Supplementary Information

Proton-Penetrable Nafion-Induced Phase Separation in Organic Semiconductor for High-Performance Organic Electrochemical Transistor

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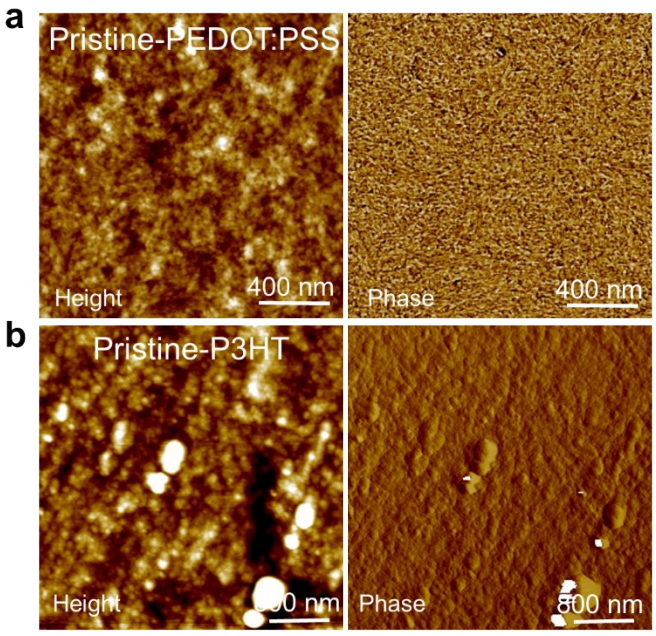
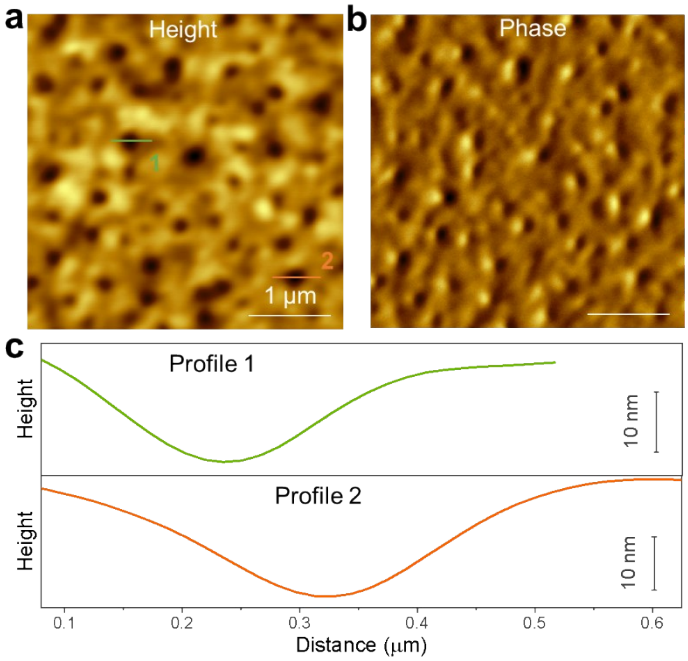
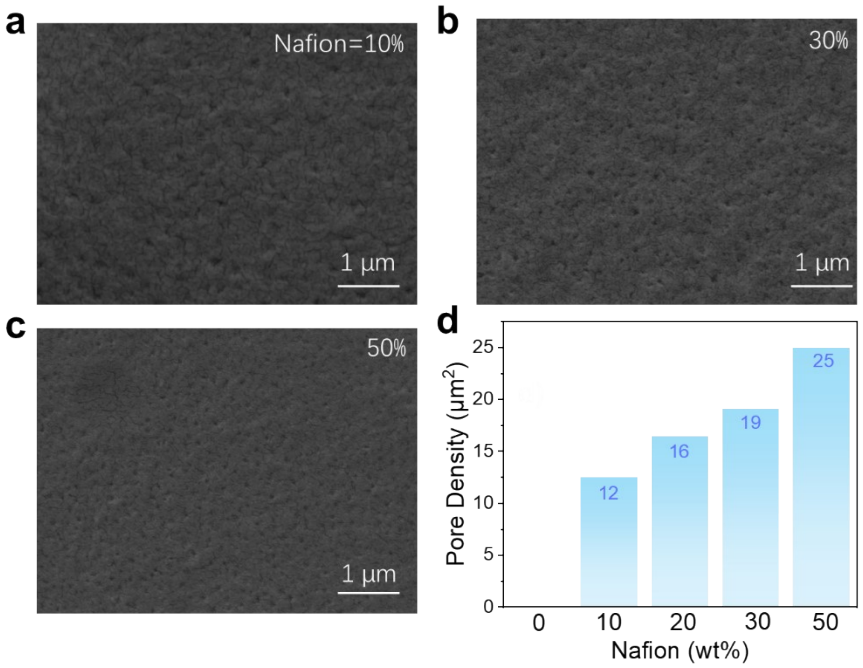
Supplementary Figures



