## **Supplementary Information**

## Fully soft organic electrochemical transistor enabling direct skin-mountable

## electrophysiological signal amplification

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## **Supplementary Notes**

#### **1. Materials Preparation**

Poly(3,4-ethyelendioxythiophene):poly(styrene sulfonate) (PEDOT:PSS, Heraeus Clevios PH1000) was purchased from Ossila. PEDOT:PSS is also known as Poly(2,3-dihydrothieno-1, 4-dioxin)-poly(styrenesulfonate). Polyethylene glycol (PEG, PEG400) was from Alfa Aeser. dodecylbenzene sulfonic acid (DBSA), 1-Ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (EMIM-TFSI), Poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-co-HFP) were purchased from Sigma-Aldrich. Silver nanowires (AgNWs) dispersed in isopropyl alcohol (IPA) were from Kechuan. Divinyl sulfone (DVS) was purchased from TCI chemicals. The elastomeric polymer, Styrene-ethylene-butylene-styrene (SEBS, G1652), was purchased from Kraton polymers. All materials were used as received without further purification process.

#### 2. Preparation of AuNPs-AgNWs/SEBS electrodes

The elastomeric AuNPs-AgNWs/SEBS composite electrodes (source and drain for soft OECTs and advanced soft OECTs, and source, drain and gate electrodes for fully soft OECTs) were prepared on a clean glass slide (glass slides cleaned with acetone, IPA, and then dried with nitrogen) by Kapton film based shadow mask. AgNWs solution was coated and heated at 60 °C to remove the solvent, followed by a welding process at 200 °C for 30 min to enhance conductivity. Thereafter, 200 mg/ml of SEBS solution was poured on patterned glass slides and kept the sample at room temperature to form a film. The porous nature of AgNWs network structure allows the SEBS solution easily permeate into the pores. The solidification of the SEBS led to partially embedded AgNWs in SEBS. The prepared film on glass was carefully peeled off. The preparation process is schematically illustrated in the Fig. S3 from i to vii (ESI). To ensure ohmic contact between soft active channel materials and electrode, the exposed AgNWs out of SEBS were coated by AuNPs through galvanic replacement using HAuCl4 •  $3H_2O$  (0.5 mM). The galvanic replacement reaction is one of facile process for the formation of metal nanostructure on the surface of different metal. In this work, the reduction potential difference between Au and Ag induces spontaneous redox reaction of Au<sup>3+</sup> and Ag.

 $HAuCl_4 + 3Ag \rightarrow Au + 3AgCl + HCl$ 

NH<sub>4</sub>OH solution was further used to remove the byproducts such as AgCl on the nanowires, followed by sequential rinsing with water and drying by nitrogen gas to finalize elastomeric AuNPs-AgNWs/SEBS electrodes.

### **3. Fabrication of soft OECTs**

PEDOT:PSS (PH1000) was mixed with 4 vol% PEG400. It is noted that 0.25% DBSA was added to reduce surface tension. In addition, 3 vol% DVS was added into the solution, followed by ultrasonication for 20 min. Soft active channel material was formed on the channel area of the electrode (namely, between source and drain) through spin casting after 30 min UV- $O_3$  treatment on the surface of the channel with Kapton film based shadow masks. The coating was repeated thrice, and 1 min annealing at 60 °C was performed after each coating process, followed by final annealing for 1 hr to ensure sufficient crosslinking between PEDOT:PSS-PEG and DVS. It is noted that channel dimension is determined by considering both device performance and potential capability for integration. Therefore, we chose the thickest film thickness (436.3 ± 33.8 nm) and the moderate planar dimension (200 µm length and 2000 µm width) from the experimental results of  $g_m$  depending on various channel geometries. Lastly, the ionic liquid was used as an electrolyte by dropping on the active channel material during characterization along with Ag/AgCl composite pellet as a gate electrode.

## 4. Preparation of iongel

Ionic liquid (EMIM-TFSI) was dehydrated at 70 °C in a vacuum oven for 24 hours. After that, PVDF-co-HFP and the ionic liquid were dissolved in acetone (1:4:7 weight ratio of polymer : ionic liquid : acetone), followed by vigorous stirring (340 rpm) at the temperature of 80 °C for 2 hours. The prepared solution was cast on glass and cured in a vacuum oven to eliminate air bubbles and solidify the gel. The obtained solid electrolyte could be peeled off from the slide and used to assemble the OECTs device.

### 5. Fabrication of advanced soft OECTs

The fabrication of advanced soft OECTs began with the abovementioned soft OECTs manufacturing process. Then, the iongel was laminated on top of the channel area instead of

applying ionic liquid to finalize the device fabrication. For the device characterization, Ag/AgCl pallet was utilized as a gate electrode.

