

Organic-Inorganic Hydrogel Strain Sensors Based on Methacryloyl Ethoxy Trimethyl Ammonium Chloride and Bentonite

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Electronic Supplementary Information

Abstract Flexible wearable electronic devices based on hydrogels have immense potential in a wide range of applications. However, many existing strain sensors suffer from significant limitations including poor mechanical properties, low adhesion, and insufficient conductivity. To address these challenges, this study successfully developed an organic-inorganic double-network conductive hydrogel using acrylic-modified bentonite (AABT) as a key component. The incorporation of AABT significantly enhanced the mechanical properties of the ATHG@LiCl hydrogel, achieving an impressive stretchability of 4000% and tensile strength of 250 kPa. Moreover, it improved the electrical conductivity of the hydrogel to a maximum of 1.53 mS/cm. The catechol structure of tannic acid (TA) further augmented the adhesive properties of the ATHG@LiCl hydrogel toward various substrates such as copper, iron, glass, plastic, wood, and pigskin. The addition of lithium chloride (LiCl) and dimethyl sulfoxide (DMSO) endowed the hydrogel with exceptional freezing resistance and flexibility, even at low temperatures of -20°C . Remarkably, the hydrogel maintained a conductivity of 0.53 mS/cm under these conditions, surpassing the performance of many other reported hydrogels. Furthermore, the ATHG@LiCl hydrogel demonstrated outstanding characteristics, such as high sensitivity (gauge factor $\text{GF}=4.50$), excellent transparency (90%), and reliable strain-sensing capabilities, indicating that the ATHG@LiCl hydrogel is a highly promising candidate for flexible wearable soft materials, offering significant advancements in both functionality and performance.

Keywords Acrylic bentonite; Organic-inorganic hydrogel; Adhesion; Strain sensor

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INTRODUCTION

Hydrogels are recognized as one of the most promising candidates for the construction of flexible sensors,^[1–3] bioactuators,^[4] in the realm of tissue engineering^[4,5], energy storage devices,^[6,7] drug delivery,^[8] and wearable electronic devices. This is ascribed to their intrinsic flexibility, tissue-analogous structures, and adjustable mechanical characteristics. The incorporation of skin-mimicking characteristics is indispensable for the development of next-generation wearable sensors. This is essential for guaranteeing the congruence of biomechanical properties with those of the human skin and achieving stable signal responses.^[9,10] Hydrogels, which are characteristic soft materials with tissue-comparable softness and excellent biocompatibility, have been extensively applied in the research domain of flexible wearable sensors.^[11–14] Nevertheless, hydrogel-based strain sensors are still confronted with challenges such as inferior mechanical properties, low electrical conductivity, and weak adhesion.

When hydrogel sensors function as electronic devices at biological interfaces, they must retain their excellent mechanical properties, good environmental adaptability, adhesion, conductivity, and biocompatibility. To enhance the mechanical properties of hydrogels, numerous researchers have incorporated inorganic materials such as SiO_2 , perovskite, graphene, and silver nanoparticles into the system. For instance, Chen *et al.* synthesized organic-inorganic interpenetrating network gels by employing materials such as clay and *N,N*-dimethylacrylamide. These gels exhibited a fracture strain of 500% and a stress of only 25 kPa.^[15] Lyu *et al.* reported that the stress and strain of the GNEC/HAPAAm gel reached 182 kPa and 1685%, respectively, owing to the incorporation of graphene nanosheets;^[16] of the F-PTD composite hydrogel synthesized by Wang *et al.* manifested as a tensile strain of 412% and a fracture stress of 95 kPa.^[17] Owing to their distinctive network architecture, organic-inorganic composite hydrogels are capable of augmenting their mechanical strength to a certain degree. However, the augmentation was circumscribed. This is attributed to the fact that the weak interfacial interaction between the polymer matrix and conductive filler restricts the enhancement of its mechanical properties.^[18]

High conductivity is an important indicator for wearable

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sensors. Numerous researchers have endeavored to enhance the mechanical properties and electrical conductivity of hydrogels by incorporating conductive fillers including graphene, carbon nanotubes, MXene, and metal nanoparticles/nanowires.^[19] Nevertheless, for conductive hydrogel sensors based on filler networks, conductivity and mechanical strength frequently exhibit incompatibility. The trade-off between these two properties represents a significant impediment that restricts their application in multifunctional sensing scenarios.^[20,21] Han *et al.* incorporated polyaniline-cellulose nanofibers (PANI@CNF) into a viscoelastic polyvinyl alcohol (PVA) network. This led to a high conductivity of 5.2 S/m. However, the maximum tensile stress was merely 36.6 kPa (approximately 5-fold that of PVA).^[22] Our research revealed that constructing a rational and robust bonding network within the hydrogel represents an efficacious approach to enhance the mechanical strength without compromising the electrical conductivity.^[23] Natural clay exhibited an extremely high in-plane elastic modulus. Moreover, its surface contains abundant exposed binding sites, which can effectively enhance the mechanical strength of the polymer composite matrix.^[24] The most representative is bentonite (BT), a layered aluminosilicate clay mineral whose main constituent is montmorillonite.^[25] BT is extensively applied in various fields, such as environmental protection,^[26] pharmaceutical excipients,^[27] catalysts and drug carriers, and petroleum,^[28] owing to its controllable interlayer structure, surface modifiability, and non-toxicity. Furthermore, its distinctive lamellar structure not only demonstrates an edge in the water absorption capacity, reaching up to 10-fold its own mass,^[29] but also promotes intercalation among water, small molecules, and polymers. Many researchers have modified it to achieve better results, such as ACBT (basic calcium bentonite). Liu *et al.* prepared a multifunctional conductive hydrogel with an acrylic acid/carboxymethyl chitosan double-network structure using basic calcium-based bentonite (ACBT) as a crosslinker, which exhibited excellent flexibility.^[30] The composite hydrogel fabricated by Yan *et al.* through the addition of clay to polyacrylamide/alginate exhibited remarkable tensile properties with a fracture strain of 1840% and enabled real-time and reliable sensing.^[31] These findings provide new insights for the design and development of mechanically robust hydrogel sensors.

Based on the aforementioned considerations, to develop highly conductive hybrid hydrogels with comprehensive properties such as enhanced mechanical performance, good adhesion characteristics, and low-temperature tolerance, modified bentonite (AABT) is incorporated into double network polymer networks composed of gelatin and poly(*N*-2-hydroxyethyl acrylamide) (PHEAA) through strong interfacial interactions among organic–inorganic components. Moreover, the addition of tannic acid (TA) can further improve the adhesive properties. The incorporation of lithium chloride (LiCl) and dimethyl sulfoxide (DMSO) endowed the hydrogel with conductivity and low-temperature resistance. As anticipated, the optimized ATHG@LiCl hydrogel demonstrated an ultra-long tensile strain of 4000%, fracture stress of 250 kPa, and good adhesion to different substrates. The lamellar structure of the material also played a crucial role in enhancing the

conductivity of the composite hydrogel, with the value increasing from 0.7 mS/cm to 1.53 mS/cm. Even at a low temperature of -20°C , the hydrogel still maintained good stretchability, flexibility, and electrical conductivity (0.53 mS/cm). In addition to the above properties, the ATHG@LiCl hydrogel also showed high sensitivity (Gauge Factor = 4.50) toward strain and good transparency (95% in the visible light region), which makes it fully qualified as a wearable electronic device that was studied in detail in this work.

EXPERIMENTAL

Raw Materials

Gelatin (Bieder Pharmaceutical), acrylic (AA, AR, >99%, RON), nanoclay, hydrophilic bentonite ($\text{Al}_2\text{O}_3 \cdot 4(\text{SiO}_2 \cdot \text{H}_2\text{O})$, Particle size $\leq 25\ \mu\text{m}$, 360.31 (MW), Picarsol), tannic acid (TA, 99.2%, Picarsol), lithium chloride (LiCl, AR, $\geq 99.5\%$, Damas-Beta), calcium oxide (CaO, AR, $\geq 98.0\%$, Hengxing reagent), absolute ethanol (AR, $\geq 99.7\%$, Hengxing Reagent), cyclohexane (AR, Tianjin Damao Chemical Reagent Factory), *N*-(2-hydroxyethyl) acrylamide (containing stabilizer MEHQ, M.W.: 115.13, Bigarsol), hydrochloric acid (HCl, M.W.: 36.5, Tianjin Kemio Chemical Reagent Co., LTD.), dimethyl sulfoxide (DMSO, AR, $\geq 99.5\%$, Tianjin Yongda Chemical Reagent Co., LTD.), (2-hydroxy-4'-(2-hydroxyethoxy)-2-methylpropiophenone (photoinitiator 2959, M.W.: 224.25, 98%, Damas-Beta). Deionized water was used in this study and the reagents used were not further treated.

