Heliyon 10 (2024) e35187

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon

Research article

High stretchable and self-adhesive multifunctional hydrogel for wearable and flexible sensors

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ARTICLE INFO

Keywords:
Polymer gels
Smart polymers
Mechanical properties
Hydrogel
Sensors

ABSTRACT

Ionic conductive hydrogel has recently garnered significant research attention due to its potential applications in the field of wearable and flexible electronics. Nonetheless, the integration of multifunctional and synergistic advantages, including reliable electronic properties, high swelling capacity, exceptional mechanical characteristics, and self-adhesive properties, presents an ongoing challenge. In this study, we have developed an ionic conductive hydrogel through the co-polymerization of 4-Acryloylmorpholine (ACMO) and sodium acrylate using UV curing technology. The hydrogel exhibits excellent mechanical properties, high conductivity, superior swelling capacity, and remarkable self-adhesive attributes. The hydrogel serves as a highly sensitive strain sensor, enabling precise monitoring of both substantial and subtle human motions. Furthermore, the hydrogel demonstrates the capability to adhere to human skin, functioning as a human-machine interface for the detection of physiological signals, including electromyogram (EMG) signals, with low interfacial impedance. This work is anticipated to yield a new class of stretchable and conductive materials with diverse potential applications, ranging from flexible sensors and wearable bio-electronics to contributions in the field of artificial intelligence.

1. Introduction

In recent years, wearable and flexible electronics have gain widespread attention due to that they can meet the demands of collecting human information, and converting it into electrical signals in real-time [1–3]. As a new type of wearable and flexible electronics, wearable and flexible sensors can convert human information including human motion [4], and physiological information [5], to electrical signals including changes of resistance [6], capacitance [7], as well as voltage [8]. Nowadays, wearable and flexible sensors have been widely applied in many fields including bioelectronics [9], robotics [10], personal health monitoring [11], and human-machine interfaces [12]. Particularly, wearable and flexible sensors are fabricated by combining conductive materials such as metal nanowire [13], carbon particles [14], graphene [15], carbon nanotubes [16], and liquid metal [17] into stretchable and flexible elastomers such as silicone rubber [15,18], and polyurethane [19,20]. For example, Zhenyu Wang et al. developed a stretchable temperature sensor consisting of cellular graphene/polydimethylsiloxane composite, the sensor shows high sensitivity, durability, and

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https://doi.org/10.1016/j.heliyon.2024.e35187
Received 7 March 2024; Received in revised form 24 July 2024; Accepted 24 July 2024
Available online 25 July 2024

stability, which can be used to monitor skin temperature accompanying an arbitrary wristwork [18]. Additionally, Yuan Gao et al. developed a carbon nanotubes (CNTs) and polyurethane (PU) nanofibers composite with superior electrical conductivity, stretchability, and high sensitivity, which can be used to monitor human motion [16]. In addition, stretchable structures such as serpentine structure [21,22] and kirigami structure [23] of non-stretchable conductors have also been greatly researched and used for wearable and flexible sensors. For example, Ziting Tan et al. developed a novel micro-foldable structure on polydimethylsiloxane (PDMS) inspired by the armadillo [24]. The as-prepared stretchable electrodes with excellent properties, show excellent performance in electromyography (EMG) signal detection, stretchable conducting wires, and capacitive stretchable sensors. Similarly, Zhenming Chu et al. reported a gradient wrinkle strain sensor by a pre-stretching method [25]. The sensor exhibits a broad strain range, ultra-high sensitivity, and stability to detect full-range human body motions, even in harsh environments. Despite many advances in this field, the challenges such as the super stretchability of wearable and flexible sensors still hinder the practical applications.

Hydrogels, are a kind of extremely hydrophilic three-dimensional physically or chemically cross-linked networks, which swells rapidly in water and can keep a large volume of water without dissolving [26]. As high water absorption and high water retention materials, hydrogels were widely used in various fields, such as smart windows [27], drug carriers in medical treatment and tissue engineering over the past decade [28,29]. Recently, conductive gels have gained significant attention within the scientific community due to their superior and distinctive properties [30], making them highly promising for various applications in soft robotics, human-machine interfaces, flexible electronics, implantable and wearable biosensors [31–35]. Ionic conductive hydrogel was also fabricated composed of hydrogel polymers and salt electrolyte materials as conductive electrolytes [36]. In recent years, ionic conductive hydrogels have gained significant attention due to their remarkable electrical properties, high stretchability, and flexibility, making them a prominent subject of research [37,38], which have been used in the field of wearable and flexible sensors [6,39,40], human-machine interfaces [41], and ionic skins [42] *etc*. However, the conventional hydrogels usually maintain single or two performances, which limits their applications in multifunctional sensors. Therefore, developing a kind of hydrogel as ideal materials with

