Title**Totally shape-conformable electrode/hydrogel composite for on-skin electrophysiological measurements**

Kuniaki Nagamine, Shun Chihara, Hiroyuki Kai, Hirokazu Kaji, Matsuhiko Nishizawa*

Department of Bioengineering and Robotics, Graduate School of Engineering, Tohoku University,
6-6-01 Aramaki, Aoba-ku, Sendai 980-8579, Japan.

*corresponding author

E-mail: nishizawa@biomems.mech.tohoku.ac.jp

Abstract

An Au film electrode supported by a conductive elastic film was tightly bonded on a stretchable double-network hydrogel sheet by means of the conducting polymer poly(3,4-ethylenedioxythiophene) (PEDOT) grown from the electrode surface into the hydrogel. This electrode/hydrogel composite showed stable resistance of $35 \pm 5 \Omega \text{ sq}^{-1}$ even during successive 20 % stretching because of the pre-formed, designed cracks in the Au film. The large interfacial electric double layer capacitance ($9.5 \pm 0.3 \text{ mF cm}^{-2}$) of the PEDOT adhesive layer at the interface of the layered composite was found to stabilize the electrode potential against external noises, and decrease the electric impedance at the frequency of 5-500 Hz, which is the typical range of electromyographic signals. The electrical robustness and shape-conformability of the composite electrode was demonstrated by monitoring electromyographic signals of the joint of a human forefinger. In addition, it was also demonstrated that an ionic liquid-containing gel (ionogel) serves as a substrate of the composite for longer-term monitoring over 3 days on air-exposed human skin.

Keywords

- Electrode/hydrogel hybrid
- Conducting polymer
- Shape-conformable device
- Electrophysiological measurements

1. Introduction

Electrophysiological measurements such as electrocorticography, electrocardiography, and electromyography on the human body surface have been widely performed for medical diagnosis, medical monitoring, and neuron-to-machine interfaces for prostheses [1,2]. These electrophysiological measurement systems have recently been integrated into wearable devices for daily health monitoring and therefore, much effort has been paid for the development of flexible and stretchable electric devices [3-5] in order to ensure the motion adaptivity. For stable monitoring, it is necessary for electrode devices to maintain electric contact with the curvature of a skin even during body motion. Therefore, soft electrolyte gels are practically used as an interfacial material between the solid electrode devices and the soft skin. In fact, a commercially available Ag/AgCl patch is combined with an adhesive electrolyte gel [6]. While these conventional hydrogels are not elastic, stretchable hydrogels such as double network (DN) hydrogel have been recently developed [7,8]. By combining stretchable electrode devices and stretchable hydrogels, a totally shape-conformable electrode device could be realized for stable electrophysiological measurement at any parts of a body with motions.

In this study, we fabricated a shape-conformable electrode/hydrogel composite shown in Fig. 1, in which a stretchable Au electrode was tightly attached to the surface of a DN hydrogel sheet. The electrochemically deposited poly(3,4-ethylenedioxythiophene) (PEDOT) serves as a stable adhesive layer [9-11]. This PEDOT adhesive layer showed the advantage of low-impedance and low-noise measurement. In addition, it was also demonstrated that a non-volatile ionic liquid-containing gel (ionogel) can be used for longer-term monitoring of electrophysiological signals on skin.

2. Experimental

2.1. Fabrication of a double network hydrogel

The double network hydrogel was prepared according to the previously reported protocols [7]. The first prepolymer solution containing 1 M 2-acrylamido-2-methyl-1-propanesulfonic acid sodium salt (NaAMPS, Sigma Aldrich) as a monomer, 40 mM N,N'-methylenebis(acrylamide) (MBAA, Wako Pure Chemicals) as a crosslinker, and 1 mM 2-oxoglutaric acid (Wako Pure Chemicals) as a photoinitiator was prepared. The solution was poured into the reaction chamber composed of two glass plates with a silicone spacer of 0.2 mm thickness between them. The reaction chamber was irradiated with UV light (365 nm, 8 W, Funakoshi) for 6 h in a nitrogen-filled glovebox. The obtained PNaAMPS hydrogel sheet was immersed in the second aqueous solution containing 3 M

N,N-dimethylacrylamide(DMAAm, Sigma Aldrich), 3 mM MBAA, and 1 mM 2-oxoglutaric acid for 2 days at room temperature in the dark. The hydrogel sheet was again irradiated with UV light for 6 h in the nitrogen-filled glovebox to polymerize the second hydrogel, followed by washing with distilled water overnight to remove residual cytotoxic monomers. The resulting DN hydrogel has a thickness of about 0.5 mm after swelling in PBS solution.