Supplementary Fig. 1 AFM height (left panel) and phase (right panel) images of the (a) Pristine PEDOT:PSS and (b) P3HT films. The scale bars are 400 and 800 nm, respectively.

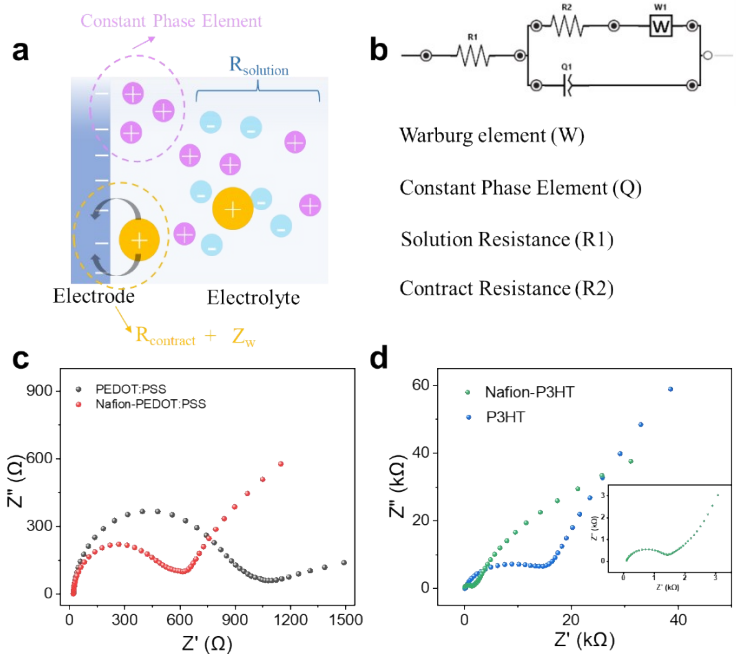


Supplementary Fig. 2 AFM height and phase images of the Nafion-included P3HT film and the depth of the pores.



Supplementary Fig. 3 (a-c) SEM images of the PEDOT:PSS films mixed with different contents of Nafion. (d) Statistical chart of the pore density versus the Nafion content.

