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Adhesive Bio-composite Electrodes on Sweaty Skin for Long-term Continuous Electrophysiological Monitoring

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ABSTRACT: *Non-invasive on-skin electrodes record the electrical potential changes from human skin, which reflect body condition and are applied for healthcare, sports management, and modern lifestyle. However, current on-skin electrodes have poor conformal properties under sweaty condition in real-life due to decreased electrode-skin adhesion with sweat film at the interface. Here, we fabricated bio-composite electrodes based on silk fibroin (SF) through interfacial polymerization which is applicable on sweaty skin. Interfacial polymerized conductive polypyrrole (PPy) and SF are structurally interlocked and endow the whole electrode with uniform stretchability. Existence of water results in similar Young's modulus of SF to the skin and enhanced interfacial adhesion. It keeps the electrodes conformal to skin under sweaty condition and allows reliable collection of ambulatory electrophysiological signals during sports and sweating. Wearable devices with these electrodes were used to acquire continuous and stable real-time electrocardiography (ECG) signals during running for 2 hours. The collected signals can provide information for sports management and are also analyzed by artificial intelligence to show their potential for intelligent human emotion monitoring. Our strategy provides opportunities to record long-term continuous electrophysiological signals in real-life conditions for various smart monitoring systems.*

On-skin electrodes, which are non-invasive and easy to apply, have been widely used for acquiring electrophysiological signals to reflect body conditions and intended movements¹⁻³. With diverse information collected, it is widely applied for healthcare (e.g. self-diagnostics of heart diseases, self-regulation of stress level), sports management, modern lifestyle (e.g. intelligent household), and human-machine interfaces⁴⁻⁵. In recent years, conformal electrodes based on gels or polymer thin films and related devices have been developed to address bio-device interfacing issues, which are crucial for monitoring continuous electrophysiological signals from the skin⁶⁻¹⁵. Their conformal contacts with deformed skin can maintain consistent contact areas between electrode and skin upon deformation and eliminate motion artifacts, according to a developed theoretical model of skin potential (Figure 1, details in Supporting Information)¹⁶. A less considered point is that sweating is inevitable in real life especially during long-term monitoring and sports. This requires the electrodes to keep conformal and adhesive in the presence of sweat. However, current non-invasive electrodes have poor conformal properties and decreased electrode-skin adhesion under sweaty condition due to the formation of sweat film at the interface (Figure 1e), which has low shear viscosity and reduces molecular interactions between skin and adhesives. It will cause detachment of the electrodes from skin, for example, com-

mercial gel electrode easily slide on sweated skin (Movie S1), leading to fake signals and even signal lost. To address this problem, the electrodes must hold conformal properties under sweaty conditions to realize stable long-term ambulatory electrophysiological monitoring.

Achieving conformal properties of electrodes under sweaty condition requires the synergy of two effects. First, the mechanical property match between the conductive layer and the adhesive layer, which ensures their uniform stretchability for conformal attachment to skin¹⁷⁻¹⁹. Compared to metals such as Au and Ag, conducting polymers such as polypyrrole (PPy) have a low impedance, excellent biocompatibility, and much lower cost, but their unstretchable property limits their applications in conformal electrodes. Through the interlocking structure between PPy and the adhesive layer formed by interfacial polymerization, the stretchability of PPy film can match with the adhesive layer to realize uniform stretchability of whole electrodes. Second, strong adhesion on wet surface of the adhesive layer that holds their conformal property under sweaty condition. The Young's modulus of silk fibroin (SF) decreases and gets similar to skin in the presence of water, achieving increased SF-skin adhesion²⁰⁻²⁵. The water responsive characteristic makes SF one of the best choices for keeping high adhesion and conformal properties on sweaty skin.

Figure 1. Conformal property of on-skin electrode enhances the stability of electrophysiological signal detection. a. The skin under the test electrode was considered as n parallel power sources as shown in the top circuit diagram. E originates from the ‘injury current’, R_c and R_t correspond to the impedances of corneum and transitional cells, respectively. b. While stretching skin, conventional electrode keeps the same area as the original state, leading to fewer power sources contacting the electrode. c. Conformal electrode deforms with skin, and E_{se} keeps the same as the original state. d. The change of ΔV obtained by conventional and conformal interface upon stretching skin based on the theory of Talhouet and Webster (See Supporting Information for detail). e. The decrease of adhesion strength with increasing humidity, which resembles the accumulation of sweat on skin. The curve is based on experimental results.