Preparation of ATHG@LiCl Hydrogel

Synthesis of acrylic modified bentonite (AABT)

(1) Raw bentonite (5 g) was mixed with 15 mL of 12% HCl solution at 80°C and stirred for 4 h. Subsequently, the dispersion was first filtered through a Buchner funnel, and the filtration cake was rinsed with deionized water several times to remove the excess hydrochloric acid and exchangeable cations. The resulting product (acid activated bentonite) was dried at 105°C for 2 h and stored in a desiccator.

(2) Acid-activated bentonite (5 g) was mixed with 1.75 g calcium oxide and a certain amount of deionized water. The temperature of the reaction mixtures was maintained at 50°C for 14 h. Then, the obtained powder was rinsed with deionized water several times to remove excess calcium hydroxide. Finally, the product was dried at 50°C for 2 h, ground, sieved through a 200-mesh sieve, and denoted as ACBT.

(3) First, at 78°C , ACBT (10.0 g) was dispersed into 100 mL of deionized water under vigorous stirring at 500 r/min. Afterward, the mixed solution (14.0 g of AA and 16.0 g of absolute ethanol) was slowly added to the mixture within 30 min. The reaction was completed by dropwise addition for 3 h. Finally, the AABT was washed with anhydrous ethanol, filtered, dried, and sieved to obtain good fluffiness.

Preparation of the hydrogels

First, 10 mg of AABT was weighed in a 5 mL beaker, followed by the addition of 100 μL of DMSO. AABT was evenly dispersed by shaking. Subsequently, 0.4 g of *N*-2-hydroxyethyl acrylamide (HEAA), 0.2 g of gelatin solution (15 wt%), 100 μL of TA (5 wt%), and 20 mg of LiCl (s) were sequentially added to the beaker. After thorough mixing, the mixture was sonicated until it became clear and transparent. Ultimately, the obtained solution was poured into a Teflon mold, and the target gel was acquired by

irradiation with a UV lamp for 30 s. Similar methods were used for other hydrogels with different elemental mass ratios.

Characterization

Fourier transform infrared (FTIR) spectra of the ATHG@LiCl hydrogels were recorded on an IR PRESTIGE-21 spectrometer (Shimadzu, Japan). The UV-Vis absorption (transmittance) of the hydrogel was measured using a UV-Vis 2550 spectrometer (Shimadzu, Japan). The microscopic morphology of the hydrogel was observed by cryo-electron microscopy (Quatro S, Semmerfeld, China) at an accelerating voltage of 20 kV. Rheological behavior of the hydrogels was measured using a controlled stress rheometer (MCR302 SN80987260, England). Tensile and adhesive strength tests of the ATHG@LiCl hydrogels were performed at room temperature or low temperatures using a microcomputer-controlled electronic universal testing machine (LD24.202, made in China) with a 200 N load cell. These hydrogels were molded into dumbbell shapes of 20 mm (length) $\times$ 4 mm (width) $\times$ 1 mm (thickness) for testing. The conductivity and electrical properties of the hydrogels were tested using an electrochemical workstation (CHI660E, China).

RESULTS AND DISCUSSION

Design of the ATHG@LiCl Hydrogel

In this study, a multifunctional hydrogel based on acrylic-modified bentonite (AABT) is investigated. This hydrogel features an organic-inorganic dual network structure and exhibits properties such as electrical conductivity and adhesion. After a series of adjustments, the final composition of the hydrogel was deter-

mined to be 10 mg of AABT, 0.2 g of 15 wt% gelatin solution, 0.5 g of HEAA, 100 μ L of 5 wt% tannic acid solution, 20 mg of LiCl (s), 100 μ L of DMSO solvent and 3 mg of photoinitiator. This hydrogel was designated as the ATHG@LiCl hydrogel. The detailed preparation process and reaction mechanism of the ATHG@LiCl hydrogel are as follows: AABT, gelatin, HEAA, TA, LiCl, and the photoinitiator were homogeneously blended according to their specific mass and volume. Subsequently, the solution was sonicated until it was clear. Thereafter, it was poured into a Teflon mold and irradiated under an ultraviolet lamp to initiate the hydrogel formation. The presence of TA not only enables better dispersion of AABT and enhances its stability within the system but also imparts good adhesion properties to the hydrogel. At the same time, $-\text{OH}$, $-\text{COOH}$, Al^{3+} , and Mg^{2+} in AABT can generate non-covalent interactions, such as hydrogen bonds, electrostatic interactions, and coordination with the hydroxyl groups of HEAA, $-\text{NH}$, $-\text{OH}$ of gelatin, and positively charged N groups. In addition, the ionic bonds of $\text{Al}^{3+}/\text{Mg}^{2+}$ and Cl^- of LiCl were included, and the mechanism of action is shown in Fig. 1. These interactions ultimately give rise to hydrogels with outstanding properties including high electrical conductivity, strong adhesion, and remarkable low-temperature resistance.

Mechanical Properties

In the domain of flexible electronic devices, desirable mechanical properties are of utmost significance. The capacity of these devices to endure strain and stress, particularly with regard to their efficacy as strain sensors, is critical. Consequently, a sequence of uniaxial tensile tests were conducted on the

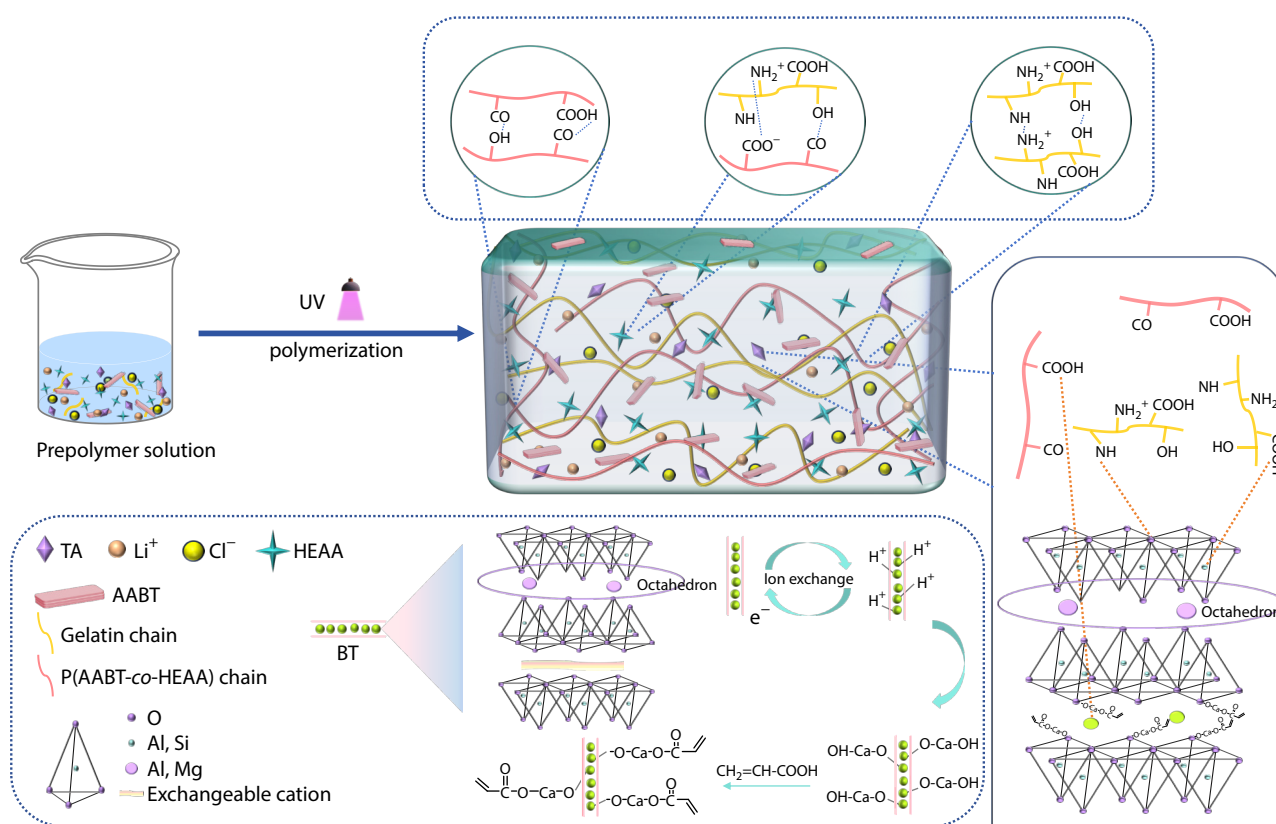


Fig. 1 The preparation mechanism of the ATHG@LiCl hydrogel.

