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Cholinium-based eutectogel electrode for high-quality dynamic EEG/ECG monitoring exceeding 48 hours



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Bioelectrical signals, especially electroencephalography (EEG) and electrocardiography (ECG), are indispensable for health assessment. However, achieving high-quality signal acquisition while ensuring long-term stability remains a significant challenge in the bioelectrical electrodes. Herein, a cholinium-based eutectogel (ChCl-egel) is reported for dynamic ECG and EEG monitoring. By utilizing hydrogen bonding of deep eutectic solvents (DESSs) to modulate conductivity, the ChCl-egel realizes an ultra-low skin impedance (4.7 k Ω at 10 Hz) with reliable stability. Additionally, the ChCl-egel exhibits excellent conformal contact and biocompatibility. During ECG monitoring, the ChCl-egel offers an 8 dB improvement in signal-to-noise ratio (SNR) compared to commercial ECG electrodes (3 M, Red Dot) over 48-hour period, enabling dynamic record under high-intensity exercises. In an 18-hour steady-state visual evoked potential (SSVEP) experiment, it can match the scalp impedance of commercial gel and support longer record. With high-quality dynamic monitoring capability, the ChCl-egel provides a promising solution for long-term bioelectrical signal acquisition.

Wearable sensor technology has greatly expanded the applications in health monitoring and human-machine interface, enhancing its relevance to demands of daily life^{1–3}. In particular, bioelectrical signal acquisition plays a vital role in internal sensing and real-time monitoring of physiological states, including various signal types such as electroencephalogram (EEG), electrocardiogram (ECG) and electromyogram (EMG)^{4–6}. High-fidelity bioelectrical monitoring can identify abnormal symptoms such as epilepsy, arrhythmia, and muscle atrophy, facilitating timely crisis alerts^{7–9}. Additionally, it enables a wide range of human-machine interactions, including task recognition, emotional regulation, and robotic control^{10–12}. These signals provide valuable insights for the diagnosis and treatment of neurological and physiological diseases. To acquire high-quality bioelectric signals, electrodes must exhibit low interface impedance and extended operational durability^{13,14}. Currently, the main types of commercially available electrodes are dry electrode and wet electrode. Dry electrode is user-friendly but often suffers from high skin contact impedance up to several hundred kilohms, which compromises signal quality^{15,16}. In contrast, the commercial wet electrode is typically Ag/AgCl electrode combined with conductive

gel¹⁷. It can offer extremely low interface impedance (<10 k Ω at 10 Hz) and is considered the industry gold standard¹⁸. Although the signal quality is significantly improved, commercial gel electrode is uncomfortable to wear, prone to short-circuiting, and typically dry out within a few hours, making it unsuitable for long-term recording^{19,20}. Signal quality is directly influenced by the interface impedance between the electrode and the skin, while the flexibility and durability of the electrode determine its suitability for long-term stability. Moreover, movement inevitably affects skin impedance and electrode fit, maintaining real-time, high-quality signal recording is very difficult during physical movement. Therefore, it remains a substantial challenge to develop bioelectrical electrode that can achieve long-term recording and maintain high-quality dynamic signal acquisition even at variable levels of physical activity.

Up to date, various materials such as hydrogels have been investigated to improve bioelectrical acquisition performance^{21–23}. As a polymer cross-linked network, hydrogels exhibit excellent biocompatibility and electrochemical stability, making it suitable for long-term monitoring^{24,25}. However, its low ionic conductivity results in higher interface impedance

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than those of commercial gel electrodes, thereby inhibiting efficient signal transmission. Two primary factors contribute to these limitations: (1) the intrinsic chemical properties, which limit conductivity and charge transfer efficiency, and (2) the high viscosity and inferior mobility, which lead to a pronounced hysteresis effect during signal acquisition^{26,27}. Enhanced conductivity can be achieved by incorporating conductive fillers such as carbon-based materials (graphene, MXene, carbon nanotubes)^{28–31}, metallic nanomaterials (silver nanowires)³² and fibers (cellulose, porous silver fibers)^{33,34}. Nevertheless, the mismatch between the rigidity of the fillers and the polymer network may reduce the flexibility of hydrogel, compromising its reliability in practical applications.

Physical crosslinking, particularly non-covalent interactions such as hydrogen bonding, is an effective enhancement strategy. It can improve the electrical conductivity while also enhancing structural stability and adhesion³⁵. Deep eutectic solvents (DESs) possess complex hydrogen-bond networks, which enhance their stability and functionality³⁶. These are environmentally friendly and easily modified solvents consisting of monomers derived from biological sources. They are composed of hydrogen bond donors (HBDs) and hydrogen bond acceptors (HBAs), forming a homogeneous liquid system with a melting point lower than that of the individual components³⁷. By adjusting the ratio of these components, the properties of DESs such as conductivity, polarity, and viscosity can be precisely modulated³⁸. Specifically, hydrogen bonds form weak interactions between hydrogen atoms and more electronegative atoms in the HBDs³⁹. These non-covalent interactions reduce the electrostatic attraction between anions and cations, leading to a lower melting point and viscosity, which in turn enhance the conductivity of the whole system. Eutectogels have been developed that possess the electrical conductivity of DESs and the mechanical strength of polymers by dispersing DESs within a polymer network^{40,41}. While DESs have been widely used for gas trapping, organic separation, and azeotropic separation, their application in bioelectrical signal sensing remains less explored^{42,43}.

In this study, we present a cholinium-based eutectogel (ChCl-egel) that demonstrates high-quality dynamic EEG/ECG monitoring. The DESs consists of glycerol and choline chloride, whose hydrogen bonding interactions reduce ionic interactions, thereby enhancing ionic conductivity. As a dispersant, it can be introduced into gelatin network to significantly improve conductivity and mechanical stability of eutectogel. The ChCl-egel achieves a skin impedance of 4.7 k Ω at 10 Hz, comparable to that of commercial wet electrode. The tensile modulus is $\sim$ 40 kPa, ensuring excellent interfacial fit and comfort. Furthermore, the initial maximum stress remains above 90% after multiple cycles of tension and compression, demonstrating high mechanical resilience. Due to the moisture retention of glycerol, the ChCl-egel can maintain 93% weight retention after 5 days at room temperature, which supports the extended working time of the electrode. Biocompatibility assessments indicate that the ChCl-egel cannot cause skin irritation or sensitization. During ECG monitoring, the ChCl-egel can offer an 8 dB

improvement in signal-to-noise ratio (SNR) compared to commercial ECG electrode (3 M, Red Dot) over a 48 h period, enabling dynamic monitoring of heart rate even under high-intensity exercises. During EEG monitoring, the signal correlation between ChCl-egel and commercial wet gel is $>98\%$. In the steady-state visually evoked potential (SSVEP) experiment, it can maintain a scalp impedance of 5.3 k Ω in 8-channel recordings over 18 h. The ChCl-egel electrode exhibits ultra-low interface impedance, excellent mechanical stability and biocompatibility, making it highly suitable for long-term and high-quality physiological electrical monitoring.

