2 was recorded. Here, ρ represents the density of anhydrous ethanol (approximately 0.789 g/cm³) and V is the volume of the MCA-LiCl hydrogel. Fig. S7 (in ESI) shows the porosities of the hydrogel scaffolds with different lithium chloride (LiCl) contents. The porosity of the MCA-LiCl hydrogel was approximately 47%. The porosity of the MCA hydrogels without LiCl reached 78%, demonstrating excellent porosity. As the LiCl content increased, the cross-linking density of the three-dimensional network rises, causing the hydrogel porosity to decrease from 78% to 47% (a reduction of 31%).

To assess the sensing performance of the MCA-LiCl hydrogel, we examined the relative resistance changes under varying strains ranging from 1% to 10% and from 100% to 500%. As Figs. 3(e) and 3(f) illustrate, when the tensile strain increases from 1% to 10% and from 100% to 500%, the relative resistance shows a stepped increase. Once the MCA-LiCl hydrogel swiftly recovered to its original shape, the relative resistance was zero. In the field of materials science, the gauge factor (GF) is extensively employed as a crucial indicator for quantifying and evaluating the sensitivity of hydrogel materials to external strain responses.^[28] According to the fitting curve calculations (Fig. 3g), the GF value is 0.66 below 300%, 1.51 during 300%–500%, and additionally, it exhibits an exponential dependence on the applied tensile strain, reaching up to 2.55 in the high-strain state (500%–800%). In practical applications, the response speed of the strain sensors is a prominent parameter. Fig. 3(h) shows that the MCA-LiCl hydrogel sensor had a very short loading response time (89 ms) and unloading recovery time (101 ms) at a strain of 200%, indicating its excellent strain response capability. To evaluate the long-term stability of the resistance response of the hydrogel system, we conducted cyclic tests on its continuous strain-

sensing performance at different stretching rates (Fig. S8 in ESI). The results indicate that during multiple loading-unloading cycles at different frequencies (100, 300, and 500 mm·min⁻¹), the resistance changes of the hydrogel remained largely consistent without significant fluctuations, demonstrating excellent repeatable stability. Furthermore, continuous stretching/releasing cycle tests were conducted on the MCA-LiCl hydrogel sensor under 50% strain for 400 cycles (Fig. S9 in ESI). The results indicate that the sensor consistently maintained a stable electrical signal output throughout the entire cycle, demonstrating excellent mechanical durability and long-term operational stability, further proving its potential as a high-performance flexible strain sensor in practical applications.

Self-healing Properties of MCA-LiCl Hydrogels

Compared to covalent bonds, non-covalent interactions possess significantly lower energies, generally ranging from 0.1 kJ/mol to 30 kJ/mol. This weak interaction allows for dynamic reversible formation and dissociation, playing a key role in processes such as molecular recognition, self-assembly, and biological regulation. Li⁺ can form ionic bonds with carbonyl groups present in MeHA, PAA, and CATAC, whereas the hydroxyl and amide groups in MeHA can form hydrogen bonds with carboxyl groups in PAA and quaternary ammonium cations (R₄N⁺) in CATAC. Owing to the dynamic ionic and hydrogen bonds, the MCA-LiCl hydrogel exhibited intriguing self-healing properties. To explore the self-healing characteristics of the hydrogel visually, as shown in Fig. 4(a), the gel was placed in a closed circuit with wires under the influence of an electric field, leading to LED light illumination. After cutting the gel, the LED light exited, which was due to the interruption of the current path that could not form a completely closed circuit. When the two cut gel pieces were brought back into contact, the LED light illuminated again. Next, as shown in Fig. 4(b), blue and pink squares (2 cm × 2 cm) were made, and the gels were symmetrically cut along the middle to form two rectangular (1 cm × 2 cm) gels. The two pieces are then stitched together and left static for approximately 1 h, during which one can observe the self-healing of the gel, and the self-healed gel can be stretched. Fig. 4(c) shows that under a microscope, the cracks in the MCA-LiCl hydrogel gradually faded and disappeared after 30 min, further demonstrating the self-healing properties of the hydrogel.