ATHG@LiCl hydrogel.

First, the influence of gelatin content on the mechanical properties of the hydrogels was examined. Gelatin is a water-soluble colloid with high molecular weight. During cooling, gelatin undergoes a transition from a random-coil conformation to a triple-helix conformation. When incorporated into the hydrogel system, gelatin can reinforce the cross-linking network of the hydrogel, consequently enhancing its mechanical properties.^[32] When the gelatin content was 0 wt%, despite the stress reaching nearly 300 kPa, the strain was less than satisfactory (approximately 2500%), as shown in Fig. 2(a). Subsequently, as the gelatin content increased, the overall mechanical properties exhibited a tendency of initially improved and then decreased. This was attributed to the fact that, as the gelatin concentration was further elevated, a

stress concentration effect might occur within the resultant gel network, thereby leading to a decline in the mechanical properties. Considering both stretchability and tensile force, hydrogels with a gelatin content of 15 wt% were chosen for subsequent experiments. The corresponding stress, elongation, and toughness of the gel were 183, 3330, and 2.91 MJ/m³ (Fig. S1a in the electronic supplementary information, ESI), respectively.

An excessively high crosslink density can decrease the molecular weight of the polymer chains between the crosslink points. Consequently, the sample became susceptible to stress concentration-induced fractures.^[33] Fig. 2(b) illustrates the alterations in the mechanical properties of the ATHG@LiCl hydrogel with varying HEAA monomer content. The best mechanical properties were observed when the

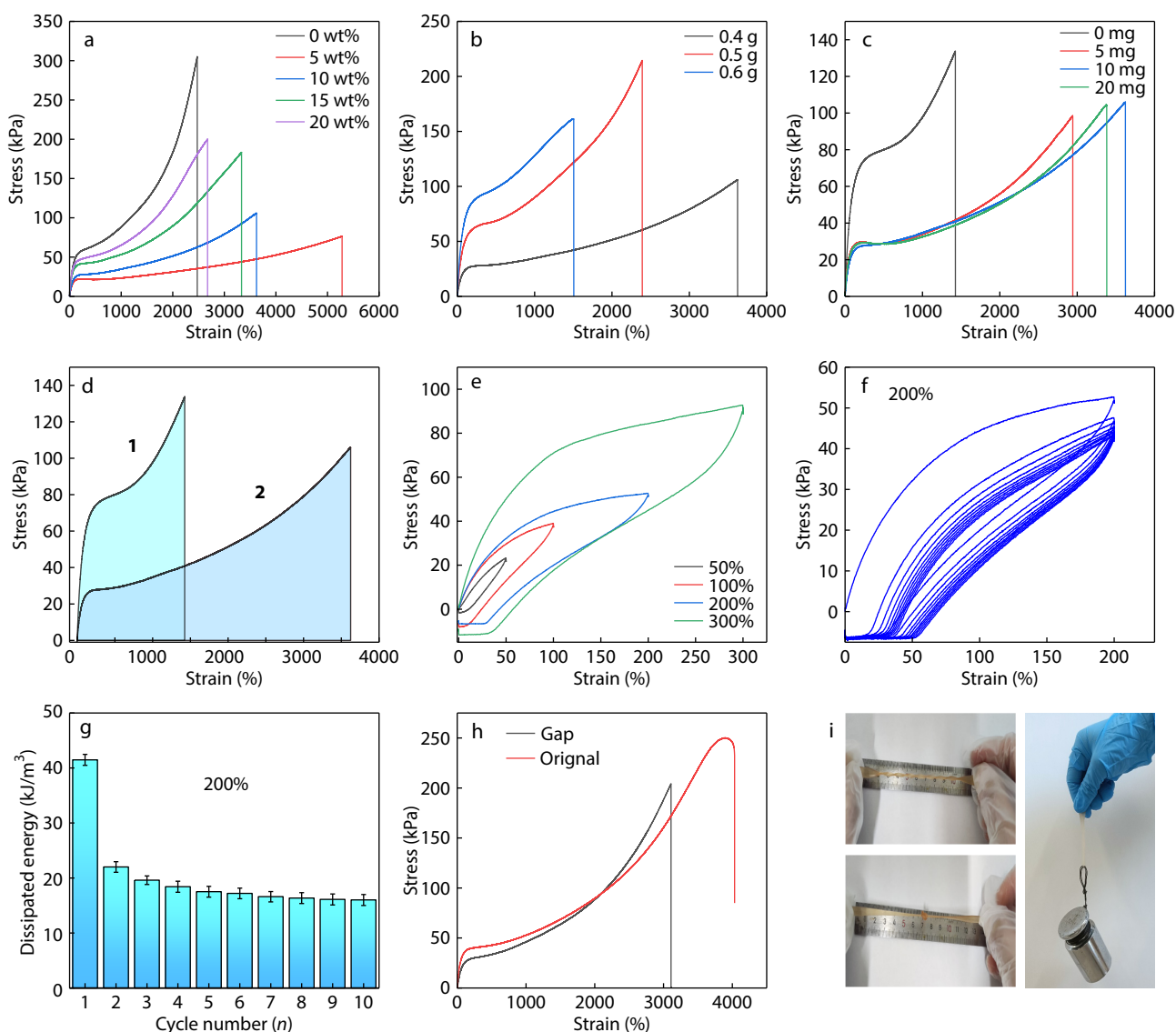


Fig. 2 Mechanical properties of ATHG@LiCl hydrogels. Stress-strain curves of the hydrogels with different contents of (a) Gelatin (0 wt%, 5 wt%, 10 wt%, 15 wt%, 20 wt%), (b) HEAA (0.4, 0.5, 0.6 g), and (c) AABT (0, 5, 10, 20 mg); (d) Toughness of the ATHG@LiCl with AABT (10 mg) and THG@LiCl hydrogel without AABT; (e) The stress-strain curves of the hydrogel under different strains (50%–300%); (f) 10 cycle strain-stress curves (200%) and (g) corresponding energy dissipation; (h) Stress-strain curves of the hydrogel before and after tearing; (i) Photos of ATHG@LiCl hydrogel strips when twisted, knotted and was used for lifting weight.

HEAA monomer was 0.5 g. Under these conditions, the stress was 214 kPa, and the tensile strain was approximately 2500% with a toughness of 110 MJ/m³ (Fig. S2b in ESI). This observation suggests that the relative content of the polymer matrix within the gel has a substantial influence on its mechanical properties. Specifically, the higher the relative content of the polymer, the more challenging it is to induce the movement of its molecular chains by external forces during stretching. Therefore, the tensile strength also increased.^[34] An insufficient amount of HEAA led to low cross-linking density and weak interaction forces. Conversely, an excessive amount of HEAA resulted in brittleness and poor stretchability of the hydrogel.

TA exhibited significant potential for forming multiple hydrogen bonds with substances, such as gelatin, within the system. To explore the impact of TA on hydrogen bond formation in hydrogels, five samples with varying TA content, namely TA₀, TA₅, TA₁₀, TA₂₀, and TA₃₀, were prepared. Uniaxial tensile tests were conducted to obtain tensile stress-strain curves, as shown in Fig. S2(c) (in ESI). The calculated toughness and Young's modulus of the hydrogels with different TA contents are presented in Fig. S2(d) (in ESI). The mechanical properties of the hydrogel were remarkably enhanced when the TA content was 5 wt%. This can be ascribed to the dendritic structure of tannic acid molecules, which possess a large number of hydroxyl groups.

As shown in Fig. 2(c), the addition of AABT led to a noticeable increase in the strain values of the ATHG@LiCl hydrogels, with variations observed across different samples. As depicted in Fig. 2(d), designations **1** and **2** represent the toughness maps of the hydrogels with and without the incorporation of AABT, respectively. The toughness of the hydrogel containing AABT was 110 MJ/m³ (Fig. 2d). This effect can be ascribed to the interactions between AABT and both HEAA and gelatin, which include hydrogen-bonding and coordination effects. Moreover, TA can intercalate into AABT layers, contributing to a more uniform and compact network structure. However, as the amount of AABT increased from 5 mg to 20 mg, the stress and elongation properties of the hydrogel initially increased and then declined. Specifically, the stress improved from 98 kPa to 106 kPa, whereas the elongation increased from 2940% to 3380% with the highest toughness. Based on these findings, AABT with a content of 10 mg was identified as the optimal proportion for achieving the desired performance.