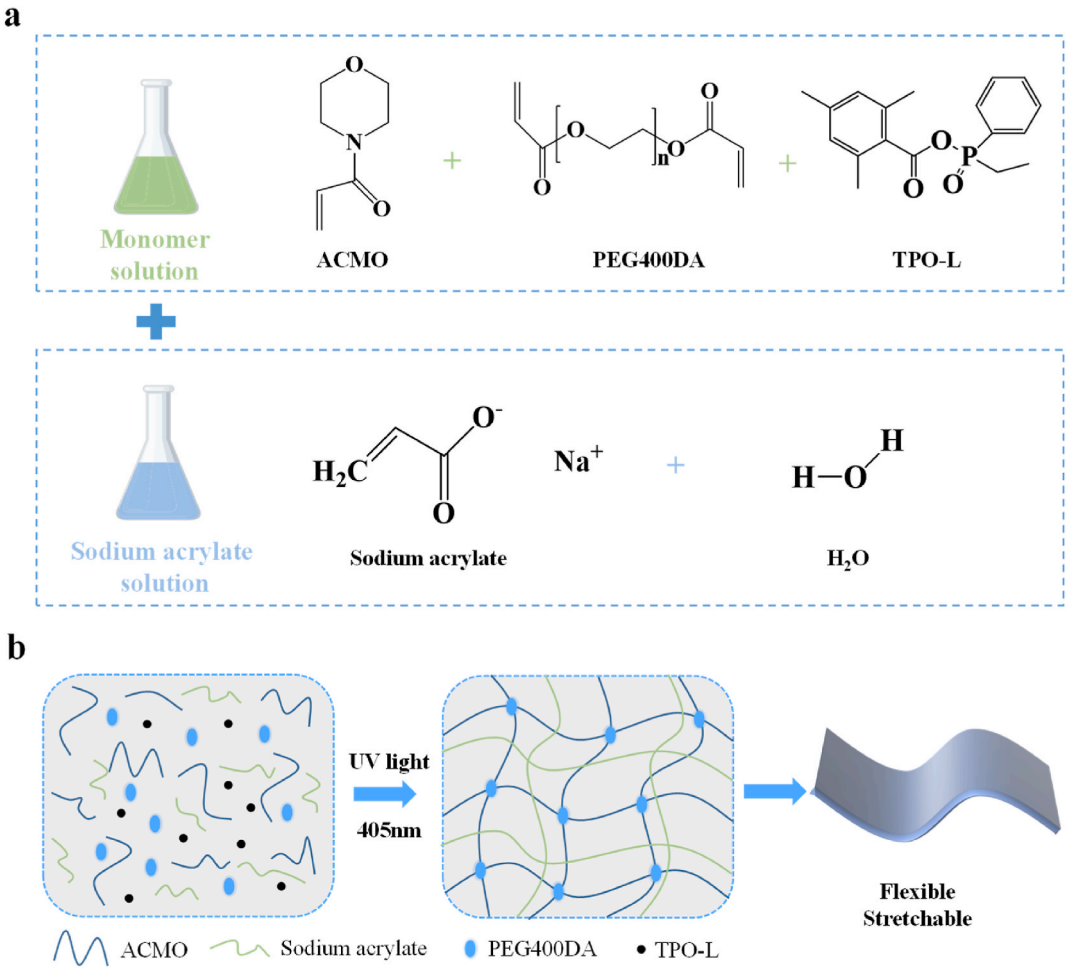

Fig. 1. Preparation of hydrogel. (a) Preparation process and chemical structural characterization of the hydrogel. (b) The polymerization process of the ACMO, sodium acrylate, TPO-L, and PEG400DA after being irradiated by the UV light of 405 nm. The flexible and stretchable hydrogel is obtained.

multiple and synergistic advantages including reliable electronic properties, high swelling properties, outstanding mechanical characteristics, and self-adhesive properties remains in increasing demand.

Herein, an ionic conductive hydrogel with multifunctional and synergistic properties was developed. Briefly, 4-Acryloylmorpholine (ACMO) and sodium acrylate as monomers were co-polymerized under the irradiation of UV light to form the as-prepared hydrogel. The as-prepared hydrogel presented outstanding mechanical characteristics with stretchability of exceed 1000 %. In addition, the hydrogel shows high conductivity, superior swelling properties, and outstanding self-adhesive properties. Owing to the multifunctional performance of hydrogel, hydrogel act as a highly sensitive strain sensor to monitor the detection of large motions and subtle motions of human. The combination of high stability, sensitivity, and rapid response time makes the hydrogel sensor an ideal choice for various applications such as biomedical monitoring, environmental monitoring, healthcare, soft robotics, or industrial process control. In addition, the hydrogel adhered to human skin as a human-machine interface, can serve as a flexible electrode for detection of electrophysiological signals such as electromyogram (EMG) signals. The results demonstrate great potential in multifunctional wearable bio-electronics.