To quantitatively assess the self-healing performance of the MCA-LiCl hydrogel, we conducted a systematic examination of the tensile mechanical properties of samples with varying healing durations. As depicted in Figs. 4(d) and 4(e), when the self-healing time was prolonged from 2 h to 12 h, the fracture stress of the hydrogel increased from 331 kPa to 849 kPa, and the fracture strain increased from 501% to 1123%. This clearly demonstrated that the self-healing capability of the hydrogel increased over time. The self-healing efficiency (η) was quantified by calculating the recovery rate of the fracture stress, defined as the ratio of the maximum tensile strength of the healed sample to that of the original sample. The self-healing efficiency of the MCA-LiCl hydrogel reached 27.75% after 2 h, and 70.88% after 12 h. The self-healing properties of the hydrogel indicate that it has a dynamically reversible cross-linked network that provides excellent mechanical recoverability and long-term durability. Fig. 4(f) shows the resistance

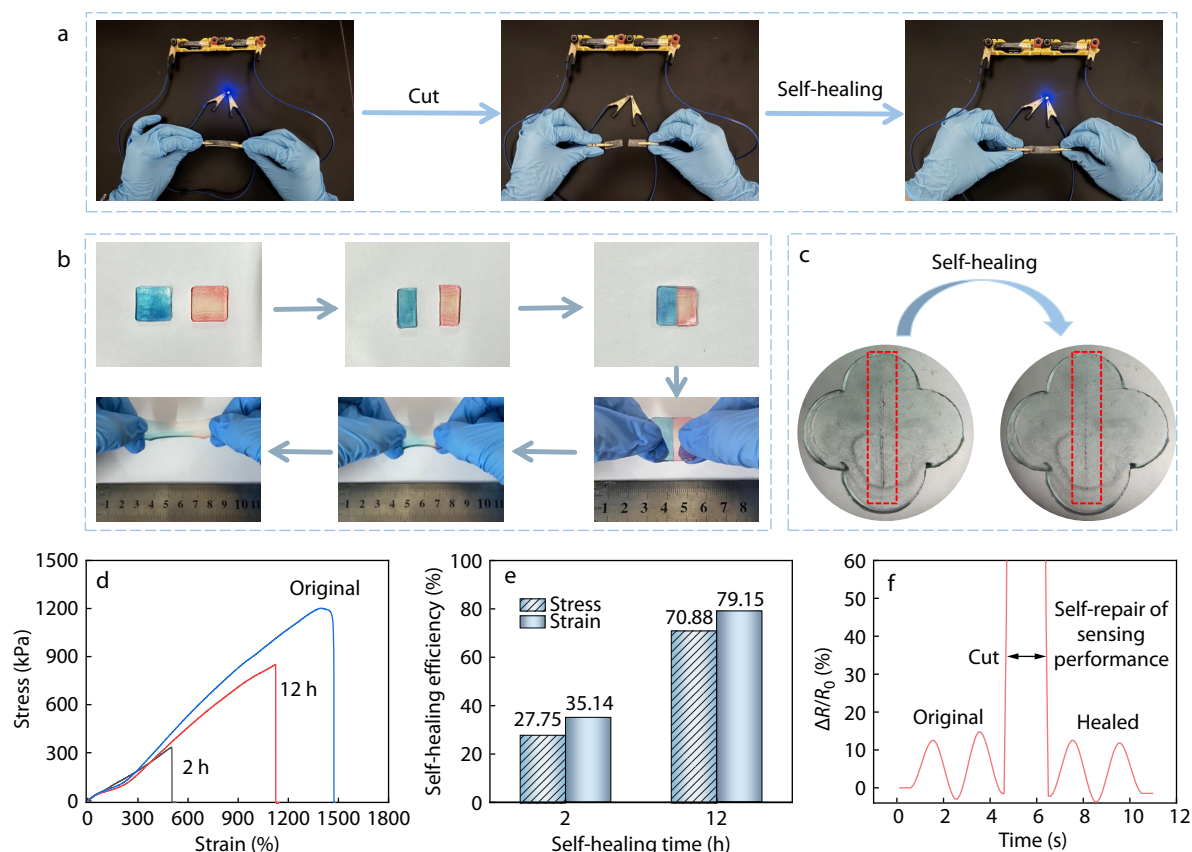


Fig. 4 Demonstration and characterization of self-healing of MCA-LiCl hydrogels. (a) Electrochemical healing process of conductive MCA-LiCl hydrogels; (b) Macroscopic self-healing ability of MCA-LiCl hydrogel without external stimuli; (c) Self-healing images of MCA-LiCl under the microscope; (d) Stress-strain curves and (e) self-healing rate of MCA-LiCl hydrogels after healing at different time; (f) Diagram of resistance recovery of MCA-LiCl hydrogel during the cleavage-healing cycle.

recovery curve of the MCA-LiCl hydrogel during the cutting-healing process. Studies indicate that this hydrogel can rapidly reconstruct its conductive network after the cross section comes into contact. This second-level self-healing sensing characteristic allows the hydrogel device to maintain continuity in signal collection, even under repeated mechanical damage, demonstrating significant advantages in practical wearable applications.