Herein, we designed a bio-composite conformal and adhesive polymer electrode (CAPE) based on SF through the interfacial polymerization with pyrrole and realized long-term monitoring of ambulatory electrophysiological signals. The interfacial polymerization produced interlocking structures between PPy and SF gel, which endowed PPy film with stretchability by well transmitting its strain to SF layer. The mechanical responses of SF to water provided an enhanced interfacial adhesion on skin with sweat and kept the conformal and adhesive properties of CAPE under sweaty condition. They could accommodate large deformation of wet skin and were capable of ambulatory electrophysiological monitoring. CAPE was employed to monitor continuous long-term ambulatory electrocardiogram (ECG) during running for sports management. The collected data was also used to train an AI to distinguish emotional status, showing its potential in intelligent health monitoring.

CAPE was fabricated by interfacial polymerization of pyrrole on the surface of an ionically crosslinked SF. In a typical experiment, a formic acid solution of SF, FeCl_3 , CaCl_2 was employed to fabricate SF gels by solvent evaporation at room temperature. CaCl_2 was used to adjust the mechanical properties of SF gels (Figure S6 and S7); FeCl_3 was the initiator for interfacial polymerization of pyrrole. A chloroform solution of pyrrole monomer and phase transfer agent tween 80 was added onto the surface of SF gel. When the chloroform solution was in contact with SF gel, ferric ions in SF gel induced the polymerization of pyrroles to form conductive PPy film. The phase transfer agent was used to promote the interfacial polymerization. Besides, the interfacial polymerization made PPy film and SF gel to form an interlocking structure, leading to uniform stretchability of CAPE. Obvious change in appearance of the film indicated successful polymerization (Figure 2a). Scanning electron microscope (SEM) images before and after interfacial polymerization also showed the

smooth SF surface turned to rough PPy surface (Figure S8a and b). ATR-IR spectra confirmed the composition of CAPE (Figure S8c).

To demonstrate the interlocking structure between PPy and SF gel, a microscopic image of the side view of CAPE showed the formation of a wavy structure at the interface between PPy and SF gel (Figure 2b), which increased the contact area between PPy and SF gel at macro level. As shown in Raman mapping at 1283 cm^{-1} (characteristic peak of PPy) for the section of CAPE (Figure 2c), the interfacial polymerization not only endowed PPy with a three-dimensional network structure but also formed an interlocking structure between PPy and SF gel^{26,27}. Besides, the surface of PPy contacting SF gel was rough (Figure 2d), which increased the frictional force between PPy and SF gel at micro level. The increase of contact area and the frictional force enhanced the interfacial adhesion between PPy and SF gel, which can efficiently transfer the strain from SF gel to PPy without delamination. The adhesive strength of 1.91 MPa between PPy and SF gel (Figure 2e) is 27 times higher than 0.07 MPa of gel and Ag/AgCl in commercial gel electrodes (product of Heal Force Biomeditech Holdings Limited). It is also higher than 0.71 MPa by directly pasting electrochemically polymerized PPy onto the SF gel without the interlocking structure. It was difficult for PPy to be removed from SF gel even when peeled off by a tape (Movie S2). Contrastingly, it was easy to peel off the PPy from SF gel for the film fabricated by pasting PPy onto SF gel (Movie S3). When transferring the strain from SF gel to PPy, the three-dimensional network structure of PPy can preferably regulate strain distribution in PPy to achieve a strain of 300 % (Figure 2f), which is much higher than 1 % of that fabricated by pasting. Additionally, SF gel provided 17 J/m² of adhesion energy between CAPE and glass substrate (52.8 % RH), which is similar to 27 J/m² of the commercial gel electrodes. The matched mechanical properties to skin and

Figure 2. Bio-composite conformal electrodes based on SF and PPy. a, Photo of SF gel and PPy/SF film before and after interfacial polymerization. b, Microscope image of CAPE displayed the wave structure between PPy and SF gel. c, Raman mapping at 1283 cm^{-1} for the section of CAPE showed the three-dimensional network structure of PPy and the interlocking structure between PPy and SF gel layer. d, SEM image showed the rough surface of PPy in contact with SF gel. e, The adhesive strength between the conductive and adhesive layer of CAPE was higher than commercial gel electrodes and PPy/SF film fabricated by pasting. f, Maximum strain of CAPE was 300%, which was much higher than 1% of PPy/SF film fabricated by pasting. g, The highly conformal property of CAPE on the human elbow. h, CAPE had a lower electrical impedance in the frequency range of 3 to 100 Hz, compared with commercial gel electrode and Au/SF electrode.

high interfacial adhesion energy resulted in a perfect fit of CAPE on the elbow (Figure 2g). The enlarged surface image of CAPE on the elbow displayed the pattern of human skin, indicating the highly conformal property.