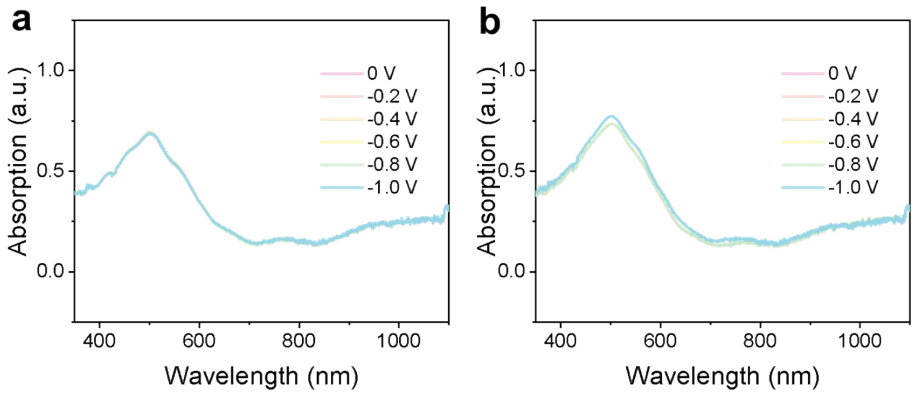
The doping/de-doping processes in organic semiconductor films in electrolyte were further investigated by EIS. We describe these impedance spectra by the equivalent circuit composed of a Warburg element, an RC parallel circuit (CDL, RDL), and a resistor in series (RS). By simulating the acquired results using the equivalent circuit, the capacitance of the systems could be obtained. After being normalized by the channel volume, the volumetric capacitance of the channel could be obtained. In the case, the ion diffusion in pristine P3HT is very slow. Thus, we tested the EIS spectra of P3HT and Nafion-P3HT films, in which a larger frequency range of 0.01 to 10^5 Hz was used.



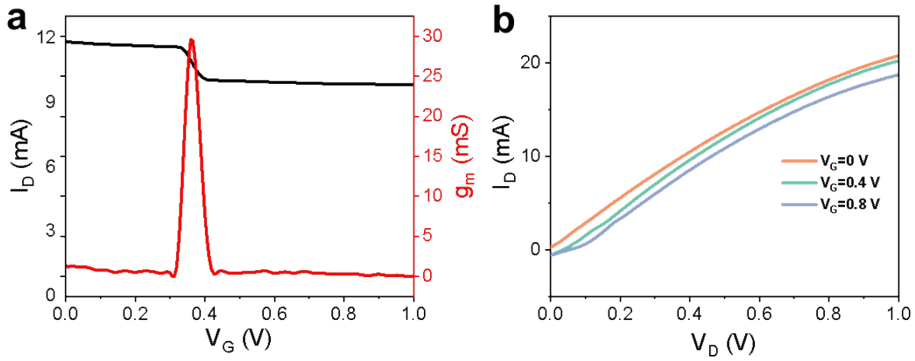
Supplementary Fig. 4 (a) Schematic diagram of the interface between semiconductor and electrolyte. (b) Electric circuit model for EIS fitting. (c) EIS spectra of PEDOT:PSS and Nafion-PEDOT:PSS. The frequency varied from 105 to 10-1 Hz during the measurements. (d) EIS spectra of P3HT and Nafion-P3HT. Inset image is a zoomed in EIS spectra of Nafion-P3HT film. The frequency varied from 105 to 10-2 Hz during the measurements.

Supplementary Table. 1. The fitted *C* value of different polymers.

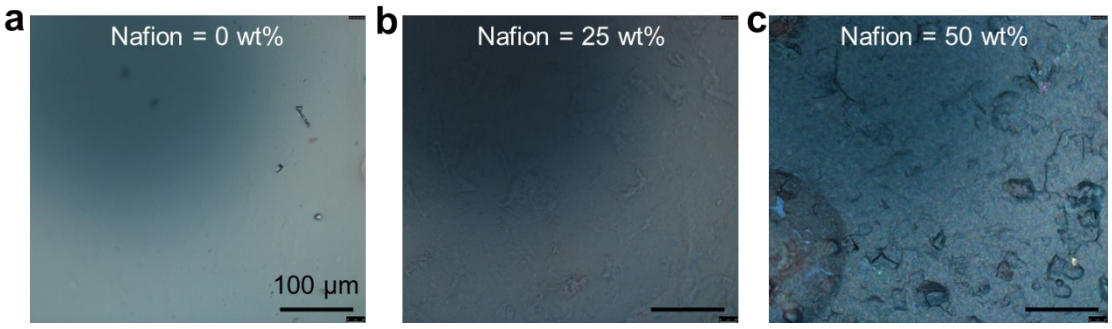
	Pristine PEDOT:PSS	Nafion-PEDOT:PSS	Pristine P3HT	Nafion-P3HT
Fitted *C* value (μF)	6.1	6.5	2.2	2.5