Adhesive Properties of MCA-LiCl Hydrogels

To assess the potential use of MCA-LiCl hydrogels as wet tissue adhesives, lap-shear tests were conducted to analyze the adhesion strength of the hydrogels on different substrates (Fig. 5a). The instantaneous adhesion of the hydrogels (0 h) and the adhesion strength after 12 h of aging on various substrate materials (paper, wood, glass, copper sheets, iron sheets, and fresh pig skin) were systematically tested. It has been demonstrated that pig skin is anatomically, physiologically, biochemically, and immunologically the most similar to human skin, making it a good dermatological model for evaluating the adhesion performance of tissue-bonding hydrogels on skin tissue.^[29] The results of the 180° peel strength test of the pressure-sensitive adhesive tape are shown in Fig. 5(b), where the adhesion strength of the substrate materials (paper, wood, glass, copper, iron, and pig skin) increase from 45.00, 36.33, 79.29, 147.14, 66.12, and 39.29 kPa to 430.00, 57.65, 166.63, 203.98, 90.00 and 188.47 kPa, respectively,

from 0 h to 12 h after dehydration. Multiple adhesion-peeling experiments were also performed to evaluate the long-term stability and practical application potential of this hydrogel as a reusable adhesive material. As shown in Fig. S10 (in ESI), after five consecutive adhesion-peeling cycles on wood, the adhesion strength of the MCA-LiCl hydrogel decreased from 57 kPa to approximately 40 kPa. These results indicated that the hydrogel retained high adhesive stability even after repeated use, exhibiting good repeatability and reusability.

Fig. 5(c) shows photographs of the adhesion of the MCA-LiCl hydrogel to the substrate materials (including paper, wood, glass, copper, iron, and rubber). As shown in Fig. 5(d), when the hydrogel is pulled to follow joint movements (such as bending of the fingers or wrists), it can adhere firmly to the human skin.^[30] Fig. 5(e) shows a schematic of the adhesion mechanism of the MCA-LiCl hydrogel, illustrating the physical bonding process between the hydrogel and substrate materials. The upper section of the figure presents the hydrogel material, whereas the lower section shows various substrate materials. At the interface, the alkyl chains of the quaternary ammonium salt interact hydrophobically with rubber. Meanwhile, the amino ($-\text{NH}_2$) and carboxyl ($-\text{COOH}$) groups found in MeHA and PAA form hydrogen bonds with glass, pigskin, paper, and wood. Additionally, these groups establish metal-coordination interactions with metallic substances.