As a result, tannic acid formed stronger hydrogen bonds with gelatin, AABT, and HEAA. Therefore, the final composition of the optimized hydrogel consisted of 10 mg AABT, 0.2 g gelatin solution (15 wt%, 0.5 g of HEAA), 100 μ L TA solution (5 wt%), 100 μ L DMSO, 20 mg of LiCl, and 5 mg of photoinitiator. The fatigue resistance of the ATHG@LiCl hydrogel was assessed using loading and unloading tests. Fig. 2(e) depicts the continuous loading-unloading cycle tests of the hydrogel under different strains (ranging from 50% to 300%), revealing distinct hysteresis loops. It can be seen that the dissipation energy gradually increased with the increase of strain, and the specific range is 2.81 MJ/m³ at 50% strain to 121.6 MJ/m³ at 300% strain, indicating that the energy required for the final synthesized hydrogel during crosslinking fracture gradual-

ly increases with the increase of strain. The presence of a hydrogen bond dynamic crosslinking effect was verified by restoration of the crosslinking effect during the unloading process. The tensile cycling stability of the ATHG@LiCl hydrogel was essential for its electrochemical cycling stability, which is of crucial importance. Figs. 2(f) and 2(g) show 10 consecutive cycles of stretching of the hydrogel at 200% strain and the energy dissipation for each corresponding cycle, respectively. In addition, Figs. S2(f)–S2(i) (in ESI) also present the cycling experiments at 50%, 100%, and 300% strains, along with the energy dissipation and total energy at different strains. As shown in Figs. 2(f) and 2(g), during the first cycle, there was substantial hysteresis and nonrecoverable deformation. However, in the second cycle, the hysteresis loop remained nearly constant, and the nonrecoverable deformation was minimal. This indicated that the stress-strain curve of the ATHG@LiCl hydrogel exhibited excellent repeatability and stability. This further corroborates our finding. Additionally, Fig. 2(h) presents the tearing energy of the ATHG@LiCl hydrogel, with a value of 270.1 J/m², which attested to its excellent tear resistance. Consequently, the ATHG@LiCl hydrogel exhibited an exceptionally high degree of flexibility (Fig. 2i). It was not only capable of being stretched under normal conditions, but could even be twisted and knotted. Notably, the ATHG@LiCl hydrogel could lift a weight of 1 kg, which was 1374 times its own mass. This phenomenon is closely related to the toughness.

Characterization of ATHG@LiCl Hydrogels

The FTIR spectra of the hydrogels were measured to verify the successful synthesis of the gel structure, as depicted in Fig. 3(a). It shows that the characteristic peaks of bentonite (BT) are located at 1031 and 3642 cm⁻¹. This phenomenon is principally associated with the stretching vibration peaks of Si—O and O—H bonds. The asymmetric and symmetric tensile vibration peaks of COO⁻ are located at 1441, 1546 and 1639 cm⁻¹, respectively. The tensile vibration peaks of C—C were located at 1441, 1546 and 1639 cm⁻¹, respectively, indicating the successful synthesis of acrylate bentonite (AABT). The tensile vibration peak of Si—O shifted from 1031 cm⁻¹ to 1040 cm⁻¹ in the direction of higher wavenumbers. This observation suggested that the groups attached to the silicon atoms on the bentonite lamellae exhibited significant steric hindrance. As a result, the asymmetric stretching vibration of the Si—O—Si moiety was impeded. Concomitantly, the sharpening of this peak indicated an alteration in the lattice structure of the material. A plausible explanation is that acrylic acid reacts with the active hydroxyl groups present in the intermediate acrylate-carboxylated bentonite (ACBT). This reaction is exothermic and releases a substantial amount of heat. Concurrently, a portion of the acrylic acid monomer was grafted onto the ACBT laminae. Simultaneously, a fraction of the acrylic monomers was intercalated between the bentonite lamellae. This intercalation event led to an expansion of the interlayer spacing of the bentonite and, in some instances, even induced exfoliation of the bentonite lamellae. These structural alterations consequently modified the lattice structure of the acrylate carboxylated bentonite (ACBT). These observations further corroborate the successful synthesis of acrylate modified bentonite (AABT).

Absorption peaks were observed at 3430 cm⁻¹ and 1451

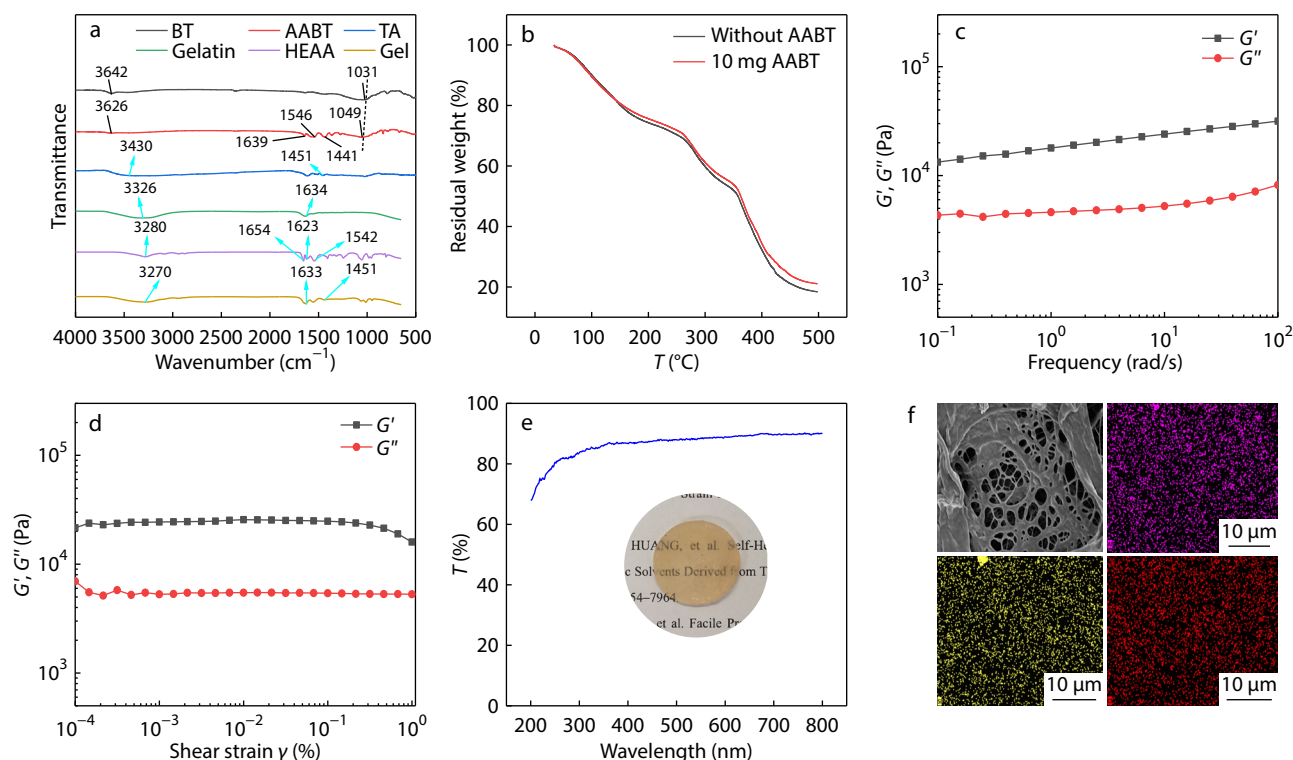


Fig. 3 (a) FTIR spectra of BT, AABT, TA, Gelatin, HEAA, and ATHG@LiCl hydrogel; (b) TG curves of ATHG@LiCl and without AABT hydrogel; (c) Frequency scanning curves of the ATHG@LiCl hydrogel; (d) Shear strain scanning curves of ATHG@LiCl hydrogel; (e) Transparency of ATHG@LiCl hydrogel; (f) SEM and mapping of the ATHG@LiCl hydrogel.

cm^{-1} , corresponding to the stretching vibration peaks of the OH of TA and the C=C of the benzene ring skeleton, respectively. A red shift was observed from 3430 cm^{-1} to 3270 cm^{-1} . This shift indicates that terephthalic acid (TA) engaged in intermolecular interactions with other substances, specifically through the formation of hydrogen bonds. The obtained hydrogel also showed characteristic peaks at 1451 cm^{-1} , which confirmed the successful doping of TA. Gelatin exhibited a characteristic peak at 3326 cm^{-1} , which was primarily attributed to the stretching vibration of the N—H bond in gelatin. The peak at 1634 cm^{-1} represents the characteristic absorption peak of the amide I band of gelatin. This peak was formed as a result of the vibrational absorption contributions from the N—H bond, carbonyl group (C=O), and C—N bond. The characteristic peaks of *N*-2-hydroxyethyl acrylamide (HEAA) carbonyl and C=C were observed at 1654 cm^{-1} and 1623 cm^{-1} , respectively. As shown in Fig. 2(a), the characteristic peak corresponding to the C=C bond of the resultant gel vanished, thereby validating its successful polymerization. The ATHG@LiCl hydrogel exhibited characteristic peaks at 3270 cm^{-1} , predominantly originating from the OH stretching vibration peaks of tannic acid (TA) and gelatin, as well as the N—H stretching vibration peaks of gelatin. This suggests that there was an interaction between HEAA, TA, AABT, gelatin, and other substances, which further confirmed their successful doping.