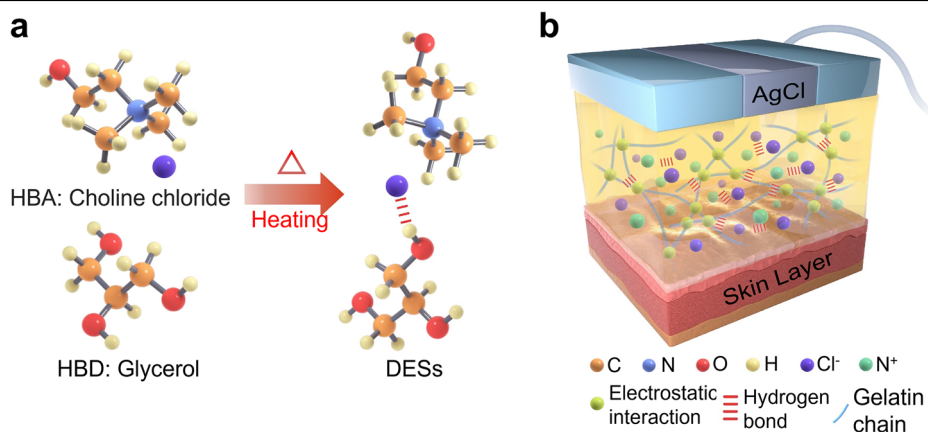
Results

Design and characterization of the ChCl-egel

Choline chloride (ChCl) is one of the most commonly used HBAs, recognized for its cost-effectiveness, non-toxicity, and biodegradability⁴⁴. As a widely utilized polyol, glycerol is particularly valued for excellent moisturizing properties and skin compatibility, which makes it an effective HBD in DESs systems⁴⁵. Consequently, ChCl and glycerol have been extensively studied as one of the earliest and most promising DESs combinations⁴⁶. As illustrated in Fig. 1a, the preparation of DESs involves heating ChCl and glycerol to form a homogeneous liquid. This process results in the formation of a complex hydrogen bonding network, where the continuous formation and breaking of hydrogen bonds between molecules significantly influences the conductivity and viscosity properties. A critical consideration in the design of eutectogel for bioelectrical signal acquisition is to enhance the flexibility and adhesion of the external crosslinked network. Gelatin is chosen as the external network material due to its thermo-reversible phase transition, which allows it to be in a liquid state at high temperatures and a gel state at room temperature. Therefore, the ChCl-egel can be formed by mixing gelatin, DESs, and deionized water without crosslinked agent at high temperatures. Figure 1b shows that multiple dynamic intramolecular and intermolecular interactions occur among the components of the ChCl-egel. The hydroxyl groups of glycerol and ChCl interact via hydrogen bonding, while the chloride anions (Cl^-) and the quaternary ammonium cations (N^+) of ChCl form electrostatic interactions. By leveraging non-covalent interactions, the melting point and viscosity can be reduced while electrical conductivity is greatly enhanced. Additionally, the hydrogen and electrostatic bonds further promote the physical crosslinking of the ChCl-egel. Its conductivity increases with higher water content, facilitating rapid ionic migration through the porous network. Gelatin serves as a network carrier, providing superior mechanical strength and structural connectivity. Combining the gelatin with DESs, ChCl-egel possesses excellent conductivity, flexibility and biocompatibility properties, making it a promising electrode material for bioelectrical signal acquisition.

As shown in Fig. 2a, the ChCl-egel is a transparent hydrogel that adheres to the fingertip and remains in place even when the finger is held vertically. Figure 2b shows that ChCl-egel maintains superior conformal contact with the pig skin substrate, with no obvious delamination at the

Fig. 1 | Schematic illustration of the ChCl-egel fabrication and its applications in bioelectrical signal acquisition. a The synthesis principle of DESs, Choline chloride (HBA) and glycerol (HBD) are combined and heated to form DESs. **b** The crosslinked interactions in the eutectogel networks, where the ChCl-egel adheres to the skin and facilitates bioelectrical signal acquisition via rapid ion movement through the network.



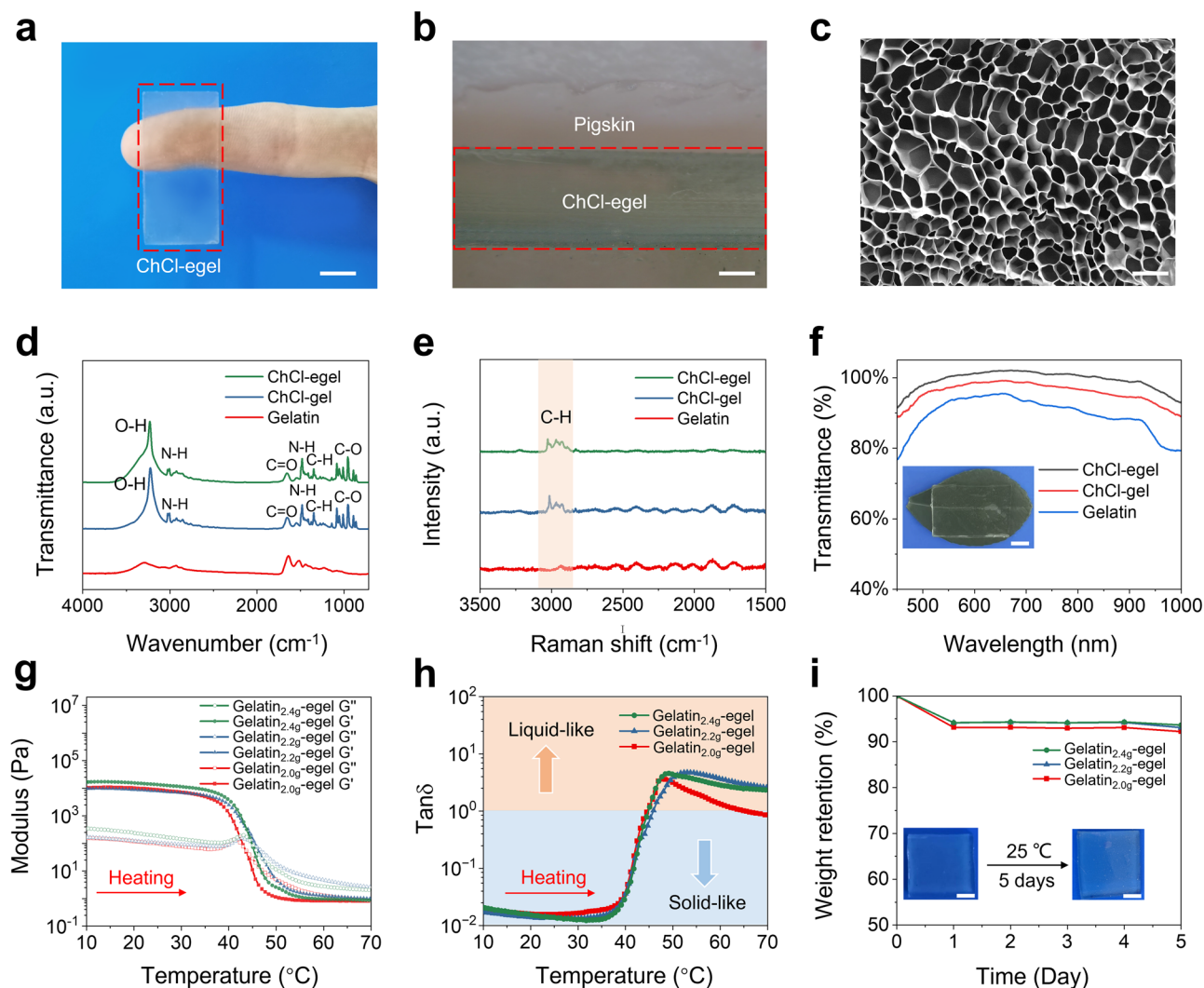


Fig. 2 | Characterization of ChCl-egel. **a** Photograph of ChCl-egel on skin. Scale bar: 2 cm. **b** Cross-sectional optical image of ChCl-egel attached to a pig skin substrate. Scale bar: 1 mm. **c** Cross-sectional SEM image of the internal structure of the freeze-dried ChCl-egel. Scale bar: 10 μ m. **d** FT-IR spectra and **e** Raman spectra of ChCl-egel with different formulations. The control group is pure gelatin without DESs. **f** Optical transmittance of ChCl-egel in the visible region. The inset photo

demonstrates that the intricate veins of the leaf can be clearly seen through the ChCl-egel. Scale bar: 1 cm. **g** Rheological characterization of ChCl-egel with different recipes at temperatures ranging from 10 °C to 70 °C. **h** The $\tan \delta$ (G''/G') calculated from the data in **(g)**. G' represents the storage modulus, while G'' represents the loss modulus. **i** The weight retention of ChCl-egel under conditions of 25 °C and 55% RH for 5 days. The inset photos depict the appearance change of the ChCl-egel. Scale bar: 0.5 cm.