2.2. Fabrication and evaluation of electrode/hydrogel composite

The Au arrays of 0.2 mm x 0.2 mm square tiles with 5 μm gaps were prepared by photolithographic patterning of sputtered Au (425.6 ± 20.6 nm ($n=3$) thickness) on a glass substrate [12]. A composite film of PEDOT and polyurethane (PEDOT/PU) was prepared on the Au arrays as described in our previous report [9], by spin coating (750 rpm, 30 s) of a precursor solution and its thermal polymerization at 100 $^{\circ}\text{C}$ for 10 min. The precursor solution used here was a mixture of butanol (2 mL, Wako Pure Chemicals), EDOT (0.88 mL of 1 M solution, Clevios MV2, Heraeus), pTSCFe(III) (6.5 mL of 0.4 M solution, Clevios C-B40V2, Heraeus) and 10 wt% PU/tetrahydrofuran solution (35 mL, Gumthane AR650, a kind gift from Okada Engineering Company). The thickness of the PEDOT/PU film, measured by a probe type step profiler (DEKTAC150), was approximately 3 μm . The Au-PEDOT/PU film on the glass plate was immersed in distilled water overnight to remove residual cytotoxic monomer and dopant, and cut out to a desired pattern by a cutting plotter (Craft ROBO Pro, Graphtec). A DN hydrogel sheet (0.5 mm thick) was then placed on the patterned Au-PEDOT/PU film. After permeation of monomer solution (50 mM EDOT and 100 mM LiClO_4 aqueous solution) into the hydrogel sheet, PEDOT was electropolymerized (300 mC cm^{-2}) to anchor the electrode to the hydrogel film [9-11]. Finally, the resulting electrode/hydrogel composite film was gently peeled from the glass substrate and further immersed in water overnight to **remove residual EDOT** monomer.

For the resistance measurement during stretching, four-point probes were attached at the two ends of the Au-PEDOT/PU film (10 mm width and 30 mm length), which was elongated at a strain rate of 0.1 mm s^{-1} using an electrically driven stage (SGSP O-35, Sigma Koki). A constant current of 10 mA was applied to the two outer probes using a Keithley 2400 SourceMeter and the potential difference was measured using the two inner probes connected to a voltmeter (ALS760C, BAS Inc., operated in the voltmeter mode) during stretching.

2.3. Interfacial AC impedance measurement and cyclic voltammetry

AC impedance at the interface between electrodes and DN hydrogels hydrated with PBS solution was measured in a three-electrode configuration using an electrochemical analyser (ALS760C, BAS Inc., operated in the AC impedance measurement mode). A Pt flag (counter

electrode) and Ag/AgCl (reference electrode) were placed in contact with the surface of the DN hydrogel to ensure ionic connection and the AC impedance was measured by applying 5 mV_{p-p} voltage at 1 Hz-100 kHz.

AC impedance on skin was measured by placing the electrodes on a human subject. Commercially available, adhesive gel-assisted Ag/AgCl electrodes (NM-31, Nihon Kodens) were used as counter and reference electrodes. For the measurement on a forearm, the working (tested) and counter electrodes were placed on the forearm 5 cm away from each other, and the reference electrode was placed on the arm 30 cm away from the working electrode. For the measurements on a forefinger joint, the working electrode was placed on the joint of a forefinger, the reference electrode 5 cm away from the working electrode, and the counter electrode on a wrist.

Cyclic voltammetry was performed in the same three-electrode configuration setup using a potentiostat (ALS760C, BAS Inc., operated in the cyclic voltammetry mode). The potential applied to the working electrode was cycled between -0.4 to 0.4 V vs. Ag/AgCl at a scan rate of 10 mV s⁻¹ while the current was measured.