Fig. 3(b) shows the thermogravimetric analysis (TGA) results of the hydrogels with and without AABT. These data revealed that the introduction of AABT had little effect on the stability of the hydrogel. The viscoelastic properties of the

ATHG@LiCl hydrogel were characterized by rheological testing. The dynamic frequency scanning test results in Fig. 3(c) indicate that the storage modulus G' and loss modulus G'' of the hydrogel are correlated with the frequency change. Moreover, throughout the entire frequency range, the storage modulus G' was significantly higher than the loss modulus G'' . This phenomenon suggests that the hydrogel possesses stable gel-like characteristics. The ATHG@LiCl hydrogel was then subjected to shear strain scanning. As depicted in Fig. 3(d), the hydrogel exhibited a linear viscoelastic region within a strain range of 0.01–100%. As the strain increased, G' of the hydrogel remained consistently higher than G'' . This observation indicates that the hydrogel demonstrated excellent structural integrity. Surprisingly, the hydrogel had good transparency of up to 90%, as shown in Fig. 3(e). As shown in Fig. 3(f), the hydrogel possessed a homogeneous porous structure. The Al, Si, and Cl elements presented in Fig. 3(f) are predominantly sourced from AABT and LiCl. Moreover, C, N, and O were primarily derived from gelatin, HEAA, and TA (Fig. S2 in ESI). The aforementioned elements were uniformly distributed in the mapping, signifying that AABT, TA, and other substances exhibited excellent dispersion within the network.

Electrical Conductivity

The electrical conductivity of hydrogels plays a pivotal role in their use as strain sensors. In this study, the electrical conductivity of a hydrogel and its influencing factors were investigated via alternating current (AC) impedance testing. The introduction of AABT not only improved the mechanical properties of the hydrogel but also significantly increased its electrical conductivity.

As depicted in Fig. 4(a), as the content of AABT increased, the conductivity of the ATHG@LiCl hydrogel exhibited a pattern of initially ascending and subsequently descending, thereby forming a "volcano"-shaped broken-line profile. The primary cause for the formation of the "volcano"-shaped broken lines was attributed to the fact that the AABT itself possesses a layered structure. This layered configuration provided a rapid pathway for the unrestrained shuttling of ions, thereby effectively enhancing their migration efficiency. Nevertheless, when the quantity of AABT in the range of 20–30 mg continued to increase, the electrical conductivity of the ATHG@LiCl hydrogel began to decline substantially. This phenomenon was ascribed to the formation of an organic-inorganic network with high cross-linking density. This network was established between a substantial amount of AABT and substances, such as HEAA and gelatin, within the system. This network impedes ion migration to a certain degree. This ultimately results in a reduction in the electrical conductivity of the hydrogel. Consequently, 10 mg of AABT was chosen for subsequent experimental investigation. At this dosage, the electrical conductivity of the ATHG@LiCl hydrogel peaked at 1.53 mS/cm. This value was five-fold higher than that of the ATHG@LiCl hydrogel without AABT and surpassed those of numerous other hydrogels (Fig. 4b).^[35–44]

The incorporation of ions is a crucial factor contributing to the enhancement of hydrogel conductivity. Subsequently, we investigated the impact of varying the LiCl dosage on con-

ductivity. The conductivity of the ATHG@LiCl hydrogel (20 mg LiCl) increased from 0.5 mS/cm to 2.5 mS/cm along with the temperature range of -20°C to 60°C (Figs. 4c and 4d). Figs. 4(e) and S3 (in ESI) present the changes in the electrical conductivity and resistance values of the hydrogel, respectively, as the LiCl content increased. At room temperature, the resistance of the ATHG@LiCl hydrogel initially increases and subsequently decreases as the LiCl content increases. When the LiCl dosage was increased from 0 mg to 30 mg, the room temperature resistances were 1328.9, 396.1, 261.4, and $300.7\ \Omega$, respectively. As expected, the electrical conductivity exhibited an analogous trend. Upon the addition of 20 mg of LiCl, the hydrogel exhibited the minimum resistance value, thereby corresponding to the maximum electrical conductivity. This phenomenon can be ascribed to the following: a relatively low concentration of ions within the system can suitably enhance ionic conductivity. However, when the ion concentration is excessive, an overabundance of ions interact with other substances, giving rise to a more compact network structure. This leads to smaller pore sizes, a reduction in the number of ions traversing the hydrogel matrix, and consequently, a decrease in conductivity.

Fig. 4(f) shows the ATHG@LiCl hydrogel serving as an interconnected conductor within a circuit, with the aim of investigating its conductivity. It was clearly observable that as the strain of the ATHG@LiCl hydrogel increased from 0% to 500%,

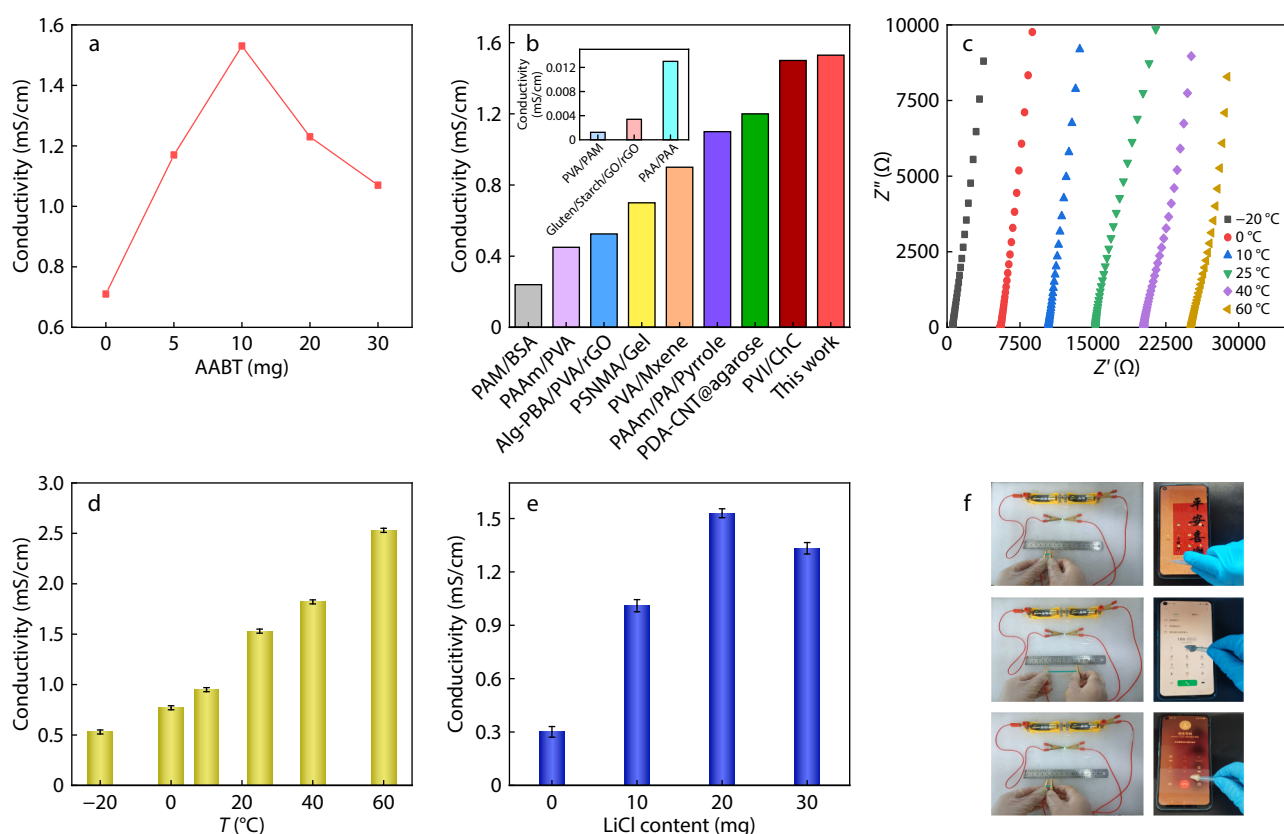


Fig. 4 Electrical properties of the ATHG@LiCl hydrogel. (a) Electrical conductivity of hydrogels with different AABT contents (0, 5, 10, 20, 30 mg); (b) Conductivity comparison at room temperature; (c) Ac impedance spectra and (d) specific values of ATHG@LiCl hydrogels at different temperatures (-20 , 0 , 10 , 25 , 40 , 60°C); (e) The electrical conductivity of hydrogel with different contents of LiCl (0, 10, 20, 30 mg); (f) Some photographs demonstrating the electrical conductivity of the ATHG@LiCl hydrogel.