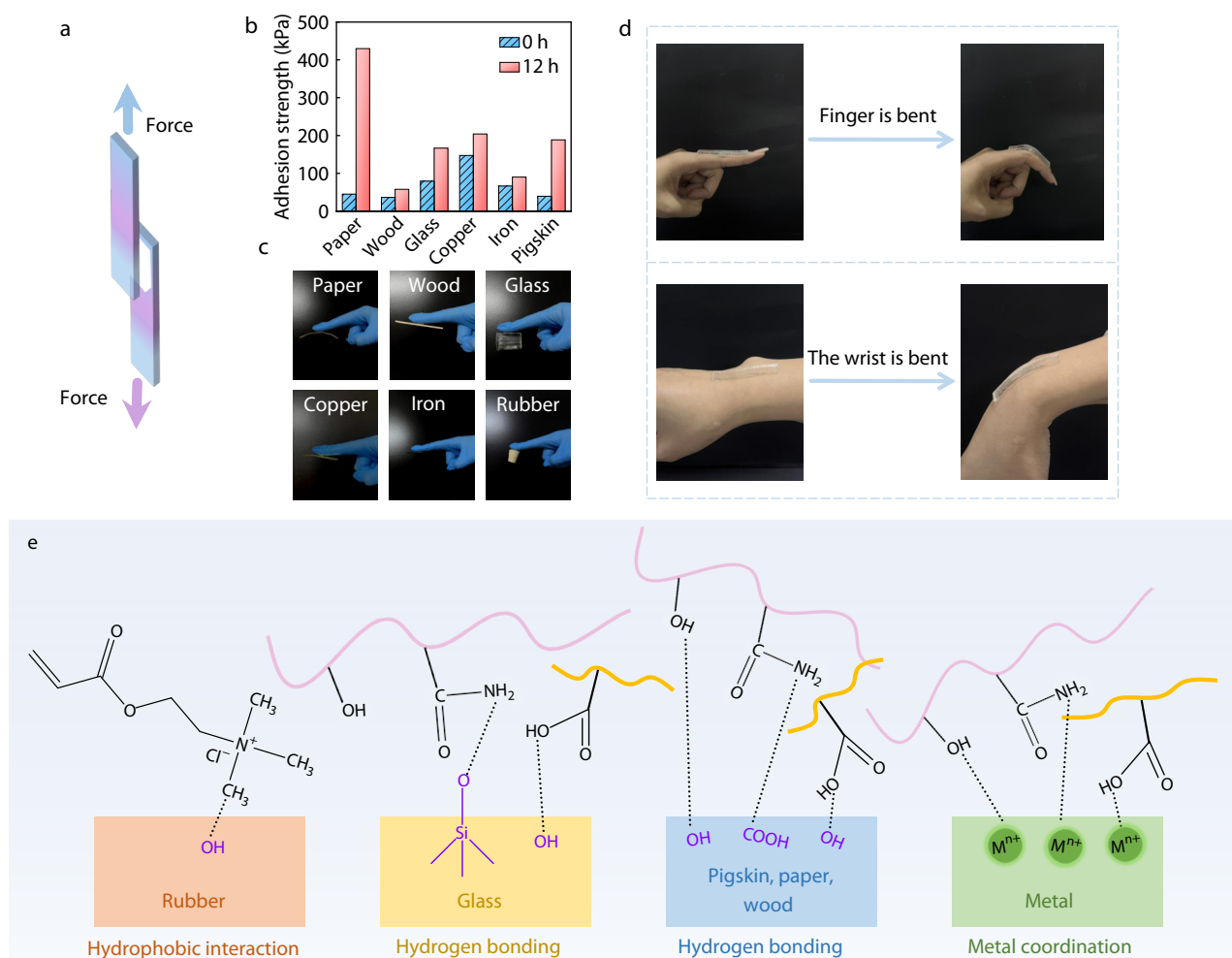


Fig. 5 Adhesion properties of MCA-LiCl hydrogels. (a) Schematic diagram of MCA-LiCl hydrogel lap shear test; (b) Adhesion strength of MCA-LiCl hydrogels to various matrices; (c) Photograph of MCA-LiCl hydrogel adhesion to different substrates; (d) Adhesion to the skin; (e) Schematic diagram of the adhesion mechanism of MCA-LiCl hydrogel toward various substrates.

Strain Sensing Performance of MCA-LiCl Hydrogel

Owing to its exceptional mechanical properties, remarkable toughness, high conductivity, strong strain-response capability, and outstanding self-healing and adhesion characteristics, the MCA-LiCl hydrogel can be employed as a strain sensor on the human epidermis for the real-time monitoring of human movement. Throughout the monitoring process, the hydrogel was attached to the surface of the skin and connected to an electrochemical workstation through wires, further converting human motion into electrical signals. As illustrated in Fig. 6, the hydrogel demonstrated excellent adaptability by adhering seamlessly to various body parts, such as the elbow, fingers, wrist, and knee, eliminating the need for additional tape. For instance, Fig. 6(a) shows that the MCA-LiCl hydrogel adhered to the neck, where a change in relative resistance was observed when looking down and up. Figs. 6(b), 6(c), 6(f), and Fig. S8(c) (in ESI) demonstrate the changes in the relative resistance waveforms during smiling, swallowing, pulse, and frowning, respectively, proving the sensor's good response capability to slight movements. Subsequently, the hydrogel was attached to a finger. When the finger was bent at 90°, as shown in Fig. S8(a) (in ESI),

the resistance change rate varied correspondingly with the bending of the finger. Figs. 6(e) and 6(h) show that the MCA-LiCl hydrogel remained attached to finger joints. As the finger was bent from 0° to 30°, then to 60°, and finally to 90°, the resistance change rate gradually and regularly increased. Bending the finger at the same angles three times results in a relatively orderly waveform change in the resistance. This suggests that the MCA-LiCl hydrogel has high sensitivity and reliability. As shown in Figs. 6(d), 6(g), and Fig. S11 (in ESI), the flexible wearable sensor can also be used to detect other body movements (elbow, knee, and wrist), and similar repetitive and stable changes in the relative resistance can be observed, suggesting potential applications for joint disease diagnosis.^[31] The integrity and continuity of the signals demonstrated that the hydrogel had high sensitivity. To further evaluate the electrical recovery performance of the MCA-LiCl hydrogel, the self-healed gel after cutting was used for strain sensing. As shown in Fig. S12 (in ESI), the self-healed hydrogel sensor retained the ability to generate clear and stable electrical signal responses when subjected to human movement (e.g., knee bending). Compared with the original sample, the self-healing sensor exhibited high consistency in re-

