

Flexible Mussel-inspired Hydrogel with Polyaniline and Polypyrrole for Transparent Wearable Strain Sensors: Investigation of Physical Properties, Self-healing, and Electrical Conductivity

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Electronic Supplementary Information

Abstract Conducting hydrogels have garnered significant interest in the field of wearable electronics. However, simultaneously achieving high transparency, high conductivity, strong adhesion, and self-healing ability within a short time remains a major challenge. In this study, a multifunctional mussel-inspired hydrogel was synthesized in only 5 min, with polydopamine (PDA)-polypyrrole (Ppy)-polyaniline (PANi) and poly(vinyl alcohol) (PVA) nanoparticles incorporated into the polyacrylamide (PAM) network. The resulting hydrogel exhibited high transparency (about 90% light transmission in the range of 400–800 nm), high conductivity ($(95.4 \pm 0.4) \times 10^{-4}$ S/cm), tensile strength (32.60 ± 1.03 kPa), strain at break ($904.46\% \pm 11.50\%$), and adhesive strength (30–60 kPa). It also demonstrated rapid self-healing properties (about 48% strength recovery within 1 h at 50 °C) and water-dependent shape memory behavior. As a wearable strain sensor, the hydrogel successfully detected finger flexion, wrist movements, facial expression changes, and breathing with high sensitivity and stability. The calculated gauge factor (GF) was 7.44 ± 0.31 , which is higher than that of many previously reported hydrogels. Compared with previous oyster-inspired or Ppy-based hydrogels, our system showed a much shorter synthesis time, higher transparency, and enhanced multifunctionality. These findings highlight the potential of the proposed hydrogel for next-generation flexible electronics, e-skin, and biomedical monitoring devices.

Keywords Electrical conductivity; Mussel-inspired hydrogel; Adhesive hydrogel; Flexible hydrogel

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INTRODUCTION

Recent societal advancements have significantly improved the quality of life, reshaped utilization patterns, and paved the way for the adoption of modern technologies. Among these advancements, wearable devices have become noticeable and have dominated healthcare, evolving from single-purpose devices to multifunctional instruments, including advanced sensors.^[1,2] Hydrogel technology has evolved into a crucial tool for biosensing applications within the medical field.^[3,4]

Hydrogels are colloidal polymeric chains with a three-dimensional cross-linked arrangement. They are hydrophilic, biocompatible, highly flexible, mimic normal tissues, and extremely water-swelling polymer networks capable of converting chemical energy into mechanical energy and responding to external stimuli.^[5,6] Hydrogels exhibit several desirable properties, including high sensitivity, flexibility, conductivity, mechanical strength, the ability to associate with biological tissues, responsiveness to stimuli, and resistance to

fouling.^[7,8] These properties make hydrogels ideal for various applications, including biosensing, tissue engineering scaffolds, wound dressings, and drug delivery platforms. Hydrogels enable early diagnosis, treatment, and disease management in biosensing. In other biomedical applications, they provide structural support, promote healing, and ensure controlled therapeutic delivery.^[9,10]

Most wearable strain hydrogel sensors are not directly attached to the human body and require additional adhesives such as tape.^[11] The lack of adhesion performance hinders the operation steps and increases interface delamination and friction between the sensor and skin, which is not conducive to detecting bioelectric signals.^[12] Ocean animals and organisms exhibit a high level of adhesion to various materials, comparable to artificial innovations such as mussels, sandcastle worms, and ocean barnacles.^[13,14] Mussels are particularly remarkable among these organisms because of their ability to strongly adhere to both organic and inorganic surfaces under wet conditions. This adhesion is primarily attributed to mussel foot proteins (Mfps), especially Mfp-3 and Mfp-5, which contain the amino acid 3,4-dihydroxyphenylalanine (DOPA), which introduces catechol functional groups and enables strong and reversible adhesion.^[15,16]

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Propelled by the chemistry of mussels, dopamine-based adhesives have gained widespread use as viable coatings.^[17] For this purpose, Huang *et al.* created a conductive and adhesive hydrogel by combining polydopamine (PDA) and carbon fibers into a polyacrylamide (PAM) hydrogel. Their research revealed that the most notable conductivity of the hydrogels was 0.394 S/m, and the greatest adhesive strength observed between the hydrogel and paper was (23.99 ± 1.08) kPa.^[18] Inspired by marine mussel-secreted protein-based adhesives^[19], researchers have prepared self-adhesive hydrogels using polydopamine and tannic acid because they possess adhesion groups similar to catechol.^[20,21]

As a typical soft material with a structure similar to that of biological tissues, conductive hydrogels are ideal candidates for smart wearable sensor devices because of their good conductivity, viscoelasticity, and high flexibility.^[22] Many conductive materials, such as metal-based materials,^[23] carbon-based materials,^[24] liquid metals,^[25] and ionic liquids,^[26] have been incorporated into hydrogels to synthesize wearable sensors. However, these conductive materials are prone to leakage from the hydrogel matrix owing to insufficient sealing, which limits their practical application in conducting hydrogels.^[27] Conjugated π -conductive polymers exhibit high conductivities, biocompatibilities, and mechanical flexibilities. They have attracted considerable attention in the field of conductive hydrogel sensors owing to their high cross-linking ability, which hinders leakage from the network.^[28] Polypyrrole (Ppy) and polyaniline (PANI), typical conjugated π -conductive polymers, have been widely explored in conductive hydrogel sensors because of their ease of preparation, strong stability, and high electrical conductivity.^[24,29] Nonetheless, Ppy and PANi is difficult to disperse Ppy and PANi entirely in water because of their inherently conjugated heterocyclic structures, which weaken the conductivity of Ppy and PANi in practical applications.^[30] In addition, the inherent dark color of Ppy and PANi leads to opacity of the conductive hydrogels, limiting their application in wearable sensors. Therefore, the preparation of Ppy- and PANi-based wearable hydrogel sensors with synergistic conductivity and transparency remains a challenge.

The arrangement of conductive hydrogels utilizing PDA-Ppy-PANI nanofillers and *in situ* polymerization created a completely conductive path and high transparency in the produced hydrogels. Specifically, PDA, inspired by mussel adhesion, enhances the hydrophilicity and dispersion of conjugated polymers, PVA contributes to hydrogen bonding and improves self-healing ability, PANi and Ppy provide electrical conductivity, and PAM forms a flexible 3D polymeric network. In other words, using polydopamine inspired by mussels due to catechol groups increases the hydrophilicity of conductive polymers and prevents their aggregation.^[31] For example, Lu *et al.* reported a mussel-inspired strategy for manufacturing conductive hydrogels based on a PDA-Ppy composite in hydrogel networks through *in situ* polymerization, resulting in hydrogels with good conductivity, adhesion, and mechanical properties.^[32] Although they succeeded in planning the hydrogel, a lengthy preparation process (9 h) and a three-day aging time may hinder its industrial application until a transparent hydrogel is achieved.

