Electrodes for Stable Electrophysiological Signal Monitoring

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Abstract. Epidermal electronic components have received much attention for their potential in personal healthcare and human-machine interaction (HMI). However, their low conductivity hinders their practical applications. Here, we developed a multifunctional flexible dry epidermal electrode for biopotential recording using modified poly(3,4-ethylenedioxythiophene) (PEDOT:PSS) doped with Polyethylene glycol(PEG). PEG was introduced into PEDOT:PSS to enhance its conductivity. We prepared electrodes with a high conductivity of 130 S/cm and 23% tensile strength. The electrodes conform well to the morphology of human skin and therefore form a stable and intimate interface with the skin, resulting in low contact impedance between the electrode and the skin and high-quality recording of biopotentials. It is capable of accurately recording electrocardiogram (ECG), electrooculogram (EOG), and electromyogram (EMG) signals.

1. Introduction

The skin is the largest organ of the human body. By placing sensors on its surface, various types of physiological information can be obtained, making it invaluable in medical diagnostics, health monitoring, and human–machine interfaces (HMIs) [1-2]. For instance, biopotentials such as electrocardiograms (ECGs), electrooculograms (EOGs), and electromyograms (EMGs) can be recorded through electrodes placed directly on the skin. ECGs are measurements of crucial vital signs, often utilized in diagnosing cardiovascular diseases [3]. EOGs are not only valuable in diagnosing retinal conditions but can also serve as control signals in HMIs [4]. Furthermore, EMGs are used to detect muscle activity, making them essential for health monitoring and HMIs [5].

Given the diverse applications of skin-based sensing, the development of multifunctional wearable devices has become a focal point for researchers in personal healthcare. Ag/AgCl electrodes do not meet the need for long-term monitoring of bioelectrical signals. Therefore, we investigated thin-film electrodes that can be used on the skin for a long period. Li et al. [6] observed that the enhancement of the conductivity of PEDOT:PSS films was primarily attributed to the addition of PEG as a solvent, which facilitated the transition of PEDOT molecules towards a highly ordered, layered arrangement. In this study, electrodes were fabricated by incorporating PEG to enhance conductivity and PVA to improve the tensile properties of the film electrodes. In the field of personal healthcare, electrodes are required to transmit bioelectrical signals accurately and rapidly, enabling precise diagnosis and treatment by doctors or medical devices. Electrodes with high conductivity can reduce resistance during signal transmission, minimizing signal attenuation and distortion, thereby improving the quality of bioelectrical signal recording. Furthermore, electrodes with high conductivity can quickly respond to bodily movements or physiological changes, converting them into machine-recognizable signals. This enhances the accuracy and response speed of human-machine interaction. Tensile strength is a crucial factor to consider for electrodes in practical applications. In healthcare and human-machine interaction scenarios, electrodes often need to maintain prolonged contact with the patient's skin, especially during movement or activity. Electrodes with high tensile strength can better resist external stress and friction, reducing the risk of electrode detachment or damage, thus ensuring stable transmission of bioelectrical signals.

2. Preparation and characterization

2.1. Preparation of test sample

An aqueous suspension of PEDOT:PSS (Clevios PH1000) was purchased from Heraeus, Ltd. (Leverkusen, Germany). Polyvinyl alcohol (PVA) (MW, ~78,000)) and PEG were purchased from Aladdin (Shanghai, China). Deionized (DI) water with a resistance of 18.2 MΩ cm^{-1} was used.

When developing wearable devices, biocompatibility is of utmost importance to guarantee safe and effective use on the skin. To this end, we employed non-toxic PEG to modify PEDOT:PSS. Here's a detailed outline of the modification process: PEG, at a concentration of 4% (volume to volume), was introduced into the PEDOT:PSS solution and vigorously stirred at 30°C for 12 hours to ensure thorough integration.