Another key point for stable ambulatory electrophysiological monitoring is electrical property. The skin-interfacial impedance of CAPE decreased from 4.8×10^6 to $2.8 \times 10^3\ \Omega\ \text{cm}^2$ in the frequency range of 0.1 to 1000 Hz (Figure 2h). Comparing with the commercial gel electrodes and the thermal evaporated Au on SF electrode, CAPE had the lowest impedance in the range of 3 to 40 Hz, which is more propitious for the collection of ECG signals²⁸. Due to the interlocking structure, CAPE (with 20 wt% initiator during preparation) kept electrical stability up to 30% strain, which is the typically encountered deformation of skin (Figure 3a). As a demonstration, LED light using CAPE as a stretchable conductor retained its luminescence when being stretched to 30% strain and relaxed (Figure 3b and Movie S4). The resistance of CAPE remained the same value even after being subjected to a hundred stretching/relaxing cycles (Figure 3b), indicating the stable electrical properties.

Increasing relative humidity (RH) was used to simulate the moisturization of skin during sweating. To show the enhanced conformal property of CAPE upon sweating, the interfacial adhesion energy of CAPE on skin mimetic PDMS substrate with increasing RH was studied in detail by mechanical tests. When the RH is below 31%, the adhesion energy of CAPE is smaller than 0.9 J/m^2 and could not adhere to skin. However, its adhesion energy increased to 40 J/m^2 upon increasing RH from 31% to 94.6% (Figure 3c), while the adhesion energy of commercial gel electrodes decreased to 2.6 J/m^2 . The enhanced adhesion

of CAPE was due to its increased contact area with substrate through decreasing Young's modulus from 236 to 2.1 MPa (Figure 3d), and led to easy attachment of CAPE on skin. Upon increasing RH to 100%, the water content of CAPE increased to 44% (Figure S9), which was the reason for the decreased Young's modulus. To further demonstrate the reason for the adhesion enhancement, the side view images of CAPE pasted on skin mimetic PDMS substrate under 52.8% and 94.6% RH were obtained by an optical microscope. Compared with plenty of interspace between electrode and substrate at 52.8% RH (the area between the white lines in the top image of Figure 3e), the contact area under 94.6% RH increased (the white line in the down image of Figure 3e) due to the decrease of Young's modulus with increasing RH. Compared with the detachment of commercial electrodes from wet skin, CAPE pasted on the wet surface of a pig heart could sustain 100 g of weight (Figure 3f). It was difficult to fall off during the simulated beating of the pig heart (Movie S5). These implied the high interfacial adhesion of CAPE on wet surfaces. Moreover, the adhesive layer of CAPE was dissolved when being immersed in water for 30 mins, implying that the highly adhesive CAPE was degradable and could be removed from skin if desired.

Dynamic modulus provides excellent insight into the dynamic mechanical properties of CAPE, which could help to understand the mechanism behind water enabled conformal property. By increasing RH from 52.8% to 94.6%, both the elastic modulus G' and the viscous modulus G'' decreased (Figure 3g), indicating the SF layer became softer and could have increased contact area with substrate. Under 52.8% RH, the phase transition point of CAPE is at ω of 32.4 s^{-1} (Figure 3g), meaning that the

Figure 3. High adhesion on wet surfaces of stretchable CAPE. a, The resistance change of different CAPE upon stretching. b, The stable conductivity of CAPE with 20 wt% of FeCl₃ upon being stretched to 30% and cycle stretched. LED light using CAPE as a stretchable conductor retained its luminescence when stretching and relaxing (also see movie S4). c, Interfacial adhesion energy of CAPE increased with increasing RH from 11% to 94.6%, compared with commercial gel electrodes. d, Young's modulus of CAPE decreased with increasing RH. e, The increase of contact area between CAPE and PDMS like skin with increasing RH. The shade indicates void space. f, The strong interfacial adhesion of CAPE on the wet surface of a pig heart. g, Dynamic modulus of CAPE with increasing RH. h, The angular frequency at the intersection point of G' and G'' increased with increasing RH.

structure of SF layer can be easily destroyed by external stress at high shearing rate. With increasing RH to 84.3%, ω at the phase transition point increased from 32.4 to 86.2 s⁻¹ (Figure 3h). It implied that the SF layer is difficult to be destroyed at high exercising frequency (Movie S6), showing good stability of dynamic mechanical property on wet surface. Thus, the enhanced conformal property of CAPE was achieved through varying their dynamic mechanical properties under sweaty condition.