This work represents significant advances compared to previous studies, which often suffer from long synthesis times (several hours to days) or trade-offs between transparency, adhesion, and mechanical strength. For example, Jia *et al.* demonstrated hydrogels that are conductive, stiff, transparent, and self-healing, but still require relatively long *in situ* polymerization steps.^[33] Yuan *et al.* reported a rapidly synthesized tough, conductive, and transparent hydrogel; however, their method focused on specific redox-based strategies and may not cover all aspects of adhesion/self-healing.^[34] Roy *et al.* developed a bioadhesive hydrogel with excellent stretchability, conductivity (about 0.29 S/m), transparency, and tissue adhesion (about 27 kPa) for wearable sensors, but combining ultrafast synthesis with all these properties remains rare.^[35] Xu *et al.* introduced a rapid gelation system that achieved mechanical strength, conductivity, and self-healing, but did not fully address the trade-offs between adhesion and transparency.^[36] Furthermore, studies on hydrogel adhesion show that achieving strong and long-term adhesion, conductivity, and mechanical durability is still a major challenge in this field.^[37]

In contrast, our method allows for the ultrafast synthesis of transparent hydrogels in only 5 min, significantly reducing the processing time. By using PDA to improve spreadability and adhesion and leveraging the synergistic effects of Ppy and PANi to create coherent conductive networks, we combine high conductivity, transparency, adhesion, and self-healing in a single system. Achieving this complete set of properties simultaneously is rare in previous studies, making our hydrogel a promising candidate for next-generation wearable sensors.

In this study, highly conductive, transparent, and flexible CHs based on PDA, Ppy, and PANi were prepared in a short time (5 min). Investigations showed that CHs exhibit self-healing behavior and shape memory effects, which facilitates their use as transparent hydrogels in electronic applications. CHs with Ppy, PANi and Ppy-PANI showed a high electrical conductivity of $(27.00 \pm 0.12) \times 10^{-4}$, $(30.40 \pm 0.50) \times 10^{-4}$ and $(95.40 \pm 0.40) \times 10^{-4}$ S/cm, respectively. After optimizing the concentration of the conductive material in the hydrogel, the possibility of using CHs as sensors to detect joint motion was evaluated.

The key innovations of this study are summarized as follows: (i) ultra-fast synthesis of transparent hydrogels within 5 min compared to hours or days in previous studies; (ii) simultaneous achievement of high conductivity, transparency, adhesion, and self-healing; (iii) elucidation of swelling behavior and shape-memory mechanisms; and (iv) demonstration of practical applications as wearable strain sensors for detecting finger bending, wrist motion, facial expressions, and breathing.

EXPERIMENTAL

Materials

All materials, including dopamine hydrochloride (DA), pyrrole (Py), aniline (ANI), acrylamide (AM), ammonium persulfate (APS), *N,N'*-methylene bis(acrylamide) (BIS), poly(vinyl alcohol) (PVA, $M_w = 7.2 \times 10^4$ Da), and ferric chloride ($\text{FeCl}_3(\text{H}_2\text{O})_6$), were purchased from Sigma Aldrich.

Fabrication of PANi-Ppy-PDA

Ppy-PDA was synthesized using the method reported by Lu *et al.* with a few modifications.^[32] We synthesized PANi-Ppy-PDA particles using the following method. First, PVA (1.5 g) was dissolved in deionized water (20 mL) and heated to 90 °C under stirring. Next, the solution was cooled to the ambient temperature. Cu(OAC)₂·H₂O (0.8 g), pyrrole monomer (140 μL), aniline monomer (131 μL), and dopamine monomer (0.05 g) were added to the solution, which was then ultrasonicated for 5 min. To remove impurities, the product was centrifuged and stored at 4 °C. Ppy-PDA and PANi-PDA particles were also synthesized for comparison.

Here, PVA acted as a stabilizer and hydrogen-bonding agent, contributing to the self-healing ability. Cu²⁺ ions catalyze dopamine oxidation and promote polymerization. PDA provided catechol functional groups to improve hydrophilicity and adhesion, whereas PANi and Ppy introduced high electrical conductivity.

Preparation of the Hydrogel

To synthesize these CHs, PANi-Ppy-PDA, PANi-PDA, or Ppy-PDA was dispersed (8 mL) in deionized water (0.026 g). Then, 2.6 g of acrylamide monomers were then added and stirred to form a homogeneous solution. Next, APS (0.104 g) and BIS (0.003 g) were added to the mixture, and the solution was subjected to ultrasonic waves using an ultrasonic bath (30 kHz) for 15 min to form a homogeneous mixture. Gelation was initiated within 5 min, whereas complete homogenization was achieved by applying ultrasonic waves (30 kHz) for 15 min using an ultrasonic bath.

Characterization of the Structure CHs

Fourier transform infrared (FTIR) spectra of PDA-Ppy-PANi containing Cu²⁺ were obtained in the wavelength range of 400–4000 cm^{−1} using an infrared spectrometer (Shimadzu) to provide information on the molecular structure. An ultraviolet-visible spectrophotometer (Shimadzu UV-2550) was used to record the UV-Vis transmission range of the hydrogel test within a wavelength range of 400–800 nm. The microstructures of the hydrogels were analyzed by scanning electron microscopy (FE-SEM, TESCAN MIRA3). Before SEM imaging, the hydrogels were freeze-dried to preserve their porous and fibrous microstructures. Additional characterization of the PDA-Ppy-PANi nanoparticles, including X-ray diffraction (XRD), was performed to confirm the crystallinity and particle size distribution (in the electronic supplementary information, ESI).

Electrical conductivity

CH was cut into square pieces (2 cm × 1 cm × 0.5 cm). CH conductivity was investigated using a four-probe tester. According to the conductivity results, ferric-containing hydrogels were found to be ideal for studying the self-healing effects. The electrical conductivity (σ) was calculated using Eq. (1).^[38]

$$\sigma = 1/\rho = L/(R \times S) \quad (1)$$

where, ρ represent the resistivity of CHs ($\Omega \cdot \text{m}$), L is the distance between adjacent probes (m), R is the relative resistance of CH (Ω), S is the CH cross-section (m²).^[39] All measurements were performed in triplicate, and the data are reported as mean $\pm$ standard deviation (SD).