2. Experimental section

2.1. Materials

4-Acryloylmorpholine (ACMO) as monomer was purchased from Chengdu Sicheng Optoelectronic Materials Co., Ltd., Sodium acrylate as monomer was purchased from Shanghai Macklin Biochemical Co., Ltd., Poly (ethylene glycol) diacrylate (PEG400DA) as crosslinker was purchased from RYOJI Chemical Co., Ltd., and Ethyl (2,4,6-trimethylbenzoyl) phenylphosphinate (TPO-L) as photoinitiator was purchased from RYOJI Chemical Co., Ltd. All materials were used as received and without any further purification. In this paper, deionized water (18.2 MΩ cm) was used throughout the experiments.

2.2. Preparation of hydrogel

The hydrogel was prepared in following steps. First, 20g ACMO monomer, 0.1 g PEG400DA crosslinker, and 0.4 g TPO-L photoinitiator were mixed and stirred for about 20min to form a monomer solution. Sodium acrylate solution was prepared by dissolving the sodium acrylate into deionized water (18.2 MΩ cm) with different concentration (Fig. 1a). Then monomer solution and sodium acrylate solution were mixed with different ratio and stirred for about 20 min (Table 1). After that, a precursor solution was completed for the following polymerization. For polymerization of hydrogel, the precursor solution was poured into a mold, then the UV light (405 nm) was irradiated to the surface of the precursor solution for about 5–10 s to form a solid hydrogel. The unreacted solution and mold were removed, then the polymerization of hydrogel was completed (Fig. 1b). The precursor solution system utilized a polymerizable sodium acrylate as the ionic monomer. This sodium acrylate serves as the ionic framework within the polymer network. Its conductive properties allowed for partial dissociation, resulting in inherent ionic conductivity in the system. It is important to highlight that the system was further strengthened by the introduction of ACMO monomer, which enhanced bond crosslinking within the network. This reinforcement contributed to the overall structural integrity of the hydrogel. Subsequently, poly(ethylene glycol) diacrylate (PEGDA) was employed as a crosslinker. With active vinyl functional groups along its molecular chain, PEGDA facilitated the covalent connection of the ionic monomers, promoting the formation of a robust gel network [43],resulting highly mechanical and adhesive performance (Table S1).

2.3. Characterization

The uniaxial tensile tests and adhesion of the hydrogels were performed on a mechanical testing machine (ZQ-990A, China). The resistance changes of the hydrogel-based sensor in different state were obtained by a sensing analysis system (DMM6500, KEITHLEY), and the relative change of the resistance was calculated using the following formula: $\Delta R/R_0=(R-R_0)/R_0$, where R_0 and R were the resistance without and with applied strain, respectively. The interfacial contact impedance between skin and electrode was obtained by an electrochemical workstation (CHI604E).

Table 1
Material used in the hydrogel precursor solution.

Constituent	Weight content of the constituent				
Monomer solution	2 g	2.5 g	3 g	3.5 g	4 g
Sodium acrylate	3 g	2.5 g	2 g	1.5 g	1 g
Deionized water	5 g	5 g	5 g	5 g	5 g
Name of solution	**AANa-30**	**AANa-25**	**AANa-20**	**AANa-15**	**AANa-10**

3. Results and discussion

3.1. Mechanical and conductive properties of hydrogels

To examine the effect of the weight content of sodium acrylate on the mechanical properties of the hydrogel. The mechanical performance of the hydrogel is influenced by the ratio of ACMO in the monomer solution and sodium acrylate in deionized water. Sodium acrylate, being a hydrophilic monomer, promotes the formation of a highly crosslinked polymer network within the hydrogel. This leads to increased entanglement of polymer chains and intermolecular interactions, resulting in different mechanical performance. The uniaxial tensile tests were performed for hydrogels with different weight contents of sodium acrylate. As shown in Fig. 2a, the typical stress-strain curves of hydrogels with different weight content of sodium acrylate were displayed. The fracture stress of AANa15 hydrogel was exceed 40 kPa, and the elongation at break of the AANa30 hydrogel was exceed 1000 %. The result showed the outstanding stretchability of as-prepared hydrogel. When the weight content of sodium acrylate increased from 10 wt% to 30 wt%, the mechanical properties of as-prepared hydrogels strongly depended on the weight content of sodium acrylate. As shown in Fig. 2b, the elongation of the hydrogel dropped slightly, and then increased when the weight content of sodium acrylate was higher than 20 wt%. As shown in Fig. 2c, the weight content of sodium acrylate also affected the mechanical modulus and toughness of the hydrogels. By increasing the weight content of sodium acrylate from 10 wt% to 30 wt%, the modulus decreased. The hydrogels demonstrate excellent cyclic stability in both stretching and compression cycles (Fig. S1). When subjected to repeated stretching and releasing cycles, hydrogels maintain their structural integrity and mechanical performance. They can endure significant elongation without suffering from permanent deformation. This cyclic stability stems from the robustness of the hydrogel network, which enables the hydrogel to undergo repetitive stretching. Similarly, in compression cycles, hydrogels exhibit excellent resilience and recover their original shape after being compressed.