the luminance of the small bulb in the circuit decreased markedly. Conversely, when the strain in the ATHG@LiCl hydrogel was altered in the reverse direction, the luminance of the LED lamp gradually recovered, demonstrating excellent stability. The underlying cause is that the porous network within the ATHG@LiCl hydrogel serves as a conduit for ion transport. As the strain increases, the conduction pathway becomes constricted and elongated. This makes the migration of ions more challenging. Conversely, upon decreasing the strain, the conductivity of the ATHG@LiCl hydrogel recovered instantaneously. The migration of ions within the ATHG@LiCl hydrogel was regulated by strain-induced deformation. This characteristic endows the hydrogel with high strain sensitivity, rapid response, and favorable electrochemical stability. Notably, owing to its high conductivity, the ATHG@LiCl hydrogel has the potential to function as an electronic skin, emulating the functions of human skin. Upon contact with wood, the phone screen was unable to respond because of its nonconductive nature. Conversely, upon coming into contact with a mobile phone, the hydrogel could manipulate the phone flexibly and successfully place a call. These outstanding properties illustrate the significant potential of the ATHG@LiCl hydrogel for applications in human-computer interactions and wearable electronic devices.

Performance of Adhesion

Flexible wearable electronics demand not only excellent flexibility and electrical conductivity but also impose stringent requirements on material adhesion. This is because material adhesion is essential for interface connectivity, which in turn is associated with the interface impedance and signal integrity.^[45]

Figs. 5(a) and 5(b) show the adhesion schematic and the mechanism diagram of the ATHG@LiCl hydrogel, respectively. Specifically, it was a process in which a ATHG@LiCl hydrogel with an adhesion area of 1 cm × 1 cm was sandwiched be-

tween two identical substrates and then tested on a tension machine. Owing to its abundance in amino groups, hydroxyl groups, and various ions, the ATHG@LiCl hydrogel can establish strong adhesion with diverse substrates *via* non-covalent interactions, including hydrogen bonding, electrostatic interactions, and metal complexation. Fig. S4 (in ESI) presents a photograph of the ATHG@LiCl hydrogel firmly adhering to a variety of materials, such as metal, glass, plastic, wood, and pigskin. This clearly demonstrates the broad applicability of the ATHG@LiCl hydrogel to diverse substrates. To quantitatively assess the adhesion characteristics of the ATHG@LiCl hydrogel, a shear test was conducted to measure the adhesion forces on the various materials. Fig. 5(c) showed that the cogel strongly adhered to several common substrates. The adhesion strengths to copper, iron, glass, plastic, wood, and pigskin were 41.3, 108.6, 75.8, 91.5, 183.6 and 29.4 kPa, respectively. The type and number of surface groups varied among different substrates, which in turn resulted in differences in the adhesion of the ATHG@LiCl hydrogel. Among these substrates, the adhesion force to wood was the greatest, and the adhesion force to pigskin was 29.4 kPa as well. This adhesion performance provided favorable conditions for the ATHG@LiCl hydrogel to be utilized as a strain sensor. Fig. 5(d) shows the bond strength after 24 h of placement. It was clearly observable that the bond strength increased significantly after 24 h of dehydration. Adhesion to wood reached as high as 1.8 MPa.

In addition, owing to the presence of tannic acid, it has a large number of catechol groups, which also contributes to the high adhesion. Fig. 5(e) depicts the adhesion strength of 100 μ L of tannic acid (TA) solutions with diverse mass fractions (10%, 20%, and 30%) after 24 h of adhesion to various substrates. The adhesion strength increased with increasing TA content. This phenomenon suggests that the hydrogel linkages established between the substrate and catechol moi-

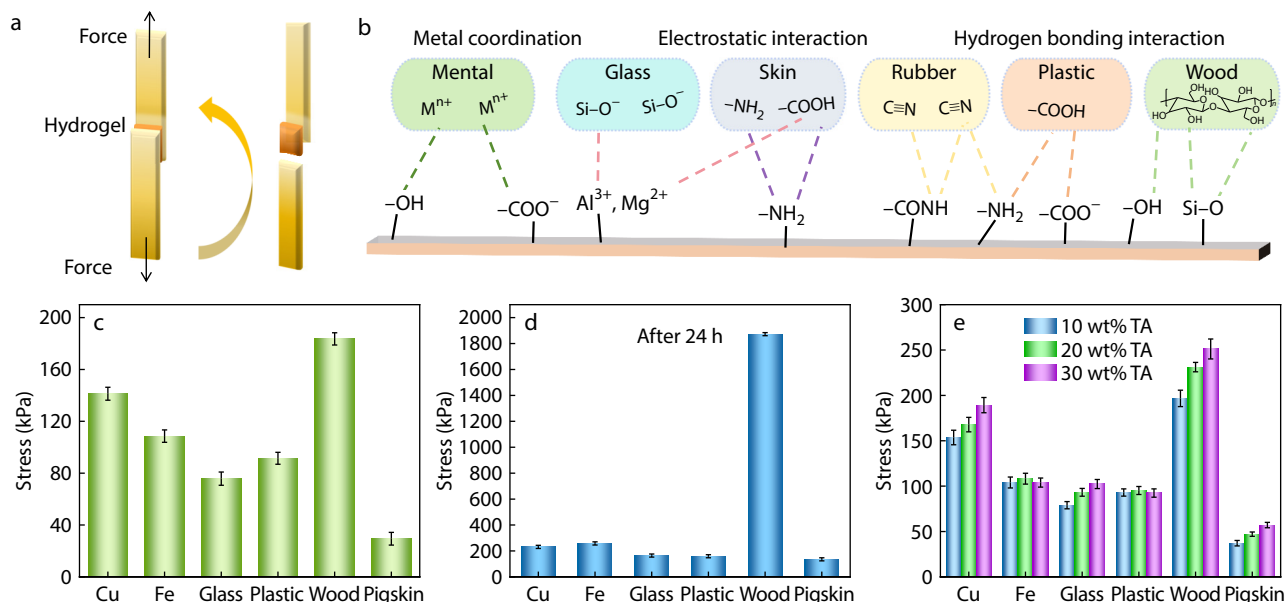


Fig. 5 (a) The adhesion diagram of the hydrogel; (b) Adhesion mechanism of hydrogel to different substrates; The instantaneous (c) and after 24 h adhesion strength (d) of the ATHG@LiCl hydrogel to different material surfaces; (e) The transient adhesion strength of ATHG@LiCl hydrogel with different TA contents (20 wt%, 30 wt%, 40 wt%).

eties of TA played a crucial role in enhancing the adhesion property. Considering all aspects, such as the tensile properties of the hydrogel and the material cost, we selected the hydrogel with 5 wt% TA content.

Frost Resistance

The synthesized ATHG@LiCl hydrogel exhibited remarkable cryoprotective properties. This characteristic endows it with distinct advantages under low-temperature conditions, thereby broadening its scope of application. To gain an intuitive understanding of the cryoprotective ability of the ATHG@LiCl hydrogel, a comprehensive investigation of its mechanical properties, optical transparency, electrical conductivity, and adhesion characteristics at low temperatures was conducted.

As depicted in Fig. 6(a), the fabricated gel demonstrated the capacity to retain its flexibility at room temperature and remained flexible even at a temperature of -20°C . It is capable of enduring diverse deformations, including stretching, torsion, and bending, without incurring fracture damage. Furthermore, it was distinctly observed that the LED bulb sustained a certain level of brightness subsequent to exposure to a low temperature environment (-20°C) for 24 h. This finding verified the persistence of a conductive pathway within the ATHG@LiCl hydrogel in such a rigorous environment, thereby

highlighting its outstanding low-temperature resistance. Next, we demonstrated its stable mechanical flexibility by testing its stretching behavior at different temperatures. As presented in Fig. 6(b), the hydrogel demonstrated the ability to be elongated to 15-fold its original length at a temperature of -20°C . Correspondingly, the fracture strengths measured under these conditions were 0.3 MPa. Remarkably, even at a temperature of -60°C , the elongation at break of the hydrogel attained 700%, and its toughness reached 7.73 MJ/m^3 , as depicted in Fig. 6(c). Fig. 6(d) presented a further quantitative characterization of the ionic conductivity of the ATHG@LiCl hydrogel by means of electrochemical impedance spectroscopy (EIS). Fig. 6(d) indicates that at -20°C , the resistance of the ATHG@LiCl hydrogel was $754.7\ \Omega$. Through calculations, its conductivity was determined to be 0.53 mS/cm . Notably, its electrical conductivity is still higher than that of many other hydrogels at low temperatures.^[46–48] This value is lower than the conductivity at room temperature. The underlying reason is that, as the temperature decreases, the mobility of ions within the polymer network diminishes, consequently leading to a reduction in the conductivity of the hydrogel.