Mechanical properties and self-healing properties of CHs

The mechanical behavior of CH was investigated using an STM-

150 tensile tester at room temperature. The thicknesses of the CH-Ppy, CH-PANi, CH-Ppy, and PANi stripes is (27 mm × 25 mm × 250 μm). The test results were obtained at a crosshead speed of 4 mm/min. The tensile stress (σ) was calculated as Eq. (2):

$$\sigma = F/A \quad (2)$$

where F is the tensile load and A is the cross-sectional area of the hydrogel. The elongation at break (ϵ) was defined as hundred percent of the extended length (L) divided by the original length (L_0).^[18]

$$\epsilon (\%) = L/(L - L_0) \times 100\% \quad (3)$$

Swelling behavior, self-healing, and water-induced shape memory elastic of CHs

To study the swelling properties of the hydrogels, CH-Ppy, CH-PANi, and CH-Ppy-PANi were completely dried, and the weight of the dried CH (W_0) was recorded. The CH was then immersed in deionized water at 25 °C, and its weight of the CH was recorded at different time (W_t). The amount of water absorbed was calculated using Eq. (4):

$$W (\%) = (W_t - W_0)/W_0 \times 100\% \quad (4)$$

To study the self-healing properties of CH, 4 pieces of CH-Ppy, CH-PANi, and CH-Ppy-PANi were cut and stained with methyl orange, methylene blue, and Rhodamine B, respectively. The pieces of CH were joined and maintained at 50 °C for 1 h. strength after healing was evaluated using a universal testing machine.

A bending test was used to assess the shape-memory effect, as described by Lendlein. Five samples of flower-shaped hydrogels (2 cm in length, 0.5 mm in width, and 0.5 mm in thickness) were bent to an angle (q_0) close to 180 °C to 80 °C and then cooled to room temperature with an external force to maintain the deformation (q_i).^[40] Herein, to investigate the fast water-induced shape memory effect of the hydrogels, after losing water, they were again placed in deionized water at 25 °C for 5 s to return to their original state. The underlying mechanism of shape-memory was attributed to the reversible hydrogen bonding of PVA, dynamic catechol-metal coordination of PDA-Cu²⁺, and elastic recovery of the PAM network.

Electromechanical Test of CHs

Resistance measurements during stretching were performed using an impedance analyzer to investigate the application of CHs as motion sensors. For stretching, the samples were cut to dimensions of 10 mm × 40 mm × 2 mm and connected to two copper electrodes. All the measurements were performed at a constant frequency of 2 kHz with a voltage in the range of 2 V. The shape change of the CHs sensor in the test is expressed by the change in the relative resistance ($\Delta R/R$, %), and its calculation formula is as follows:

$$\Delta R/R (\%) = (R - R_0)/R_0 \times 100\% \quad (5)$$

where R represents the real-time recorded relative resistance of the CHs (Ω) when stress is applied and R_0 represents the initial relative resistance (Ω) of the CHs sensor when no stress is applied.^[41]

Additionally, the gauge factor (GF) was calculated to compare the effects of CH-Ppy, CH-PANi, and CH-Ppy-PANi on the sensitivity of the hydrogels. Herein, ϵ is the strain, and R_0 and

R are the resistance at 0% strain and tensile strain, respectively [42].

$$GF = ((R - R_0)/R_0)/\varepsilon \quad (6)$$

All electromechanical measurements were conducted in three independent replicates, and the results are presented as the mean $\pm$ SD.

Motion-sensing Tests

To demonstrate the application of CHs as wearable sensors, additional tests were conducted on human subjects. The following tests were performed and the resistance changes were recorded.

Facial expression (frowning) test

The hydrogel was placed on the forehead and the resistance change was recorded during frowning. Respiration test: The hydrogel was attached to the abdomen, and the resistance change during normal inspiration and expiration was measured.

Wrist bending test

The hydrogel was mounted on the wrist, and resistance changes were recorded during flexion and extension. Finger bending test: The hydrogel was placed on a finger joint and resistance changes were measured during finger bending and straightening.

These tests confirmed the high sensitivity of CHs sensors to subtle human motions, including facial expressions, breathing, and joint movements.

RESULTS AND DISCUSSION

Design Strategy for the CHs

Ppy and PANi have been introduced as excellent conductive polymers for sensing applications; however, these polymers have problems owing to their hydrophobicity.^[43,44] Ppy and PANi nanoparticles doped with PDA were prepared using Cu^{2+} as an oxidant to increase hydrophilicity. Py, ANi, and DA monomers can be doped together through non-covalent interactions such as π - π interactions or hydrogen bonding. Acrylamide monomers were mixed with PDA-Ppy-PANi, and APS, as a radical initiator, formed fibers along the acrylamide chains, as confirmed by FE-SEM. As a result, conductive fibers were formed inside this flexible hydrogel, providing a transparent hydrogel. Unlike rigid metallic sensors, flexible sensors based on hydrogels can convert subtle mechanical deformations into electrical signals with a higher sensitivity. Therefore, our strategy was to integrate the conductivity of conjugated polymers with the flexibility and transparency of the hydrogels to achieve multifunctionality. However, our efforts have been to combine the exceptional conductivity of metals and hydrophobic polymers with the flexibility of hydrogels to create a sensitive and flexible sensor.^[43,45] Owing to the incomplete oxidation of catechol groups in DA, they are responsible for the adhesion of the hydrogel to different surfaces.^[32,46] Additionally, the $-\text{OH}$ groups in the PVA chains can form hydrogen bonds with various substrates.^[47] The construction of hydrogels using non-covalent bonds, such as coordination, Hydrogen Bonding, and host-guest interactions of metals, results in reversible and dynamic networks. These interactions can be dissociated and reassociated under the influence of external forces. Therefore, hydrogels can exhibit self-healing properties.^[48,49] Moreover, the synergistic effects of PDA (adhe-

sion and hydrophilicity), PVA (hydrogen bonding and elasticity), and PAM (flexible 3D backbone) contribute to both shape-memory and self-healing abilities. Additionally, using sonication to prepare the hydrogel eliminates the need for radical initiators such as TMEDA, as reported in the literature. The final product was a transparent hydrogel prepared in a short period, as compared to previous studies.^[32]

Optical and Structural Properties of Hydrogels (UV-Vis, SEM and FTIR)

Fig. 1(a) presents the UV-Vis spectra for CH in the range of 400–800 nm for the different polymers. Following the *in situ* formation of PDA-Ppy-PANi fibrils, highly transparent CHs were obtained because of the entanglement of the fibrils, which formed a network that allowed light to pass through. Consequently, the light transmission spectra indicated nearly 100% transmission for these hydrogels, measured at a thickness of 5 mm across wavelengths ranging from 400 nm to 800 nm. Additionally, Fig. 1 shows the transparency of the gels. Figs. 1(c)–1(k) present FE-SEM images of the CHs, revealing the internal structure of the gel.