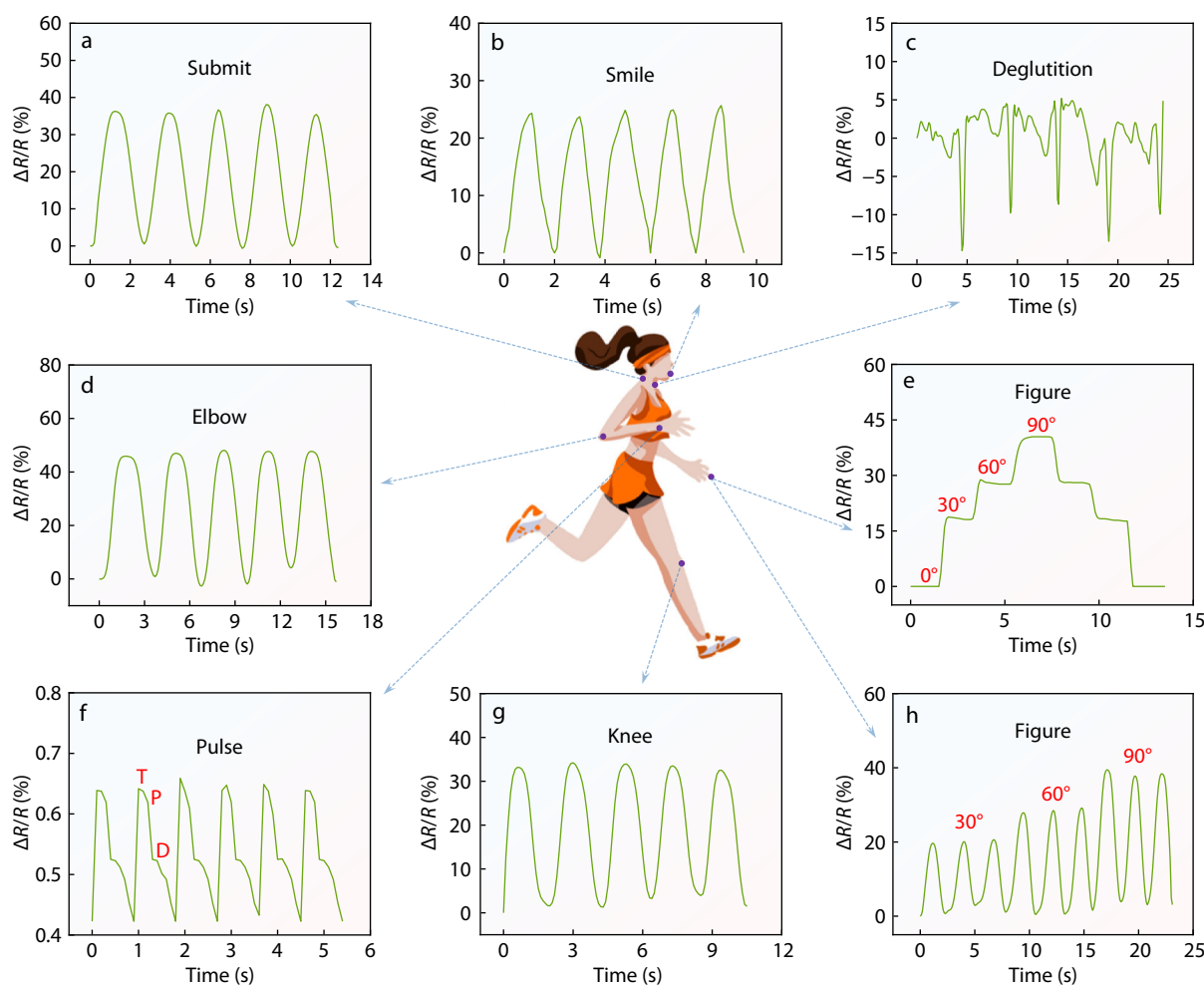


Fig. 6 Sensing performance of MCA-LiCl hydrogels. Hydrogel sensor relative resistance over time to monitor real-time human activity signals, including (a) neck nodding, (b) smile, (c) swallowing, (d) elbow flexion, (e) finger flex 30°, 60°, 90°, 60°, 30°, (f) pulse, (g) knee flexion, (h) bend the finger 3 times in a row at 30°, 60°, 90°.

sponse sensitivity and signal stability. These results demonstrate that the hydrogel not only achieves effective structural self-repair, but also undergoes nearly complete recovery of sensing functionality, thereby providing critical reliability support for its practical application in wearable electronic devices, and that the strain sensors successfully convert various types of movements into different electrical signals, showing a fast and reliable response.

CONCLUSIONS

In summary, this study successfully prepared chemically cross-linked MCA-LiCl polymer hydrogels using a convenient one-pot method. Notably, the MeHA in the polymer network not only participates in the formation of the chemically cross-linked network, but the robust network structure also benefits from various supramolecular interactions, such as hydrogen bonding and coordination covalent bonds. The obtained hydrogel exhibited excellent mechanical properties, with a maximum elongation at break of 1390%, a tensile strength of 1200 kPa, and a toughness of 9.4546 MJ/m³. It also shows strong adhesion to various substrates, particularly the adhesion strength to wood, which can

reach 430 kPa after 12 h. Additionally, this hydrogel has a high conductivity of 5.33 mS/cm and high strain-sensing sensitivity (GF=2.55). Based on these characteristics, we integrated the MCA-LiCl hydrogel into a flexible strain sensor, and tests showed its ability to accurately detect physiological activities ranging from micro-expressions (smiling, swallowing) to pulse signals, as well as large-scale joint movements (wrist, elbow, knee, fingers, etc.), to achieve real-time monitoring of full-scale human movements. This work not only provides new ideas for the design of multifunctional conductive hydrogels, but also lays a material foundation for applications in fields such as electronic skin, artificial muscles, and wearable devices.

