SCIENCE CHINA
Information Sciences

• LETTER •

February 2025, Vol. 68, Iss. 2, 129402:1–129402:2
https://doi.org/10.1007/s11432-024-4246-9

A stretchable, ionic conductive, and adhesive patch electrode with ultra-low on-skin impedance for electrophysiological signal recording

Yang LI[1,2*†], Yuzhe GU[1†], Sheng QIAN[2†], Yuncong PANG[2], Aoxi YU[2], Shuwen ZHENG[1], Wenjie XIA[1], Yuan LIAO[1], Baoguang LIU[1], Shujuan LIU[2] & Qiang ZHAO[1,2*]

[1]*College of Electronic and Optical Engineering & College of Flexible Electronics (Future Technology), Nanjing University of Posts & Telecommunications (NJUPT), Nanjing 210023, China*
[2]*State Key Laboratory of Organic Electronics and Information Displays & Jiangsu Key Laboratory for Biosensors, Institute of Advanced Materials (IAM), Nanjing University of Posts & Telecommunications (NJUPT), Nanjing 210023, China*

Received 10 June 2024/Revised 25 August 2024/Accepted 28 November 2024/Published online 15 January 2025

Citation Li Y, Gu Y Z, Qian S, et al. A stretchable, ionic conductive, and adhesive patch electrode with ultra-low on-skin impedance for electrophysiological signal recording. Sci China Inf Sci, 2025, 68(2): 129402, https://doi.org/10.1007/s11432-024-4246-9

Flexible electrodes have significant potential in fields such as healthcare and neuroscience, as they enable comprehensive recording and analysis of human electrical signals, providing deeper insights into physiological conditions and the state of internal organs and tissues [1,2]. However, commercial Ag/AgCl wet electrodes face issues such as dehydration, poor comfort, and insufficient mechanical strength, while other types of electrodes, like dry and semi-dry electrodes, also have their own limitations, including the higher contact impedance. In contrast, ionically conductive electrodes offer potential advantages such as transparency, stability, and low impedance, making them a promising alternative to existing electrically conductive electrodes [3]. Acrylic acid (AA) and 2-acrylamido-2-methylpropanesulfonic acid (AMPS) are commonly used in the synthesis of ionic gels, as they ionize in water to produce a large number of ions, helping to maintain moisture balance and reduce impedance caused by dehydration [4]. However, the presence of freely moving ions in the gel network can interfere with conductivity, highlighting the need to introduce new conductive networks to enhance performance [5].

Design and modeling of ionic gels. The ionic gel (AMPI) was synthesized via a one-pot photopolymerization process, involving a precursor mixture of AA, AMPS, polyethylene glycol (PEG), ionic conductive components, the crosslinker N,N'-Methylenebisacrylamide (MBA), and the initiator potassium persulfate (KPS) (Figure 1(a)). PAA and PAMPS were employed to construct a complex hydrogel network, enhancing both mechanical strength and ionic conductivity. To mitigate challenges associated with water loss and inherent brittleness, PEG was incorporated to enhance the flexibility and stability of the PAA/PAMPS hydrogel. For the ionic conductive component, we evaluated sodium chloride (NaCl), hygroscopic salts lithium chloride (LiCl) and lithium bromide (LiBr), and the biocompatible ionic liquid choline chloride (ChCl). After a comprehensive assessment of impedance, adhesion, and stability, LiCl emerged as the optimal choice for incorporation into the AMPI gel. For more detailed information and a discussion of the results refer to our supplementary information.

Results and discussion. To meet the demands of flexible sensors, hydrogels need excellent mechanical properties like toughness, elongation, and strength. The ratio of monomers AA and AMPS (A_xM_y) was adjusted to control the formation of the PAA and PAMPS network structure. The (A_1M_1) hydrogel exhibited the highest tensile strength of 23.47 kPa and a commendable elongation at break of 329%, along with a peak Young's modulus of ∼426.7 kPa. Given PEG's excellent hygroscopic properties, it was introduced (10 wt%) into the (A_1M_1) hydrogel (AMP hydrogel) to enhance its moisture retention and stability under atmospheric conditions. PEG could form interactions with PAA/PAMPS chains, making the network softer and more stretchable, while also improving elasticity (toughness of ∼42 $kJ \cdot m^{-3}$ and elongation at break of ∼390%). However, the non-conducting nature of PEG caused the ionic conductivity to decrease from $(3.4\pm0.2)\times10^{-2}$ to $(2.8\pm0.3)\times10^{-2}$ $S\cdot cm^{-1}$, but further experiments were chosen due to the excellent tuning of the properties.

To further decrease the impedance of gels, representative ionic conductive components like NaCl, LiCl, LiBr, and ChCl were added to the AMP hydrogel, forming the AMPI

* Corresponding author (email: yli@njupt.edu.cn, iamqzhao@njupt.edu.cn)
† These authors contributed equally to this work.