FTIR spectra were utilized to investigate the chemical structure of samples containing poly(vinyl alcohol) (PVA), dopamine, polyaniline (PANi), and polypyrrole (Ppy) (Fig. 1b). In all the 3 samples, a broad band can be detected in the region of 3400 cm^{-1} , which is associated with the vibrational stretching of the hydroxyl ($-\text{OH}$) groups present in PVA and dopamine. This peak may also overlap with the stretching of amino groups ($-\text{NH}$) found in the structures of polyaniline or polypyrrole.^[50,51]

In the sample containing polypyrrole (red sample), characteristic bands were observed in the regions of 1550 cm^{-1} ($\text{C}=\text{C}$ stretching in the pyrrole ring), 1300 cm^{-1} ($\text{C}-\text{N}$ stretching), as well as bending bands in the region of $800\text{--}600\text{ cm}^{-1}$, confirming the presence of Ppy.^[51] In contrast, in the sample containing polyaniline (green sample), two essential bands can be observed at 1580 and 1490 cm^{-1} , corresponding to the quinoid ($\text{C}=\text{N}$) and benzenoid ($\text{C}=\text{C}$) structures, respectively, which are characteristic of oxidized and reduced polyaniline structures.^[50,52]

In the mixed sample (blue) containing both PANi and Ppy polymers, simultaneous presence of peaks related to both polymers was observed, along with a slight shift in some bands. This could be due to the interactions between the polymer chains or the formation of hybrid structures between them.^[52]

The presence of a $\text{C}-\text{O}$ stretching band in the $1080\text{--}1040\text{ cm}^{-1}$ region in all three samples indicates the presence of PVA in the structure. Additionally, the high intensity of the $\text{O}-\text{H}$ band in the samples, particularly in the red and green samples, suggests the presence of dopamine in monomeric or unpolymerized form. However, in the blue sample, which contains both aniline and pyrrole monomers, the possibility of polydopamine (PDA) formation through the oxidation of dopamine and interaction with other chains is increased by a change in the position of the $\text{C}-\text{N}$ and $\text{C}=\text{C}$ bonds.^[53,54]

The UV-Vis spectra (Fig. S2a in ESI) shows the characteristic absorption features of polypyrrole (about 320 nm) and polyaniline (in the region of $320\text{--}350\text{ nm}$ and extending towards the visible region). In the case of the PANi/Ppy hybrid

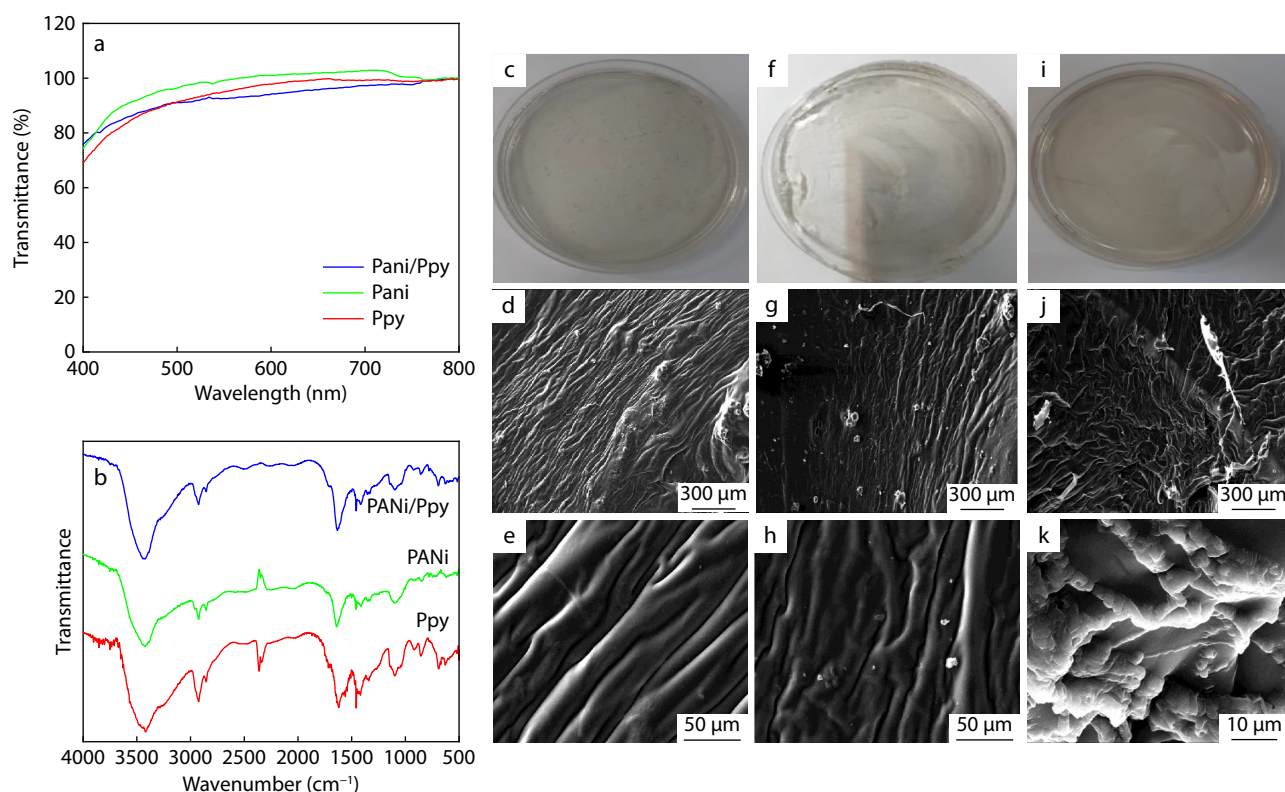


Fig. 1 (a) Transparency of the CHs with a thickness of 2 mm in the UV-Vis spectra; (b) FTIR spectra of PDA-Ppy-PANi; (c, f, i) Transparency of CH-PANi, CH-Ppy, and CH-Ppy-PANi; FE-SEM images of (d, e) CH-PANi, (g, h) CH-Ppy, and (j, k) CH-Ppy-PANi.

sample, the absorption intensity in the range of 280–320 nm was significantly enhanced. In addition, the broadening and change in the intensity of the bands in the visible region indicated strong interactions between the PANi and Ppy chains and the formation of a stable hybrid structure. These spectral changes are consistent with previous reports on hydrogels inspired by hybrid conductive compounds.

