

• Supplementary File •

# A Stretchable, Ionic Conductive, and Adhesive Patch Electrode with Ultra-low On-skin Impedance for Electrophysiological Signal Recording

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## Appendix A Introduction

The rapid proliferation of wearable bioelectronics has underscored the potential utility of flexible electrodes across diverse domains such as healthcare and neuroscience, driven by their mechanical properties akin to human skin tissues [1–5]. Leveraging electrodes facilitates the comprehensive recording and analysis of human electrical signals, including but not limited to electrocardiogram (ECG), electromyogram (EMG), electroencephalogram (EEG), and electrooculogram (EOG), thereby furnishing deeper insights into physiological conditions and the state of internal organs and tissues [6–8]. Securing persistent and reliable detection of electrophysiological signals requires a stable interface between electrodes and the scalp, demanding that electrodes fulfill rigorous criteria, including high signal-to-noise ratio (SNR), strong adhesion, and enduring stability [9, 10]. Nonetheless, commercial Ag/AgCl wet electrodes suffer from issues such as water loss, poor comfort, and inadequate mechanical strength, resulting in elevated interfacial impedance and the inability to withstand multiple peel-adhere cycles. Consequently, wet electrodes fail to consistently record high-fidelity biosignals over extended periods, significantly diminishing their utility. Therefore, contemporary research focuses on overcoming these obstacles by designing electrodes with enhanced mechanical strength, improved stability, superior conductivity, and stronger adhesion, which constitute the core objectives and inherent challenges of current investigations [11].

To address the limitations of commercial Ag/AgCl wet electrodes in long-term monitoring, several dry electrodes have been proposed due to their independence from liquid conductive mediums for extended use [12–15]. However, the rigidity of dry electrodes presents challenges, including inadequate contact with the scalp, particularly in hairy regions, resulting in suboptimal SNR, potential skin damage, and elevated impedance, all of which impede signal acquisition [16]. Certain semi-dry electrodes, crafted from flexible materials like silicone, rubber, and polyurethane, offer a means to enhance electrode SNR by maintaining scalp electrolyte moisture. Liu et al. achieved impedance reduction in the stratum corneum and minimized ECG signal attenuation by integrating a continuous slow-release electrolyte, penetrating the stratum corneum, into a hydrogel containing poly(vinyl alcohol) (PVA) and silver nanowires [17]. In contrast to dry electrodes, fluctuations in fluid release levels can induce signal instability, while the necessity for manual salt solution replenishment and discomfort persist as limiting factors hindering the widespread adoption of semi-dry electrodes [18].

The inherent abundance of ions in ionic gels obviates the necessity for manual electrolyte replenishment during application, offering a comfort and conductivity level surpassing that of semi-dry electrodes, matching the skin’s modulus. Typically, ionic gels are prepared by selecting a gel matrix containing ions or directly incorporating inorganic salts or ionic liquids, facilitating the free movement of ions within the gel and establishing a highly efficient ionic conductive network, thereby enhancing charge conduction and electrical conductivity [19–21]. Acrylic acid (AA) and 2-acrylamido-2-methylpropanesulfonic acid (AMPS) are frequently used monomers in the synthesis of ionic gels due to their ability to protonate in water, yielding a significant number of ions [22–24]. Moreover, the high hygroscopicity of AA and AMPS, along with the non-volatility of the ionic components, facilitate the maintenance of a dynamic equilibrium between water loss and absorption during use, thereby mitigating impedance increase resulting from gel dehydration. In a prior investigation, we attained a notable improvement in hydrogel conductivity  $\sim 1.5 \text{ S m}^{-1}$  through the incorporation of a conducting polymer into a PAMPS gel network with a high water content ratio (97.4%), resulting in ultra-low on-skin impedance and sustained stability exceeding one month in EEG-based brain-computer interface (BCI) devices [25]. AMPS, however, shows insufficient mechanical properties independently, making it challenging to preserve the original morphology through repeated bonding-dissociation cycles, necessitating the introduction new monomers to bolster the gel’s mechanical integrity [26]. Recent studies revealed that the mechanical strength of PAMPS gel can be augmented by incorporating another gel network including PVA, polyacrylamide (PAM), and poly(dimethylacrylamide) (PDMAAm), forming an interpenetrating gel matrix [27–29]. Nevertheless, the conductivity of this composite gel remains relatively low due to the interference arising

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from the presence of free-moving ions within the hydrogel network structure.

In this study, we polymerized AA and AMPS to form a PAA/PAMPS network hydrogel, incorporating inorganic salts or ionic liquids, resulting in a stretchable, robust ion-bonded electrode with ultra-low skin impedance. By optimizing the monomer ratio, we not only obtained an electrode with high ionic conductivity  $(2.8 \pm 0.3) \times 10^{-2} \text{ S cm}^{-1}$ , but also excellent mechanical properties with an elongation at break of about 330% and a maximum stress of  $\sim 24 \text{ kPa}$ . The further enhancement of ionic conductivity in the electrodes was achieved through the incorporation of electrolytes such as various salts or ionic liquid, resulting in an impedance level  $\sim 5\%$  of that observed with commercial Ag/AgCl electrodes in the low-frequency range. Furthermore, the presence of multiple functional groups on the surface of the our AMPI electrode, along with the hygroscopic nature of both the gel matrix and salts or ionic liquids, contributed to the hydrogel's favorable adhesion strength ( $\sim 75 \text{ kPa}$ ) and sustained stability for a duration exceeding two weeks. When interfaced with an ECG sensing module, our electrodes demonstrated the capability to reliably record ECG signals with a SNR of  $\sim 25 \text{ dB}$ . These ionic electrodes, characterized by a straightforward fabrication process, superior mechanical and electrical properties, and commendable stability, hold promise for diverse wearable bioelectronics.

