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Supplementary Materials for

Strong adhesion of wet conducting polymers on diverse substrates

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Published 20 March 2020, *Sci. Adv.* **6**, eaay5394 (2020)
DOI: 10.1126/sciadv.aay5394

The PDF file includes:

Other Supplementary Material for this manuscript includes the following:

(available at advances.sciencemag.org/cgi/content/full/6/12/eaay5394/DC1)

Movie S1 (.mp4 format). Adhesion stability of wet PEDOT:PSS under cyclic bending deformations.

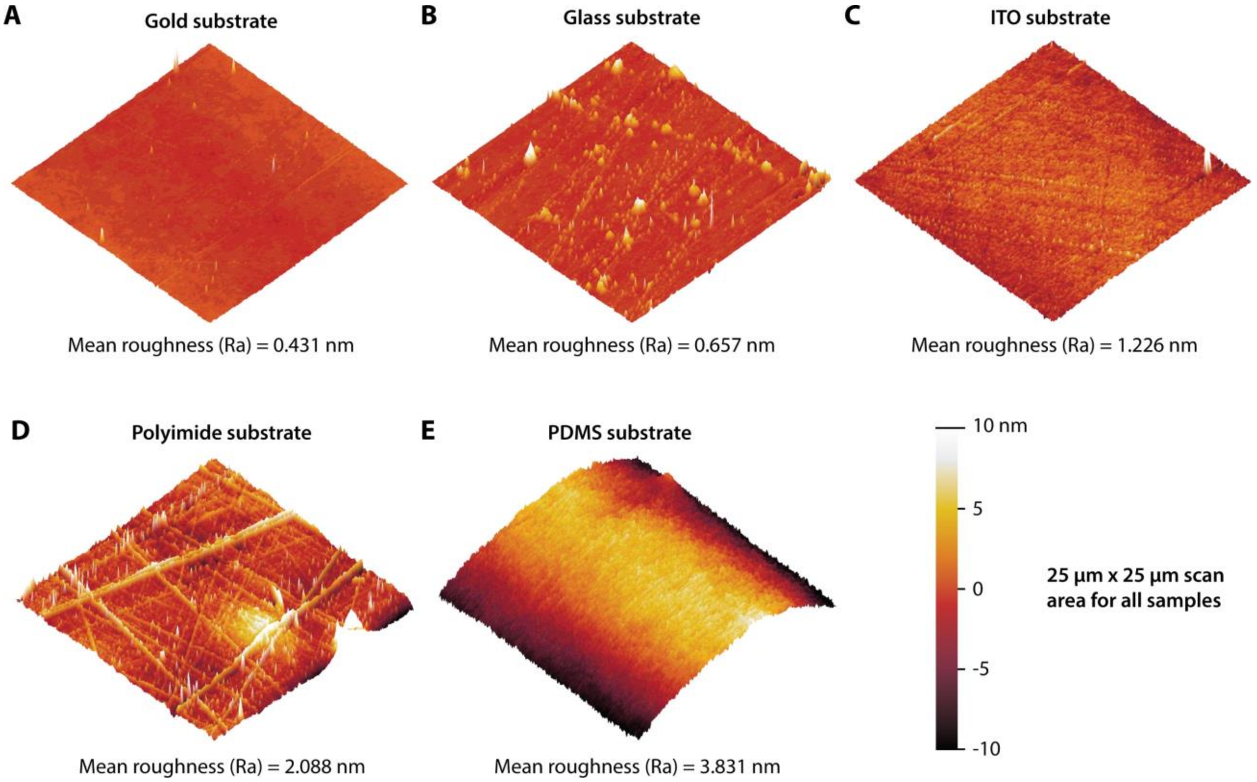

Fig. S1. Surface roughness of diverse substrates. (**A-E**) AFM characterizations of diverse substrates used for adhesion tests with small mean surface roughness (Ra) as 0.431 nm for gold (A), 0.657 nm for glass (B), 1.226 nm for ITO-glass (C), 2.088 nm for polyimide (D), and 3.831 nm for PDMS (E).