Fig S2(b) (in ESI) shows the X-ray diffraction (XRD) patterns of Ppy, PANi, and the PANi/Ppy hybrid hydrogel. In the spectrum of Ppy, a broad peak can be observed at approximately 2θ of approximately 24° , indicating the amorphous nature and disordered structure of Ppy chains.^[55] In the PANi sample, 2 distinct peaks appear at 2θ of approximately 20° and 25° , which were attributed to the presence of a semi-crystalline structure owing to the relative arrangement of the polymer chains.^[56] In the PANi/Ppy hybrid sample, both peaks can be observed, but their intensities were reduced and broadened, indicating interchain interactions and the formation of an amorphous hybrid structure owing to simultaneous doping.^[57] These changes implied a decrease in crystallinity and an increase in the uniformity of the chain distribution in the hybrid hydrogel. The XRD results are fully consistent with the FTIR and UV-Vis data, confirming the successful formation of the PDA-PANi-Ppy structure.

Swelling Behavior, Self-healing Properties, Adhesion Characteristics, and Water-Induced Shape Memory Elasticity of CHs

Functional groups and non-covalent interactions play key roles in the self-healing and swelling behavior of hydrogels. The

swelling behavior was examined by immersing the CH-Ppy, CH-PANi, and CH-Ppy-PANi hydrogels in water for 9 h (Fig. 2a). The higher swelling of CH-PANi compared to CH-Ppy was attributed to more hydrophilic amine groups and stronger water-polymer interactions, whereas CH-Ppy-PANi exhibited intermediate swelling due to balanced hydrophilic/hydrophobic domains.

Self-healing was studied by cutting the hydrogels into four pieces and then rejoining them (Fig. 2b). Hydrogels healed at 50°C within 1 h owing to dynamic hydrogen bonds, π - π stacking, and catechol-metal coordination. Mechanical tests on healed samples confirmed about 48% recovery of the tensile strength (Fig. S3 in ESI). The self-healing behavior of the hydrogels after mechanical damage and tearing increases their lifespan. This property arises through dynamic interactions, including hydrogen bonds and covalent and non-covalent bonds. The excellent self-healing behavior of CHs is attributed to the catechol and $-\text{COOH}$ groups, which are involved in non-covalent interactions, such as π - π stacking and hydrogen bonding^[58]. Self-healing CHs prepared based on the hydrogen-bonding mechanism are less stable than self-healing CHs prepared by covalent and ionic interactions. However, it has been demonstrated that incorporating hydrophobic interactions and size effects into hydrogels can enhance their self-healing properties.^[59] On the other hand, PVA has been shown to play a positive role in self-healing due to its abundant hydroxyl groups and the formation of a hydrogen bonding network.^[60] Dopamine, inspired by mussels, is vital for self-healing because of its catechol groups.

A design inspired by the adhesion of mussels was proposed for self-adhesive hydrogels to enhance the adhesion of

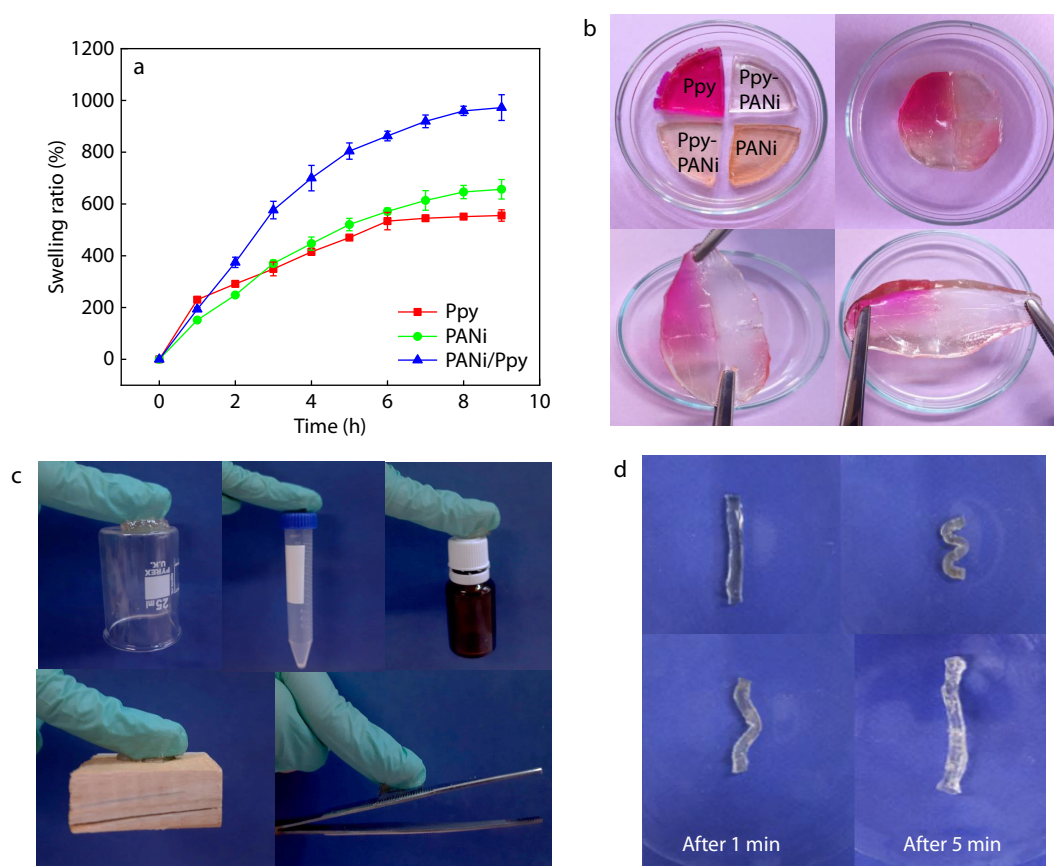


Fig. 2 (a) Swelling ratios of hydrogels at 25 °C in 9 h; (b) The self-healing properties after cutting into four parts; (c) Photographs of CHs stuck on different substrates such as glass, metal, wood, and plastic; (d) The property of shape memory, drying the hydrogel with a specific shape and its return to its original state.

biosensors to the skin. These hydrogels are reinforced with polydopamine, which features catechol groups and provides strong adhesion to polar surfaces such as glass, metal, wood, and plastic (Fig. 2c). The edge-cutting test on aluminum substrates showed that the CH-Ppy-PANi hydrogel exhibited the highest adhesion strength (66.1 ± 1.4 kPa) compared to CH-PANi (56.7 ± 0.9 kPa) and CH-Ppy (32.8 ± 1.2 kPa). The significant increase in the adhesion performance is attributed to the synergistic effect of the catechol groups in PDA and the π -conjugated system of Ppy and PANi, which together enhance the interfacial interactions with the metal substrate (Table S1 in ESI). The use of additional adhesives can increase the resistance and reduce the accuracy of sensors. Consequently, developing self-adhesive hydrogels is crucial for enhancing the signal transmission and efficiency of sensors.