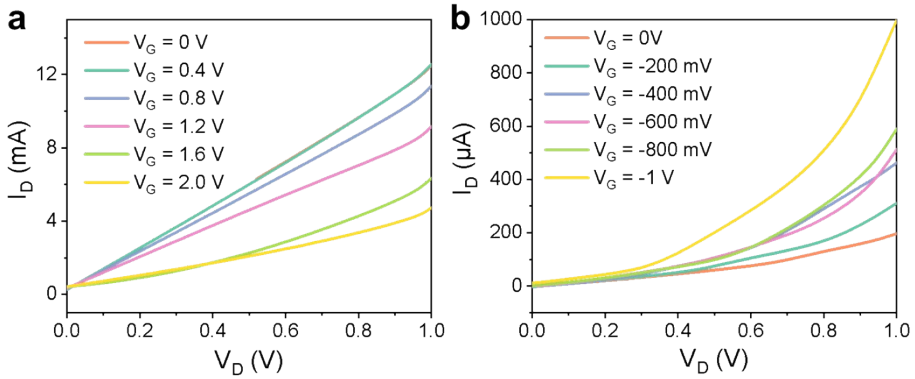
Supplementary Fig. 5 UV-Vis-NIR spectra of the (a) pristine and (b) Nafion-included P3HT films at biases ranging from -1.0 to 0 V.



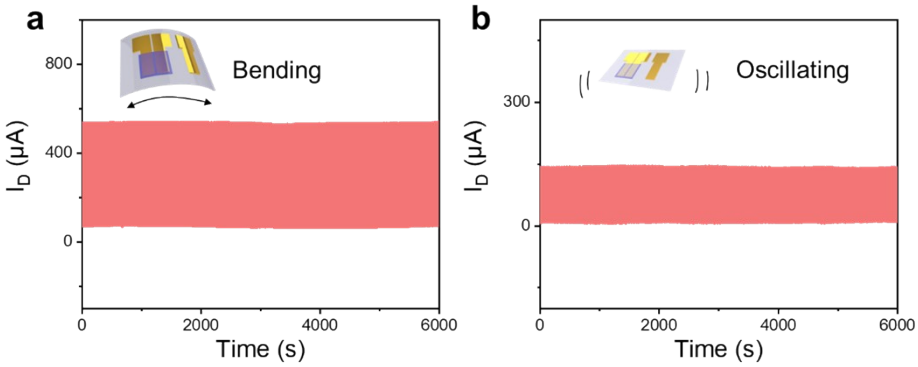
Supplementary Fig. 6 (a) Transfer curve and transconductance of the OECT based on Nafion-included .PEDOT:PSS. (b) Output curves of the Nafion-PEDOT:PSS OECT at the gate voltages of 0, 0.4, and 0.8 V.



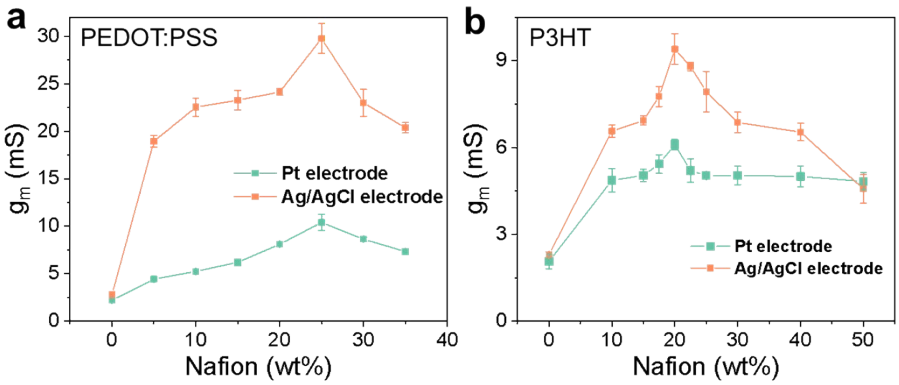
Supplementary Fig.7 Optical photographs of pristine PEDOT:PSS (Nafion = 0%) and the Nafion-PEDOT:PSS films at Nafion contents of 25 wt% and 50 wt%.