#### 6. Fabrication of fully soft OECTs

The fabrication of advanced soft OECTs involves the aforementioned AuNPs-AgNWs/SEBS composites electrodes preparation and soft active channel preparation. Then, the soft gate electrode was prepared by a twice coating of PEDOT:PSS through Kapton film based shadow mask. It is worth noting that each coating involves two-step coating process, including 2000 rpm for 60 sec to spread materials uniformly over the target place and 3000 rpm for 30 sec to arrest the polymer structure. Finally, the prepared iongel was placed on the gate electrode and was fully covered the channel region as schematically shown in Fig. 4a.

#### 7. Characterization of materials and devices

The absorption spectra of PEDOT:PSS with PEG and PEDOT:PSS with PEG and DVS films were measured using Jasco V-670 spectrophotometer under room temperature. Electrical characteristics of all devices and film were performed by using Keithley 4200. AFM measurements were carried out using Bruker Dimension ICONXR in tapping mode. The ECG signals were obtained using Sparkfun AD8232 and Arduino UNO-3. IR spectra were recorded using FT-IR spectrometer (Spectrum Two, Perkin Elmer) with an attenuated total reflectance (ATR) accessory. <sup>1</sup>H NMR spectra were recorded at room temperature using 400 MHz FT-NMR (Avance III HD, Bruker) spectrometer.

#### 8. Analyses of ATR-FTIR and NMR for crosslinking between PEG and DVS.

Fig. S10 (ESI) shows the ATR-FTIR spectra, which present the loss of O–H stretch peak (~3450 cm<sup>-1</sup>) from PEG, loss of C–H stretch peak (~3050 cm<sup>-1</sup>)/band peak (~977 cm<sup>-1</sup>) from DVS, and enhanced C–O stretch peak (~1100 cm<sup>-1</sup>) from the crosslinked product. The results clearly indicate the crosslinking reaction between DVS and PEG. Moreover, the <sup>1</sup>H NMR spectra prove the reaction. Compared to spectra of DVS only and PEG only, the spectrum of PEG:DVS shows the apparent disappearance of proton peaks from DVS (around 6.9, 6.5, and 6.35 ppm). In addition, new triplet peaks at 3.97 ppm and 3.53 ppm corresponding to the CH<sub>2</sub>

proton from the crosslinked product are observed. Therefore, both ATR-FTIR and <sup>1</sup>H NMR analyses are experimental evidence for the bond formation between PEG and DVS.

# Supplementary Figures



**Fig. S1** Conductivity change of PEDOT:PSS with PEG and DVS depending on mechanical strain (n = 5).



**Fig. S2** (a) Schematic illustration of the galvanic replacement process. SEM images of (b) AgNWs/SEBS and (c) AuNPs-AgNWs/SEBS. (d) Resistance change of the AuNPs-AgNWs/SEBS under mechanical strain (n = 5).



Fig. S3 Schematic fabrication process of the OECTs



**Fig. S4**  $g_m$  summary of soft OECTs with variation in channel geometries (n = 5).



**Fig. S5** Average thickness of the soft active channel depending on the number of spin-casting (n = 5).



Fig. S6 Single cycle of dynamic current switching response of the soft OECTs.



**Fig. S7** (a) Schematic illustration of the advanced soft OECTs. (b) Output and (c) transfer characteristics of the device. (d) Dynamic current switching response of the soft OECTs.



**Fig. S8** Transfer characteristics of the advanced soft OECTs under mechanical strain (a) perpendicualr and (b) parallel to channel.



**Fig. S9** Conductivity changes of PEDOT:PSS and PEDOT:PSS with PEG under mechanical strain (n = 5).



Fig. S10 ATR-FTIR spectra of DVS, PEG and PEG-DVS mixture.



**Fig. S11** <sup>1</sup>H NMR spectra of DVS, PEG and PEG-DVS mixture, recorded in D<sub>2</sub>O.



Fig. S12 Water soaking test of active channel films with and without crosslinking between PEG and DVS.



**Fig. S13** UV-vis-NIR absorption spectra of PEDOT:PSS with PEG thin films and PEDOT:PSS with PEG and DVS.



**Fig. S14** Normalized resistivity change of PEDOT:PSS with PEG and PEDOT:PSS with PEG-DVS (n = 5).



Fig. S15 (a) Output and (b) transfer characteristics of fully soft OECTs.



**Fig. S16** Voltage transfer curves of fully soft OECTs based amplifier under with and without strain ( $\varepsilon = 20\%$ ) at supplied voltage of (a) -4 V and (b) -6 V.



**Fig. S17** Sinusoidal output signals without amplifier, with amplifier under no strain, and with amplifier under strain of 20%.



Fig. S18 (a) Circuit diagram and (b) camera image of the data acquisition setting for ECG measurement.