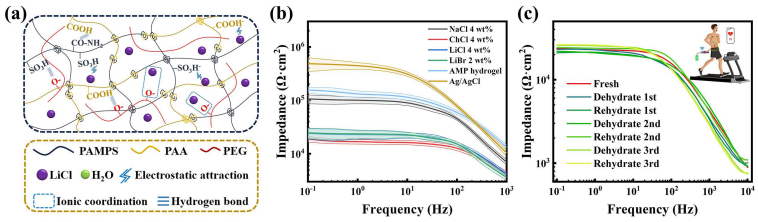

Figure 1 (Color online) (a) Schematic diagram of the AMPI hydrogel's internal network structure; (b) on-skin impedance versus frequency of Ag/AgCl wet electrodes, AMP hydrogels, and AMPI hydrogels containing different salts and ionic liquids; (c) on-skin impedance versus frequency of the AMPI hydrogel throughout a dry hydration cycle.

hydrogel. The added ions engaged in ion-exchange reactions with the carboxyl and sulfonic acid groups in PAA and PAMPS, enhancing ionic concentration and electrical conductivity, with a positive correlation at low concentrations (1 wt%–4 wt%). The AMPI hydrogels showed significantly lower epidermal impedance than commercial Ag/AgCl electrodes (∼485.6 $k\Omega \cdot cm^2$), with the lowest body impedance at the highest salt or ionic liquid concentrations. At 0.1 Hz, impedance was reduced to ∼19 $k\Omega \cdot cm^2$ for 4 wt% ChCl, ∼24 $k\Omega \cdot cm^2$ for 4 wt% LiCl, and ∼25 $k\Omega \cdot cm^2$ for 2 wt% LiBr, corresponding to only ∼12%, ∼15%, and ∼16% of the AMP hydrogel's impedance, respectively (Figure 1(b)).

However, adhesion is also critical for epidermal electrodes to capture accurate electrophysiological signals, especially during activities like exercise. Although the adhesion performance of AMP hydrogels reached ∼11 kPa, the addition of inorganic salts or ionic liquids led to a decrease in hydrogel adhesion, with the reduction in LiCl-added AMPI hydrogels being less pronounced, remaining at ∼7.5 kPa even with 4 wt% addition, a decrease of only ∼30%. Additionally, the inherent water loss of the hydrogel affected its mechanical properties, electrical conductivity, and stability. The 4 wt% LiCl-added AMPI hydrogel showed higher stability, and the human body impedance slightly decreased to 12.2 $k\Omega \cdot cm^2$ after 5 h of use, which was attributed to continuous moisturization of the skin by the hydrogel and subsequent ion-exchange reactions that enhanced electrical conductivity.

Due to the excellent overall performance of AMPI hydrogels with 4 wt% LiCl, they can accurately monitor human activities and detect electrophysiological signals, such as electrocardiogram (ECG), electromyogram (EMG), electroencephalogram (EEG), electrooculogram (EOG), with a high signal-to-noise ratio (SNR) (20–25 dB), outperforming commercial Ag/AgCl wet electrodes. Since most electrophysiological signals are weak, signal amplification and noise filtering are critical during signal acquisition. To address this, we also developed an internal ECG sensing circuit system. After amplifying the signal 1000 times and filtering, our AMPI electrodes further improved the SNR in capturing ECG signals. Under long-term exercise testing, we unexpectedly found that the AMPI hydrogel absorbed sweat without impacting the recording performance. To investigate this phenomenon in more detail, we monitored the mass change of AMPI hydrogels in PBS solution at room temperature. Due to the strong water absorption of PAA and PAMPS, the mass increased tenfold within one hour before slowing down. When simulating sweat through intermittent hydration and drying, the skin contact impedance remained stable, demonstrating strong resistance to sweat and water loss (Figure 1(c)). This stability maintained a consistent SNR, adhesion, and robustness under dynamic hydration conditions, supporting long-term monitoring. Although prolonged sweating could introduce impurities, no such effect was observed in our 2-hour test. This dynamic recharge strategy opens up tremendous possibilities for long-term monitoring applications.

Conclusion. 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 PAA/PAMPS hydrogel, we achieved a high ionic conductivity (∼0.028 $S \cdot cm^{-1}$) alongside exceptional mechanical properties (toughness of ∼42 $kJ \cdot m^{-3}$ and elongation at break of ∼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, exhibits an ultra-low impedance of ∼24 $k\Omega \cdot cm^2$ in the low-frequency range (only ∼5% of that of commercial Ag/AgCl electrodes) and good adhesion properties and stability. These characteristics facilitated excellent and stable sensing capabilities, enabling the recording of human electrophysiological signals with a high SNR of ∼25 dB and no significant degradation after 5 h 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.

Acknowledgements This work was financially supported by National Natural Science Foundation of China (Grant Nos. 62304112, 62288102), Natural Science Foundation of Jiangsu Province of China (Grant No. BK20230359), and Natural Science Research Start-up Foundation of Recruiting Talents of Nanjing University of Posts and Telecommunications (Grant No. NY221111).

Supporting information The supporting information is available online at info.scichina.com and link.springer.com. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the authors.

References

1 Li N, Li Y, Cheng Z, et al. Bioadhesive polymer semiconductors and transistors for intimate biointerfaces. Science, 2023, 381: 686–693
2 Li Y, Gu Y, Qian S, et al. An injectable, self-healable, and reusable PEDOT:PSS/PVA hydrogel patch electrode for epidermal electronics. Nano Res, 2024, 17: 5479–5490
3 Mogli G, Reina M, Chiappone A, et al. Self-powered integrated tactile sensing system based on ultrastretchable, self-healing and 3D printable ionic conductive hydrogel. Adv Funct Mater, 2024, 34: 2307133
4 Cui J, Xu R, Dong W, et al. Skin-inspired patterned hydrogel with strain-stiffening capability for strain sensors. ACS Appl Mater Interfaces, 2023, 15: 48736–48743
5 Kiyama R, Yoshida M, Nonoyama T, et al. Nanoscale TEM imaging of hydrogel network architecture. Adv Mater, 2023, 35: 2208902