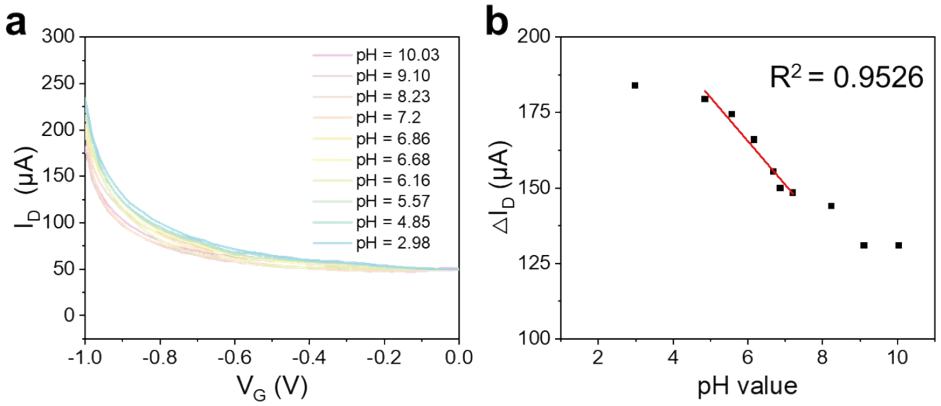
Supplementary Fig. 8 (a) Output curves of the OECT based on pristine PEDOT:PSS at the gate voltage ranging from 0 to 2.0 V. (b) Output curves of the OECT based on pristine P3HT at the gate voltage ranging from -1.0 to 0 V.



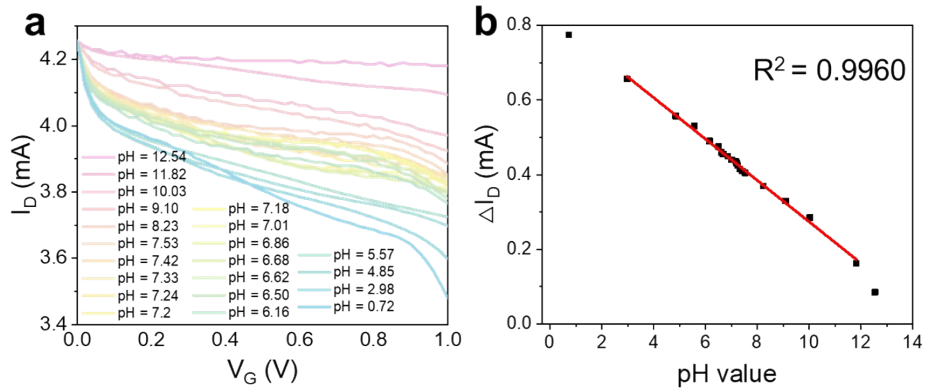
Supplementary Fig. 9 Mechanical stabilities of the OECT based on Nafion-included PEDOT:PSS film in (a) bending and (b) mechanical oscillating. The gate voltage was varied within the range of 0 to 100 mV. Note that the amount of electrolyte was less to avoid spilling during the oscillating test.



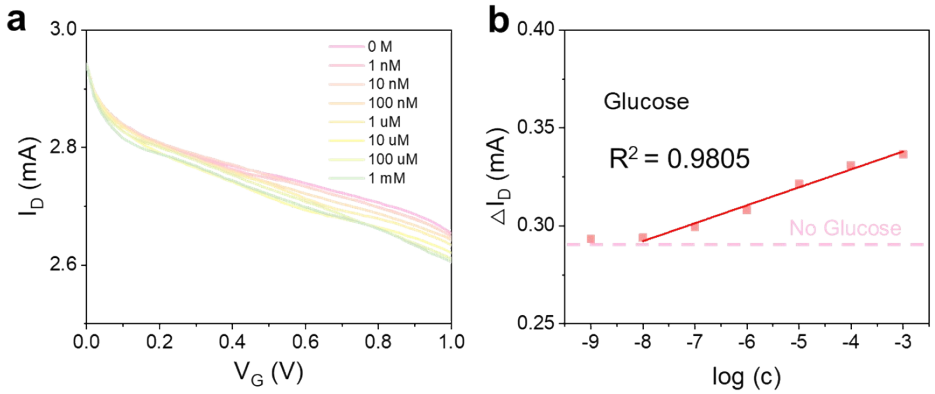
Supplementary Fig. 10 Transconductance of the OECTs based on (a) Nafion-included PEDOT:PSS (Nafion-PEDOT:PSS) and Nafion-included P3HT (Nafion-P3HT) at different Nafion contents. The Ag/AgCl (orange line) and Pt (green line) were used as the gate electrodes.