Conflict of Interests

The authors declare no interest conflict.

Electronic Supplementary Information

Electronic supplementary information (ESI) is available free of charge in the online version of this article at <http://doi.org/>

10.1007/s10118-025-3482-8.

Data Availability Statement

The data supporting the findings of this study are available from the corresponding author upon reasonable request.

ACKNOWLEDGMENTS

This work was financially supported by the National Natural Science Foundation of China (No. 22271074), Natural Science

Foundation of Hebei Province (Nos. B2023208042, B2022208032, B2021208066, E2024208084, and E2024208088), Science Research Project of Hebei Education Department (No. JZX2024013), Special Fund for Local Scientific and Technological Development under the Guidance of the Central Government (No. 236Z3704G), and Hebei Province High Level Talent Funding (No. A202001010).

REFERENCES

- 1 Zhang, H.; Zhang, D.; Cai, H.; Ma, Y.; Li, K.; Zhang, P.; Guo, Y. A bacterial cellulose-based multifunctional conductive hydrogel

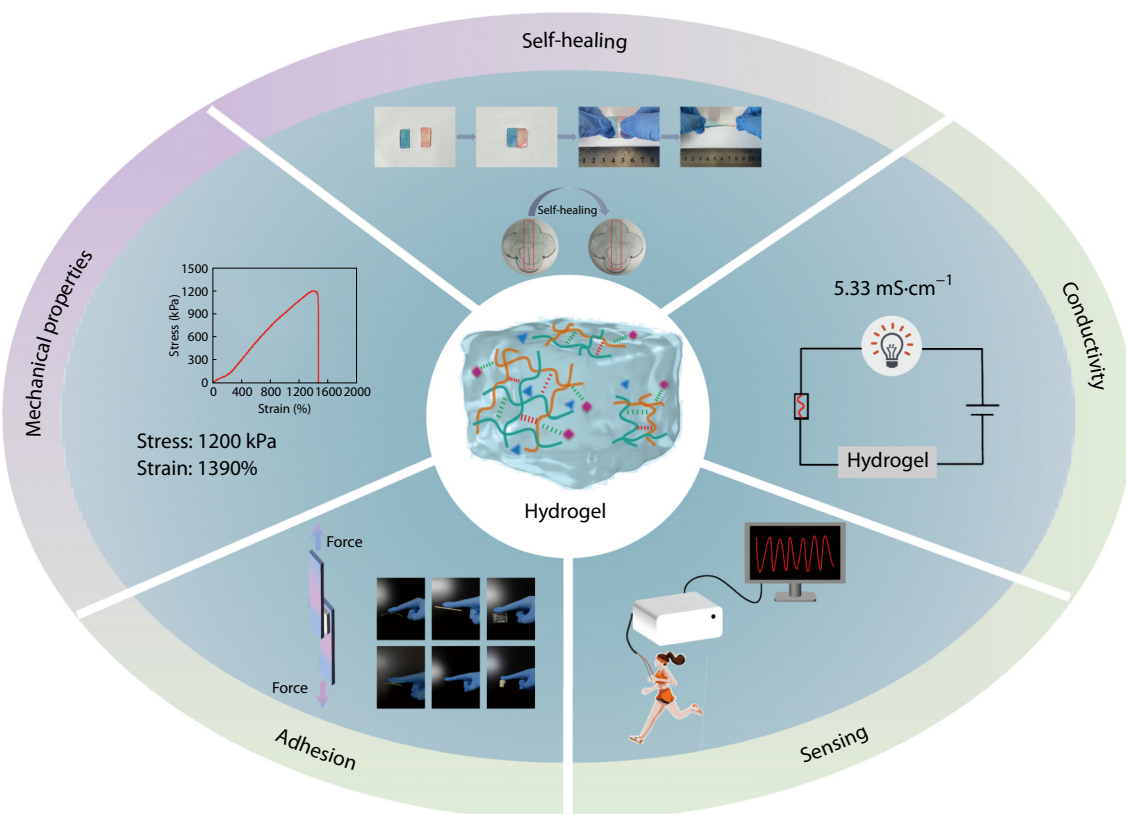
Graphical Abstract

Highly Conductive and High-tensile Methacrylated Hyaluronic Acid Hydrogels for Strain Sensing Applications

Meng Zhang and Xu-Dong Yu

Hebei University of Science and Technology

A novel hydrogel sensor by copolymerizing modified hyaluronic acid, olefin monomers, and acrylic acid. The hydrogel exhibits excellent mechanical properties (stress 1200 kPa, strain 1390%), high electrical conductivity (5.33 mS/cm), good sensitivity (GF=2.55) and efficient self-healing properties.