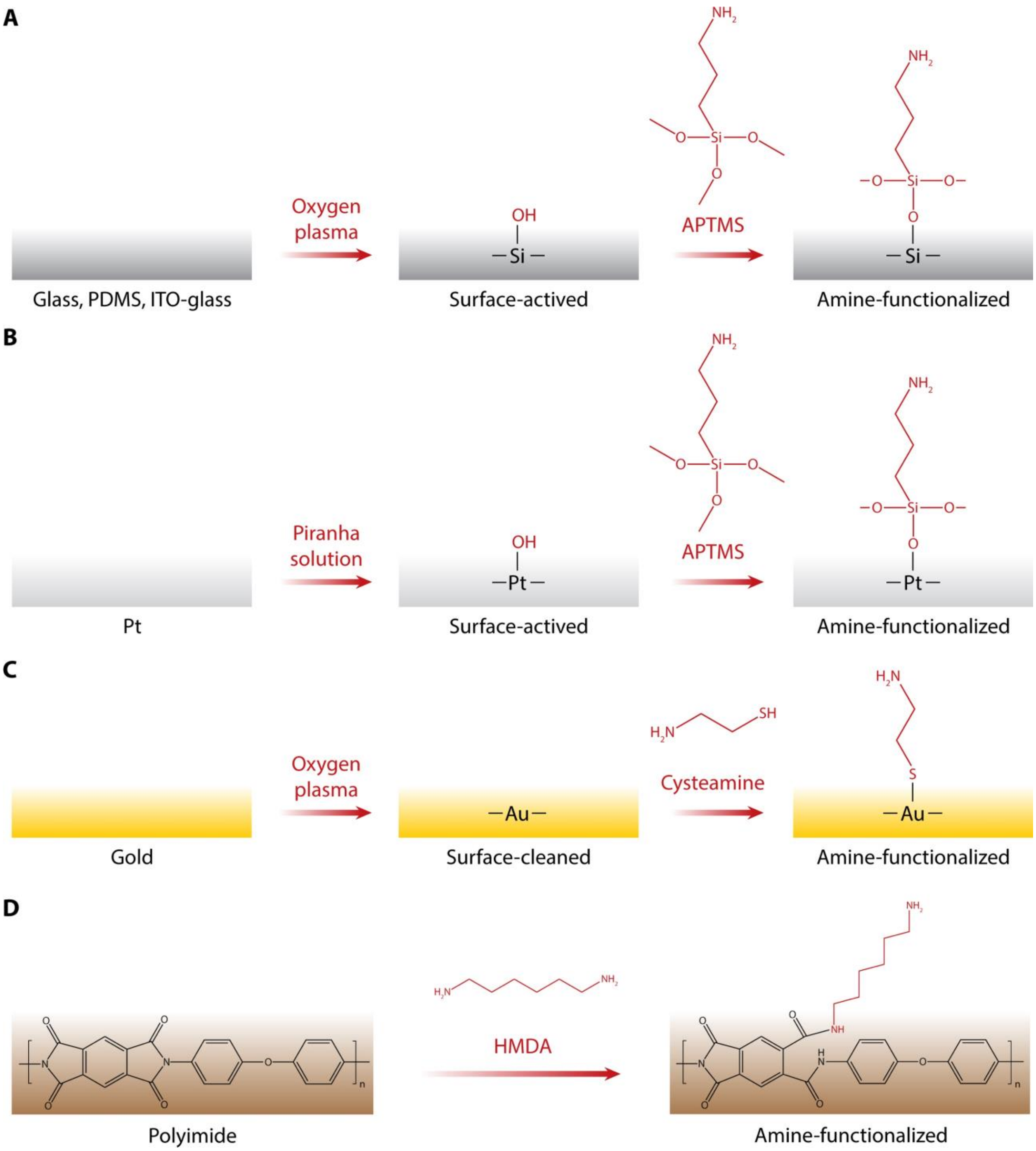

Fig. S2. Amine functionalization of diverse substrates. (**A-D**) Schematic illustrations for primary amine functionalization for glass (A), PDMS, ITO-glass, Pt (B), gold (C), and polyimide (D).