ECG records the tiny electric potential changes on the skin that arise from the heart muscle's electrophysiological pattern of depolarization and repolarization during each heartbeat and is widely used for diagnosis of heart disease in clinic²⁹. CAPE was employed to obtain ECG of a male (32 years old) with 0.3 mV of relative voltage in T wave (Insert image of Figure 4a) for two hours under 50% RH. The sensitivity of ECG electrodes is defined as the relative voltage ratio of T wave to R wave measured. The sensitivity of CAPE is 0.50 due to the conformal electrodes, compared to 0.31 achieved by commercial gel electrodes (Figure 4a). Specifically, ECG obtained by CAPE was highly stable upon deformation of the skin, including being squeezed, pressed or stretched (Figure 4b and Movie S7). The sensitivity of CAPE was always higher than that of commercial gel electrodes, regardless of various skin deformation (Figure 4c). Under sweaty condition, ambulatory ECG achieved by commercial gel electrodes became inaccurate and unstable by extending the time to 60 minutes (Upper image in Figure 4d). This is a result of sweat production which in turn decreased the adhesive

energy between electrodes and skin (Figure 4f). Conversely, ambulatory ECG is highly stable under the sweaty condition when being recorded by CAPE (lower image in Figure 4d), resulted from the highly conformal property on wet surface. CAPE kept similar sensitivity after different sport times (Figure 4e), showing accurate ECG under sweaty condition.

Ambulatory ECG aid in early diagnosis of potential coronary heart disease through analyzing the ST segments and T waves³⁰. For example, the inversion of T wave in ambulatory ECG shows a negative value of the intensity ratio of T to R, implying myocardial dyskinesia and an unhealthy state of the heart muscle³¹. We recorded ambulatory ECG signals of the same young man in different running speeds by CAPE (Figure 4g and Movie S8). Upon increasing speed to 8 km/h, the intensity ratios of T to R were all positive (Figure 4h), indicating the subject has a healthy heart at the present stage. Besides, exercise heart rate (EHT) provides accurate information on exercise intensity referred to as the energy expended by various sport^{32,33}. It can be a reference for the scientific arrangement of exercise for a healthy lifestyle, such as the energy consumed by the body and the adaptations made by the body after various exercises³⁴. If the subject wants to lose weight through burning the fat, the heart rate of running should be kept in the range of 140 to 160 bpm, matching with 60 to 70% of maximal heart rate^{35,36}. According to the relationship between the heart rate and running speed (Figure 4i), we could know that 6.5 to 8.0 km/h of running speed at this moment should be kept for burning

Figure 4. Ambulatory ECG monitoring by CAPE under sweaty condition. a, ECG obtained by CAPE and commercial gel electrodes. Insert image showed one heartbeat. b, ECG upon different skin deformation (squeezed, pressed, and stretched). c, The sensitivities of CAPE and commercial gel electrodes under different skin deformation obtained by the intensity ratio of T to R. d, ECG after different sport time obtained by CAPE and commercial gel electrodes under sweaty condition. e, The sensitivities of CAPE and commercial gel electrodes under sweaty condition. f, Photos of sweat on the chest acquired after running for different times. g, ECG at different running speeds obtained by CAPE under sweaty condition. h, The sensitivities of CAPE at different running speeds. i, The increase of heart rate with increasing running speed obtained through ECG.

fat, confirming the application of CAPE in sports management.

Besides, CAPE was applied to collect other ambulatory electrophysiological signals, such as electroencephalography (EEG), electromyography (EMG) and electrooculography (EOG). For example, during the act of making a fist, the intensity of EMG increased and kept the same intensity with prolonged clenching time (Figure S10a). After relaxing the hand, the intensity of the signal recovered to the original state. While we did nine times act of making a fist in 12 seconds, their EMG was recorded by CAPE. Besides, upon the subject blinking eyes, a sharp peak of the EOG was easily observed (Figure S10b). Our electrodes accurately recorded seven sharp peaks in EOG signals, corresponding to seven times act of blinking. Moreover, CAPE was pasted on the forehead to collect various EEG such as alpha, beta, theta, and delta waves when the subject was taking a nap (Figure S10c). The EEG showed 8% of wake time, 18% of sleep time, and 72% of motion time in the whole sleep process of the subject, which can be used to evaluate sleep quality of the subject.