interface, thereby enhancing the reliability of electrophysiological signal transmission. To further investigate the microstructure, cross-sectional scanning electron microscopy (SEM) characterization is performed. The SEM image in Fig. 2c illustrates the porous microstructure of the ChCl-egel after absorptive swelling and freeze-drying, which can facilitate the migration of conductive ions in the conductive path formed by the gelatin network. The interactions between the components of ChCl-egel are investigated using Fourier-transform infrared (FT-IR) spectra, as shown in Fig. 2d. ChCl-egel is composed of ChCl and gelatin. Due to the absence of glycerol, it does not exhibit the characteristics of deep eutectic solvents. Compared with the gelatin, the characteristic signals of ChCl-egel and ChCl-gel at ~ 1000 cm⁻¹ (C-O), 1481 cm⁻¹ (C-H), 1540 cm⁻¹ (N-H), 1650 cm⁻¹ (C = H) are generated. Additionally, a band broadening is observed in the 3100–3500 cm⁻¹ region, corresponding to (N-H) and (O-H) stretching vibrations, indicating the formation of strong hydrogen bonds between DES and gelatin. As shown in Fig. 2e, Raman spectral data show peaks in the range of 2850 to 3100 cm⁻¹ corresponding to C-H tensile vibrations, demonstrating the involvement of hydroxyl groups in hydrogen bonding or other molecular interactions. Figure 2f demonstrates that ChCl-egel can achieve nearly 100% transmittance in the 500–900 nm range compared to

ChCl-gel and pure gelatin. The high transmittance allows for clear visual inspection of the underlying skin, making it easier to detect potential irritation or sensitization during long-term wear.

Due to its temperature-dependent property, ChCl-egel transitions from a fluid state at elevated temperatures to a gel state at room temperature (Supplementary Fig. 1). The thermo-reversible property of ChCl-egel enables it to be reshaped into various shapes to conform to different areas of the human body (Supplementary Fig. 2). This property also facilitates reshaping without compromising its structural integrity. Additionally, the human skin temperature is ~ 37 °C. To prevent liquefaction and ensure stable performance during daily activities, the phase transition temperature of the ChCl-egel must exceed this value. It should be noted that gelatin concentration significantly affects DESs dilution and crosslinked strength. Excessive gelatin concentration can disrupt the uniform distribution during ion exchange, causing phase segregation of the colloids and negatively impacting performance. To verify the phase transition temperature of the eutectogel, the rheological properties of ChCl-egels with varying gelatin concentrations (Gelatin_{2.0g}-egel, Gelatin_{2.2g}-egel, Gelatin_{2.4g}-egel) are characterized, as shown in Fig. 2g. During the heating process, the storage modulus (G') and the loss modulus (G'') of ChCl-egels gradually decrease

with increasing temperature. This thermal response reflects the disruption of physical crosslinks and weakening of the internal structure at elevated temperatures. Figure 2h presents $\tan\delta$ (G''/G') calculated from the data in Fig. 2g, revealing phase transition temperatures of $\sim 44.4^\circ\text{C}$, 45.9°C , and 45.1°C for Gelatin_{2.0g}-egel, Gelatin_{2.2g}-egel, and Gelatin_{2.4g}-egel, respectively. Gelatin_{2.2g}-egel exhibits slightly higher thermal stability compared to the other formulations, owing to its denser crosslinked network. To prevent thermal softening above 45°C , the ChCl-egel should be stored in a cool, shaded environment, which ensures its structural stability during both storage and application. The cooling process exhibits similar behavior, with viscosity decreasing as the temperature rises (Supplementary Fig. 3 and Fig. 4). The ChCl-egel can reversibly transition between liquid and gel states, driven by thermally induced disruption and recovery of physical interactions.

In frequency scanning rheological measurements, the storage modulus (G') consistently exceeds the loss modulus (G'') across the frequency range of 0.1 Hz–150 Hz at room temperature, indicating that ChCl-egel remains in a gel state with a stable crosslinked network (Supplementary Fig. 5). Hydrogen bonding enables the ChCl-egel to undergo self-healing processes (Supplementary Fig. 6). To investigate this property, a strain step rheological test is performed in which the strain amplitude is alternated between low strain (5%) and high strain (250%). Under 250% strain, the storage modulus (G') drops sharply, indicating significant disruption or collapse of the internal hydrogen-bonded network within the gel. When the strain is subsequently reduced to 5%, the G' value rapidly recovers to its initial level, demonstrating that the hydrogen bonding interactions are re-established. This recovery reflects the reversible nature of the non-covalent crosslinking in the ChCl-egel, confirming its ability to autonomously restore mechanical integrity after deformation. Additionally, the ChCl-egel containing glycerol maintains stable electrical and mechanical properties over extended periods, effectively preventing hardening. As shown in Fig. 2i, ChCl-egels placed at 25°C and 55% humidity exhibit a 5.8% water loss on the first day, with only a 6.9% loss over 5 days (Supplementary Fig. 7). The inset photographs specifically show no significant changes in the appearance or volume of the ChCl-egel, highlighting its excellent weight retention. We further investigate the weight retention of ChCl-egels under physiological conditions. When ChCl-egels are placed in a humid environment with phosphate-buffered saline (PBS, pH 7.0 $\sim$ 7.2), the Gelatin_{2.2g}-egel exhibits 5.3% weight loss on the first day, and maintains over 94% of its original weight after 5 days (Supplementary Fig. 8). The weight change of ChCl-egel during 12 h of skin contact is shown in Supplementary Fig. 9. A slight increase is initially observed due to moisture absorption from the skin. After 12 h, the ChCl-egel retains 92.2% of its original weight, demonstrating good moisture retention and stability in humid or skin-contact environments, which supports its potential for long-term monitoring. Compared to ChCl-egels with different gelatin concentrations, Gelatin_{2.2g}-egel exhibits the highest crosslinked density due to lower swelling rates (Supplementary Fig. 10a). Supplementary Fig. 10b shows the visual appearance of the ChCl-egels after swelling and freeze-drying, confirming their ability to retain structural integrity and shape.

Electrical, mechanical, and biocompatible properties

The sensing responses of electrode are attributed to the electronic conduction pathways established by the ChCl-egel. Its electrode-skin interface can be modeled as containing a half-cell potential, with the double-layered structure represented by a parallel combination of a capacitor and a resistor (Supplementary Fig. 11). To obtain high-fidelity, long-term electrophysiological signals, an essential criterion is to construct a low-impedance, long-lasting interface between the sensor and human skin. DESs can be dispersed in gelatin to form an ionic conductive network, enhancing its electrical conductivity. However, incorporating excessive DESs may cause eutectogel instability and swelling, compromising its mechanical integrity and stability. There is a trade-off between conductivity and the amount of DESs. An optimal concentration of DESs in the eutectogel maximizes conductivity while maintaining the structural integrity and functionality of the gelatin (Supplementary Fig. 12). The electrical conductivity of the ChCl-