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Subsequently, an appropriate quantity of PVA was added to the PEG-modified PEDOT:PSS mixture and stirred further at 70°C for four hours, yielding a stable modified PEDOT:PSS solution. This step was crucial in enhancing the solution's homogeneity. To further refine the material, the mixture was degassed in a vacuum environment to eliminate any residual air bubbles. The degassed solution was then poured into silicone molds and allowed to solidify naturally. Once dried at room temperature, the electrodes embedded within the modified PEDOT:PSS were carefully extracted from the molds. In the next article, we use MPE to refer to modified PEDOT:PSS electrodes.

2.2. Characterization

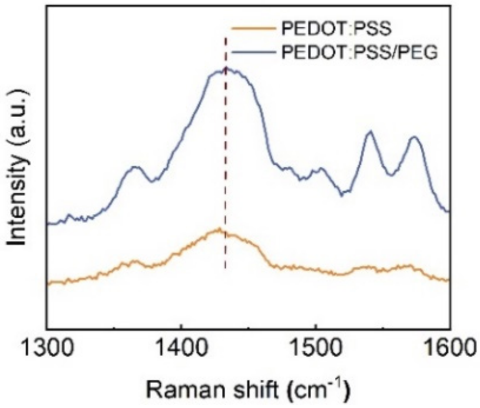

Fig. 1. Raman spectra of PEDOT:PSS and PEDOT:PSS/PEG.

In the Raman spectrum depicted in Figure 1, a notable shift in the $C_{\alpha}=C_{\beta}$ peak, representative of the thiophene structure within PEDOT:PSS, is observed. Specifically, this peak shifts from 1432 cm^{-1} to 1428 cm^{-1}. This shift suggests that the introduction of PEG into the PEDOT:PSS matrix results in a rearrangement of the PEDOT:PSS chains. This rearrangement is indicative of a transition from a more disordered state to a more ordered one, which is generally considered favorable for enhancing the material's properties.

The rearrangement of the PEDOT:PSS chains, facilitated by the introduction of PEG, is likely to have positive implications for the material's conductivity and mechanical properties. A more ordered structure can lead to improved charge transport within the material, resulting in higher conductivity.

To determine the optimum ratio of PEDOT:PSS and aqueous solution of PVA at 1% content, four modified PEDOT:PSS samples with different volume ratios (1:1, 7:1, 9:1, and 11:1) PEDOT:PSS/PEG to PVA were prepared for further characterization and testing.

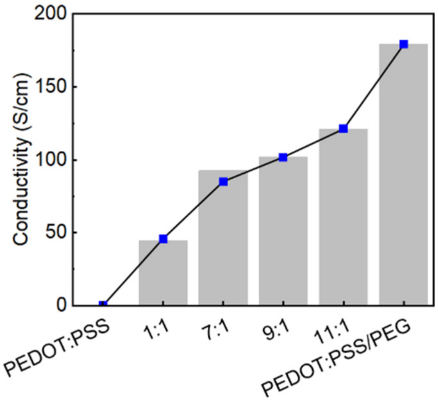

Fig.2. The conductivity of PEDOT:PSS, PEDOT:PSS/PEG, and different formulations of modified PEDOT:PSS.

Figure 2 shows the electrical conductivity of films with different contents of PEDOT:PSS. As the volume fraction of PEDOT:PSS increases, the conductivity of the films shows an increasing trend. This is due to the formation of more conductive paths as the amount of PEDOT:PSS increases. The conductivity of unadded PEDOT:PSS grows from 0.18 S/cm to 180 S/cm as compared to the PEG-added films. This is because the addition of PEG tends to linearize the PEDOT molecular arrangement, which facilitates carrier mobility. It also induces a conformational change of PEDOT molecules from benzene to quinone structure.

Subsequently, we compared the tensile strengths of electrodes with ratios of 9:1 and 11:1, as well as electrodes composed of PEDOT:PSS with and without the addition of PEG, as shown in Figure 3. Notably, the addition of PEG resulted in a significant decrease in Young's modulus of the electrodes, which aligns with the observation in previous literature that PEG incorporation renders the PEDOT:PSS film more flexible [6]. Furthermore, the incorporation of PVA further enhanced the stretchability of the electrodes. In pursuit of achieving a balance between electrode performance and tensile strength, we comprehensively selected a ratio of 9:1.