- for flexible strain sensors and supercapacitors. *Carbohydr. Polym.* **2025**, 360, 123601.
- 2 Yang, D. Recent advances in hydrogels. *Chem. Mater.* **2022**, 34, 1987–1989.
 - 3 Yu, J. Y.; Moon, S. E.; Kim, J. H.; Kang, S. M. Ultrasensitive and highly stretchable multiple-crosslinked ionic hydrogel sensors with long-term stability. *Nano-Micro Lett.* **2023**, 15, 51.
 - 4 Zhao, X. J.; Xu, J. H.; Zhang, J. Y.; Guo, M. R.; Wu, Z. L.; Li, Y. Y.; Xu, C.; Yin, H. Z.; Wang, X. L. Fluorescent double network ionogels with fast self-healability and high resilience for reliable human motion detection. *Mater. Horiz.* **2023**, 10, 646–656.
 - 5 Wu, M.; Pan, M. F.; Qiao, C. Y.; Ma, Y. H.; Yan, B.; Yang, W. S.; Peng, Q. Y.; Han, L. B.; Zeng, H. B. Ultra stretchable, tough, elastic and transparent hydrogel skins integrated with intelligent sensing functions enabled by machine learning algorithms. *Chem. Eng. J.* **2022**, 450, 138212.
 - 6 Ding, X.; Xu, L.; Wang, Y.; Liu, T.; Yang, L.; Gao, F.; Hu, D. Facile synthesis of conductive PVA/HA/PEDOT composite hydrogel for human motion monitoring. *Mater. Today Commun.* **2025**, 43, 000.
 - 7 Hu, Z.; Ma, R.; Li, D.; Wang, T.; Elmalki, M.; Li, C.; Xue, Z.; Nie, G. Hierarchically constructed a noncovalent chitosan/hyaluronic acid hydrogel with entanglement-enhanced toughness. *Int. J. Biol. Macromol.* **2025**, 318, 145055.
 - 8 Liu, Y.; Wang, Y.; Fu, Y.; Wang, N. N.; Zhan, S. Q.; Sheng, L.; Yang, H. Y.; Liu, C. Healable and transparent ionic conductive hydrogels based on pnatf as multiple-signal sensors. *ACS Appl. Polym. Mater.* **2025**, 7, 2529–2540.
 - 9 Zhao, L.; Zhou, Y.; Zhang, J.; Liang, H.; Chen, X.; Tan, H. Natural polymer-based hydrogels: from polymer to biomedical applications. *Pharmaceutics*. **2023**, 15, 2514.
 - 10 Bao, Z.; Xian, C.; Yuan, Q.; Liu, G.; Wu, J. Natural polymer-based hydrogels with enhanced mechanical performances: preparation, structure, and property. *Adv. Healthc. Mater.* **2019**, 8, 1900670.
 - 11 Ryy, J.; Kim, S.; Oh, I.; Kato, S.; Kosuge, T.; Sokolova, A. V.; Lee, J.; Otsuka, H.; Sohn, D. Internal structure of hyaluronic acid hydrogels controlled by iron(III) ion-catechol complexation. *Macromolecules* **2019**, 52, 6502–6513.
 - 12 Jensen, G.; Holloway, J. L.; Stabenfeldt, S. E. Hyaluronic acid biomaterials for central nervous system regenerative medicine. *Cells* **2020**, 9, 2113.
 - 13 França, C.; Leme, K.; Luzo, A.; Hernandez-Montelongo, J.; Santana, M. Oxidized hyaluronic acid/adipic acid dihydrazide hydrogel as cell microcarriers for tissue regeneration applications. *Polymers* **2022**, 22, 949–958.
 - 14 Shi, W.; Hass, B.; Kuss, M. A.; Zhang, H.; Ryu, S.; Zhang, D.; Li, T.; Li, Y.; Duan, B. Fabrication of versatile dynamic hyaluronic acid-based hydrogels. *Carbohydr. Polym.* **2020**, 233, 115803.
 - 15 Tavsanlı, B.; Okay, O. Mechanically robust and stretchable silk/hyaluronic acid hydrogels. *Carbohydr. Polym.* **2019**, 208, 413–420.
 - 16 Murray, J. S.; Politzer, P. Hydrogen bonding: a coulombic σ -hole interaction. *J. Indian Inst. Sci.* **2020**, 100, 21–30.
 - 17 Xiong, J.; Wang, X.; Li, L.; Li, Q.; Zheng, S.; Liu, Z.; Li, W.; Yan, F. Low-hysteresis and high-toughness hydrogels regulated by porous cationic polymers: the effect of counteranions. *Angew. Chem. Int. Ed.* **2024**, 63, 202316375.