egel during mechanical stretching is demonstrated, with the lamp remaining consistently illuminated even after multiple stretching cycles (Supplementary Mov. 1). The effects of gelatin concentration on electrical conductivity are exhibited in supplementary Fig. 13. Due to the lower crosslinking density in the 2.0 g gelatin sample and the excessive crosslinking in the 2.4 g gelatin sample, both exhibit suboptimal conductivity. As shown in Fig. 3a, the optimized ChCl-egel with 2.2 g gelatin exhibits a low impedance (below 6 k Ω) over a frequency range of 0.1–100,000 Hz. Figure 3b compares the impedance of ChCl-egels with various gelatin concentrations at 1 Hz, 10 Hz, and 100 Hz, indicating that the 2.2 g gelatin sample has the highest crosslinked density and ion mobility. Additionally, the electrochemical impedance spectroscopy (EIS) test is carried out to demonstrate the ionic conductivity of the ChCl-egel (Supplementary Fig. 14). According to the Nyquist plots, the conductivities of Gelatin_{2.0g}-egel, Gelatin_{2.2g}-egel and Gelatin_{2.4g}-egel are 0.84, 0.96, and 0.82 mS cm⁻¹ respectively, as shown in Fig. 3c. In addition, cyclic compression tests are performed on ChCl-egels with varying gelatin contents to assess their mechanical properties. As shown in Fig. 3g and Supplementary Fig. 15, higher gelatin content leads to a greater peak stress. The peak stress reaches ~ 60 kPa for Gelatin_{2.4g}-egel, ~ 50 kPa for Gelatin_{2.2g}-egel, and ~ 40 kPa for Gelatin_{2.0g}-egel. These results indicate that increasing gelatin content enhances the load-bearing capacity. As the number of cycles increases, the compressive stress retention reflects better cyclic stability, which is critical for the long-term and reliable monitoring of wearable electrodes under repeated deformations.

To evaluate the mechanical performance of the ChCl-egel, the tensile and compression tests are performed to quantify the mechanical stability, including ultimate strain, elastic modulus, and energy dissipation. The experimental setup of the tensile test is shown in Fig. 3d. The ChCl-egel exhibits excellent tensile properties and can endure rapid stretching. Elastic modulus ranges from 40 to 60 kPa, which increases with tensile speed (Supplementary Fig. 16a and Mov. 2). Fatigue resistance is evaluated by applying multiple stress cycles with no dwell time. As shown in Fig. 3e, a large hysteresis loop is observed in the first stress cycle, which may be due to the breakage of hydrogen bond crosslinks in the network. After the first cycle, the hysteresis loops almost overlap in the subsequent stress cycles. Specifically, the peak tensile stress remains almost above 95% and the energy dissipation is maintained at 10% under repeated tensile loading (Fig. 3f). Compression tests at different strains demonstrate that the mechanical properties of the hydrogel remain highly stable (Supplementary Fig. 16b). The hysteresis is not obvious until 60% compression (Supplementary Fig. 17). Figure 3g shows the compressive stress curves under 50% strain, where the hysteresis loops almost overlap at 6 cycles. Correspondingly, the maximum compressive stress decreases from 48.5 kPa to 44.6 kPa, with an average energy dissipation of $\sim 8.4\%$, as shown in Fig. 3h. These results indicate that the ChCl-egel exhibits excellent elasticity, attributable to the homogeneous stress distribution within its internal network. Adhesion properties are crucial for maintaining a stable electrode-epidermal interface. Weak adhesion allows the ChCl-egel to be easily removed from skin or surfaces, preventing damage or residue, which is crucial for long-term medical use. T-peel tests are performed to assess the adhesion strength of ChCl-egel on different substrates (Supplementary Fig. 18). As shown in Fig. 3i, the measured adhesion strengths follow the order: steel (6.18 N/m), pig skin (4.37 N/m), plastic (3.73 N/m), wood (2.63 N/m). Relatively stable and high peeling forces are observed for steel and pig skin substrates, as shown in the force-displacement curves. Lap-shear tests (Supplementary Fig. 19) further confirm that ChCl-egel adheres most strongly to steel among all tested substrates. This strong adhesion is attributed to the high hydrophilicity of steel, which facilitates the formation of hydrogen bonds at the interface with the ChCl-egel. These bonds provide effective intermolecular interactions that enhance adhesion strength. In contrast, the softness and microscopic roughness of pig skin hinder intimate contact and lead to uneven stress distribution during peeling. Wood shows the weakest adhesion, primarily due to its porous structure, which disperses applied stress and reduces the effective bonding area. Notably, the ChCl-egel can be peeled off without leaving residue or causing allergic reactions.

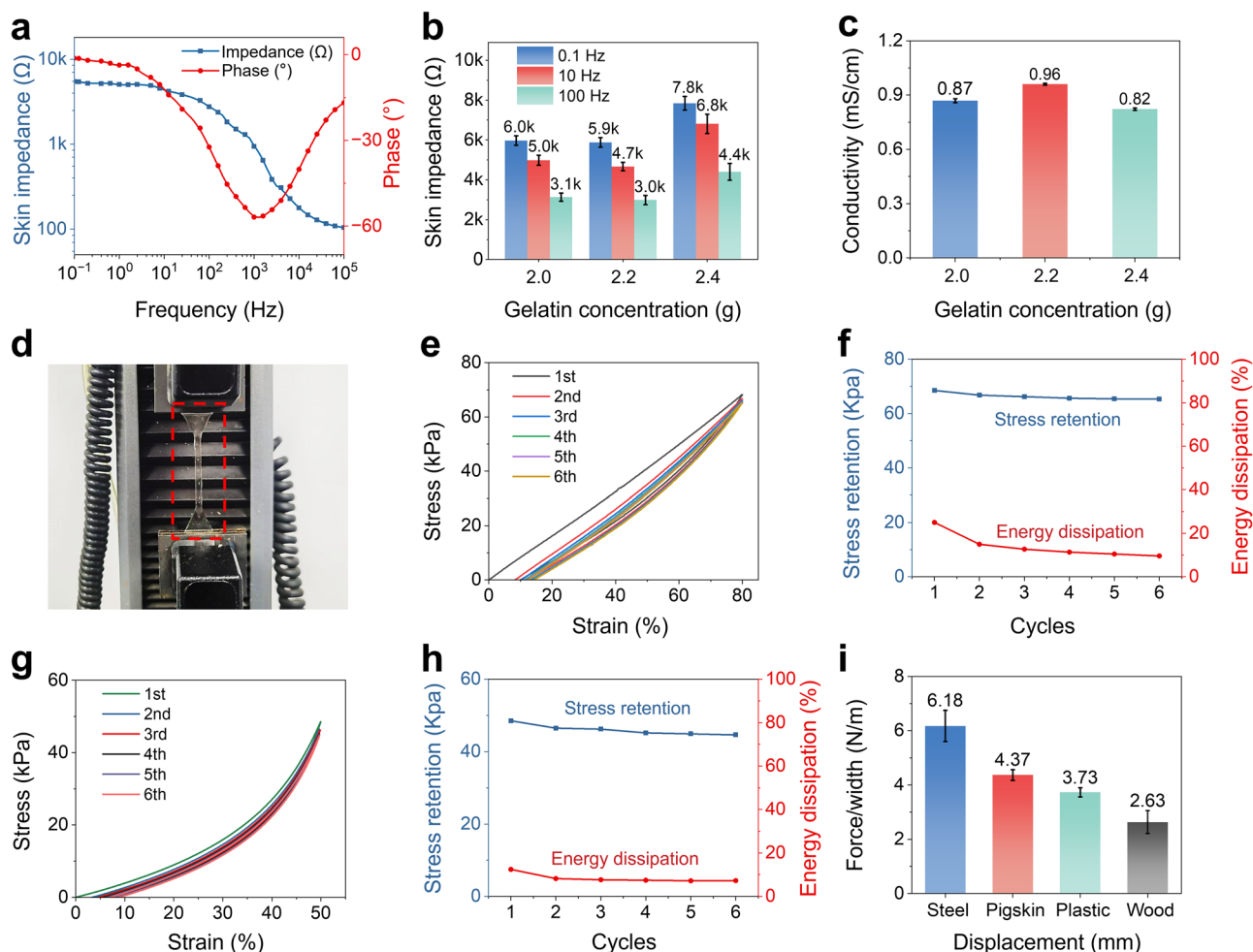


Fig. 3 | Electrical and mechanical properties. **a** The skin-electrode contact impedance and phase spectra of ChCl-egel. **b** Comparison of impedance of ChCl-egels with various concentrations of gelatin at 1 Hz, 10 Hz, and 100 Hz. **c** Comparison of ionic conductivity of ChCl-egels with various concentrations of gelatin. **d** Image of a mechanical tensile testing device. **e** Tensile stress under 50% strain and (f) energy

loss coefficient $\Delta U/U$ and stress maintenance variation under different cyclic tensile conditions. **g** Compressive stress curves under 50% strain and (h) energy loss coefficient $\Delta U/U$ and stress maintenance variation under different cyclic compressive conditions. **i** Comparison of T-peel forces per width of ChCl-egels on different substrates.