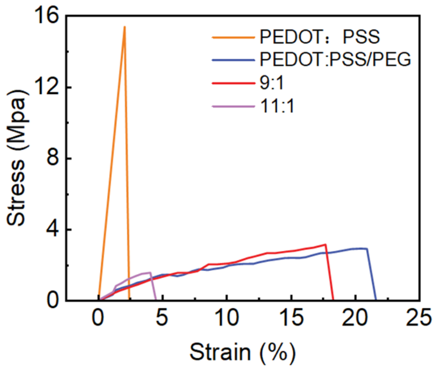

Fig. 3. tests the stress strain at different ratios.

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Figure 4 shows a cross-sectional SEM image of the MPE. Its thickness of approximately only 21 µm allowed the electrode to conform well to the morphology of the skin. Because the electrode is thin enough, it is able to adhere to the skin through van der Waals forces.

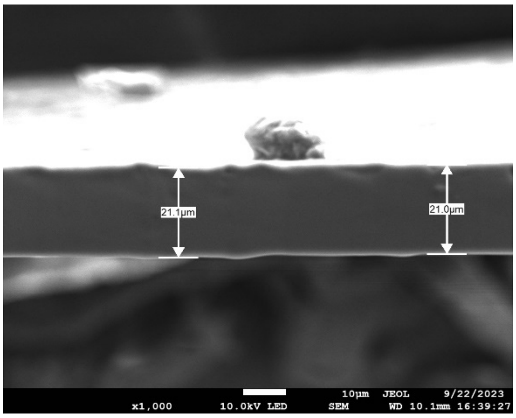

Fig. 4. Cross-sectional SEM image of modified PEDOT:PSS film.

Figure 5 shows the dimensions of the MPE fabricated according to our procedure compared to the AgCl electrode. From the comparison between MPE and Ag/AgCl electrodes, we can see that relative to our electrodes, the Ag/AgCl electrodes are larger in size but have similar results, which means that our electrodes have better results at a smaller size.

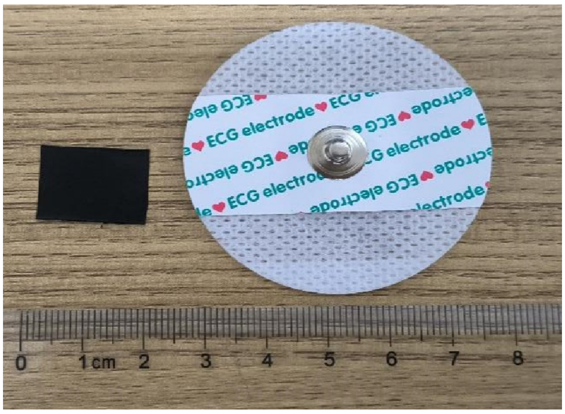

Fig. 5. Flexible dry and Ag/AgCl electrodes

3. Results and Discussion

3.1. ECG recording

For the ECG recording, measurements were obtained simultaneously from two ECG channels. The MPE (Micropatterned Electrode) and the Ag/AgCl electrode were positioned as closely as possible on the skin of the subject's chest. Additionally, two Ag/AgCl electrodes, designated as the reference electrode and the ground electrode, were placed on the left and right legs, respectively. This configuration allowed for a comprehensive and accurate recording of the electrical activity of the heart.

The ECG waveforms recorded by both the MPE and the Ag/AgCl electrode exhibited all the characteristic peaks, including the P wave, QRS complex, and T wave. These peaks represent distinct electrical events during a heartbeat and are crucial for diagnosing various cardiac conditions. The clarity and distinctness of these peaks in the recorded waveforms indicate the high quality of the ECG data obtained.