Shape memory was demonstrated by drying hydrogels into predefined shapes and immersing them in water for recovery (Fig. 2d). This mechanism arises from the reversible hydrogen bonding of PVA, catechol-metal coordination, and elastic recovery of PAM, which is consistent with previous models of dynamic networks.^[61,62]

All the 4 properties were related to the functional groups of the hydrogel molecules. The hydrogel healed independently without the need for self-healing agents. The excellent self-healing behavior is attributed to the catechol and $-\text{COOH}$ groups, which are involved in non-covalent interactions, such

as hydrogen bonding and π - π stacking. The presence of groups that can form hydrogen bonds, such as $-\text{NH}$ in hydrogels, is a key factor in creating memory properties. It has been shown that water molecules can act as adhesives. During compression and deformation of the gel, newly formed contact points form hydrogen bonds that lock the deformation in place. However, recontacting the gel with water causes the formation of H-bonds between water and the gel, releasing the formed lock.^[61,62] It should be noted that the formation of this hydrogen bond leads to high water absorption and swelling of CHs.

Mechanical Properties of CHs

Hydrogels exhibit unique mechanical properties, including excellent recovery, elasticity, and flexibility. For specific functional applications, it is essential to strike a balance between strength and elasticity. As shown in the Fig. 3, CH-PANi hydrogels can be stretched up to $904.46\% \pm 11.50\%$ and return to their original state without tearing, which occurs in only 1 s (Fig. 3d). The tensile strength of the CH-Ppy-PANi hydrogel is superior to that of other types of hydrogels, with a value of (32.66 ± 1.03) kPa (Fig. 3c). The elongation at break (EB) of CH-PANi, CH-Ppy, and CH-Ppy-PANi are reported to be $904.46\% \pm 11.50\%$, $652.27\% \pm 10.40\%$, and $669.79\% \pm 20.11\%$, respectively (Fig. 3b).

These outstanding mechanical properties are attributed to the presence of numerous hydroxyl groups and the double-network structure in the CHs. Metal ions help strengthen the

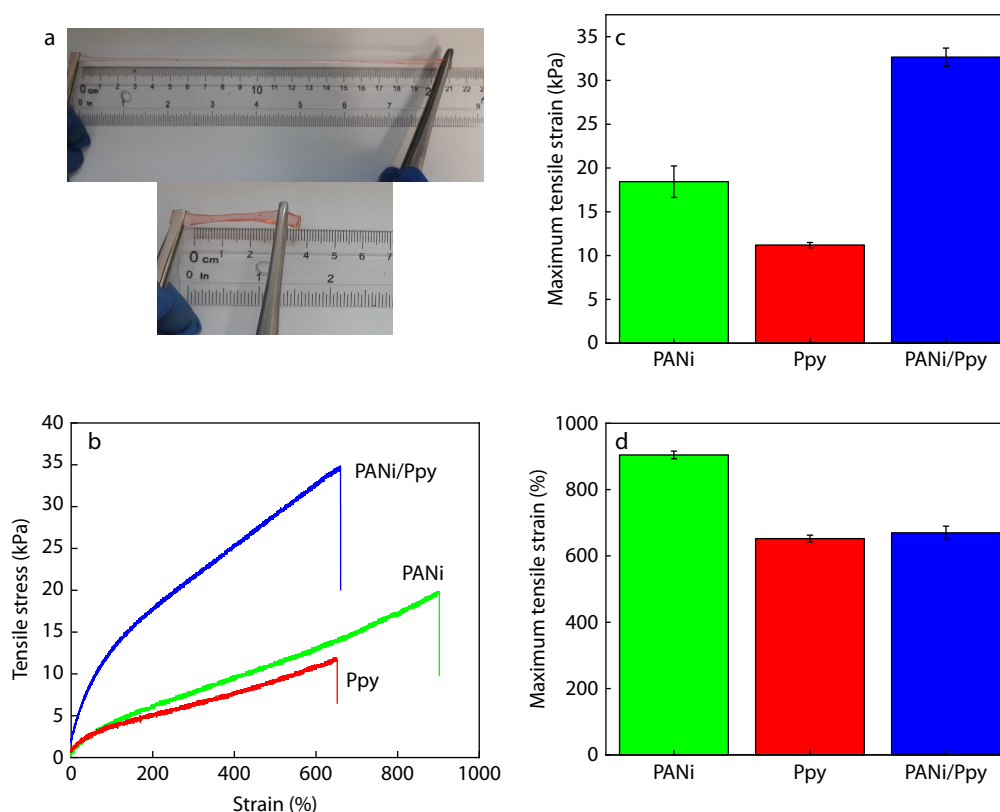


Fig. 3 (a) Photographs of the CHs showing high stretchability; (b) Tensile stress-strain curves of CH-Ppy, CH-PANi, and CH-Ppy-PANi; (c) Maximum tensile stress; (d) Maximum tensile strain.

gel network by forming non-covalent interactions with the amine and hydroxyl groups and by reducing the catechol groups to facilitate oxidation. These hydrogels can also withstand mechanical changes such as twisting, tearing, kinking, and puncturing. These capabilities render them suitable for use as sensitive sensors.

Electrical Test of CHs

These adhesive hydrogels can be easily applied to or stretched on flat and curved surfaces and exhibit conductive properties. The good electrical conductivities of the hydrogels are shown in Fig. 4(a). CH-Ppy-PANi ($(95.4 \pm 0.4) \times 10^{-4}$ S/cm) exhibits higher conductivity than CH-Ppy ($(27.00 \pm 0.12) \times 10^{-4}$ S/cm) and CH-PANi ($(30.4 \pm 0.5) \times 10^{-4}$ S/cm). With the increase in CH-Ppy-PANi particles, the conductivity of the hydrogel increased compared to the CH-Ppy and CH-PANi hydrogels, but their transparency did not change significantly, as shown in Fig. 4(b). Increasing the PDA-Ppy-PANi content enhanced the conductivity while maintaining a high transparency (about 93%–97%), demonstrating synergistic improvement over single-component systems. Fig. 4(c) shows that the hydrogels exhibit conductivity even when stretched and twisted, but their conductivity changes, making them suitable for sensor applications. The conductivity and self-healing ability of the CH-Ppy-PANi hydrogel make it a potential conductive material for circuit repair. Because of the conductivity and stretchability of the hydrogel, we connected it to a circuit with a light-emitting diode (LED) as an indicator to detect strain-dependent responses. As the hydrogel sample was stretched, the brightness of the LED gradually decreased.