Biocompatibility is a crucial attribute for bioelectrical signal acquisition. The pHs of DESs and ChCl-egel exhibit neutral values ($6 \sim 7$) without causing significant irritation on skin (Supplementary Fig. 20). To evaluate the cytotoxicity of ChCl-egel, a 3 day dynamic monitoring assay is conducted using L929 fibroblasts, with cell viability assessed via the CCK-8 assay at 24 h, 48 h, and 72 h post-incubation. As shown in Supplementary Fig. 21, ChCl-egel maintains cell viability above 90% across all time points (98.2% at 24 h, 92.0% at 48 h, and 99.8% at 72 h) without significant decline, indicating no cumulative cytotoxicity. The negative control group exhibits over 99% viability throughout, confirming minimal interference from culture conditions. In contrast, the positive control group (20% DMSO) shows $\sim 1\%$ viability, confirming the assay sensitivity to cytotoxic stimuli. Moreover, skin irritation tests are conducted on rabbits, and sensitization tests are performed on rats. Neither the negative control group nor the test sample group exhibits edema or erythema at 0, 24, 48, or 72 h of exposure, indicating that no skin irritation occurred (Supplementary Fig. 22). Furthermore, at the end of the 14th day, neither the negative control group nor the test sample group in guinea pigs displays significant changes in responses at 24 or 48 h (Supplementary Fig. 23). The sample also does not cause sensitization.

Continuous and dynamic ECG monitoring in daily life

The ChCl-egel, with low impedance and excellent skin contact, is capable of recording high-quality electrophysiological signals in daily life, athletic

training, and clinical diagnostics. Figure 4a illustrates the ECG signal acquisition process using a single chest lead configuration. Two ChCl-egels are attached to the right chest and stomach as ground and reference electrodes, while a third is placed over the heart to record real-time signals as the working electrode (Supplementary Fig. 24). As shown in Fig. 4b and Supplementary Fig. 25, ECG signals are simultaneously recorded for 48 h using both ChCl-egel and commercial 3 M electrodes. The P wave, QRS complex, and T wave are clearly visible in Fig. 4b. ECG signals obtained using the ChCl-egel are of higher quality compared to those acquired with the commercial 3 M electrode, which is essential for the diagnosis of cardiac abnormalities. In long-term ECG monitoring, ECG signals captured by the ChCl-egel show no significant change in characteristic peaks after 48 h of continuous recording. The SNR and mean squared error (MSE) of the ECG recordings, obtained using both the ChCl-egel and commercial 3 M electrodes, are calculated based on the signals before and after filtering. Figure 4e compares the average SNR and MSE of ECG signals of multiple subjects, revealing that the ChCl-egel achieves a high SNR of 47.8 ± 1.0 dB, whereas commercial 3 M exhibits a lower SNR of 39.8 ± 2.0 dB. This indicates that the ChCl-egel provides high-quality ECG monitoring over extended periods without interfering with daily activities.

During dynamic ECG monitoring, human activity and sweating can increase impedance at the electrode-skin interface, leading to poor contact. This can also introduce motion artifacts that interfere

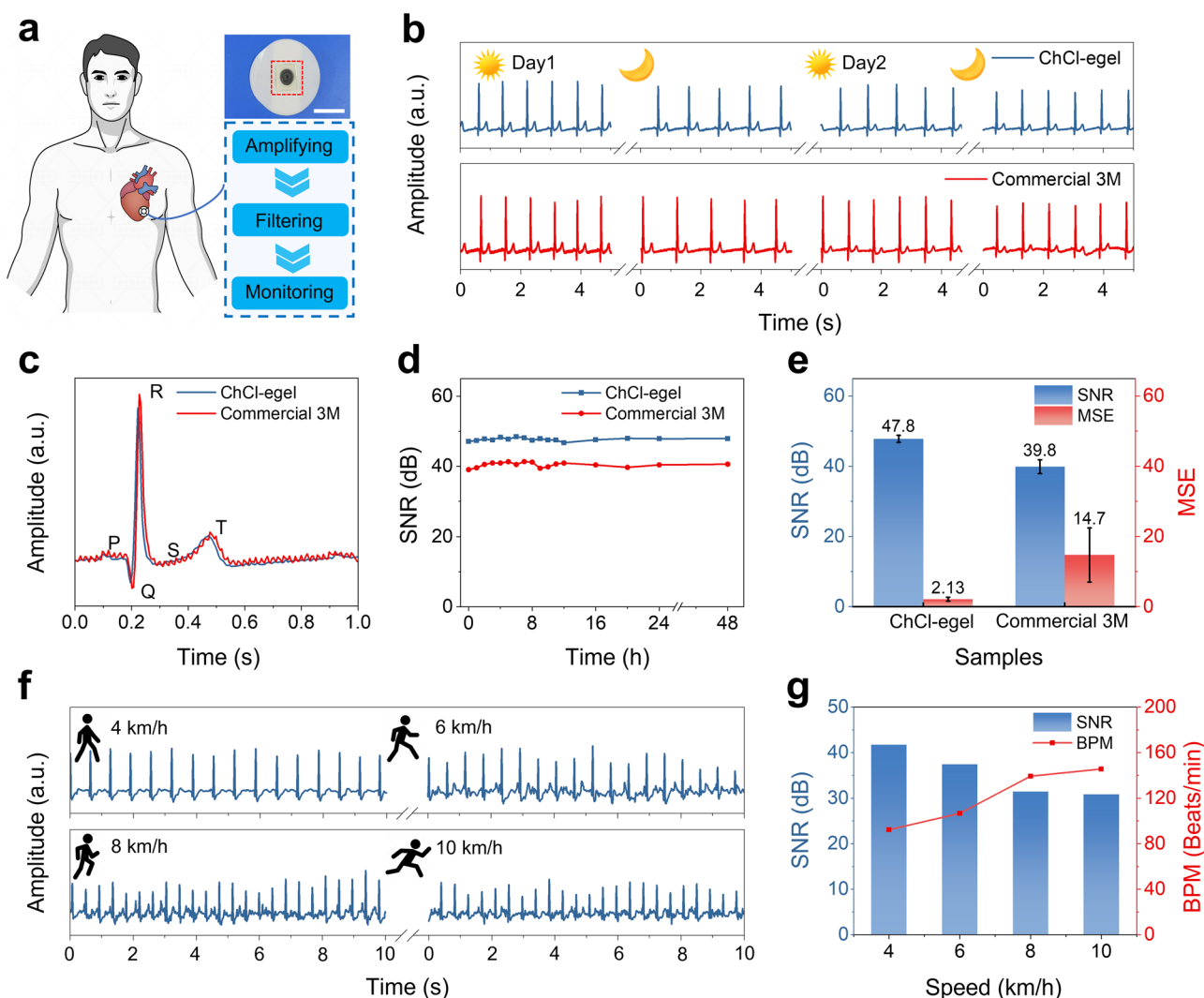


Fig. 4 | Continuous and dynamic ECG monitoring in daily activities. **a** Schematic of the ECG signal acquisition. **b** Comparison of ECG signals acquired by the ChCl-egel and commercial 3 M electrode over 48 h. **c** Characteristic peaks in ECG signals, P, QRS, and T waves. **d** Corresponding SNR changes. **e** Comparison of average SNR

and MSE for ECG signals. **f** Continuous ECG monitoring during dynamic human motion: slow walking (4 km/h), brisk walking (6 km/h), jogging (8 km/h), and running (10 km/h). **g** Corresponding SNR and BPM values.

