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Ultraflexible Vertical Corbino Organic Electrochemical Transistors for Epidermal Signal Monitoring

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Skin-conformal organic electrochemical transistors (OECTs) have attracted significant attention for real-time physiological signal monitoring and are vital for health diagnostics and treatments. However, mechanical harmonization amid the inherent dynamic nature of the skin surface and the acquisition of intrinsic physiological signals are significant challenges that hinder the integration of the ultimate skin interface. Thus, this study proposes a novel 4-terminal (4-T) vertical Corbino OECT, exhibiting high transconductance (>400 mS) and offering remarkable resilience and operational stability at an extremely low voltage of 10 mV (1.9% of minimal current change after 10^4 biasing cycles and endurance up to 10^3 cycles of repetitive deformation with a 5 µm bending radius). Consequently, ultralow-power, motion-resistant epidermal electrocardiogram, electromyogram, and electrooculogram sensors are developed with an exceptional signal-to-noise ratio of 40.1 dB. The results of this study present a significant stride in non-invasive, skin-interfaced health-monitoring technologies and herald a new era in integrative health technologies.

1. Introduction

Ultraflexible organic electrochemical transistors (OECTs) are considered potential pivotal elements that facilitate physiological signal monitoring owing to their distinct attributes, including high sensitivity,[1–3] mechanical flexibility,[4–7] and biocompatibility.[8–11] Their unique ability to interlink ionic and electronic charges allows their effective operation at the ion-rich interfaces of skin and tissue, wherein, subtle electrochemical physiological signals are converted into electrical outputs with instrument compatibility.[12–15] For optimal bio-interfacing, several key factors must be addressed. 1) Enhanced amplification at minimal operating voltages, achievable by eliminating parasitic resistances to minimize the energy loss and foster an effective conduction path, 2) A rapid response time paired with an expansive cutoff frequency range where an optimized spatial design in the volumetric occupancy of the active layer can enhance ion transport and redox reaction efficacy, 3) robust mechanical resilience, given the dynamic nature of human skin, that remains unaffected by motion-induced distortions, necessitating isotropic channel designs to withstand multidirectional stresses and finally, 4) seamless adhesion to intricate skin topographies facilitated with an ultrathin design of minimal flexural rigidity, enabling a reliable and seamless human-machine interface to adhere to various skin textures without bending or fractures. Thus, to achieve optimal bio-interfacing with electronic devices, it is essential to strike a balance between electrical performance and geometrical constraints within a limited space while ensuring consistent functionality even under mechanical stress.

State-of-the-art OECTs have achieved profound advancements, with high transconductance (g_m) values of 384 mS and cutoff frequencies (f_c) of 1.2 kHz.[16] Such advances are attributed to the innovative use of redox-active semiconducting polymers blended with a redox-inert and photocurable polymer component in a vertical architecture, which facilitates long-term and rapid switching operations, demonstrating high-performance complementary circuits on a rigid substrate. Considering the flexibility benchmark, several flexible OECTs have been manifested on few-µm-thick polymeric substrates with transconductance values of a few tens of millisiemens (g_m <70 mS)[17] and cutoff frequencies lower than 560 Hz.[4] These devices also show potential for accurate sensing of physiological signals at skin and tissue surfaces.[8,18–22]

Despite these advancements, efforts to improve device performance often face limitations due to a trade-off related to channel

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channel radius of 70 μm and a channel length of 500 nm using a Tecnai G2 F30 S-Twin microscope operated at 300 kV. The SEM image of the ultraflexible vcOECT was obtained using a JSM-7900F microscope (Jeol, USA) operating at 15 kV. For the measurement, the ultraflexible vcOECT, laminated with a polymer elastomer (3 M VHB Y-4905J) pre-stretched to 200% and then compressed to a 50% strain, was loaded into the chamber. The Confocal images were obtained using an OLS3000 microscope with a 300 mm auto-stage. The analyses and plots were generated using Origin software.

Device Characterization and Analysis: Electrical characterization of the 2-terminal and 4-terminal OECTs was performed in two source-measure units (Keithley, 2400) under ambient conditions. All the electrical measurements were conducted in a phosphate-buffered saline (PBS) solution, the Ag/AgCl reference electrode immersed in KCl solution was used as the gate electrode. For both 2-terminal and 4-terminal measurements were conducted using identical device geometry regardless of pOECT or vcOECT, while measurement conditions were different. Detailed measurement setup is presented in Figure S1 (Supporting Information). To conduct 2-terminal measurement, the drain and source electrodes of the OECT devices were connected to the HI terminal and LO terminal of the source–measure units, respectively, for applying the drain voltage. The drain current of the device is measured by this source–measure unit. The Ag/AgCl gate electrode is connected to the HI terminal of another source-measure unit, which is used for applying the gate voltage to the OECT. The ground of the source–measure unit is shared by connecting both LO terminals to the source electrode of OECT. The measurement mode of source-measure unit for applying drain current is set to 2-wire sense mode. All the measurements were conducted by controlling the operation of source-measure unit using LabVIEW software. To conduct 4-terminal measurement, source, drain, and gate electrode connection is identical to the case of 2-terminal measurement, while the voltage probes ($V_{Sense,1}$ and $V_{Sense,2}$) are connected to the 4-wire sense connector of source-measure units for applying drain current, and the measurement mode of source–measure unit for applying drain current is set to 4-wire sense mode. All the source–measure unit and measurement parameters were controlled using LabVIEW software.