The as-prepared hydrogel possessed superior conductivity due to the presence of Na^+ ions. Fig. 2d shows the hydrogel acts as a conductor to connect the circuit with a light-emitting diode (LED) bulb. The brightness of LED bulb decreased as a function of the applied strains, and the brightness of LED bulb returned to its original level when hydrogel was released to its original length.

3.2. Swelling behaviors of hydrogels

Swelling ratio is one of the most significant parameters in hydrogel technology. The water molecules gradually enter the hydrogel matrix, resulting in an increase in the weight of the hydrogel (Fig. 3a). To analyze the swelling behavior of the hydrogel, measurements were conducted at room temperature. At specific time intervals, the hydrogels were taken out of the water, quickly blotted with filter paper to remove excess moisture, and weighed. The obtained results were quantified as the swelling ratio (S), calculated using the equation: $S = (W_t/W_0)$, where W_0 represents the initial weight of the hydrogel, and W_t corresponds to the weight of the swollen hydrogel at time t. The swelling measurements of as-prepared hydrogels were performed in water and solution with different pH values. The swelling ratios of the hydrogels as a function of the weight content of sodium acrylate were exhibited in Fig. 3b. As the as-

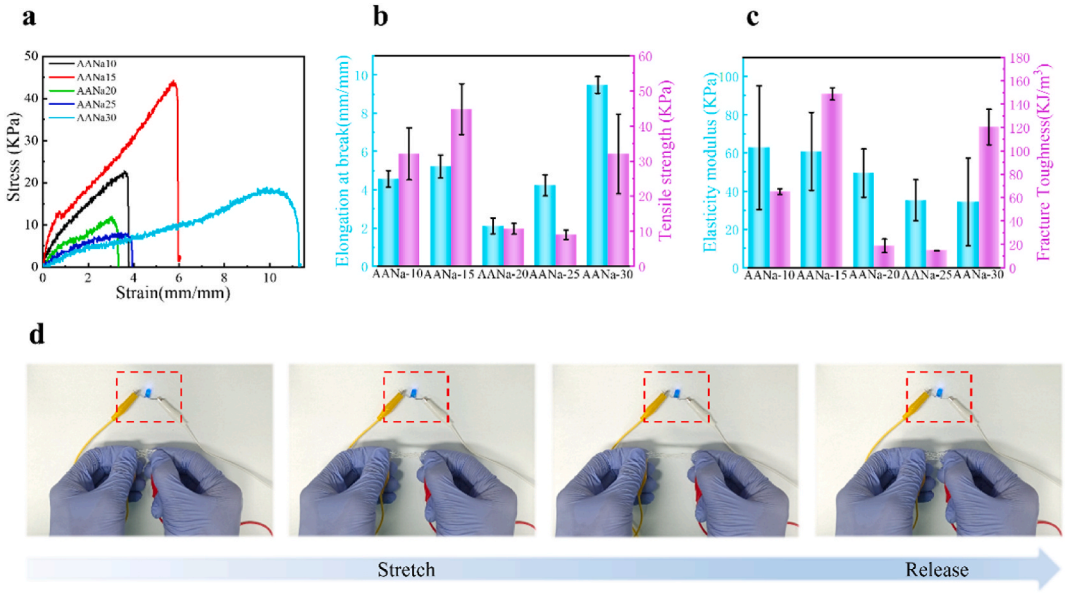

Fig. 2. Uniaxial tensile tests of hydrogels at room temperature. (a) The stain-stress curve of hydrogel with the 30 wt% of sodium acrylate. (b) The calculated elongation and tensile strength of hydrogels with different weight content of sodium acrylate. (c) The calculated modulus and toughness of hydrogels with different weight content of sodium acrylate. (d) Circuit containing the hydrogel with a LED indicator. The images show the luminance variation of a LED as a function of the applied strains.