## Appendix B Materials and methods

### Appendix B.1 Materials

Acrylic acid (AA, >99%), 2-acryloylamino-2-methyl-1-propanesulfonic acid (AMPS, 98%), Potassium persulfate (KPS, 99.90%), N,N'-Methylenebisacrylamide (MBA, >99%), Poly(ethylene glycol) (PEG, 400 average molecular weight), lithium bromide (LiBr, 99%), lithium chloride (LiCl, 99%), sodium chloride (NaCl, 99%) and choline chloride (ChCl, 99%) were purchased from Macklin (China). All materials were used without further purification.

### Appendix B.2 Preparation of AMPI hydrogel and gel electrode

The preparation of the AMPI hydrogel precursor solution involved combining 1 mL of AA, 1 g of AMPS, 0.00375 g of MBA, 0.075 g of KPS, 0.5 mL of PEG, and inorganic salts (or ionic liquids) with 4.5 mL of H<sub>2</sub>O. The mixture underwent thorough mixing for 30 minutes using the one-pot method, followed by a 30-minute period to remove air bubbles. Subsequently, the hydrogel precursor solution was poured into a PTFE mold, and the AMPI hydrogel was synthesized via UV illumination for 15 minutes. Before use, the surface of the AMPI hydrogel should be quickly rinsed with deionized water to remove any trace residues of unreacted substances, and the surface moisture should then be promptly absorbed with a lint-free cloth. The content of inorganic salts and ionic liquids was calculated as the ratio of the added inorganic salts or ionic liquids to the solutes' content, which is the sum of the content of deionized water and PEG. The AMPI hydrogel patch electrode was fabricated by modifying the 3M electrode. Initially, the original hydrogel on the 3M electrode was carefully removed using tweezers, followed by thorough wiping of any residual hydrogel on the AgCl electrode with anhydrous ethanol. Following this, cylindrical AMPI hydrogels, measuring 1 cm in radius and 0.2 cm in thickness, were fabricated and affixed onto the surfaces of AgCl electrodes to create AMPI hydrogel electrodes.

### Appendix B.3 Characterization

#### Appendix B.3.1 FTIR spectroscopy

FTIR spectroscopy was employed to analyze the prepared hydrogels using an infrared spectrometer (PE-Spectrum Two, USA). The spectral resolution was set at  $4 \text{ cm}^{-1}$ , and the scanning wave numbers ranged from 400 to  $4000 \text{ cm}^{-1}$ . Prior to testing, hydrogel samples were lyophilized and ground into powder using a vacuum freeze dryer (LGJ-10, Beijing Songyuan Huaxing Technology Development Co., Ltd., China). The powdered samples were then mixed with potassium bromide powder at a ratio of approximately 1:100 and pressed into transparent sheets to facilitate infrared spectrogram testing.

#### Appendix B.3.2 Mechanical characterization

Adhesion and stress-strain tests were conducted using a universal testing machine, specifically the Instron 3365 (USA). For the tensile test, hydrogel samples were prepared into long strips measuring  $60 \text{ mm} \times 15 \text{ mm} \times 2 \text{ mm}$ . These strips were clamped in the testing machine chuck with a center-to-center spacing of 40 to 50 mm, and the setup speed was adjusted to  $200 \text{ mm min}^{-1}$  for stretching the hydrogel. Subsequently, tensile stress-strain curves were generated to analyze the results, and parameters such as strain at break, Young's modulus, and toughness were calculated for different samples. Adhesion experiments were performed using the lap-shear method between hydrogels and vertically shaped slides. The hydrogel samples were shaped into circles with a radius of 1.2 cm. Initially, the hydrogel was securely fixed between the two slides to ensure complete contact without air bubbles. Then, the ends of the slides were clamped with the collet of the universal testing machine to achieve fixation. The tensile speed was set at  $50 \text{ mm min}^{-1}$ . The adhesion force was determined as the maximum force normalized to the area of skin covered by the hydrogel. Error values correspond to the standard deviation.

#### Appendix B.3.3 Electrochemical impedance spectroscopy

The electrochemical impedance spectroscopy experiments were conducted using a CS310H electrochemical work-station (Wuhan Kesite Instrument Co., Ltd., China), employing the three-electrode method to measure impedance with the skin at 1 mV and frequencies ranging from 0.1 Hz to 10 kHz. During the test, hydrogel electrodes with a diameter of 1.2 cm were positioned on the subject's left forearm, arranged in a straight line with a spacing of 7 cm. It was ensured that the working

electrode, reference electrode, and counter electrode were all from the same hydrogel sample. Before electrode placement, the skin surface was cleansed with anhydrous ethanol to eliminate any potential effects of skin surface contaminants.

### Appendix B.3.4 Stability measurement

The stability test involves placing the prepared hydrogel electrode in a natural, unsealed environment and recording its residual mass at different time intervals. The mass retention was calculated as follows:  $m = m_1/m_0 \times 100\%$ , where  $m_0$  represents the original mass, and  $m_1$  denotes the residual mass at various time points.

### Appendix B.3.5 Biocompatibility

Biocompatibility was assessed through the 3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyl-2H-tetrazolium bromide (MTT) cell in vitro toxicity assay. Gel samples were placed in the upper compartment of transwell plates, with serum-free Dulbecco's modified eagle medium (DMEM) in the lower compartment, and left to soak for 24, 48, and 72 hours. For the MTT colorimetric assay, L929 cells in logarithmic growth phase were suspended at a density of  $5 \times 10^3$  cells per milliliter and seeded into 96-well culture plates (100 L per well, in 5 replicate wells). After incubating for 12 hours to allow cell adhesion, 200  $\mu$ L of the 2PEDOT/PVA gel soak solution was added to the test sample wells, while a blank control DMEM group was established. The plates were placed in a 5% CO<sub>2</sub> incubator at 37 °C for 24, 48, and 72 hours. Subsequently, 20  $\mu$ L ( $5 \text{ g L}^{-1}$ ) of MTT solution was added to each well and incubated for an additional 4 hours. After removing the culture solution, 150 L of dimethyl sulfoxide (DMSO) was added to dissolve the crystals, and the absorbance (A) value was measured at 570 nm using an enzyme marker.