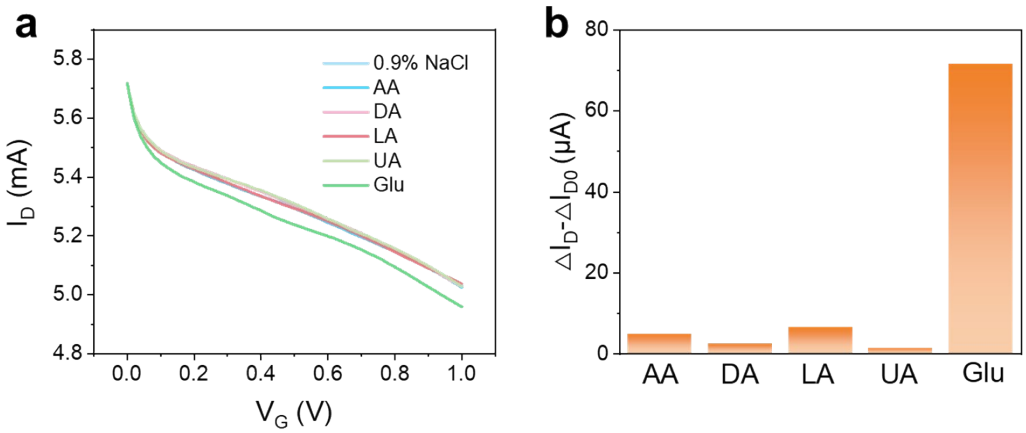
Supplementary Fig. 11 (a) Transfer curves of the pristine P3HT OECT at different pH values. Every pH was tested for four times and the results were highly consistent. (b) The change of ΔID versus the pH value.



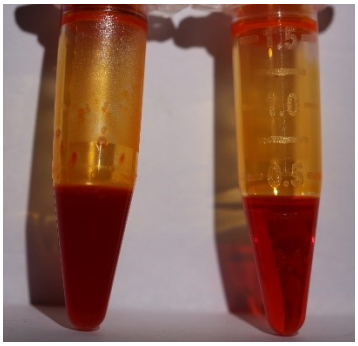
Supplementary Fig. 12 (a) Transfer curves (VD = 0.2 V) of the Nafion-PEDOT:PSS OECT at different pH values. Every pH was tested for four times and the results were highly consistent. (b) The change of ΔID versus the pH value. The pH of 0.72 was excluded in the linear fitting.



Supplementary Fig. 13 (a) Transfer curves of the pristine PEDOT:PSS OECT at different glucose concentrations. The glucose was dissolved in 0.9% NaCl solution. Every concentration was tested for four times and the results were highly consistent. (b) The change of $^{\Delta I_D}$ versus the concentration of glucose.



Supplementary Fig. 14 (a) Selectivity of the OECT based on Nafion-included PEDOT:PSS. (b) The change in $^{\Delta I_D}$ of the OECT during sensing of ascorbic acid (AA), dopamine (DA), lactic acid (LA), uric acid (UA), and glucose (Glu). $^{\Delta I_{D_0}}$ was defined as the change in ^{I_D} of the control sample (0.9% NaCl).



Supplementary Fig. 15 Photograph of the Nafion-P3HT mixture (left) and pristine P3HT solution (right).