Interestingly, the ATHG@LiCl hydrogel retained its adhe-

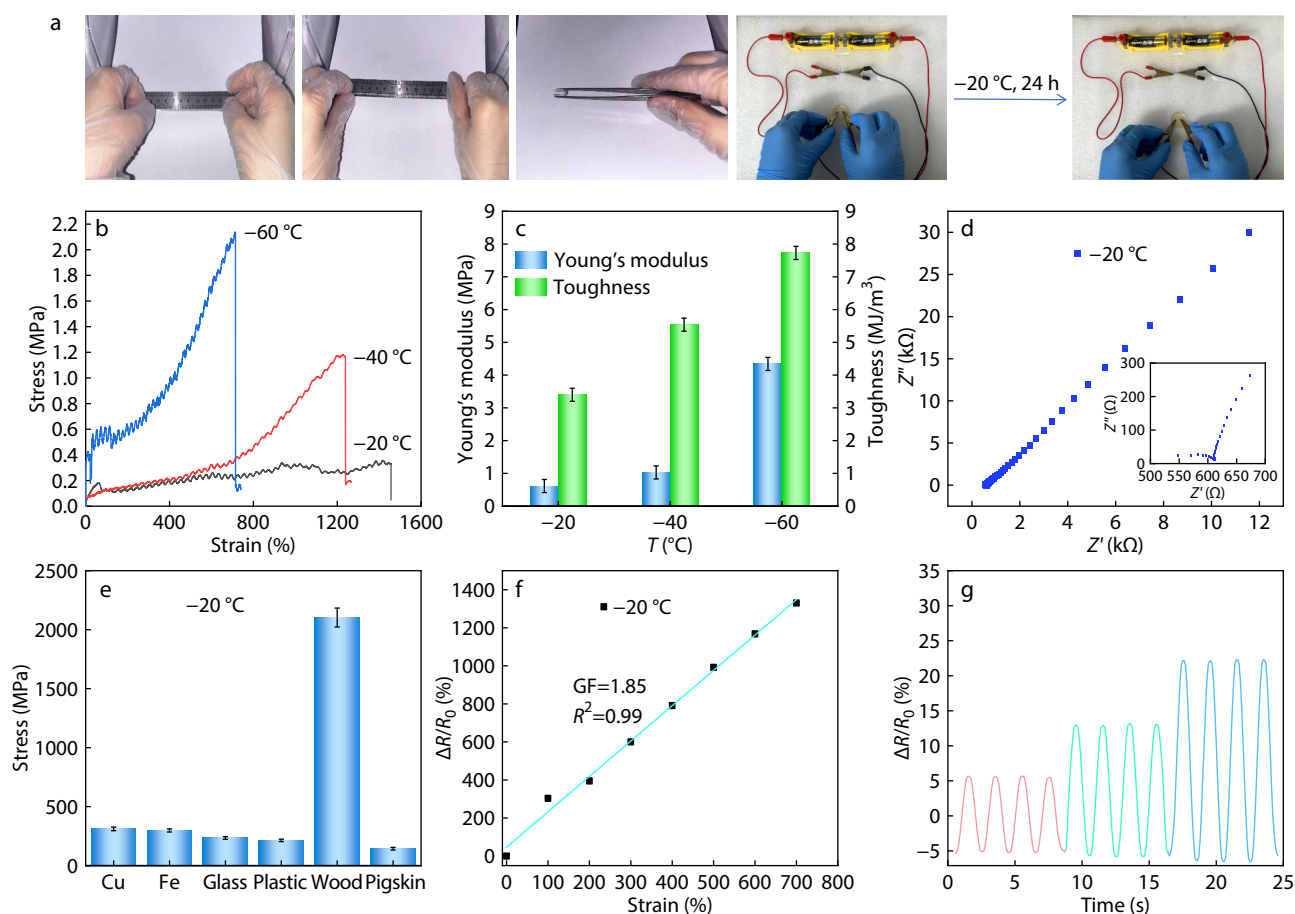


Fig. 6 Freezing resistance of the ATHG@LiCl hydrogel. (a) The stretching, bending and twisting images of the hydrogel at -20°C and the comparison of LED brightness before and after the ATHG@LiCl hydrogel was placed at -20°C for 24 h; (b) Stress-strain curves and (c) corresponding Young's modulus and toughness at low temperature (-20 , -40 , -60°C); (d) The conductivity of hydrogel at -20°C ; (e) Adhesion of the hydrogel to different substrates at -20°C ; (f) Sensitivity of hydrogel at -20°C ; (g) Sensing performance of ATHG@LiCl hydrogel at -20°C .

sion capability, even at low temperatures. The material was exposed to a variety of substrates such as metals, rubber, pigskin, and plastic. Subsequently, it was placed at $-20\text{ }^{\circ}\text{C}$ to evaluate its adhesive strength. As can be discerned from Fig. 6(e), this value is notably higher than that at room temperature. This phenomenon could be ascribed to the strengthened hydrogel bonding between the substrate and the gel at $-20\text{ }^{\circ}\text{C}$. Among these substrates, its adhesion to wood attained an impressive 2.1 MPa, and its adhesion to pigskin surpassed 100 kPa, reaching 143.4 kPa. This adhesion performance unveiled the potential for its utilization as a cryogenic sensor. Sensitivity is a crucial parameter for evaluating the applicability of hydrogels in low-temperature sensing scenarios. In this study, the hydrogel exhibited a sensitivity value of 1.85 at $-20\text{ }^{\circ}\text{C}$, as depicted in Fig. 6(f). This result validated its potential for low-temperature sensing applications.

The excellent durability and fatigue resistance of the strain sensor are shown in Fig. 6(g), which demonstrates the resistance changes exhibited by the hydrogel attached to the rubber tube at bending angles of 30° , 60° , and 90° at $-20\text{ }^{\circ}\text{C}$. In summary, the aforementioned results demonstrate that the ATHG@LiCl hydrogel strain sensor has broad application prospects in freezing environments.

Strain Sensing Performance and Application of the ATHG@LiCl Hydrogel

Owing to the excellent mechanical properties, high electrical conductivity, strong adhesive strength, and remarkable low-temperature resistance of the ATHG@LiCl hydrogel, it was anticipated to serve as a highly sensitive and stable flexible strain sensor.

Subsequently, we evaluated its performance as a strain sensor, with a focus on assessing its sensitivity. The relationship between the relative resistance change ($\Delta R/R_0$) and strain was fitted using linear regression to determine the gauge factor (GF). Specifically, the GF value was calculated from the slope of the linear fit of $\Delta R/R_0$ with respect to strain.^[49]

Fig. 7(a) depicts the changes in the relative resistance of the ATHG@LiCl hydrogel under diverse strain conditions. Owing to the homogeneous and stable micronetwork architecture of the hydrogel, the gauge factor (GF) of the measured ATHG@LiCl hydrogel exhibited a distinct linear correlation with the strain.^[50] Evidently, within the strain ranges of 0%–400% and 400%–700%, the gauge factor (GF) values of the synthesized cogel strain sensor were 2.86 and 4.50, respectively. This clearly demonstrates that the strain sensor possesses a high gauge factor and broad strain-sensing range.

Furthermore, among the crucial performance parameters of the designed hydrogel sensor, response time is of significant importance. As presented in Fig. 7(b), the response and recovery times of the cogel strain sensor during the loading and unloading processes at 100% strain were measured to be 0.1 s. This rapid response indicates the capacity of the ATHG@LiCl hydrogel for the real-time monitoring of the sensing signal. Subsequently, a detailed investigation of the cyclic stability of the cogel under diverse strain conditions was conducted. This involved an examination of the relative resistance variations ($\Delta R/R_0$) for both large strains, ranging from

100% to 400%, and small strains, specifically 1%, 5%, and 10%, as shown in Fig. 7(c).

Remarkably, the ATHG@LiCl hydrogel sensor exhibited a distinct electronic signal, even when subjected to 1% strain. This finding revealed that its detection limit was as low as 1% strain, demonstrating outstanding repeatability and stability.

In addition to its excellent thermal stability, remarkable stretchability, and outstanding sensitivity, the synthesized ATHG@LiCl hydrogel also possesses the advantages of being non-toxic and harmless upon contact with the human body. This is attributed to the green and pollution-free properties of the gelatin polymer. Its ability to function as a wearable strain sensor was verified by fixing the ATHG@LiCl hydrogel sensor.