### Appendix B.3.6 Ionic conductivity

Electrochemical impedance spectroscopy (EIS) tests were conducted on AMPI hydrogels to assess their ionic conductivity utilizing an equivalent circuit model comprising three elements: resistive ( $R_\Omega$  and  $R_{ct}$ ), constant phase element (CPE), and Warburg element (ZW). The calculation formula for ionic conductivity ( $\sigma$ ) was determined as:  $\sigma = l/(R \times A)$ , where  $\sigma$  is ionic conductivity, A is the cross-sectional area of the sample, and l is the thickness of the sample. All experiments were repeated at least three times.

## Appendix B.4 Human movement monitoring

The hydrogel sensor was fabricated in a rectangular shape, featuring dimensions of 1.5 cm  $\times$  6 cm  $\times$  0.2 cm. The hydrogel sensor was initially affixed to various regions of the human body. Subsequently, the Model 2450 Digital Source Meter was linked to both terminals of the hydrogel sensor to facilitate real-time monitoring of body movements, such as finger, wrist, arm, and overall body movements, by observing changes in resistance exhibited by the Digital Source Meter.

## Appendix B.5 Electrophysiological signals recording

To capture electrophysiological signals (ECG, EMG, and EEG), a biosensing board (16-channels OpenBCI V3 open-source Arduino EEG brainwave module, China) was utilized, with an amplifier gain set to 24. For ECG signal recording, the left forearm of the volunteers was connected with the AMPI hydrogel electrode serving as the grounding electrode and positive electrode, while the right forearm was connected with the same electrode serving as the negative electrode. Real-time signals from the heart were recorded utilizing a biosensor monitor. In a similar fashion, ECG signals were concurrently recorded using commercial Ag/AgCl wet electrodes, which were placed in identical locations for comparison. A bandpass filter ranging from 5 to 50 Hz was applied, and a 50 Hz notch filter was utilized to eliminate power frequency during the ECG signal sampling (at a rate of 125 Hz). Surface electromyography (sEMG) signals were acquired as the volunteers clenched and relaxed their fists. Three AMPI hydrogel electrodes were positioned on the volunteers left forearm with a spacing of 3 cm. Distinct electromyographic signals were detected during fist clenching, while no muscle activity was observed during relaxation. EEG was used to monitor signals related to blinking and teeth biting. The AMPI hydrogel electrodes were employed for tests with the working electrodes positioned at FP<sub>1</sub> on the forehead and O<sub>1</sub> on the pillow, while the reference and ground electrodes were situated on both ears. EOG was employed to detect signal changes during blinking, with the working electrode placed above the left eye and the reference and ground electrodes positioned to the left and below the left eye, respectively. Similar filtering settings, including a bandpass filter from 5 to 50 Hz, removal of the 50 Hz power frequency, and a sampling rate of 125 Hz, were applied.

The SNR of each type of electrode was calculated according to the following equation:

$$SNR(dB) = 20 \times \log_{10} \frac{\sqrt{\sum_{k=1}^N V_{signal}(k)^2}}{\sqrt{\sum_{k=1}^N V_{noise}(k)^2}}$$

, where N is the number of samples,  $V_{signal}(k)$  and  $V_{noise}(k)$  are the voltage values of the signal.

## Appendix C Results and discussion

### Appendix C.1 Modelling and characterization of AMPI hydrogels

The AMPI hydrogels investigated in this study were synthesized through one-pot photopolymerization involving a mixture of precursors comprising AA, AMPS, PEG, ionic conductive components, cross-linker N,N'-Methylenebisacrylamide (MBA),

and initiator potassium persulfate (KPS) (Figure S1(a)). Biocompatible PAA and PAMPS were employed to establish hydrogel structure, thereby enhancing mechanical strength and ionic conductivity. However, to mitigate issues related to water loss and the inherent brittleness in hydrogel network structures, we introduced the plasticizer PEG to enhance flexibility and stability in the PAA/PAMPS hydrogel [30]. Regarding the ionic conductive components, we evaluated sodium chloride (NaCl), hygroscopic salts lithium chloride (LiCl) and lithium bromide (LiBr), and biocompatible ionic liquid choline chloride (ChCl), where the incorporation of LiCl emerged as the optimal choice after the following comprehensive assessment of the AMPI gel's performance, encompassing impedance, adhesion, and stability. The resulting AMPI hydrogel forms numerous hydrogen bonds with the skin and exhibits an electrostatic effect conducive to its effective adhesion to the skin surface as a patch electrode (Figure S1(b)). Furthermore, the abundance of ions within the hydrogel endows it with high ionic conductivity, rendering it suitable for applications such as strain sensors and monitoring various electrophysiological signals in the human body (Figure S1(c)).