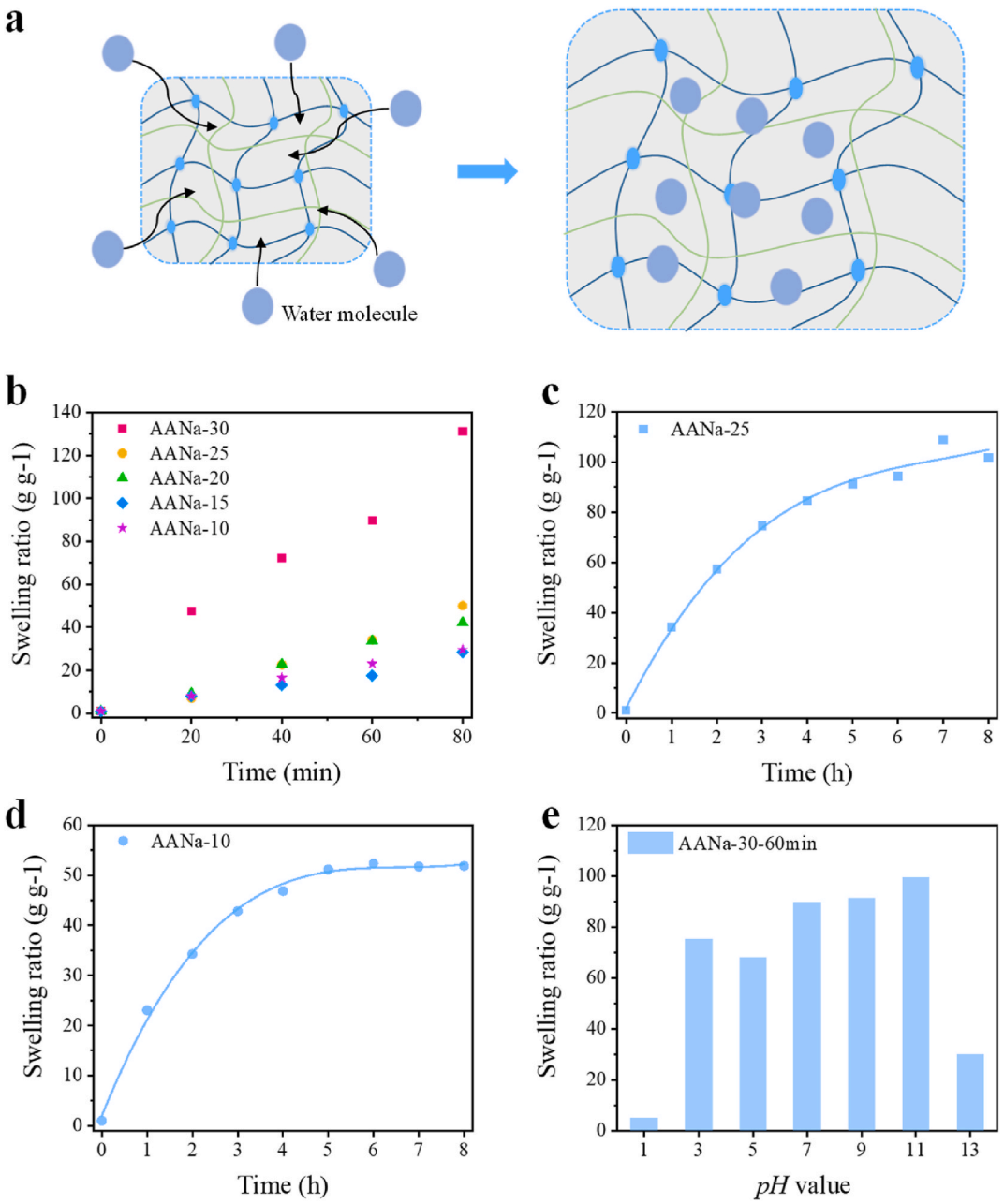

Fig. 3. Swelling behaviors of as-prepared hydrogels. (a) The schematic diagram shows the swelling process of as-prepared hydrogel. (b) The swelling kinetics of the hydrogels in water plotted against the weight content of sodium acrylate. (c) Equilibrium swelling ratio of the hydrogel with the 25 wt% of sodium acrylate. (d) Equilibrium swelling ratio of the hydrogel with the 10 wt% of sodium acrylate. (e) The swelling ratios of the hydrogels in solution with different pH values. ***3.3 Adhesive performance of hydrogels.***

prepared hydrogels were immersed into deionized water (18.2 MΩ cm), the swelling ratios gradually increase with the swelling time at room temperature. This is mainly because water molecules would enter the hydrogel network to increase the sample weight of the hydrogel. It can be seen that the weight content of sodium acrylate affects the swelling ratios. With the increasing weight content of sodium acrylate, the swelling speed and ratios gradually increased. This is mainly due to sodium acrylate enhancing the interaction between the water and polymer matrix in hydrogel. As shown in Fig. 3c, the equilibrium swelling ratio of hydrogel with 25 wt% of sodium acrylate was studied. It can be seen that the hydrogel absorbed water quickly and obtain the swelling ratio of 90 g g^{-1} within 5 h, the swelling ratio achieve to the equilibrium of about 100 g g^{-1} from 5 to 8 h. Interestingly, the same result was achieved in the hydrogel with 10 wt% of sodium acrylate. Note that the equilibrium swelling ratio for this hydrogel is about 50 g g^{-1} (Fig. 3d). The effect of pH values on the swelling behavior of as-prepared hydrogel with 30 wt% of sodium acrylate was investigated at room

temperature for 60 min (Fig. 3e). The swelling ratio is lower at pH = 1. This is that the H^+ ions from the solution form additional physical cross-linking with the hydrogel network at lower pH levels, which decreases the swelling ratio. With increasing the pH value from 5 to 11, the swelling ratio increases rapidly. However, for a strong alkaline solution at pH = 13, the destruction of crosslinking between polymer molecules occurs, which results in a relative lower swelling ratio.