Furthermore, a correlation analysis was conducted to compare the signals recorded by the MPE and the Ag/AgCl electrode. The correlation between the two signals was calculated to be 97.2% in figure 6, which is a remarkably high value. This implies that the MPE is capable of recording ECG signals with almost the same level of accuracy and reliability as the conventional Ag/AgCl electrode.

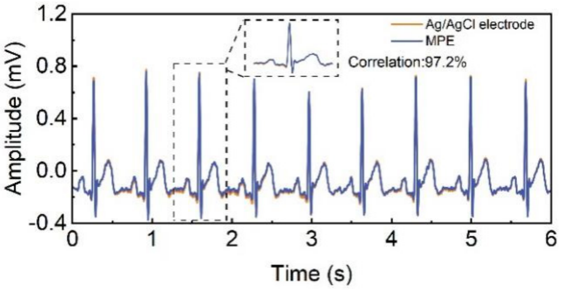

Fig.6. 6 s segments of ECG recording by Ag/AgCl electrode and MPE.

3.2. EOG recording

The performance of the MPE in recording EOGs was subsequently evaluated. In Figure 7, it is evident that the electrode was able to clearly distinguish and record different eye movements, including upward glances, downward glances, and blinking. The waveform patterns corresponding to these movements are distinct and recognizable, indicating the high sensitivity and accuracy of the MPE in detecting eye movements.

This evaluation demonstrates the utility and effectiveness of the proposed MPE in EOG recording. The ability to accurately capture and differentiate eye movements is crucial for various applications.

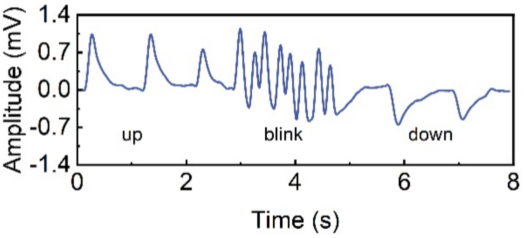

Fig. 7. Segments of EOG recording by MPE.

3.3. EMG Recording

Two MPEs were securely attached to the upper arm of a healthy adult to capture the electromyography (EMG) signals of the biceps muscle during activity. EMG is a technique that measures the electrical activity of muscles and can provide valuable insights into muscle function and performance. As the subject performed a cycling motion of lifting and lowering their arm, the EMG signals were continuously recorded. The waveforms generated by the EMG signals, as depicted in Figure 8, exhibit distinct patterns that vary with the lifting motion. These patterns reflect the activation and relaxation of the biceps muscle as it contracts and relaxes during the lifting and lowering movements. Upon careful observation, subtle amplitude changes can be discerned within the EMG waveforms. These amplitude variations are indicative of the intensity and force exerted by the muscle during different stages of the lifting cycle. For instance, the amplitude may increase as the muscle contracts more forcefully to lift the arm, and decrease as it relaxes during the lowering phase.

By analyzing this EMG waveform, we can gain insights into the muscle's behavior, such as its activation patterns, force generation capabilities, and any potential abnormalities or asymmetries that may indicate muscle dysfunction. Such information is crucial for understanding muscle performance in healthy individuals and for diagnosing and treating musculoskeletal disorders.

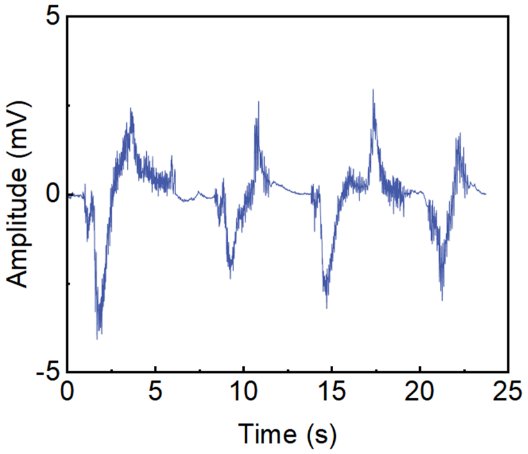

Fig. 8. Segments of EMG recording by MPE.