Recurrent neural networks (RNNs) was combined with continuous recordings of ECG to build an intelligent electrophysiological monitoring system for distinguishing the emotional status of the test subject (Figure 5a). The ECG signals obtained by CAPE were classified according to two emotional states: positive and negative feelings. First, we trained the model through manually annotated ECG signals from the DREAMER and Ascertain databases^{37,38}. Subjects were asked to watch a video clip for a few minutes, where a particular emotional state was induced and the corresponding ECG was recorded. For everyone, 60 seconds of ECG data accordingly classified as 'happy' (i.e. positive feelings) or 'sad' (i.e. negative feelings) was used. Then, we obtained 169 features from each sample, which were used as the input to our neural network³⁹. The model is robust to ECG problems such as flipped signal, varying heart rate transformations and translation of heartbeats. After the model has been trained, the intelligent electrophysiological monitoring system can be used to classify ECG signals from different subjects (Figure 5b). To validate such an approach, we obtained different ECG

Figure 5. Intelligent perception of emotions through perceiving and classifying ECG obtained by CAPE. **a**, The machine learning methodology for ECG. **b**, Perceiving and classifying emotions of a subject through real ECG collected by CAPE.

recordings by CAPE following the procedure described above and used such data to validate our approach as a testing set. In particular, we presented 5 individuals with 4 different videos and asked them to classify their feelings, following the procedures described in the literature. Competitive results ($f\text{-score} = 0.73$) were achieved on the unknown emotional data collected by CAPE from individuals watching video clips inducing happy or sad emotional states. This ECG-AI system could be helpful for emotion management to maintain good mental and physical conditions, showing their potentials in smart health monitoring and management fields.

Sweating is inevitable in daily life and will greatly decrease adhesion between electrodes and skin, leading to malfunction of healthcare devices. To overcome this problem, we took advantage of the water responsiveness of silk fibroin. With increasing humidity, Young's modulus of SF decreased and became similar to skin, leading to increased adhesion in the presence of sweat. Upon this water enabled adhesive layer, a conducting PPy layer was polymerized and the resulted interlocking structure, endowing the whole film with good stretchability. The water enabled conformal properties resulted in increased skin-electrode adhesion energy at high RH. With these electrodes, stable and reliable ECG signals were collected during sports with sweat and long-term monitoring. The collected signals could provide useful information for sports management and healthcare. The long-term and large numbers of signals also provide the opportunity for AI analysis of the data for a smarter lifestyle such as emotion perception and management.

Our strategy for on-skin electrodes fitted the real-life scenario where sweat is generated on the skin. The stable adhesion and conformal properties under sweaty condition ensured reliable long-term monitoring of ambulatory electrophysiological signals during routine activities, which is crucial for continuous health-monitoring, intelligent self-diagnostics systems of disease, and smart human-machine interfaces.

ASSOCIATED CONTENT

Supporting Information

The experimental method, theoretical assumption for the effect of stretchability to on-skin signal collection, and additional characterization data. (PDF)

Movie S1: commercial gel electrode on sweated skin. (MP4)

Movie S2-S3: interfacial adhesion between PPy and SF gel. (MP4)

Movie S4: conductivity of CAPE during the stretch. (MP4)

Movie S5-S6: conformal adhesion of CAPE on pig heart and skin. (MP4)

Movie S7-S8: ECG recoding with CAPE under skin deformation or during running. (MP4)

This material is available free of charge via the Internet at [HTTP://pubs.acs.org](http://pubs.acs.org).

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Author Contributions

X.C. and Y.S.O. conceptualized the project. X.C. and H.Y. designed the experiments. H.Y. and S.J. performed experiments and optimized the method. H.X., L.P. and D.Q. participated characterizations. T.W. and C.W. performed some of the ECG tested and data extraction. G.C. performed some fabrication experiments. I.C. performed RNNs analysis. X.C., H.Y., Y.S.O., I.C. and S.J. wrote the paper. All authors have approved the final version of the manuscript. ‡These authors contributed equally.

Notes

The authors declare no competing financial interest.

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