In the hydrogel network structure of the AMPI hydrogel, AA and AMPS, serving as the two monomers, undergo substantial crosslinking to enhance the mechanical properties significantly. Fourier transform infrared spectroscopy (FTIR) was employed to examine the characteristic peaks of PAA gel, PAMPS gel, and PAA/PAMPS (AM) gel, enabling the assessment of the degree of integration in the hydrogel network hydrogels (Figure S2(a)). The bands observed at  $1725\text{ cm}^{-1}$ ,  $1640\text{ cm}^{-1}$ ,  $1555\text{ cm}^{-1}$ ,  $1222\text{ cm}^{-1}$  and  $1027\text{ cm}^{-1}$  correspond to the  $\text{-C=O}$  carbonyl band of AA, the  $\text{-C=O}$  band of AMPS, the N-C stretching, and the symmetric and asymmetric stretching of the  $\text{O=S=O}$  group in the  $\text{-SO}_3\text{H}$  moiety of AMPS, respectively, thereby confirming the formation of the hydrogel network [31]. It is noteworthy that a slight reduction in band intensity was observed in the bi-network hydrogel compared to the hydrogel devoid of AA, possibly attributable to the robust interactions between PAA and PAMPS chains.

To cater to the demands of forthcoming flexible sensors, it is imperative that they exhibit superior mechanical characteristics, encompassing toughness, elongation, and strength, which are crucial for applications involving elastic substrates or for enhancing reusability potential [32–34]. To achieve robust and resilient hydrogels, we modulated the ratio of monomers AA and AMPS to govern the formation of the hydrogel network structure comprising PAA and PAMPS, utilizing uniaxial tensile testing at room temperature to assess the mechanical properties of AM gels. A range of AMPI hydrogels with varying AA/AMPS mass ratios (3:1, 5:3, 1:1, 3:5, 1:3) were synthesized to examine the impact of net-work cross-linking degree on gel mechanics (Figure S2(b)). The elongation at break of the hydrogel peaked at  $\sim 408\%$  with a 3:1 ratio of AA to AMPS ( $A_3M_1$ ), whereas it diminished to  $\sim 248\%$  with a 1:3 ratio ( $A_1M_3$ ). Furthermore, hydrogel toughness gradually declined from  $\sim 55\text{ kJ/m}^3$  to  $\sim 17\text{ kJ/m}^3$  as the AA/AMPS ratio decreased. Notably, augmenting the AA/AMPS ratio substantially heightened the degree of hydrogen bonding cross-linking, effectively dissipating energy within the elastomeric network. While the overall change in Young's modulus was minimal, it reached a maximum of  $\sim 6.7\text{ kPa}$  at an AA and AMPS ratio of 1:1 ( $A_1M_1$ ), which was selected for the further investigation (Figure S2(c)).

Due to PEG's favorable hygroscopic properties, we introduced it into the AMI hydrogel to enhance its moisturizing ability and stability under atmospheric conditions. To evaluate the tensile properties and strength of the hydrogel, different proportions (0 wt%, 5 wt%, 10 wt%, 15 wt%, and 20 wt%) of PEG were integrated into the PAA/PAMPS network. For ease of reference regarding hydrogels comprising AA, AMPS, and PEG, we designate them as AMP hydrogels. With increasing PEG content, it was observed that the modulus gradually decreased, elongation at break increased steadily, and the toughness of the hydrogel remained largely unchanged (Figure S2(d-e)). By interacting with the PAA/PAMPS chains, PEG is inserted or embedded between the polymer chains, reducing the interaction forces between the polymer chains (e.g., van der Waals forces and hydrogen bonding), which reduces the rigidity of the polymer chains, making the entire network softer and easier to stretch [35]. Furthermore, the high molecular weight and effective dispersion of PEG contribute to increased hydrogel elasticity, while its exceptional hydration properties and formation of hydrogen bonds with water molecules alter the structure and organization of AMP hydrogels, rendering them softer and more elastic.

In addition to mechanical properties, electrical conductivity is also crucial for hydrogels to serve as epidermal electrodes. Utilizing electrochemical impedance spectroscopy, the ionic conductivity of AM hydrogel was determined, revealing a notable value of  $(3.4 \pm 0.2) \times 10^{-2}\text{ S cm}^{-1}$  (Figure S2(f)), likely attributed to the substantial presence of ionic charge carriers generated by the protonation of AA and AMPS, which bestows the AM hydrogels with exceptional ionic transport capabilities. Nevertheless, the addition of 10 wt% PEG to the AM hydrogels, chosen as an intermediate value, resulted in a decrease in ionic conductivity to  $(2.8 \pm 0.3) \times 10^{-2}\text{ S cm}^{-1}$  due to PEG's nonconductive nature, despite the improvement in stretching properties.

## Appendix C.2 Impedance of AMPI hydrogels

To assess the impedance of these hydrogels, a double-plate measurement setup was employed, where two parallel copper electrode plates were connected to an electrochemical workstation while securing the sample under test (Figure S3(a)). To standardize impedance comparisons across various electrodes, their values were normalized by multiplying with the electrodes area, recognizing that larger electrodes generally exhibit lower impedance. We measured the impedance of AM hydrogels at different monomer ratios, along with AMP hydrogels featuring varying PEG contents when keeping the ratio of AA to AMPS constant at 1:1. While adjusting the monomer ratio, we observed an impedance of  $\sim 146\ \Omega\text{ cm}^2$  at a low frequency of 0.1 Hz when AA and AMPS contents were comparable (Figure S4). The incorporation of PEG dispersed the internal network of the hydrogel, imparting softness but resulted in a gradual increase in hydrogel impedance with higher PEG content, likely due to the non-conductive nature of PEG (Figure S3(b)). While a moderate amount of PEG could enhance precursor solubility, excessive PEG softened the hydrogel excessively, affecting its strength; hence, a PEG addition of 10 wt% was utilized in subsequent experiments, resulting in an impedance of  $\sim 314\ \Omega\text{ cm}^2$  for the AMP hydrogel at 0.1 Hz.