Frequency response of the 2-terminal and 4-terminal OECT devices was characterized by using a source-measure unit and oscilloscope/function generator equipment (Keysight Infinitives DSOX2002A). To perform the 2-terminal measurement, the drain and source electrodes of the OECT devices were connected to the HI terminal and LO terminals of the source–measure units, respectively, for applying the drain voltage. The Ag/AgCl gate electrode is connected to the oscilloscope/function generator equipment, which is used for applying the gate voltage to the OECT. To perform the 4-terminal measurement, source, drain, and gate electrode connection is identical to the case of 2-terminal measurement, while the voltage probes ($V_{Sense,1}$ and $V_{Sense,2}$)) are connected to the 4-wire sense connector of source–measure units, and the measurement mode of source–measure unit is set to 4-wire sense mode. A sinusoidal V_G signal (frequency range from 1 Hz to 10 kHz) with an amplitude of 50 mV was applied at offset potential where the maximum g_m was observed using a function generator. The drain current trace under a constant V_D was recorded using an SR570 preamplifier (Stanford Research Systems) and an oscilloscope. The transconductance was calculated as $\Delta I_D/\Delta V_G$, and the cutoff frequency was defined as the point where the transconductance value decreases by −3 dB from its initial value.

Mechanical Stability Test of Stretchable vcOECT: The freestanding ultraflexible vcOECT device was laminated with 200% outward pre stretched polymeric elastomer and placed in a compression machine. To accurately measure the electrical signal of the flexible device after peeling, the device pad was connected to an external wire using a Cr/Au (10/50 nm) line pattern. External wiring was deposited on a 3-μm-thick polyimide film through thermal evaporation. The contact between the device and the Au wiring was made with electrically conductive double-sided tape (3 M 9703-0.25″-36YDS). All mechanical stability tests were performed in PBS solution on polymer elastomers, and the Ag/AgCl reference electrode immersed in KCl solution was used as the gate electrode. Mechanical strains were measured using a compression machine, with the edges of the Au wires connected to alligator clips. A drain bias was applied via a source meter (Keithley 2400) connected to the Au wire.

ECG, EMG, and EOG Signal Measurement: To monitor ECG measurements, an ultraflexible device was attached to the right index finger using a drop of PBS solution, and source contact was established with the left chest using a gel electrode. For EMG measurements, the ultraflexible device was attached to the inner left wrist using a drop of PBS solution, and source contact was established with the inner right wrist using a gel electrode. To monitor EOG measurements, the ultraflexible device was attached to the upper and left areas around the eye using a drop of PBS solution, and source contact was established with the lower and right areas around the eye using a gel electrode. The connection to the gel electrode was made by linking the device pad to an external wire featuring a Cr/Au (10/50 nm) line pattern, fabricated using the same manufacturing method as the stretchable vcOECT measurements. The edge of the Au wire was connected to an alligator clip, which was attached to the gel electrode. The drain bias for the ECG, EMG, and EOG signals was applied using a source meter (Keithley 2400) connected to the Au wire. The ECG, EMG, and EOG signals were recorded at a sampling rate of 1 kHz using an SR570 preamplifier (Stanford Research Systems) and a data acquisition system (DAQ). The SR570 preamplifier was connected to the Au wire attached to the gel electrode. ECG signals were collected using a signal recording setup processed with a low-pass filter at 30 Hz. EMG signals were collected using a signal recording setup processed with a band-pass filter ranging from 1 to 300 Hz. EOG signals were collected using a signal recording setup processed with a low-pass filter at 30 Hz. The SNR was calculated as the ratio of the peak signal current to the standard deviation of the current between two peaks. All participants for the skin-conformal biosignal sensing experiments were co-authors of this manuscript and provided informed consent, who were approved by the Ajou University Institutional Review Board.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

I.L., J.H.K., and Y.K. contributed equally to this work. M.-H.Y. and S.P. conceived the idea for this work. I.L., Y.K., J.H.K., M.-H.Y., and S.P. designed the experiments and wrote the manuscript. I.L., D.S., H.L., J.-G.C., and J.W. fabricated the pOECT and vcOECT samples and conducted related electrical and mechanical experiments. I.L. fabricated the ultrathin vcOECT sample and conducted physiological signal acquisition. M.-H.Y., K.K., and S.P. oversaw the project, revised the manuscript, and led the effort to completion.