The hydrogels show outstanding adhesive properties. The hydrogel was able to adhere to a variety of surfaces including PMMA and glass. The adhesion strength of the hydrogels with different content of sodium acrylate on PMMA surface was further quantified by a tensile adhesion test (Fig. 4a). The hydrogel was affixed to the surface of the specimens, and two surfaces were then joined together in a face-to-face manner. The adhesion strength (A) of our hydrogel was determined by dividing the maximum force (F) applied during the adhesion process by the contact area (S) between the two surfaces, using formula $A = F/S$. It can be seen that the hydrogel with 20 wt% of sodium acrylate shows the highest adhesion strength of 62.3 kPa. In addition, the adhesion strength of the hydrogels with different content of sodium acrylate on glass surface was also measured. As shown in Fig. 4b, with decreasing weight content of sodium acrylate to 20 wt%, the adhesion strength of the hydrogels increased obviously. After further decreasing the weight content of sodium acrylate to 10 wt%, a small reduction of the adhesion strength to glass was obtained. The hydrogel with 20 wt% of sodium acrylate also had superior adhesiveness to a variety of substrates. The adhesion strength to PMMA, glass, wood, copper and pig skin was 62.3 kPa, 29.0 kPa, 101.9 kPa, 2.62 kPa, and 11.32 kPa respectively (Fig. 4c). In addition, the hydrogels were adhered to a variety of materials, such as plastic, tape, glass, clip, screw, and battery, indicating superior self-adhesive properties of hydrogel (Fig. 4d). The remarkable mechanical and adhesion properties observed in the system can be attributed to the synergistic effects of the polymer chains, solvent, and ionic framework. This combination plays a vital role in achieving outstanding properties. The hydrogels demonstrate excellent stability in adhesion cycling (Fig. S2). In adhesion cycling, the hydrogel exhibits remarkable durability and reliability. it can adhere to glass substrate and maintain its adhesion strength even after undergoing numerous cycles of attachment and detachment. This cyclic stability is crucial in applications where repeated adhesion is required, such as in the field of wearable electronics, medical devices, and adhesive materials. T-peeling tests were conducted to evaluate the interfacial strength between the hydrogel layer and the substrates. The results indicate robust interfacial adhesion, suggesting significant potential for the hydrogel in self-adhesive device applications (Fig. S3).

3.3. Hydrogel-based stain sensor

Owing to the presence of sodium acrylate in water, the hydrogel could exhibit great conductivity, also the hydrogel with superior mechanical properties and adhesive performance made it a highly sensitive strain sensor. The strain sensing mechanism of an ionic hydrogel primarily relies on alterations in its electrical resistance when subjected to mechanical strain or deformation. The hydrogel's ionic conductive network, comprising mobile ions, facilitates ion migration in response to mechanical stress. As the hydrogel undergoes deformation, the movement of polymer chains induces a redistribution of the ions within the network. This redistribution

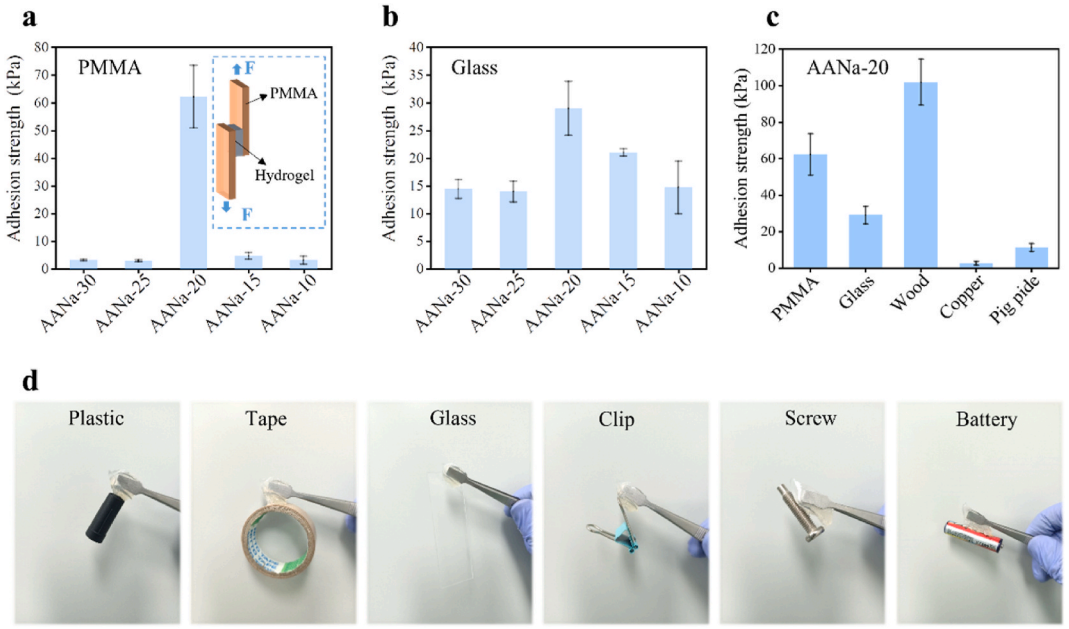

Fig. 4. Adhesive performance of hydrogels. (a) The adhesive strength of as-prepared hydrogels with different weight content of sodium acrylate to PMMA film. (b) The adhesive strength of as-prepared hydrogels with different weight content of sodium acrylate to glass. (c) The adhesive strength of the hydrogel with 20 wt% of sodium acrylate to various surfaces of materials. (d) The as-prepared hydrogels were adhered to a variety of materials.