Sensor Behavior of CHs

Hydrogels are excellent candidates for wearable body-monitoring sensors owing to their outstanding adhesion, conductivity, and mechanical properties. CH hydrogels were tested as pressure sensors and their responses were recorded as relative resistance changes ($\Delta R/R_0$) over time (Figs. 5a–5f). To assess the sensitivity of these hydrogels, resistance changes ($\Delta R/R_0$) were measured at a strain rate of three-tenths over a 100-s interval (Fig. 5g). The gauge factor (GF) was calculated to evaluate the effect of the conductive polymer on the performance of the hydrogel sensors; the GF values are 3.57 ± 0.06 , 6.04 ± 0.25 , and 7.44 ± 0.31 for CH-Ppy, CH-PANi, and CH-Ppy-PANi, respectively (Fig. 5h). The ability of these hydrogels to adhere directly to the device without the need for additional adhesives is crucial. Moreover, even after repeated and prolonged stress, no significant changes in the baseline electrical signal can be observed (Fig. 5g), indicating the high stability of the conductivity of the CH sensor. The combination of high strain sensitivity and stable conductivity suggests a substantial expansion of the application range of the CH sensor. Small changes in strain resulted in significant increases in the resistance, demonstrating the high accuracy and sensitivity of these sensors.^[63,64] The hydrogels were tested on various body parts including the forehead, throat, wrist, fingers, and chest. The results show that movements such as bending of the wrist or fingers caused changes in $\Delta R/R_0$, indicating the presence of pressure within the structure (Figs. 5a and 5b). Facial expressions and breathing changes also affected the sensor resistance, confirming its high sensitivity (Figs. 5c and 5e). These findings suggest that CH hydrogels are a

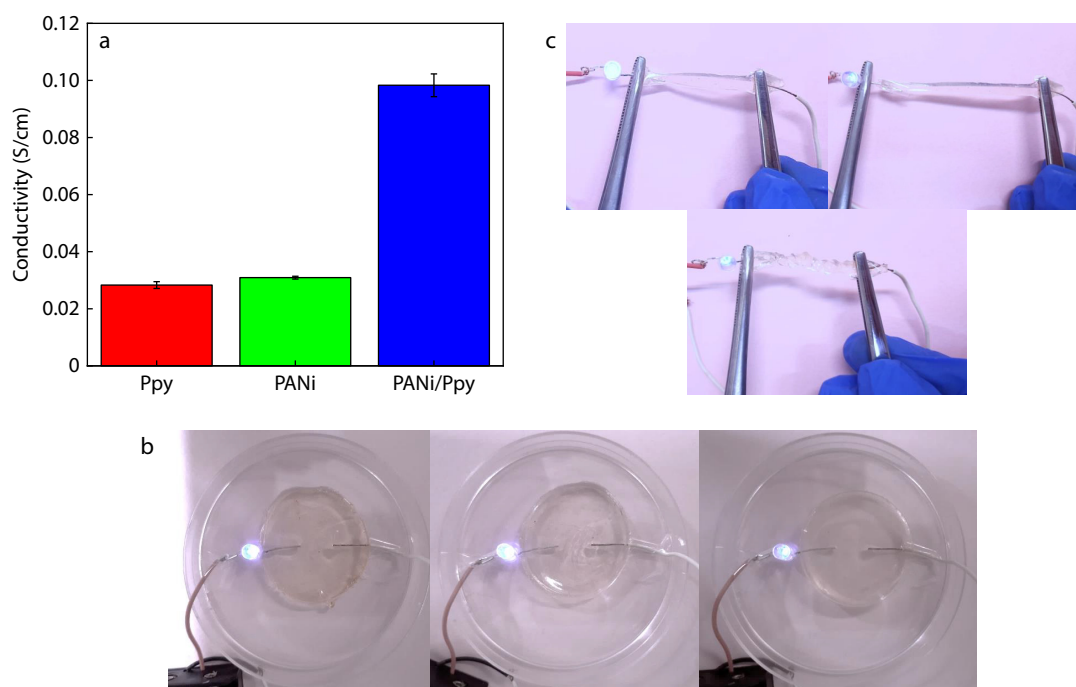


Fig. 4 Electrical properties of hydrogels. (a) Electrical conductivity diagram of hydrogels; (b) Conductivity imaging of CH-Ppy, CH-PANi, and CH-Ppy-PANi hydrogels, respectively; (c) Photographs of CH response during stretching and twisting.

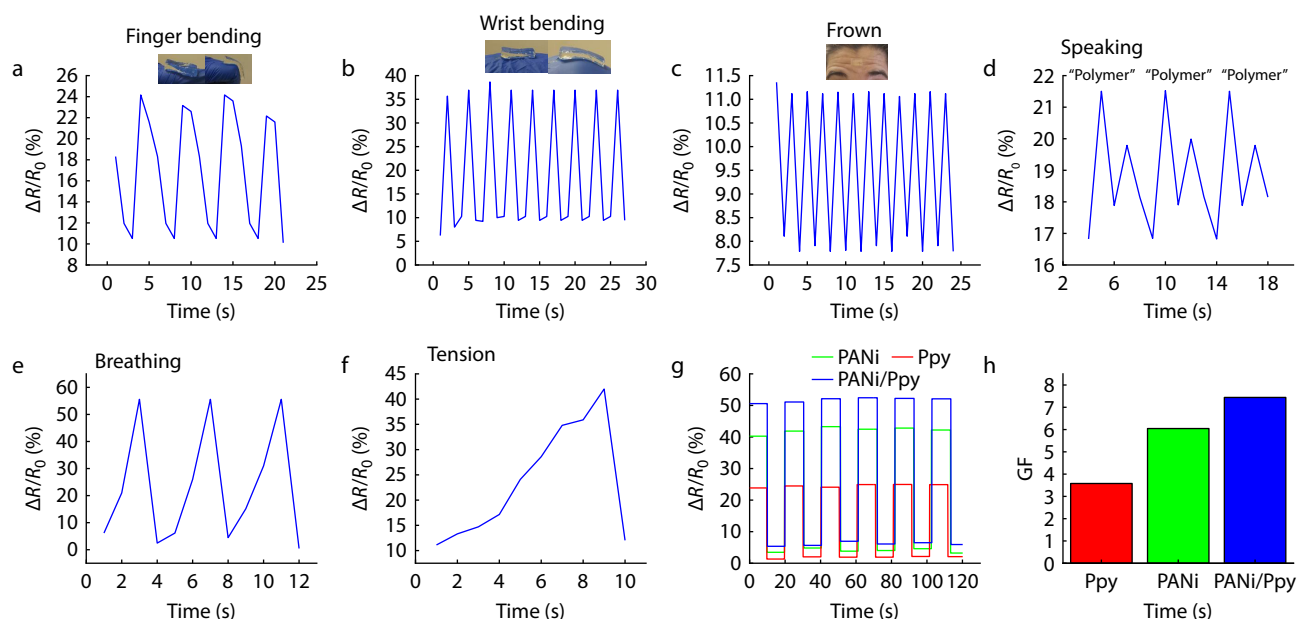


Fig. 5 (a) The relative change in resistance of the hydrogel adhered to a finger to monitor its bending; (b) The relative change in resistance of the hydrogel adhered to a wrist to monitor its bending; (c) The relative change in resistance of the hydrogel during a frown; (d) The relative change in resistance of the hydrogel while speaking the word "Polymer"; (e) The relative change in resistance of the hydrogel during breathing monitoring; (f) The relative change in resistance of the hydrogel under various tension levels; (g) The relative change in resistance of the hydrogel during cyclic testing; (h) Different hydrogels' gauge factor (GF) at varying tensile strains.

promising option for use in electronic skins and flexible electronic devices.