2.4. Evaluation of the potential stability

The potential difference between the test electrode and the reference Ag/AgCl electrode was measured using a voltmeter (ALS760C, BAS Inc., operated in the voltmeter mode). The test electrodes were a **bare** Au film covered by a DN hydrogel hydrated with PBS solution and the Au-PEDOT/PU electrode/DN hydrogel composite with the entangling PEDOT adhesive. The reference Ag/AgCl electrode was placed in contact with the surface of the DN hydrogel to ensure ionic connection. A Keithley 2400 SourceMeter was connected to the circuit in series to inject ± 1 nA of constant current as a model of external noise. After waiting for 30 s, a constant current of +1 nA was applied on the test electrode for 30 s, then the current was turned off for 30 s, followed by a current of -1 nA for 30 s during the recording of the potential difference.

2.5. Electromyographic (EMG) signal measurements

EMG signal measurement was performed by placing the Au-PEDOT/PU electrode/DN hydrogel composite on the joint of a forefinger to ensure the contact between the hydrogel surface and the skin, and the gel-assisted Ag/AgCl electrode on the back of a subject's hand 5 cm away from the composite. Another gel-assisted Ag/AgCl electrode was placed on the wrist as a ground electrode. These electrodes were connected to the wireless EMG logger (input impedance: 10 T Ω , SPORTS SENSING Co., LTD) to detect EMG signals during repeated bending motion of the joint of a forefinger.

3. Results and discussion

The stretchable conductors are the key component of the present electrophysiological sensing device. We employed an Au film supported by an elastic conductor that is the composite of conducting polymer PEDOT and polyurethane (PU) [9], which is noted as Au-PEDOT/PU electrode in this manuscript. The attachment of Au on the PEDOT/PU was strong enough as checked by a scotch tape test. At first, we tried to use a continuous Au film. However, the resistance of this Au-PEDOT/PU electrode randomly changed depending on the number and shape of the cracks in the Au film although these cracks can be bridged by the underlying conductive PEDOT/PU support to prevent insulation. Therefore, in order to ensure stable, reliable electric property, we designed the pre-formed cracks in the Au film. Fig. 2a exhibits the array of 0.2 mm x 0.2 mm square Au tiles with 5 μm gaps on the PEDOT/PU, which shows resistance of $19.1 \pm 2.5 \Omega \text{ sq}^{-1}$ ($n=3$) that can be reasonably calculated from an equivalent circuit (Suppl. Fig. S1) using resistance of the Au film ($2.0 \pm 0.6 \Omega \text{ sq}^{-1}$, $n=3$), PEDOT/PU ($127.3 \pm 9.0 \Omega \text{ sq}^{-1}$, $n=3$), and the contact resistance between Au and PEDOT/PU (6.8Ω) that were separately measured. The electrode of smaller tiles (0.1 mm x 0.1 mm) showed larger resistance due to the increased number of the gaps. The larger Au tiles than 0.2 mm x 0.2 mm often cleaved upon stretching. Fig. 2b shows the sheet resistance of Au-PEDOT/PU during the first two cycles of 20 % stretching. After resistance increased to $40 \Omega \text{ sq}^{-1}$ by the first stretching, 10 cycles of reversible changes between 30 and $40 \Omega \text{ sq}^{-1}$ were observed (Fig. 2c). The irreversible increase caused by the first elongation is probably due to the partial rupture of PEDOT/PU substrate. As explained later, the $10 \Omega \text{ sq}^{-1}$ fluctuation does not pose a problem for electrophysiological measurements such as electromyography (EMG).

The Au-PEDOT/PU electrode was attached to a hydrogel sheet by means of entanglement of PEDOT polymer chains that are electrochemically grown from the surface of the electrode into the hydrogel, as described in our previous report.⁹ Fig. 3a shows the cross sectional image of the PEDOT (300 mC cm^{-2}) grown into the DN hydrogel up to approximately 50 μm depth as an anchor to fix the electrode. Fig. 3b shows the interface impedance between the hydrogel containing PBS and three kinds of electrodes: a bare Au film, an Au film with a prior coating of PEDOT (300 mC cm^{-2}), and the present electrode with the entangling PEDOT adhesive. As has already been reported [13-17], the pre-coating of the electrode with PEDOT is effective in decreasing the interface impedance between an electrolyte gel and an electrode. Importantly, the entangling PEDOT grown into the DN hydrogel shows the same or higher effect as that of the conventional PEDOT coating at the frequency of 5-500 Hz, which is the typical range of electrophysiological signals [18]. These effects of the PEDOT in

lowering impedance can be attributed to the large double-layer capacitance of the fibril structure of PEDOT. Cyclic voltammograms in Fig. 3c shows that both of the PEDOT electrodes (PEDOT coating and PEDOT adhesive) have larger capacitive currents than the bare Au electrode: $9.5 \pm 0.3 \text{ mF cm}^{-2}$ for the PEDOT electrodes and $0.077 \pm 0.04 \text{ mF cm}^{-2}$ for the bare Au electrode.