To further decrease the impedance of gels, various representative ionic conductive components were introduced into the

AMP hydrogel network, including NaCl, hygroscopic salts LiCl and LiBr, and biocompatible ionic liquid ChCl, forming the AMPI hydrogel. The relatively limited presence of free ions in AMP hydrogels prompted these added ions from inorganic salts and ionic liquids to engage in ion-exchange reactions with the carboxyl and sulfonic acid groups in the PAA and PAMPS hydrogels, thereby enhancing the ionic concentration and electrical conductivity of the AMPI hydrogels, displaying a positive correlation at low concentrations (1 wt% - 4 wt%) (Figure S5). However, when the addition exceeded a certain threshold, these free ions hindered the formation of the hydrogel network, potentially compromising its conductive network formation and biocompatibility. For instance, when additions exceeded 2 wt% LiBr, the excess salt hindered the formation of the gel network, prompting the decision to maintain the maximum LiBr content at 2 wt%. The impedance of various AMPI hydrogels at maximum addition was compared, revealing lower impedance values  $\sim 30 \Omega \text{ cm}^2$  in the high-frequency range (100-1000 Hz) across all samples (Figure S3(c)). Conversely, in the low-frequency range (1-10 Hz), similar impedance values ranging from 50 to 150  $\Omega \text{ cm}^2$  were observed for all AMPI hydrogels.

Lower on-skin impedance, typically indicating a higher probability of detecting weak signals, is a crucial parameter for evaluating electrophysiological recording capabilities and effectively utilizing hydrogels for epidermal electronics, considering the inherently poor conductivity of the skin stratum corneum due to its lipid matrix [36]. Hydrogel electrodes attached to the skin were tested using an electrochemical workstation, with the three electrodes arranged equidistantly as counter electrode (CE), reference electrode (RE), and working electrode (WE) (Figure S3(d)). It is evident that the on-skin impedance of all AMPI hydrogels was significantly lower than that of the commercial Ag/AgCl wet electrodes ( $\sim 485.6 \text{ k}\Omega \text{ cm}^2$ ), and the on-skin impedance decreased with increasing salt or ionic liquid content (Figure S6). The 4 wt% NaCl addition resulted in an impedance of ( $\sim 106 \text{ k}\Omega \text{ cm}^2$ ) at 0.1 Hz, which was  $\sim 58\%$  of the AMP's impedance of ( $\sim 156 \text{ k}\Omega \text{ cm}^2$ ) at 0.1 Hz. Similarly, the addition of 4 wt% ChCl, 4 wt% LiCl, and 2 wt% LiBr led to more significant changes, with impedances of ( $\sim 19 \text{ k}\Omega \text{ cm}^2$ ), ( $\sim 24 \text{ k}\Omega \text{ cm}^2$ ), and ( $\sim 25 \text{ k}\Omega \text{ cm}^2$ ) at 0.1 Hz, representing only  $\sim 12\%$ ,  $\sim 15\%$ , and  $\sim 16\%$  of the AMP hydrogel impedance, respectively. The significant decrease in impedance can be attributed to the enhanced ionic conductivity resulting from the introduction of salt and ionic liquid, while in their absence, the capacitive coupling process between ionic and electronic currents at the electrode-skin interface is considerably weakened, leading to a higher bioelectrical interface impedance. Additionally, ionic conductivity measurements on different AMPI hydrogels with a thickness of 0.1 cm and a cross-sectional area of 0.2  $\text{cm}^2$  were conducted (Figure S3(e)). Consistent with the on-skin impedance results, gel with 4 wt% ChCl exhibited the highest ionic conductivity of  $(3.8 \pm 0.2) \times 10^{-2} \text{ S cm}^{-1}$ , while the gel with 4 wt% LiCl and 4 wt% NaCl displayed ionic conductivities of  $(3.3 \pm 0.5) \times 10^{-2} \text{ S cm}^{-1}$  and  $(2.9 \pm 0.4) \times 10^{-2} \text{ S cm}^{-1}$ , respectively. However, the ionic conductivity of gel with 2 wt% LiBr was only  $(1.4 \pm 0.2) \times 10^{-2} \text{ S cm}^{-1}$ , potentially due to LiBr's greater water absorption, likely resulting in heightened contact area with the skin by absorbing sweat and consequently reducing on-skin impedance. We conducted a comparative analysis of the ion conductivity of ionic hydrogels over the past two years, revealing that our current work has achieved a higher level of performance (Figure S7) [37–45]. To ensure precise and stable recording of electrophysiological signals, the on-skin impedance at low frequencies should ideally be below 100  $\text{k}\Omega \text{ cm}^2$  [46]. Comparing the impedance of our hydrogel with other recent hydrogel electrodes against this criterion places our hydrogels at the forefront of performance (Figure S3(f)) [36, 47–56].

Notably, all AMPI hydrogel electrodes exhibited secure adhesion to the skin surface without causing any observable adverse effects such as rashes or sensitization after 1 hour of use (Figure S8(a)). To further validate the biocompatibility of AMPI hydrogels, cytotoxicity tests were conducted using the MTT assay. The cell viability in the culture medium immersed in the AMPI hydrogel was approximately 90.26% on day 3, exceeding the recommended cell viability requirement of 70% according to the United States Pharmacopeia (USP) standard (ISO 10993-5), indicating the biocompatibility of AMPI hydrogel with cells and its suitability for long-term skin contact studies (Figure S8(b)).