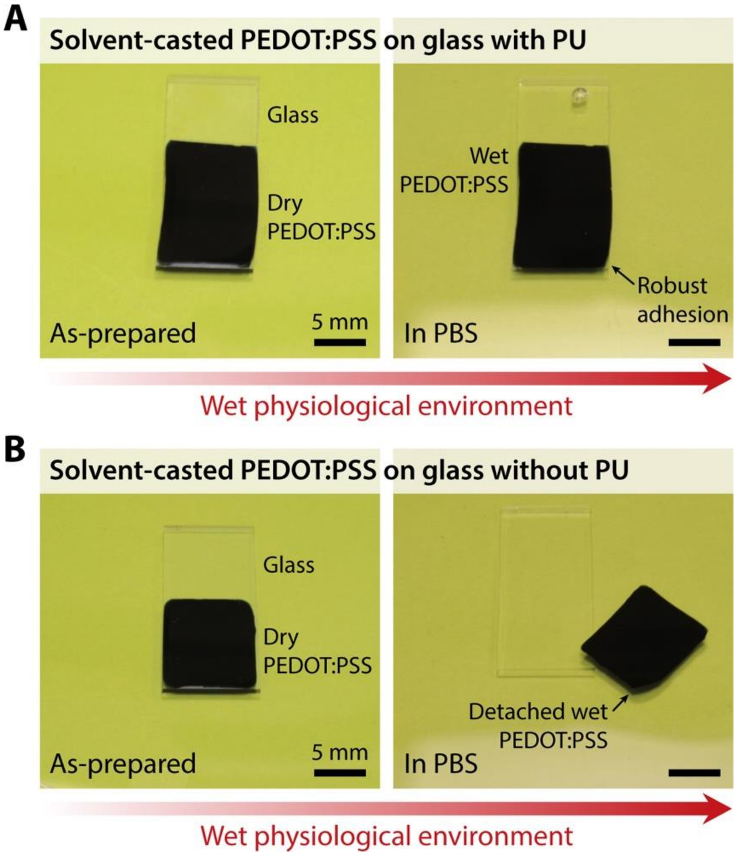

Fig. S3. Strong adhesion of wet conducting polymer by PU adhesive layer. (**A**,**B**) Images of a solvent-casted wet PEDOT:PSS on glass substrates with (A) and without (B) the PU adhesive layer in PBS. Conducting polymers with 50 µm thickness and PU adhesive layers with 60 nm thickness were used for all experiments. Photo Credit: Hyunwoo Yuk, MIT.

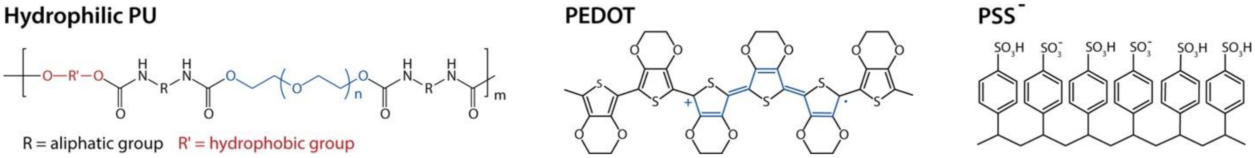

Fig. S4. Chemical structures of hydrophilic PU, PEDOT, and PSS.

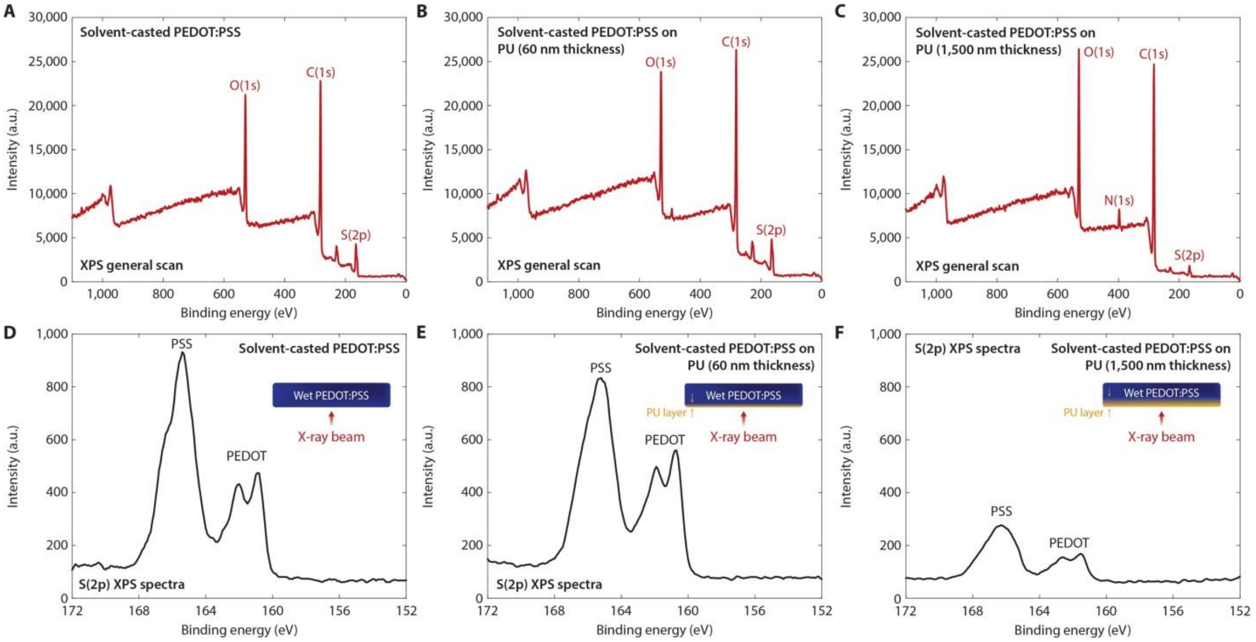

Fig. S5. XPS spectra of solvent-casted PEDOT:PSS with varying thickness of PU adhesive layer. (**A-C**) XPS general scan spectra for PEDOT:PSS without the PU adhesive layer (A), with the 60 nm PU adhesive layer (B), and the 1,500 nm PU adhesive layer (C). (**D-F**) XPS S(2p) (sulfur) spectra for PEDOT:PSS without the PU adhesive layer (D), with the 60 nm PU adhesive layer (E), and the 1,500 nm PU adhesive layer (F). Conducting polymers with 100 µm thickness were used for all experiments.