In addition to the advantage of low-impedance, the capacitive PEDOT adhesive can show the low-noise property because the potential drift is related to the capacitance of an electrode surface (C) and the noise current (I) as follows [19]:

$$\text{Potential drift} = dV/dt = I/C$$

Fig. 3d shows the change of the electrode potentials when a constant current of $\pm 1 \text{ nA}$ was injected as a model of external noise [19]. The bare Au electrode showed intense potential drift ($78.6 \text{ } \mu\text{V s}^{-1}$ for $+1 \text{ nA}$, $53.7 \text{ } \mu\text{V s}^{-1}$ for -1 nA) upon the noise injection. On the other hand, the potential of the electrode with the PEDOT adhesive was stable with negligible drift due to the larger surface capacitance.

Fig. 4a shows photographs of the electrode-hydrogel composite that was just put on the joint of a forefinger. The totally stretchable composite stably adhered to the skin surface even upon 90-degree bending of the finger without any additional adhesive material (the strain at the forefinger joint: 20 %). Fig. 4b shows the spectra of contact impedance without significant change during 10 cycles of bending. On the other hand, the spectra of the commercial electrode with adhesive gel often changed by bending, which is accompanied by partial detachment because of the lack of stretchability of the solid Ag/AgCl electrode and the stiff silicone substrate (Suppl. Fig. S2). The robustness of the electrode/hydrogel composite for EMG measurement was demonstrated on the moving forefinger joint as shown in Fig. 4c (Suppl. Movie S1). The spike signals were obtained, which was synchronized with the 90-degree bending motion of the forefinger. This proves stable EMG monitoring even at the drastically moving parts of a human body. Since the input impedance of the EMG logger is typically $10 \text{ T}\Omega$, the resistance change of the Au-PEDOT/Pu electrode during reversible 20 % strain ($10 \text{ } \Omega \text{ sq}^{-1}$, Fig. 2c) corresponds to the negligible 10^{-15} V fluctuation.

The stability of the contact impedance of the stretchable electrode/hydrogel composite on the subject's arm is shown in Fig. 5a. The measurement was conducted every hour at 10 Hz, which is the typical operation frequency of a commercial EMG sensing system for the contact impedance checking circuit [20]. The impedance was stable around $10^5 \text{ } \Omega$ for the first 2 h, but drastically increased at 3 h because of delamination of the dried hydrogel. Re-swelling of the dried gel satisfactorily recovered the original impedance, suggesting the

possible longer-term monitoring by periodic water supply. On the other hand, when the ionogel containing non-volatile ionic liquid was used as a substrate, the sensor was stable for longer time without refilling ionic liquid. Recently, Leleux et al. have reported the effective use of the ionogel-assisted conformal electrode for long-term on-skin EMG measurement [13,14]. Here the stretchable electrode/hydrogel composite was prepared by exchanging the aqueous solution in the hydrogel for the ionic liquid (1-ethyl-3-methylimidazolium ethyl sulfate) by soaking the device overnight until its weight became constant (Suppl. Fig. S3). The size of the device did not change significantly during the solvent exchange. Another possible approach to prepare the electrode/ionogel hybrid could be re-swelling of the previously dried hydrogel with the ionic liquid. However, this swelling process took more than 5 days, and therefore we adopted the former technique (overnight soaking of a wet hydrogel). The skin contact impedance of the electrode/ionogel composite at 10 Hz was almost constant even after 3 days of measurement without drying of the film (Fig. 5b). Non-volatility of the ionic liquid was advantageous for continuous long-term monitoring, although it should be noted that biocompatibility of ionic liquid has remained to be discussed [21]. **Future progress in ionic liquid synthesis is expected to provide more biocompatible ionic liquids.**