 - 18 Shahi, S.; Roghani-Mamaqani, H.; Talebi, S.; Mardani, H. Chemical stimuli-induced reversible bond cleavage in covalently crosslinked hydrogels. *Coord. Chem. Rev.* **2022**, 455, 214368.
 - 19 Wang, Y.; Zhang, X.; Zhao, X.; Liu, Y.; Wu, D.; Huang, H. Tailoring carboxyl-rich micro-environment of metal-organic framework via mixed linkers strategy for enhanced adsorption of lithium ion. *Chem. Eng. J.* **2025**, 511, 162116.
 - 20 Wang, Y.; Pang, B.; Wang, R.; Gao, Y.; Liu, Y.; Gao, C. An anti-freezing wearable strain sensor based on nanoarchitectonics with a highly stretchable, tough, anti-fatigue and fast self-healing composite hydrogel. *Compos. Part A Appl. Sci. Manuf.* **2022**, 160, 107039.
 - 21 Yan, X.; Zhao, R.; Lin, H.; Bao, X.; Zhao, Z.; Song, S. Dual-network conductive hydrogel with rapid self-healing ability and great fatigue resistance as strain sensor for human motion monitoring. *Eur. Polym. J.* **2023**, 201, 112570.
 - 22 Li, C.; Wang, T.; Lai, H. C. J.; Park, S. W.; Chan, W. Y. K.; Li, Q.; Zhao, Y.; Fan, J.; Pei, Z.; Zhi, C. Hydrophobic-unit-regulated hydrogel electrolytes with high water content and low salt concentration for high-voltage aqueous batteries. *Joule* **2025**, 9, 101827.
 - 23 Cheng, W.; Xu, C.; Zhang, Q.; Yue, Y.; Lei, D.; Wen, L.; Wang, S. Ion concentration gradient induced efficient ion migration in hydrogen-bonded organic frameworks for high-performance, self-powered humidity sensing. *Small* **2025**, 21, 2412497.
 - 24 Sui, X.; Guo, H.; Cai, C.; Li, Q.; Wen, C.; Zhang, X.; Wang, X.; Yang, J.; Zhang, L. Ionic conductive hydrogels with long-lasting antifreezing, water retention and self-regeneration abilities. *Chem. Eng. J.* **2021**, 419, 129478.
 - 25 France-Lanord, A.; Grossman, J. C. Correlations from ion pairing and the nernst-einstein equation. *Phys. Rev. Lett.* **2019**, 122, 13601.
 - 26 Yan, Y.; Guan, S.; Wang, S.; Xu, J.; Sun, C. Synthesis and characterization of protocatechuic acid grafted carboxymethyl chitosan with oxidized sodium alginate hydrogel through the Schiff's base reaction. *Int. J. Biol. Macromol.* **2022**, 222, 2581–2593.
 - 27 Alsaïd, Y.; Wu, S.; Wu, D.; Du, Y.; Shi, L.; Khodambashi, R.; Rico, R.; Hua, M.; Yan, Y.; Zhao, Y.; Aukes, D.; He, X. Tunable sponge-like hierarchically porous hydrogels with simultaneously enhanced diffusivity and mechanical properties. *Adv. Mater.* **2021**, 33, 2003995.
 - 28 Wei, J.; Xie, J.; Zhang, P.; Zou, Z.; Ping, H.; Wang, W.; Shen, J. Z.; Lei, L.; Fu, Z. Bioinspired 3D printable, self-healable, and stretchable hydrogels with multiple conductivities for skin-like wearable strain sensors. *ACS Appl. Mater. Interfaces*. **2021**, 13, 2952–2960.
 - 29 Fan, X.; Wang, S.; Fang, Y.; Li, P.; Zhou, W.; Wang, Z.; Chen, M.; Liu, H. Tough polyacrylamide-tannic acid-kaolin adhesive hydrogels for quick hemostatic application. *Mater. Sci. Eng. C*. **2020**, 109, 110649.
 - 30 Han, L.; Yan, L.; Wang, K.; Fang, L.; Zhang, H.; Tang, Y.; Ding, Y.; Weng, L. T.; Xu, J.; Weng, J.; Liu, Y.; Ren, F.; Lu, X. Tough, self-healable and tissue-adhesive hydrogel with tunable multifunctionality. *NPG Asia Mater.* **2017**, 9, e372.
 - 31 Zeng, H.; Ma, H.; Xu, L.; Gao, J.; Yan, M.; Wang, Q. Tough, Highly conductive and frost-resistant chitosan based hydrogel for flexible sensor. *Int. J. Biol. Macromol.* **2025**, 297, 139847.