with the stability and accuracy of the ECG signal. To evaluate the influence of sweat and exercise, we conducted an experiment to examine the electrode capability for dynamic monitoring during variable levels of physical activity. Figure 4f exhibits continuous ECG monitoring under slow walking (4 km/h), brisk walking (6 km/h), jogging (8 km/h), and running (10 km/h). The ECG signals remain clear, with key waveforms intact during slow and brisk walking, indicating that ChCl-egel electrodes provide stable signal acquisition during low-intensity exercise. Under strenuous exercise conditions, although the key waveforms in the ECG signals remain legible, certain artefacts may manifest in the signals. As illustrated in Fig. 4g, the beats per minute (BPM) increase with increasing velocity, while SNR is >30 dB even at 10 km/h running speed. Another subject also performed EEG acquisition while running, demonstrating that the electrodes have a high SNR and can be used for both continuous and dynamic ECG monitoring (Supplementary Fig. 26). Due to the ultra-low impedance and the soft-fit interface design, the ChCl-egel can effectively mitigate sweat interference with signal quality.

High-fidelity EEG record and SSVEP classification

EEG signals provide an effective and non-invasive approach to monitoring real-time brain electrical activity. Compared with ECG, high-fidelity EEG

recording is much more challenging because of their microvolt-level amplitude and the density of hair on the scalp. The quality of EEG signals can be affected by the potential drift of electrode. To assess this effect, the open-circuit potential (OCP) of the ChCl-egel and commercial gel electrode is measured (Supplementary Fig. 27). The OCP of the ChCl-egel is significantly lower than that of commercial gel electrode. In a 10 min test, the ChCl-egel electrode exhibits equilibrium potentials and potential drifts of 0.84 ± 0.015 mV and 0.31 ± 0.23 μ V/min, respectively, whereas the commercial gel electrodes show values of 1.31 ± 0.013 mV and 0.25 ± 0.19 μ V/min. Both the equilibrium potentials and potential drifts for the ChCl-egel and commercial gel electrode fall within the recommended guidelines (≤ 0.5 mV for equilibrium potential and ≤ 10 μ V/min for potential drift). To verify the correlation between EEG signals acquired with the ChCl-egel and commercial gel electrode, both types of electrodes are placed in the occipital region of the scalp during an eye-opening and eye-closing task (Supplementary Fig. 28a). The EEG signals recorded using the ChCl-egel exhibit no significant differences from those obtained with the commercial gel, in terms of waveform, amplitude, and power spectral density (PSD) during the same task period. A distinct central peak at 8–13 Hz, characteristic of EEG alpha rhythms, can be observed in the PSD of the eyes-closed task, while no alpha rhythms are generated during the eye-opening recording (Supplementary Fig. 29 and Fig. 30).

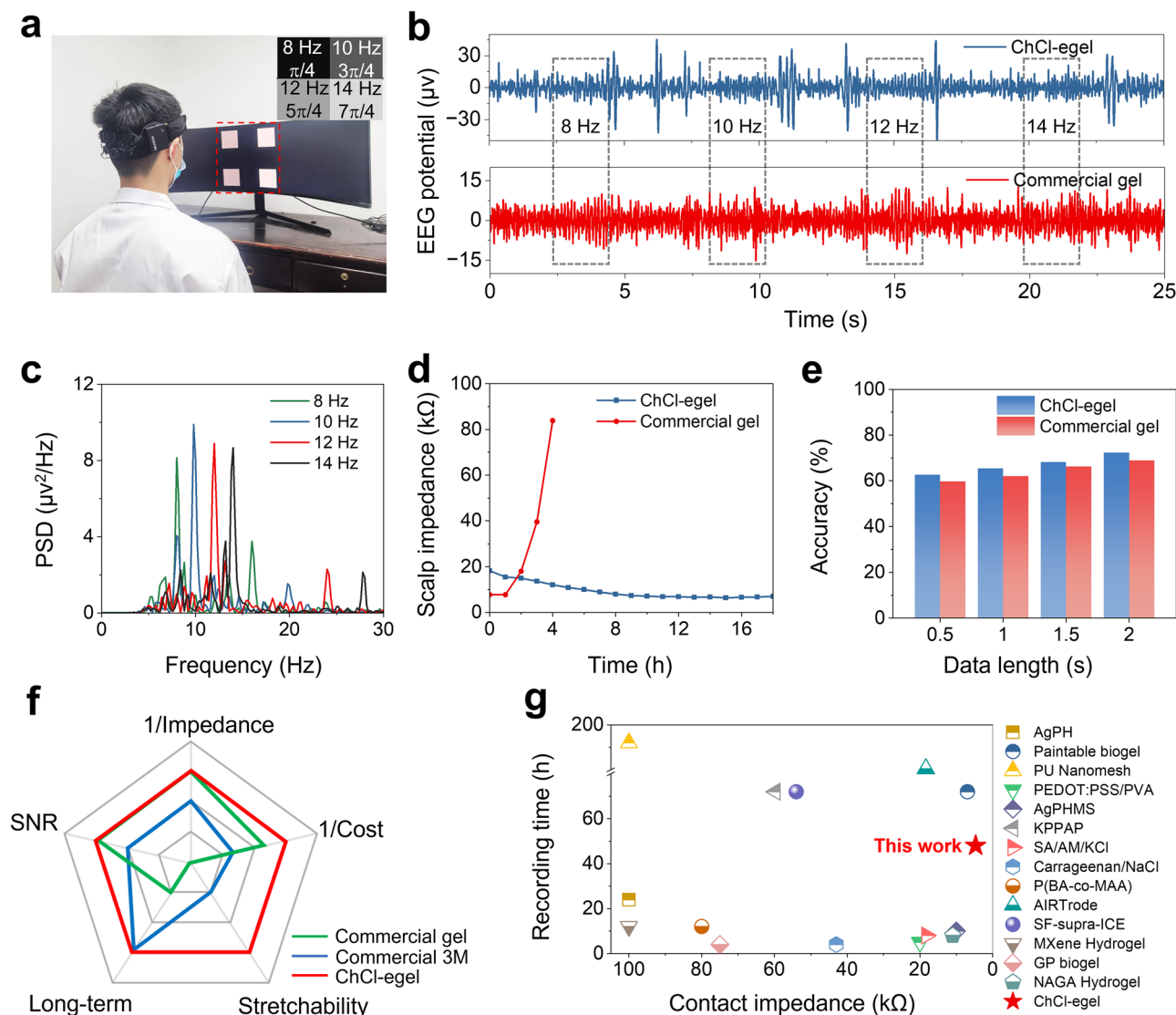


Fig. 5 | High-fidelity EEG recording and SSVEP Classification. **a** Photograph of a subject being tested to evoke SSVEP response with 8 channels. SSVEP paradigm: 8 Hz, $\pi/4$; 10 Hz, $3\pi/4$; 12 Hz, $5\pi/4$; 14 Hz, $7\pi/4$. **b** SSVEP signals recorded when the subject gazed at four goals. **c** PSD for SSVEP signals. **d** Scalp impedance using ChCl-egel for 18 h and commercial gel for 4 h. **e** SSVEP classification accuracy at different

time scales by the ChCl-egel and commercial gel. **f** Comparison of characteristics among commercial 3 M, commercial gel and ChCl-egel, presented as radar plots that illustrate their properties and preferences. **g** Comparison of contact impedance and recording time with other materials from the literature.