### Appendix C.3 Adhesion of AMPI hydrogels

For epidermal electrodes, adhesion is crucial for accurately capturing electrophysiological signals from the human body, especially during activities like exercise, as it determines the electrodes' ability to securely adhere to the skin surface throughout the testing period. The abundance of hydroxyl, carboxyl, and sulfonic acid groups in AMPI hydrogels facilitates the formation of robust adhesion through hydrogen bonds or electrostatic forces with corresponding functional groups on the human skin surface (Figure S9(a)) [57]. The strong adhesion between the hydrogel and skin interface was evident upon peeling from a subject's arm (Figure S9(b)). Adhesion of AMPI hydrogels to glass substrates was verified using the lap-shear method with a universal testing machine (Figure S10). While the adhesion performance of AMP hydrogels reached  $\sim 11 \text{ kPa}$ , incorporation with inorganic salts or ionic liquids resulted in a reduction in hydrogel adhesion (Figure S9(c)). Specifically, the adhesion stresses of ChCl- and LiBr-added hydrogels decreased dramatically with increasing amounts, reaching  $\sim 5.2 \text{ kPa}$  and  $\sim 4.5 \text{ kPa}$  at 4 wt% and 2 wt% additions, respectively. Conversely, LiCl-added AMPI hydrogels exhibited a less pronounced decrease in adhesion, maintaining a level of  $\sim 7.5 \text{ kPa}$  even after a 4 wt% addition, representing a decrease of  $\sim 30\%$ . Notably, although the adhesion force of hydro-gels substantially decreased to  $\sim 5.1 \text{ kPa}$  with a 1 wt% NaCl addition, it exhibited a slight increase with higher doping amounts. The adhesion mechanism of AMPI hydrogels is primarily governed by the presence of carboxyl and hydroxyl groups on the surface. The introduction of inorganic salts and ionic liquids modifies the surface chemistry by inter-acting with these groups, diminishing surface interactions and adhesion. Specifically, ions from additives like NaCl, ChCl, LiCl, and LiBr engage in ion exchange reactions, altering the charge distribution and ionic composition of the hydrogel surface, thereby reducing its interaction and adhesion with external substances, which is more pronounced with increasing additions [58]. Additionally, the presence of salt or ionic liquid creates a solvent layer on the hydrogel surface, resulting in smoother surface characteristics and reduced contact area with other substances, further contributing to decreased adhesion.

Furthermore, the magnitude of the adhesion force is correlated with skin impedance and ample adhesion force ensures proper attachment of skin electrodes during testing, potentially reducing skin impedance by increasing contact area. Optical

images depicting the adhesion of AMPI hydrogels on various substrates illustrated their adhesive properties, attributed to dynamic hydrogen bonding, thereby promoting close skin contact and potentially lowering impedance, laying the groundwork for electrophysiological signal-recording devices (Figure S9(d)).

#### Appendix C.4 Stability of AMPI hydrogels

Hydrogels, with their advantageous properties, are prone to water loss, impacting mechanical properties, conductivity, and stability. Thus, for their use as electrodes in detecting bioelectrical signals, ensuring prolonged stability is essential. Incorporating suitable amounts of inorganic salts and ionic liquids aids in attracting water molecules to form hydrogen bonds, enhancing moisturizing ability, and markedly improving the hydrogel's ionic conductivity [59]. The stability of AMPI hydrogels under confined, low-humidity conditions with varying levels of salts or ionic liquids, including NaCl, ChCl, LiCl, and LiBr, was evaluated (Figure S11). Over a 14-day observation period, hydrogels supplemented with these additives showed significantly reduced water loss, with greater additions correlating with enhanced hydrogel stability, reaching dehydration equilibrium by the fourth day. Comparative analysis of mass changes for the highest-added amounts of these substances after 1 and 12 hours of exposure to air revealed that the addition of hygroscopic salt LiCl notably benefited hydrogel stability, with mass changes of  $\sim 71\%$  and  $\sim 54\%$ , respectively (Figure S9(e)). Conversely, AMPI hydrogels supplemented with NaCl and LiBr displayed mass changes of  $\sim 67\%$  and  $60\%$  respectively after 1 hour, with these figures rising to  $\sim 51\%$  and  $\sim 47\%$  respectively after 12 hours. While AMPI hydrogels incorporating the biocompatible ionic liquid ChCl exhibited the better electrical conductivity, they displayed limited effectiveness in enhancing hydrogel stability, manifesting rapid water loss with mass reductions to  $\sim 60\%$  and  $\sim 47\%$  after 1 and 12 hours respectively. Moreover, immersing AMP hydrogels in various concentrations of LiCl solution for 24 hours greatly improved hydrogel stability, preserving over 85% of their mass after two weeks; nevertheless, excessive ionic doping disrupted the hydrogel's electrical conductivity (Figure S12). The pronounced hydration of Li<sup>+</sup> and Cl<sup>-</sup> ions facilitates enhanced water retention, attributed to their strong attraction to water molecules [60]. However, the significant influence of Li<sup>+</sup> ions on the Hofmeister effect during hydrogel immersion, owing to their high hydration number, low charge number, and small atomic radius, results in reduced pore size within the hydrogel, thereby substantially impacting ion transport rates [43].