4. Conclusion

A flexible dry epidermal electrode, utilizing a modified version of the conductive polymer PEDOT:PSS, has been developed for biopotential recording. This innovation introduces PVA (polyvinyl alcohol) and PEG (polyethylene glycol) into the PEDOT:PSS matrix, significantly enhancing its stretchability and conductivity. The epidermal electrode, designed to conform intimately to the contours of human skin, forms a stable interface that minimizes electrode-skin impedance. This optimized contact ensures the accurate and reliable capture of biopotential signals, making it suitable for recording high-quality ECG, EOG, and EMG data. The electrode's flexibility and dry design offer significant advantages over traditional wet electrodes, enhancing wearer comfort and facilitating long-term monitoring. Its potential applications in personal healthcare are vast, enabling real-time physiological monitoring and analysis for individuals in various settings. Our developed PME is capable of monitoring biopotential signals, including electrocardiograms, electrooculograms, and electromyograms, and detecting subtle changes in electrical potentials. The potential impact of these electrodes on biopotential recording is immense. The use of dry electrodes offers new and powerful support for the development of healthcare monitoring and diagnostic technologies. Users can record their bioelectric signals over a long period, providing doctors with accurate information for assessing patients' health status.

Moreover, this approach may pave the way for the creation of more advanced and user-friendly wearable devices that seamlessly integrate into our daily lives, revolutionizing the way we monitor and manage our health.

References

1. X. Li, P. Zhu, S. Zhang, X. Wang, X. Luo, Z. Leng, H. Zhou, Z. Pan, Y. Mao. (2022) A Self-Supporting, Conductor-Exposing, Stretchable, Ultrathin, and Recyclable Kirigami-Structured Liquid Metal Paper for Multifunctional E-Skin, Acs Nano, 16(4) 5909-5919. https://doi.org/10.1021/acsnano.1c11096
2. J. Xu, X. Li, H. Chang, B. Zhao, X. Tan, Y. Yang, H. Tian, S. Zhang, T.-L. Ren. (2022) Electrooculography and Tactile Perception Collaborative Interface for 3D Human- Machine Interaction, Acs Nano, 16(4) 6687-6699. https://doi.org/10.1021/acsnano.2c01310
3. N. Verweij, J.-W. Benjamins, M.P. Morley, Y.J. van de Vegte, A. Teumer, T. Trenkwalder, W. Reinhard, T.P. Cappola, P. van der Harst. (2020) The Genetic Makeup of the Electrocardiogram. Cell Syst. 11(3) 229-238.e5. DOI: 10.1016/j.cels.2020.08.005
4. Y. Won, J.J. Lee, J. Shin, M. Lee, S. Kim, S. Gandla. (2021) Biocompatible, Transparent, and High-Areal-Coverage Kirigami PEDOT:PSS Electrodes for Electrooculography-Derived Human-Machine Interactions, ACS Sens. 6(3) 967-975. DOI: 10.1021/acssensors.0c02154
5. J.L. de Lacalle, M.L. Picchio, A. Dominguez-Alfaro, R.R.-M. Serrano, B. Marchiori, I. del Agua, N. Lopez-Larrea, M. Criado-Gonzalez, G.G. Malliaras, D. Mecerreyes. (2023) Hydrophobic Eutectogels as Electrodes for Underwater Electromyography Recording, ACS Mater. Lett. ,5(12) 3340-3346. DOI: 10.1021/acsmaterialslett.3c00938
6. Li, Y., Li, X., Zhang, S., Liu, L., Hamad, N., Bobbara, S.R., Pasini, D. and Cicoira, F., 2020. Autonomic self-healing of PEDOT: PSS achieved via polyethylene glycol addition. Advanced Functional Materials, 30(30), p.2002853. DOI: 10.1002/adfm.202002853