Figs. 7(d)–7(i) depicted the real-time monitoring of relative resistance alterations by the ATHG@LiCl hydrogel sensor during body movements at various anatomical sites of the human body. The precise deformation-sensing capability of the hydrogel was evaluated through cyclic finger bending at different angles, and the degree of variation in the relative resistance was analyzed. As depicted in Fig. 7(d), the relative resistance of the hydrogel exhibited a gradual increase with progressive bending angles of the finger, specifically from 30° to 60° and then to 90° . This phenomenon can be attributed to the elongation of the hydrogel as the fingers bend. With an increase in the length of the hydrogel, its resistance correspondingly increases according to the fundamental principles of electrical resistance, where resistance is directly proportional to the length of the conducting material under constant cross-sectional area and resistivity conditions.

In addition, their ability to monitor other parts of the body was tested. For example, the relative resistance changed in the state of wrist flexion and knee extension (Figs. 7e and 7f). The hydrogel sensors exhibited the potential to monitor subtle physiological movements, including facial expressions such as frowning and smiling, as well as physiological signals such as the heartbeat. As depicted in Figs. 7(g)–7(i), the corresponding real-time signals exhibited distinct and stable characteristics. This further enhances the sensitivity of the hydrogel sensor. In conclusion, the above results demonstrate that the ATHG@LiCl hydrogel sensor fabricated in this study exhibits rapid, reproducible, and stable responsiveness. Moreover, it displays outstanding capabilities in monitoring human motion and transmitting information, rendering it an ideal candidate for flexible wearable strain sensors.

Biocompatibility Evaluation

The biocompatibility of the ATHG@LiCl hydrogel was systematically evaluated using a Cell Counting Kit-8 (CCK-8) assay (Fig. S5 in ESI). At concentrations of 5 and 10 $\mu\text{g/mL}$, cell viability remained high at 99% and 96%, respectively, after 3 days of culture. Notably, even at a higher concentration (20 $\mu\text{g/mL}$), the hydrogel maintained 94% cell viability over the three-day period, demonstrating the excellent cytocompatibility of the hydrogel.

CONCLUSIONS

In summary, we developed an ATHG@LiCl hydrogel prepared by free radical polymerization of HEAA monomers in the presence of gelatin, tannic acid, AABT, and LiCl. Among these compo-

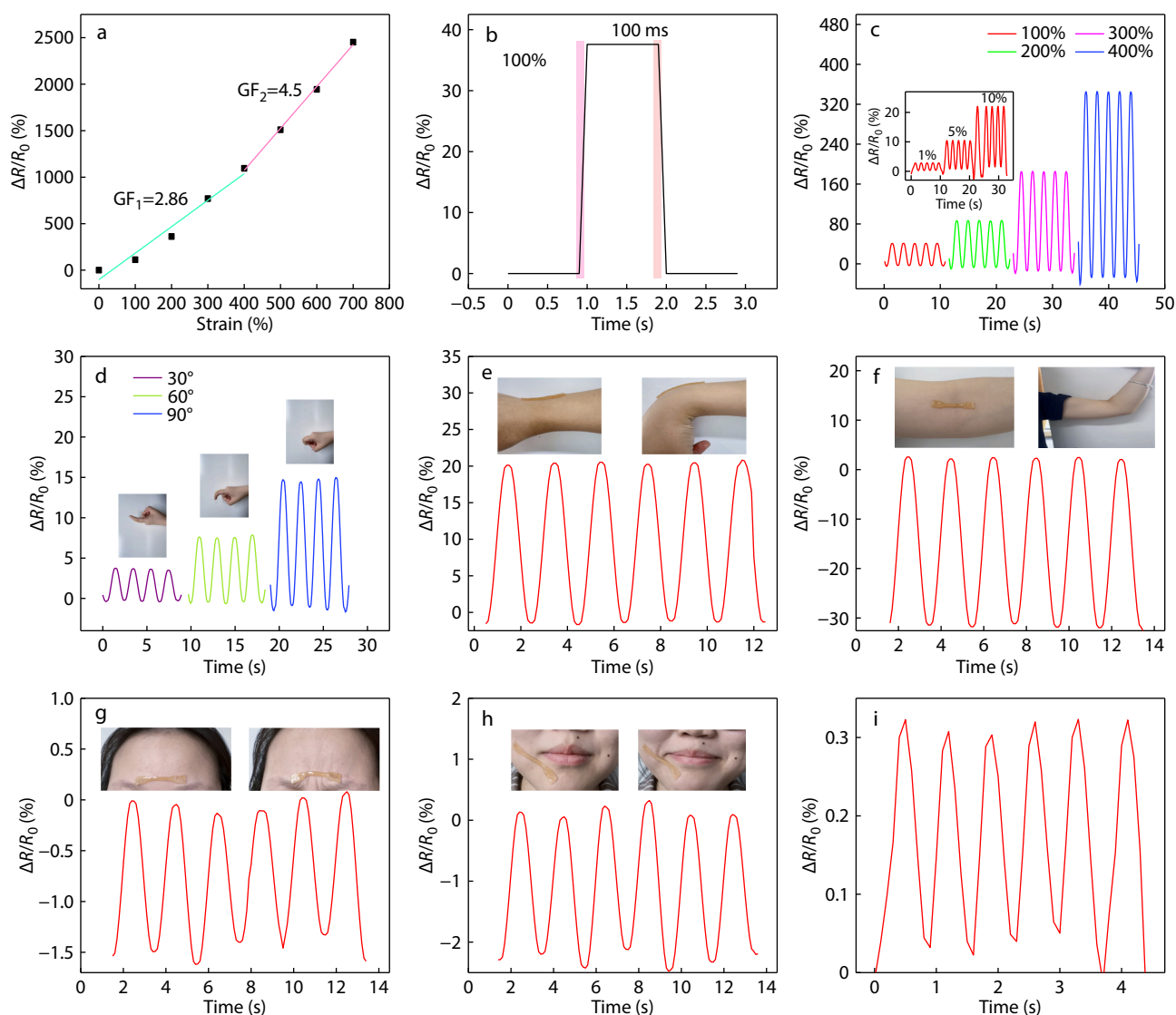


Fig. 7 Strain sensing performance and signal monitoring sensor of ATHG@LiCl hydrogel. (a) The sensitivity of the ATHG@LiCl hydrogel under different strains; (b) Tensile and recovery response times of electrical signals under 100% strain; (c) The sensing performance of ATHG@LiCl hydrogel under different tensile strains (including large strain: 100%–400% and small strain: 1%, 5%, 10%, 20%); Flexion of finger (d) and wrist (e), elbow (f), frowning (g), smile (h), and heartbeat (i) were monitored.

nents, gelatin, which has abundant active functional groups, can enhance the stability of the ATHG@LiCl hydrogel. This is achieved by interacting with other substances through non-covalent bonds, including hydrogen bonds and electrostatic interactions. The incorporation of AABT augmented the mechanical characteristics of the composite gel and enhanced the electrical conductivity of the ATHG@LiCl hydrogel. This enhancement in conductivity can be attributed to the layered structure of AABT. Simultaneously, tannic acid (TA) can be homogeneously dispersed within the interlayers of AABT, further strengthening the intermolecular interaction forces between them. The results show that the prepared gel strain sensors have good tensile properties (approximately 4000%), high transparency (90%), excellent ionic conductivity (up to 2.53 mS/cm), and a wide operating temperature range (−20 °C to 60 °C). At the same time, the addition of LiCl and DMSO give the cogel excellent freezing resistance, and the LED light could still be lit even when placed at

−20 °C for 24 h, solving the problem of the conductive hydrogel freezing at low temperatures. Therefore, the ATHG@LiCl hydrogel possesses comprehensive mechanical and strain-sensing properties, superior to many kinds of inorganic-organic hybrid hydrogels reported in the literature (Table S6 in ESI).^[51–55] In particular, its adhesion force, ranging from 29.4 kPa to 183.6 kPa, meets the requirements for wearable electronic devices. Collectively, these results suggest that the ATHG@LiCl hydrogel has potential as an artificial e-skin, emulating the functionality of human skin. In the context of the future wearable sensing domain, it can be used to monitor human movement, thereby facilitating human exercise training and health management. Finally, it is expected to obtain gel with more excellent mechanical properties through confined polymerization in layered bentonite with qualitative and regular arrangement; Through structural optimization of olefin derivatives, the adaptability between organic and inorganic components is enhanced, which is expect-

ed to further improve various performance of gel.

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/s10118-025-3407-6>.

Data Availability Statement

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

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