Subsequently, we conducted 14-day storage experiments on hydrogels containing the inorganic salt LiCl, renowned for its superior performance, and hydrogels enhanced with the ionic liquid ChCl, under normal temperature and sealed conditions. The groups stored at room temperature were denoted LiCl-1 and ChCl-1, while those under sealed storage were labeled LiCl-2 and ChCl-2. Sealed storage primarily resulted in volume shrinkage rather than drying, indicating reduced susceptibility to water loss (Figure S9(f)). This phenomenon could be ascribed to the rapid loss of water prompting closer interactions among internal polymer chains, thereby tightening the hydrogel structure, and resulting in subsequent volume reduction, which was notably pronounced during the initial three days and gradually subsided upon reaching dehydration equilibrium. Notably, our hydrogel showed sensitivity to ambient humidity; during days 5 to 8, increased air humidity caused by rain resulted in water absorption by the hydrogel from the atmosphere (Figure S9(g)). However, the encapsulated AMPI gels remain unaffected by environmental conditions, retaining their water content and maintaining their properties as in their initial state. By the 14th day, hydrogels containing ChCl exhibited a light yellow and transparent appearance, with light transmittance measurements indicating 75-80% for LiCl added gel and 60-65% for ChCl added gel (Figure S9(h)). Besides water loss, we assessed hydrogel stability during operation by monitoring the on-skin impedance of hydrogels with 4 wt% LiCl during extended wear. After approximately 5 hours, skin contact impedance decreased from 24.0 k $\Omega$  cm<sup>2</sup> to 12.2 k $\Omega$  cm<sup>2</sup> due to continuous skin moisturization by the hydrogel and subsequent ion exchange reactions enhancing electrical conductivity (Figure S9(i)). However, after 1 day of use, water loss from the hydrogel led to a significant rise in contact impedance to 278.9 k $\Omega$  cm<sup>2</sup>. Despite prolonged wear, the excellent biocompatibility of the AMPI hydrogel facilitated easy removal of electrodes from the skin without causing rash or residue (Figure S13). While the incorporation of significant quantities of inorganic salts can potentially harm the skin, small amounts of such salts can enhance the physicochemical properties of hydrogels, rendering them more compatible with biological tissues [61]. Notably, LiCl exhibits remarkable antimicrobial properties, mitigating bacterial proliferation and inflammation associated with prolonged wear [62, 63]. Consequently, this presents the prospect of employing AMPI hydrogels for extended periods in electrophysiological signal monitoring and sensing applications.

#### Appendix C.5 Stability of AMPI hydrogels

As hydrogel electrodes containing ChCl and LiCl exhibited lower impedance, after considering the stability and adhesion properties, we selected an AMPI hydrogel with 4 wt% LiCl for potential applications due to its overall excellent performance. To validate the sensing capabilities of AMPI hydrogels, various human activities were monitored. Hydrogel ends were affixed with copper tape, and real-time resistance was monitored using a digital source meter connected to the hydrogel wires. Finger bending at different angles was tested for 5 cycles, demonstrating increased relative resistance with bending angle while maintaining stability over cycles (Figure S15(a, b)). Additionally, the hydrogel sensor accurately responded to elbow and arm bending, reflecting the change in bending angle (Figure S15(c, d)).

Electrophysiological signals, including ECG, EMG, and EEG, provide vital physiological insights by detecting electrical activity in specific body regions, aiding in assessing human health status and facilitating therapeutic interventions through electrical stimulation. In ECG signal acquisition, a 16-lead biosensor plate is employed, with the ground electrode connected to the tester's left forearm's back end, the positive electrode to the left forearm's front end, and the negative electrode to the right forearm. Signal acquisition necessitates the tester's relaxation to ensure accuracy and validity while minimizing interference from other electrical appliances. A typical ECG signal comprises a P-wave (atrial depolarization), a QRS wave (ventricular depolarization), and a T-wave (ventricular repolarization). Compared to the commercial Ag/AgCl wet

electrodes, our developed AMPI hydrogel electrode exhibits lower impedance and superior adhesion. ECG signals acquired from the same volunteers and filtered using MATLAB software demonstrated significantly higher signal quality with the AMPI hydrogel electrode compared to Ag/AgCl wet electrodes (Figure S14(a)), with a calculated SNR of  $(24.6 \pm 0.5 \text{ dB})$  for the AMPI hydrogel electrode, surpassing the Ag/AgCl wet electrodes ( $23.2 \pm 0.8 \text{ dB}$ ).

In addition, we assessed the AMPI gel electrode's EMG detection capability by connecting all electrodes, including the grounding, positive, and negative electrodes, to the tester's left forearm's ascending and flexor muscle groups to optimize EMG signal acquisition. Notable potential changes were observed each time the fist was clenched during data acquisition, as evidenced by recordings in both clenched and relaxed states (Figure S14(b)). However, due to susceptibility to interference during electromyographic signal collection, the SNR for EMG signal acquisition ( $22.4 \pm 1.1 \text{ dB}$ ) was slightly lower than that for ECG signal acquisition. Unlike ECG and EMG signals, EEG signals are highly susceptible to environmental and subject interference, posing a challenge to electrode sensitivity due to their relatively small signal amplitudes. To assess our AMPI hydrogel electrode's effectiveness in EEG monitoring, we integrated it into an EEG acquisition system, positioning reference and ground electrodes on both earlobes and working electrodes at FP<sub>1</sub> in the frontal region and at O<sub>1</sub> in the occipital region. Signal tests involving biting and blinking in volunteers yielded robust signal fluctuations with high SNR ( $21.3 \pm 0.6 \text{ dB}$ ) (Figure S14(c)). Electrophysiological signals were also recorded around the eyes for EOG signal acquisition, a region less prone to external interference. By inducing regular blinking, high-quality EOG signals with an SNR of  $(26.4 \pm 0.7 \text{ dB})$  were obtained (Figure S14(d)).

Given that most electrophysiological signals produce weak body signals, signal amplification and noise filtering during acquisition are crucial. To address the limitations of commercially available 16-lead biosensor boards, which often yield signals with insufficient amplitude, we developed an in-house ECG sensing circuit system. This system comprises an ECG Active Front End (AFE) Module, Microcontroller unit (MCU), and Bluetooth Low Energy (BLE), assembled and powered to enable ECG acquisition through electrodes (Figure S14(e)). The actual circuit connection diagram is provided in the supplementary information (Figure S16). Following signal amplification and filtering at  $1000\times$ , our AMPI electrodes exhibited relatively higher magnitudes than commercial Ag/AgCl wet electrodes in capturing ECG signals with a SNR of  $(24.5 \pm 1.0 \text{ dB})$  (Figure S14(f)).