The Pearson correlations coefficient between the ChCl-egel and the commercial gel is calculated over 6 h period. A Pearson correlation coefficient close to 1 indicates a high degree of similarity between the two signals, suggesting that they capture nearly identical neural activity patterns. Supplementary Fig. 31 shows significant correlations >0.98 in 6 h, confirming that the ChCl-egel provides signal acquisition capability comparable to that of the commercial gel electrode.

SSVEP is a crucial paradigm of EEG task, the recording and classification of SSVEP is of great importance for EEG-based system. By using visual stimuli that flash at a specific frequency, SSVEPs enable higher accuracy and information transmission rates (ITRs) in signal acquisition (Supplementary Mov. 3). The ChCl-egels are placed according to the 10–10 international standards on PO5, PO3, POz, PO4, PO6, O1, Oz, and O2, the reference and ground electrodes are placed on the forehead (Supplementary Fig. 28b). As illustrated in Fig. 5a, a subject is tested to elicit SSVEP responses using an 8-channel system. The presented stimuli included frequencies and phases set as follows: 8 Hz ($\pi/4$); 10 Hz ($3\pi/4$); 12 Hz ($5\pi/4$); 14 Hz ($7\pi/4$). SSVEP signals could be evoked when the subject concentrated on the flicker with the same frequency in Fig. 5b. The topography maps are generated to

depict the spatial distribution of EEG responses (Supplementary Fig. 32). This spatial pattern is consistent with visual cortex involvement and demonstrates the effectiveness of the 8-channel EEG in capturing SSVEP components. As shown in Fig. 5c, their PSD peaks are at the corresponding and harmonic frequencies, which is important for high classification accuracy of SSVEP task. Figure 5d shows the scalp impedances of multiple subjects using ChCl-egel for 18 h and commercial gel for 4 h. ChCl-egel can achieve more conformal contact with the scalp over time, resulting in a gradual decrease in impedance to ~ 5.3 k Ω during 18 h monitoring. In contrast, commercial gel electrode gradually dries out, leading to increasing impedance and making it unsuitable for long-term monitoring. Specifically, the scalp impedance variation of one subject in different channels is shown in Supplementary Fig. 33. To further assess the classification accuracy, we use task-related component analysis (TRCA) for the analysis. Figure 5e shows that longer time lengths lead to larger classification accuracies. For acquisition via ChCl-egel, the accuracies are 59.7% (0.5 s), 62.0% (1 s), 66.2% (1.5 s), and 68.9% (2 s), which are higher than those obtained with commercial gel. It is worth noting that not all participants had prior experience with SSVEP-based tasks, which may introduce inter-subject variability but

reflects more realistic usage conditions. Furthermore, the EEG classification accuracy of one subject is tracked over time (Supplementary Fig. 34). Commercial gel exhibits increasing impedance during long-term wear, accompanied by a gradual decline in classification accuracy. In contrast, the ChCl-egel maintains low and stable impedance over 18 h, with consistently high accuracy, demonstrating its superior long-term signal fidelity and suitability for continuous EEG monitoring.

A comprehensive comparison of ChCl-egel, commercial gel and commercial 3 M with respect to some other preparation and performance parameters such as contact impedance, cost, stretchability, long-term and SNR is summarized in Fig. 5f. To provide a more objective interpretation of the electrode performance, a quantitative analysis is included in Supplementary Tab.1. The commercial gel is effective for only a few hours, while 3 M electrode exhibits high impedance and poor signal quality. The preparation of ChCl-egel involves a heating and mixing process with a production cost of ~\$ 0.18 per 10 grams. As shown in Supplementary Fig. 35, 3 M electrodes exhibit a maximum elongation of 50%, whereas the ChCl-egel reaches 200%, enabling better conformity to curved skin surfaces and thus improving contact stability during monitoring. Notably, ChCl-egel can achieve impedance levels similar to commercial gel and offer a comparable operational lifespan to 3 M electrodes. Compared with previously reported electrodes^{47–60}, the ChCl-egel simultaneously offers ultra-low impedance and long-term recording as shown in Fig. 5g. Furthermore, the impedance, SNR and temporal performance with various other type electrodes are summarized (Supplementary Tab. 2). Its conductivity is compared with that of other eutectic gels, confirming that ChCl-egel has higher conductivity (Supplementary Tab. 3). These results suggest that the ChCl-egel has significant performance advantages, making it ideal for maintaining high-quality dynamic physiological signal acquisition in daily life.

Discussion

In summary, we design a ChCl-egel electrode for long-term and high-quality monitoring of physiological electrical signal. Incorporating a deep eutectic solvent into the gelatin network enhances its mechanical properties through hydrogen bonding, ensuring a comfortable fit across all body regions. The rapid ion mobility within the ChCl-egel facilitates the formation of ionic crosslinked, thereby achieving skin impedance of 4.7 k Ω and scalp impedance of 5.3 k Ω at 10 Hz. Additionally, the inclusion of glycerol improves the moisturizing capability, retaining 93% of weight over 5 days, which guarantees long-term recording in physiological electrical signal record. For ECG monitoring, the ChCl-egel can realize the high-quality recording for over 48 consecutive hours, with an 8 dB higher SNR compared to commercial 3 M electrode. It also supports dynamic monitoring of heart rate under high-intensity exercises. In EEG applications, the acquisition quality of the ChCl-egel is comparable to that of commercial gel, and the recording time is significantly extended. Moreover, the ChCl-egel electrode is environmentally friendly with ultra-low toxicity, causing no skin irritation or sensitization. This makes it a high-quality physiological electrical acquisition platform for daily monitoring and medical diagnostic applications.

Methods

Materials

Gelatin (~100 g Bloom), choline chloride (AR, 98%), and glycerol (AR, 99%) were purchased from Aladdin Tech. Co., Ltd. Commercial ECG electrodes were purchased from 3 M Co., Ltd. (2223CN, American). Commercial EEG electrodes were purchased from Greentek Tech. Co., Ltd. (GT5, China). Phosphate-buffered saline (1 $\times$ PBS, pH 7.0~7.2) was purchased from Biosharp (BL302A, China). All chemicals were used without further purification. Deionized water was used for all experiments.

Preparation of ChCl-egel

Firstly, Choline chloride and glycerol (molar ratio 1:0.8) were heated at 80 °C for 3 h, stirring until uniform deep eutectic solvents (DESSs) were

formed. Then, 2.2 g gelatin powder was added to 4 mL deionized water and heated at 70 °C for 3 h, stirring until fully dissolved. Finally, 7 mL of DESSs was added to the gelatin solution and heated at 70 °C for 4 h, with stirring until the mixture was uniform and bubble-free.

General characterizations

The microscope image of ChCl-egel attached to a pig skin substrate was recorded by an industrial microscope (AW31, All Ways, China). SEM images of the freeze-dried ChCl-egel's cross-sectional microstructure were obtained using a LEO-1530 field emission scanning electron microscope (FE-SEM, Zeiss, Germany) and a LabRAM Odyssey Raman spectrometer (Horiba, Japan). The dehydration test was conducted in a temperature- and humidity-controlled chamber (DHTHM-27-20-P-ES, DHT, China) at 25 °C and 55% RH.

Rheological experiments

Rheological properties were characterized by a rheometer (Kinexus Pro +, NETZSCH, Germany). The dynamic temperature sweep mode ranged from 10 °C to 70 °C and then from 70 °C to 10 °C at a rate of 2 °C min⁻¹, while the oscillation frequency was 2 π rad s⁻¹. The dynamic frequency sweep mode was conducted from 0.1 Hz–150 Hz at a constant temperature of 25 °C. The self-healing behavior was investigated by alternating strain (5% or 250%) at a frequency of 0.5 Hz.