influences the hydrogel's resistance, leading to modifications in its electrical properties. Specifically, during deformation, the resistance of the ionic hydrogel changes due to alterations in ion channels and migration path lengths. Tensile deformation caused the ion channels to narrow and the migration paths to lengthen, which slowed the migration of Na^+ and AAc^- ions, thereby increasing resistance. As a result, the ionic hydrogel demonstrated a strain-dependent response in its resistance, and thereby acted as strain sensor. Fig. 4 shows the applications of hydrogel strain sensors for monitoring the detection of large motions and subtle motions of human. The relative change of the resistance of the hydrogel stain sensor was calculated using the following formula: $\Delta R/R_0=(R-R_0)/R_0$, where R_0 and R were the resistance without and with applied strain, respectively (Fig. 5a). To create a strain sensor, we employed a method wherein two separate copper electrodes were attached to opposing sides of a printed hydrogel structure using conductive silver pastes. This results in effective transduction between ions and electrons at the ionic hydrogel and copper electrode interface, characterized by low interfacial impedance achieved through non-faradaic processes (formation of an electrical double layer, EDL). Subsequently, the hydrogel sensor was connected to a digital meter to assess its performance. The hydrogel sensor demonstrates exceptional stability over multiple cycles, exhibits high sensitivity with a gauge factor (GF) of 6.563, and exhibits a rapid response time (Fig. S4). As shown in Fig. 5b, the hydrogel stain sensor was adhered to the knee for monitoring the motion of knee bending. It can be seen that the relative change of the resistance rapidly increased as knee bending, and decreased when we completed the knee bending. As shown in Fig. 5c, the hydrogel sensor was attached to the finger, the strain sensors could precisely monitor the signal of a bending finger. In addition, the hydrogel strain sensor attached to the wrist and ankle also could monitor the corresponding motion (Fig. 5d and e). The hydrogel sensors can also accurately detect slightly human motions. As shown in Fig. 5f, When the hydrogel strain sensor was attached to the human throat, the strain sensors could monitor the motion of swallowing. It can be seen that the curves of the relative change of the resistance of hydrogel stain sensor displayed similar characteristic peaks when the person swallowed, which has great potential in the field of wearable devices for healthcare.

3.4. Hydrogel-based human-machine interface

The as-prepared hydrogel, which has combination of superior conductivity, mechanical properties, and great adhesiveness, can adhere to human skin as a human-machine interface. The hydrogel can also serve as a flexible electrode for the detection of electromyogram (EMG) signals. For physiological signal measurements, the interfacial impedance between skin and electrode is one of the most significant parameters. The interfacial impedance of the hydrogel electrode on the skin was measured using the electrochemical workstation. It can be seen that the hydrogel electrode has low interfacial impedance over a broad frequency ranging from 1 Hz to 1 MHz (Fig. 6a). We used the hydrogel electrodes adhered to our arm to measure the EMG signals. As shown in Fig. 6b, when we relaxed, the amplitudes of the EMG signals collected by our hydrogel electrode were stable. when we clenched repeated six times, the