## Appendix C.6 Continuous and stable recording of ECG signal based on AMPI hydrogels

To underscore the potential of AMPI hydrogels in continuous real-time health monitoring, heart rate fluctuations during exercise in healthy volunteers were tracked (Figure S17(a)). Volunteers equipped with AMPI hydrogel electrodes connected to the ECG sensing module maintained a baseline heart rate of around 77 during relaxation (Figure S17(b)). Following 30 minutes of exercise and subsequent relaxation, the heart rate escalated to 104 (Figure S17(c)). Real-time heart rate assessments were performed on volunteers with varying exercise proficiency, revealing distinct responses. Despite similar baseline heart rates of 70-75 during a 10-minute relaxation period, non-proficient individuals experienced a rapid heart rate increase, surpassing 120 within 30 minutes of exercise, while proficient individuals showed a more gradual rise and quicker recovery post-exercise (Figure S17(d)).

In practical long-term monitoring scenarios, frequent removal of electrodes from the skin is unlikely, thus exposing them to sweat or water during activities such as showering, which can degrade the electrodes' performance due to the swelling [64]. To further understand the swelling ability of AMPI gel electrodes, monitoring the mass change of AMPI hydrogel in PBS solution at room temperature revealed rapid water absorption, increasing its mass tenfold within the first hour, slowing thereafter, and reaching 121 times the original mass within 48 hours (Figure S18). This behavior is attributed to the high charge density resulting from phosphates in AA, which promotes swelling [65]. Additionally, ionization of carboxyl groups in PAA/PAMPS network enhances water absorption by elevating osmotic pressure, while hydrophobic groups aid in creating water-retaining voids [66–68]. Investigation into water loss after 48 hours of immersion in PBS solution showed rapid desiccation and crack formation within one hour upon exposure to  $80^\circ\text{C}$ , likely due to structural damage during rapid swelling preventing network restoration, leading to crack formation (Figure S19). Similarly, when AMPI hydrogels undergo rehydration following substantial water loss, such as upon reaching equilibrium, they not only absorb water but also tend to develop cracks. This occurs because the contraction and condensation of polymer chains during water loss lead to a more rigid and fragile structure, making the hydrogel susceptible to rupture under mechanical stress. Nevertheless, during simulations simulating sweat effects with low and intermittent water content, AMPI hydrogel electrodes with PBS solution demonstrated minimal impact, showing a SNR of  $\sim 23.9 \pm 0.8 \text{ dB}$  during light exercise (Figure S17(e)). Despite slight mass variations due to water absorption and loss, the quality of ECG signal detection remained unaffected ( $\sim 105\%$  and  $\sim 95\%$  by mass, respectively) (Figure S17(f)). Rehydration or post-drying maintained stable on-skin contact impedance of the AMPI hydrogel, underscoring its resilience to sweat and water loss during exercise (Figure S17(g-h)). This aspect is paramount as it underscores the AMPI electrode's capacity to withstand a certain degree of water interference while upholding stable characteristics such as SNR, adhesion, and robustness, thereby preserving recording performance. Moreover, considering the inevitable water loss in hydrogel electrodes during prolonged health monitoring, attaining a dynamic equilibrium in gel water content through sweat or artificial supplementation could bolster monitoring stability. Hence, the swelling capacity of our AMPI hydrogel enables sweat or artificial hydration to offset water evaporation, establishing a dynamic equilibrium or retaining water within the electrodes without significant performance degradation, thus holding considerable promise for ensuring stable, long-term health monitoring. However, prolonged sweating may introduce impurities such as skin cuticle or dust into the AMPI electrodes, potentially causing signal attenuation, although this effect was not observed during our 2-hour test.

## Appendix D Conclusions

In this study, we have developed and evaluated AMPI electrodes with remarkable attributes, including ultra-low on-skin impedance, good ionic conductivity, adhesion, and stretchability. Through the incorporation of plasticizer PEG into the protonic network PAA/PAMPS hydrogel, we achieved a high ionic conductivity ( $\sim 0.028 \text{ S cm}^{-1}$ ) alongside exceptional mechanical properties (toughness of  $\sim 42 \text{ kJ m}^{-3}$  and elongation at break of  $\sim 390\%$ ). Furthermore, the addition of ionic conductive components, such as salts (NaCl, LiCl, and LiBr) or the ionic liquid ChCl, significantly reduced impedance. Specifically, our AMPI hydrogel with 4 wt% LiCl demonstrated an ultra-low impedance of  $\sim 24 \text{ k}\Omega \text{ cm}^2$  in the low frequency range, only  $\sim 5\%$  of commercial Ag/AgCl electrodes, while preserving transparency and biocompatibility on skin surface. Despite a decrease in adhesion with the introduction of additional ionic components, the abundance of hydroxyl, carboxyl, and sulfonic acid groups in the AMPI hydrogel formed robust hydrogen bonds with the skin, resulting in excellent adhesion force ( $\sim 7.5 \text{ kPa}$ ). Moreover, the hygroscopic nature of LiCl contributed to the stability of AMPI hydrogels, retaining  $\sim 54\%$  mass after 12 hours. These characteristics facilitated excellent and stable sensing capabilities, enabling the recording of human electrophysiological signals with a high SNR of  $\sim 25 \text{ dB}$  and no significant degradation after 5 hours of monitoring. Additionally, leveraging the swelling ability of AMPI gels, the prepared ECG sensing module enabled continuous and stable monitoring of ECG signals during exercise, maintaining a dynamic equilibrium between water loss and hydration. Overall, our AMPI hydrogels amalgamate a straightforward fabrication process with exceptional properties, offering promising prospects for the field of epidermal electronics and long-term health monitoring devices.

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