4. Conclusions

This study showed performance of the totally stretchable electrode/hydrogel composite as an electrophysiological sensor, which is prepared by binding a stretchable electrode and a hydrogel with the electrochemically deposited conductive PEDOT. The large electric double layer capacitance of the PEDOT adhesive contributes to lowering electrode/hydrogel interfacial impedance for sensitive measurement. **Especially at 5 Hz which is in the typical range of electrophysiological signals, 27 times lower electrode/hydrogel interface impedance was obtained for the PEDOT adhesive than that of bare Au film.** Additionally, the capacitive PEDOT adhesive stabilizes the electrode potential against external noise. The electrical robustness and shape-conformability of this composite electrode ensure the stable electrophysiological measurement even on the parts of human **finger during 10 cycles of drastic 90-degree bending movements without detachment from the skin.** The use of ionogel enables the longer daily medical diagnosis and health monitoring on air-exposed human skin.

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Biographies

Kuniaki Nagamine is an assistant professor of Department of Bioengineering and Robotics, Graduate School of Engineering, Tohoku University. He received his PhD degree in 2007 from Tohoku University. His current interests include tissue engineering, organ on a chip, and hydrogel-based electric devices.

Shun Chihara is currently a MS student of Tohoku University. He received his BS degree in 2014 from Tohoku University. His current research is the development of the hydrogel/ electrode hybrids.

Hiroyuki Kai is an assistant professor of Department of Bioengineering and Robotics, Graduate School of Engineering, Tohoku University. He received his PhD degree in 2015 from University of California. His current research has been focused on the development of flexible biosensors based on material science and electrochemistry.

Hirokazu Kaji is an associate professor of Department of Bioengineering and Robotics, Graduate School of Engineering, Tohoku University. He received his PhD degree in 2005 from Tohoku University. His current interests include organ on a chip and the application of micro- and nanotechnologies in the fields of cell and tissue engineering.

Matsuhiko Nishizawa is a professor of Department of Bioengineering and Robotics, Graduate School of Engineering, Tohoku University. He received his PhD degree in 1994 from Tohoku University. His current interests include biofuel cells and hydrogel-based electric devices.

Figures and their captions

Fig. 1. Schematic illustration of the developed shape-conformable electrode/hydrogel layered composite on a forefinger.

Fig. 2. (a) The Au arrays of 0.2 mm x 0.2 mm square tiles with 5 μm gaps on a PEDOT/PU film imaged by a scanning electron microscopy. (b, c) Sheet resistance of the Au-PEDOT/PU during the two cycles of 20 % stretching (b) and the following 10 cycles (c).

Fig. 3. (a) A cross sectional image of the PEDOT (300 mC cm^{-2}) grown into the DN hydrogel. (b) Interface impedance between the hydrogel and the electrodes: **red** Au film (gray), the Au film with a prior coating of PEDOT (blue), and the electrode with the entangling PEDOT adhesive (red). (c) Cyclic voltammograms measured with the **red** Au film (gray), the Au film with a prior coating of PEDOT (blue), and the electrode with the entangling PEDOT adhesive (red). (d) Change of potential of the **red** Au film (gray) and that with the entangling PEDOT adhesive (red) when a constant current of $\pm 1 \text{ nA}$ was injected as a model of external noise.

Fig. 4. (a) Photographs of the electrode/hydrogel composite that was put on the joint of forefinger. (b) AC impedance spectra measured by the electrode/hydrogel composite before (blue) and after (red) the 1st cycle (dashed lines) and the 10th cycle (solid lines) of 90° bending of the joint. (c) The EMG signal monitored during the repeated bending of the forefinger joint.

Fig. 5. (a) The contact impedance at 10 Hz of the electrode/hydrogel composite measured every hour on the subject's arm. Each point represents average value $\pm$ standard deviation ($n=3$). Note that two of the three dried samples could not be laminated on the skin at 3 h and their contact impedances was not measured. (b) The contact impedance at 10 Hz of the electrode/hydrogel (blue) and the electrode/ionogel (red) composites. The y-axes in this figure represent the value of impedance at 10 Hz from AC impedance spectra measured by applying 5 mV_{p-p} voltage at 1 Hz-100 kHz.

Graphical Abstract

Totally Shape-Conformable