Electrical experiments

Electrochemical impedance spectroscopy (EIS) was measured using a computer-controlled electrochemical workstation (PGSTAT101, Autolab, Switzerland). The skin impedance was measured by placing two ChCl-egel electrodes (2 cm $\times$ 4 cm $\times$ 2 mm) on the forearm with a separation of 5 cm. The electrical impedance was measured by sandwiching a ~2 mm thick piece of ChCl-egel. All applied voltages were set to 50 mV, and the frequency of the device was varied from 0.1 Hz–100 kHz.

Mechanical experiments

Compression tests were conducted using a dynamic thermomechanical analyzer (qqDMA850, TI Instruments, USA) at a stretching speed of 10% mm min⁻¹. Tensile and adhesion tests were performed using a universal tensile tester (5942, INSTRON, USA). The adhesive strength of the ChCl-egel to the substrate was evaluated through a lap shear test. All mechanical experiments on the ChCl-egel were conducted at room temperature.

Biocompatibility tests

Cytotoxicity Detection: The cytotoxicity of ChCl-egel was evaluated using L929 fibroblasts treated with extraction solution (10 mg/mL in MEM medium). Three groups were tested: ChCl-egel, a positive control (10% DMSO), and a negative control (blank). Cells were incubated for 24 h, 48 h, and 72 h, and viability was quantified using the CCK-8 assay after 2 h of reagent incubation, followed by absorbance measurement at 450 nm. Calcein-AM/PI staining combined with confocal microscopy further confirmed the absence of significant cytotoxic effects. Data are expressed as mean $\pm$ standard deviation ($n = 3$).

Skin Irritation Test: Six rabbits were exposed to ChCl-egel, saline (negative control), and 10% SDS (positive control) on shaved dorsal skin for 72 h. The back of each rabbit was divided into four regions: two regions received ChCl-egel (parallel test sites), one region served as the negative control (saline), and one as the positive control (10% SDS). Skin reactions were evaluated at 24 h, 48 h, and 72 h post-application for signs of erythema and edema. Scoring was performed using a five-point scale (0–4) based on symptom severity. Cumulative irritation scores were classified as follows: no irritation (0.00–0.49), mild irritation (0.50–2.99), moderate irritation (3.00–5.99), and severe irritation (6.00–8.00). The negative control group was scored 0, while the positive control group showed erythema and edema with scores ≥ 1 . ChCl-egel was consistently scored below 0.33, indicating no skin irritation.

Skin Allergy Test: Six guinea pigs were treated with ChCl-egel, saline, or 10% SDS on shaved skin on days 1, 7, and 14. Each treatment was applied to a 2 cm × 2 cm area. Skin reactions, including erythema and edema, were photographed and evaluated at 24 h and 48 h post-application using the Magnusson and Kligman grading scale: Grade 0 (no visible reaction), Grade 1 (scattered or patchy erythema), Grade 2 (moderate confluent erythema), and Grade 3 (severe erythema or edema). At both 24 and 48 h following the final application on day 14, the test sample and negative control groups showed no visible skin reactions (Grade 0), while the positive control group exhibited Grade ≥1 responses, confirming the occurrence of skin sensitization.

Euthanasia Procedure: Anesthesia was not involved in the irritation and allergy tests. Euthanasia was performed using carbon dioxide (CO₂) inhalation. A dedicated euthanasia chamber with a transparent observation window (volume ≥10 L) was used, connected to a CO₂ source with ≥99.9% purity. A pressure-reducing valve and a precision flowmeter were used to control the gas flow rate. Prior to CO₂ exposure, animals underwent humane pre-sedation by being placed in a transfer cage within a quiet environment for 10 min. A cotton ball lightly soaked with <1% isoflurane vapor was placed in the cage without direct contact with the animal. The transfer cage was then placed into the euthanasia chamber, and CO₂ was introduced at a rate of 20% of the chamber volume per minute (e.g., 2 L/min for a 10 L chamber), maintaining a CO₂ concentration of ≥70% for 5 min. Death was confirmed using three criteria: cessation of respiration (observed for at least 60 seconds), absence of heartbeat (verified via stethoscope), and loss of corneal reflex (tested with a sterile cotton swab).

ECG and EEG signal acquisitions

ECG signals at rest were recorded by a wired system consisting of an AD8232 sensor and an Arduino-UNO R3, the sampling rate is 9600 Hz. One electrode was used for acquisition and the other two electrodes served as reference and ground. During exercise, ECG signals were recorded using a wireless system consisting of a BMD101 sensor and JDY-18 Bluetooth module, the sampling rate is 512 Hz. One electrode was used for acquisition and the other electrode served as reference. The ECG data is processed using wavelet transform. First, the raw ECG signal is imported and subjected to multi-level wavelet decomposition using the wavelet basis. A thresholding algorithm is then applied to the wavelet coefficients to reconstruct a denoised signal. Subsequently, the MSE and SNR are calculated based on Eqs. (1) and (2). These quantitative indices are used to evaluate signal fidelity of the denoising process.

The filtered signal was obtained through wavelet decomposition and compared with the original

$$MSE = \frac{1}{N} \sum_{n=1}^N [y(n) - x(n)]^2 \quad (1)$$

where $y(n)$ is filtered signal, $x(n)$ is original signal, N is sampling number.

$$SNR = 10 \log_{10} \frac{P_{\text{signal}}}{P_{\text{noise}}} = 10 \log_{10} \left[\frac{\sum_{n=1}^N y^2(n)}{\sum_{n=1}^N [y(n) - x(n)]^2} \right] \quad (2)$$

where P_{signal} is the power of filtered signal, and P_{noise} is the power of the original signal minus filtered signal.

EEG signals were recorded by 8-lead EEG acquisition system (NeuSen W, Neuracle, China) and the sampling rate is 1000 Hz. The EEG signals processing was performed on the collected data using MATLAB for fundamental signal analysis. To eliminate power-line interference at 50 Hz and suppress both high-frequency and low-frequency noise components, a 5–45 Hz bandpass Butterworth filter was applied during preprocessing. This filtering step effectively preserved the EEG frequency bands of SSVEP stimulation frequency, while enhancing signal quality for subsequent analysis. TRCA is a spatial filtering technique designed to enhance task-relevant EEG

components, especially in SSVEP. It works by identifying a spatial filter that maximizes the inter-trial reproducibility of EEG signals corresponding to the same task. This approach improves the signal-to-noise ratio by reinforcing consistent neural responses and attenuating unrelated noise.

Ethics declarations

Animal-related experiments were performed at The Animal Experimental Center of Zhengzhou University, with approval from the Ethics Committee (Approval No. ZZU-LAC20241014[01]). The human study participant experiments were approved by the ethical committee of Beijing University of Posts and Telecommunications (Approval No. BUPT-P-2025001). All subjects had normal hair volume, no history of hair loss, and provided written informed consent.

Statistical analysis

Matlab software was used for statistical analysis and the data were represented as the mean ± standard deviation. Each data was come from at least three independent experiments.

Data availability

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

Code availability

The code used in this study is available from the corresponding author upon reasonable request.

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Author contributions

G.X., K.H., and H.W. designed and supervised the research. W.W. carried out material synthesis and characterizations. C.R., and Y.W. performed the experiments. Z.C. and Y.Z. helped in data processing. W.W. and C.R. designed ECG and EEG experiments. S.Z., C.Y., and H.L. designed Matlab code to analyze performance in EEG. W.W. and G.X. co-wrote the manuscript. S.X., Y.L., and M.L. revised the manuscript. All authors analyzed the experimental data and discussed the results.

Competing interests

The authors declare no competing interests.

Additional information

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