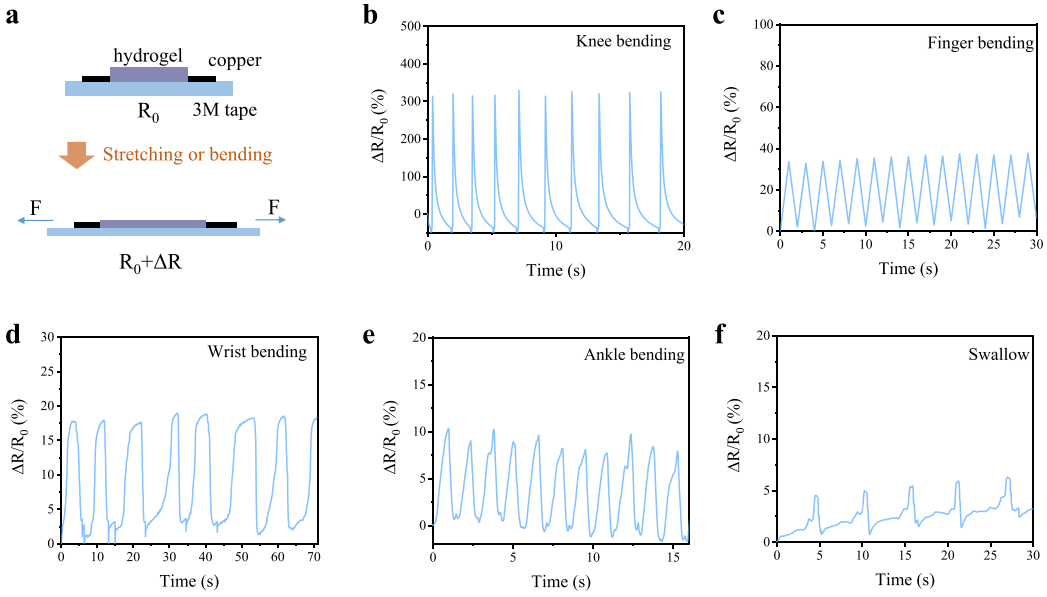

Fig. 5. Hydrogel as stain sensor. (a) The schematic underlying mechanism for the hydrogel strain sensor. 3M™ VHB™ acrylic adhesive tape is used as the stretchable substrate, upon which the copper foil electrode is placed and contacted with conductive silver paste. The hydrogel is then applied over the electrode, and then in-situ UV curing. This ensures that there is no slippage between the hydrogel and the copper electrode, thereby preventing any resistance changes due to bonding issues. (b) The relative resistance changes when bending and unbending the knee. (c) The relative resistance changes when bending and unbending the finger. (d) The relative resistance changes when bending and unbending the wrist. (e) The relative resistance changes when bending and unbending the ankle. (f) The relative resistance changes when the hydrogel strain sensor was adhered to the throat, and the person swallowed.

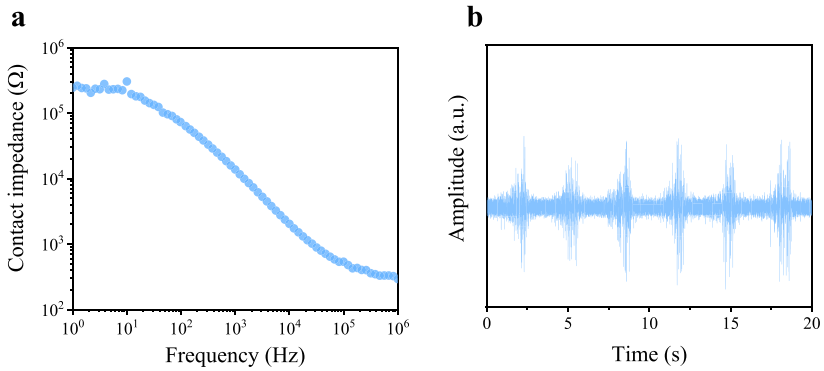

Fig. 6. Hydrogel based human-machine interface. (a) The interfacial impedance between skin and electrode over a broad frequency ranging from 1 Hz to 1 MHz. (b) EMG signals collected by our hydrogel electrode.

amplitudes of the collected EMG signals increased, and showed repeated sharps. The results show that our hydrogel electrode could be used in on-skin electronics as human-machine interface to collect physiological signal, and expected to be applied in the field of artificial intelligence potentially.

4. Conclusion

In conclusion, we developed a kind of hydrogel with multifunctional properties including high stretchability (elongation at break exceed 1000 %), high conductivity, superior swelling ratio, and outstanding self-adhesive properties (62.3 kPa to PMMA). The as-prepared hydrogel with a combination of superior conductivity, mechanical properties, and great adhesiveness, can work as a highly sensitive strain sensor for monitoring the detection of large motions and subtle motions of the human. Moreover, the hydrogel can adhere to human skin as a flexible electrode for the detection of electromyogram (EMG) signals. In combination with all of the properties of the hydrogel, the as-prepared hydrogel exhibited promising applications in various fields, such as flexible sensors, wearable devices, and even artificial intelligence.

Data availability statement

Data will be made available on reasonable request.

CRediT authorship contribution statement

Hao Zhong: Writing – original draft, Conceptualization. **Wubin Shan:** Writing – original draft, Data curation. **Lei Liang:** Writing – original draft, Data curation. **Xianzheng Jiang:** Data curation. **Linmei Wu:** Writing – review & editing, Supervision, Project administration, Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

H. Z., W. S. and L. W. conceived the project, carried out the experimental work, analyzed the data and wrote the manuscript. L. L and X. J helped to prepare the samples and revised the manuscript. W. S. and L. W. supervised the whole project. All the authors discussed the results and commented on the manuscript. This work was supported by the Natural Science Foundation of Jiangsu Province (BK20210868), the Natural Science Foundation of Hunan Province (21A0404), the Natural Science Foundation of Hunan Province(2022JJ60022), the Scientific Research Fund of Hunan Provincial Department of Education (2021C1014).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2024.e35187.

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