Data Availability Statement

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

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Sterilizable vertical n-type organic electrochemical transistors for skin-conformal ECG monitoring

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ABSTRACT

The development of stable, high-performance epidermal biosignal monitoring devices is critical for advancing wearable healthcare technologies. Here, we present a novel electrochemical transistor-based biosignal sensor utilizing a 4-terminal vertical Corbino configuration and an n-doped poly(benzodifurandione) (n-PBDF) polymer. The 4-terminal device configuration effectively reduces the parasitic resistance, enabling a high transconductance of 374 mS at a low operational voltage, and one of the highest reported μC^* values of 1787 F cm^{-1} V^{-1} s^{-1} for *n*-type OECTs. In addition, this device achieves exceptional operational stability, maintaining consistent performance over extended periods, and demonstrates a superior shelf-life stability under ambient conditions. Furthermore, the sensor exhibits robust sterilization capabilities, withstanding both UV and thermal sterilization processes without performance degradation. Mechanical flexibility, a key requirement for on-skin applications, is ensured by the intrinsic properties of the n-PBDF polymer and the ultra-thin device architecture. The combination of these features makes this device an ideal candidate for monitoring of biosignals such as electrocardiograms, addressing practical challenges in wearable biosensing technologies.

1. Introduction

Organic electrochemical transistors (OECTs), which utilize organic mixed ionic−electronic conductors (OMIECs), have gained significant attention in bioelectronics due to their high transconductance (g_m), operational stability, and low-noise biosignal transduction, positioning them as a promising platform for advanced healthcare technologies [1–3]. Despite remarkable performance and response speed advancements enabled by novel OMIECs, research has predominantly centered on device metrics, often neglecting the broader requirements for real-world applicability. To enable practical use of OECT-based biosignal monitoring devices, efforts must prioritize both long-term reliability and high performance. This requires accurate, sustained signal monitoring alongside electrical and mechanical stability, extended shelf-life, and sterilization resilience to ensure consistent and secure skin attachment. Three key factors must be addressed for effective long-term biosignal monitoring: i) low power consumption with high-amplification across varying signal polarities, ii) robust stability during operation, storage, and sterilization, and iii) mechanical softness for reliable adhesion to the complex skin surface upon dynamic mechanical stimuli. Addressing these challenges during the device development phase is essential for the successful application of OECTs as reliable and durable biosignal recorders.

State-of-the-art organic electrochemical transistors (OECTs) for biosignal recording have gained attention by utilizing depletion-mode PEDOT:PSS, which enables peak transconductance (g_m) to be achieved

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Skin-adhesive stretchable conductors for wireless vital diagnostics

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ABSTRACT

Continuous physiological signal monitoring and diagnosis are crucial for proactive health management and timely interventions. Key challenges include achieving non-toxic adhesion of stretchable conductors to dynamic skin and integration with lightweight, wearable circuits equipped diagnosing algorithms. We propose wireless physiological monitoring with vital diagnosis, featuring octopus-inspired micromembrane structure electrodes that enhance both adhesion and permeability. These stretchable electrodes exhibit a conductivity of over 2700 S/cm and maintain stretchability up to 1000 %, with minimal degradation after 1000 cycles of deformation. Adhesion reaches 12 kPa, ensuring durability for over 1000 attachment-detachment cycles and long-term attachment exceeding 24 h without skin toxicity. The system, connected to a miniaturized wireless circuit (2.8 g), facilitates real-time, accurate collection of electrocardiography (ECG), electromyography (EMG), electrooculography (EOG), and electroencephalography (EEG) signals. As proof of concept, ECG signals from real subjects processed with a transfer-learning algorithm achieved over 93.3 % diagnostic accuracy, paving the way for reliable, personalized health monitoring.

1. Introduction

Monitoring and diagnosing long-term physiological signals have gained significant attention over the past decade due to their ability to deliver real-time health insights [1–5]. Continuous monitoring is essential, as abnormal physiological signals often occur intermittently rather than periodically, requiring long-term data collection for accurate detection. This capability enables prompt intervention and optimized treatment when such irregular health indicators are identified based on extended measurements [6,7]. To ensure the robust performance of

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

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Data availability

Data will be made available on request.

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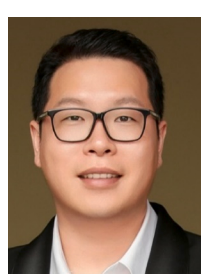

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