CONCLUSIONS

Herein, we report an efficient and rapid method for producing CH hydrogels with unique properties. The synthesis time was

only 5 min, which is significantly shorter than previously reported mussel-inspired hydrogels, which often require several hours to days for completion. This hydrogel exhibited high conductivity and transparency owing to the simultaneous formation of fibers within its structure. Additionally, they exhibit outstanding mechanical properties, including excellent elasticity and structural stability. The presence of π - π interactions and hydrogen

bonds contributed to the self-healing properties of this material. The self-adhesive ability of this hydrogel enables it to be directly attached to various surfaces and functions as a sensor to detect changes, such as pressure, bending, and torsion. The remarkable sensitivity of this hydrogel to strain combined with its excellent repeatability provides a suitable field for use in wearable sensors. The strain sensors fabricated from CH hydrogels demonstrated gauge factors (GF) of 3.57 ± 0.06 , 6.04 ± 0.25 , and 7.44 ± 0.31 for CH-Ppy, CH-PANi, and CH-Ppy-PANi, respectively, highlighting their high sensitivity compared to many previously reported hydrogel-based strain sensors. These sensors can be directly attached to the skin and can accurately track the precise movement of body joints, including the shoulder, wrist, neck, and elbow. By benchmarking against prior mussel-inspired conductive hydrogels, the present CHs simultaneously achieve higher conductivity (up to $(95.4 \pm 0.4) \times 10^{-4}$ S/cm), rapid synthesis (5 min), excellent adhesion, and self-healing performance, thereby addressing the limitations of opacity, long preparation times, and poor dispersion of conjugated polymers reported in earlier studies. Their excellent adhesion properties and stability under repeated stresses make these hydrogels ideal for flexible electronics.

In addition, studies on the different polymers used in the CH hydrogel structure have shown that the type of these materials can significantly affect the electrical and mechanical properties. This suggests the high potential of this technology for developing advanced sensors and creating novel features. The results of this research can help in the design and fabrication of CH hydrogels more efficiently, providing new insights for their application in innovative and flexible devices. In addition to recording body movements, these hydrogels can aid in the development of applications in electronic skins and wearable devices. Beyond monitoring body movements, these hydrogels show promise for applications in electronic skin, biomedical monitoring, and next-generation wearable electronics.

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/s10118-025-3473-9>.

Data Availability Statement

All data generated or analyzed during this study are included in this published article and its supplementary information files.

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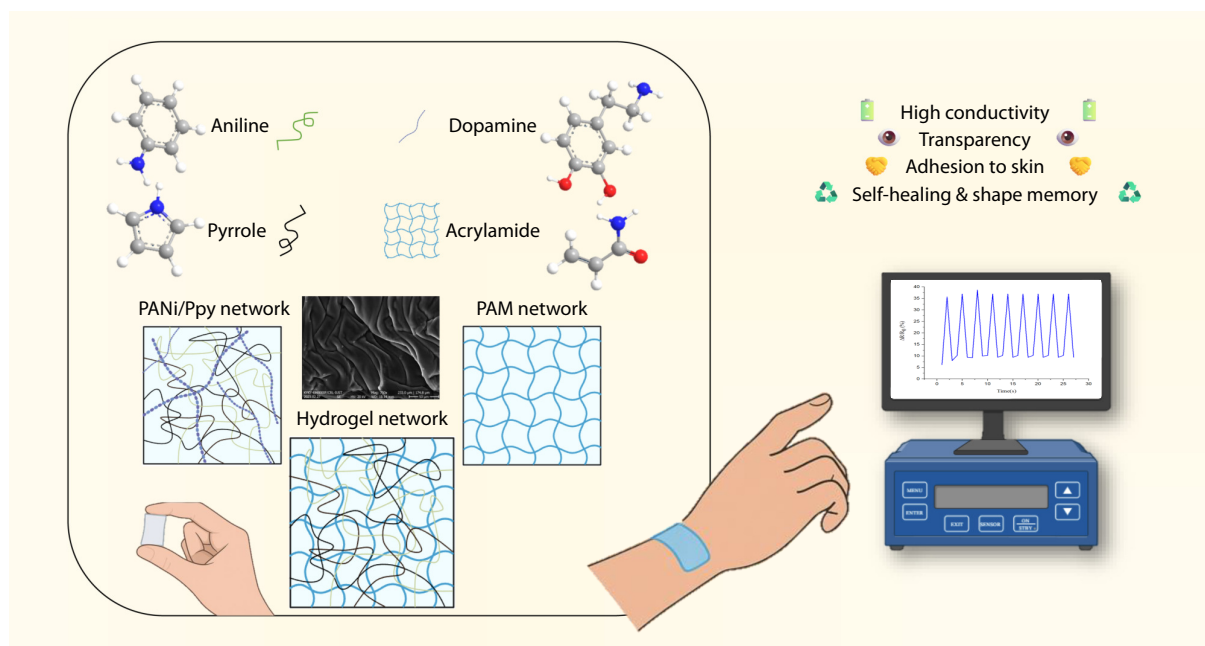
Graphical Abstract

Flexible Mussel-inspired Hydrogel with Polyaniline and Polypyrrole for Transparent Wearable Strain Sensors: Investigation of Physical Properties, Self-healing, and Electrical Conductivity

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A mussel-inspired hydrogel incorporating polydopamine (PDA), polypyrrole (Ppy), polyaniline (PANI) and poly(vinylalcohol) (PVA) within a polyacrylamide (PAM) network delivers high transparency (about 90%), electrical conductivity (about 95.4×10^4 S/cm), tensile strength (about 32.6 kPa), and rapid self-healing (about 48% recovery in 1 h at 50 °C). It functions effectively as a flexible, wearable strain sensor with GF of approximately 7.44.



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