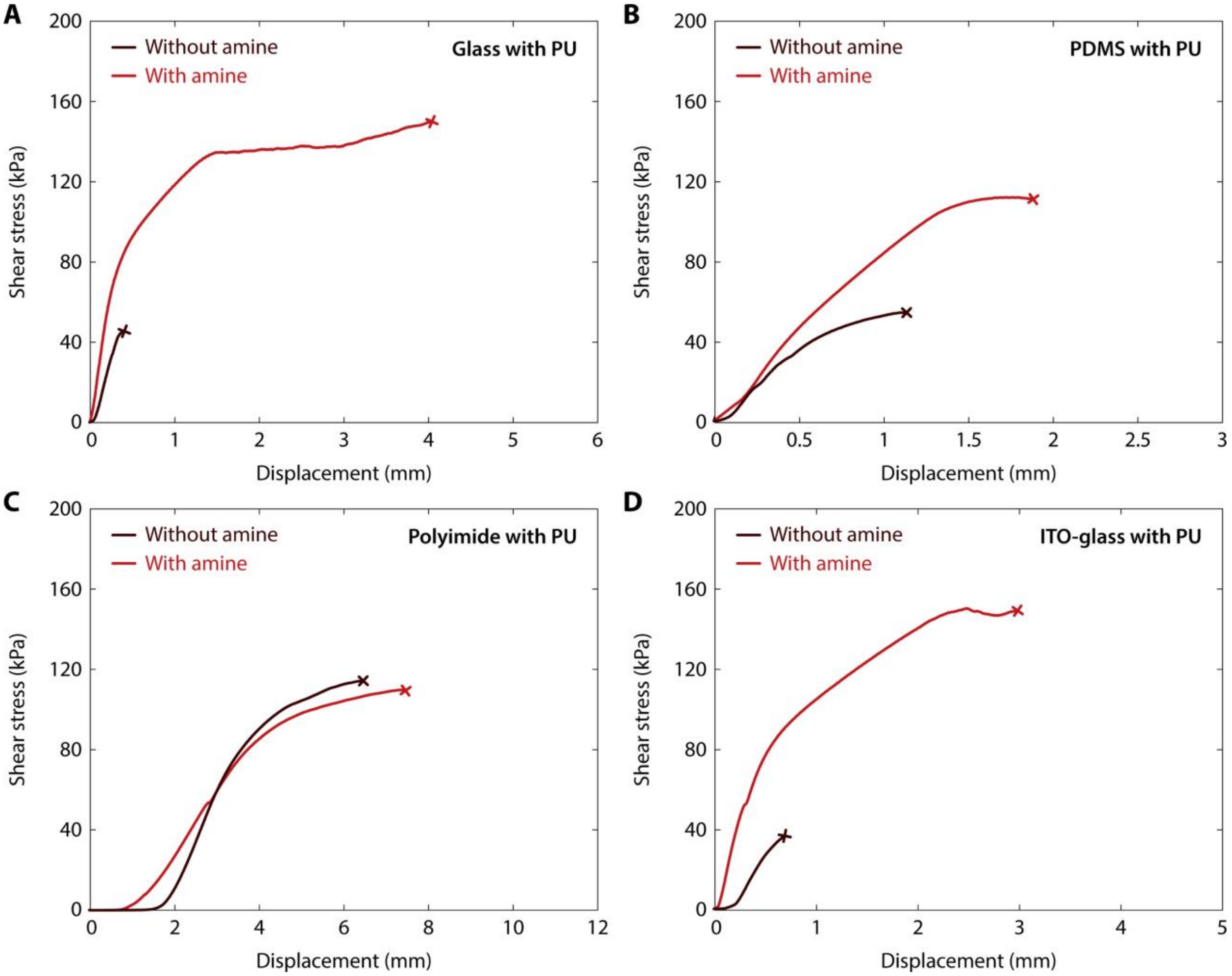

Fig. S6. Amine functionalization effect on lap-shear strength for diverse substrates. (**A-D**) Displacement vs. shear stress curves of lab-shear tests of wet PEDOT:PSS on PU-coated glass (A), PDMS (B), polyimide (C), and ITO-glass (D) substrates with and without primary amine functionalization. Conducting polymers with 10 µm thickness and PU adhesive layers with 60 nm thickness were used for all experiments.

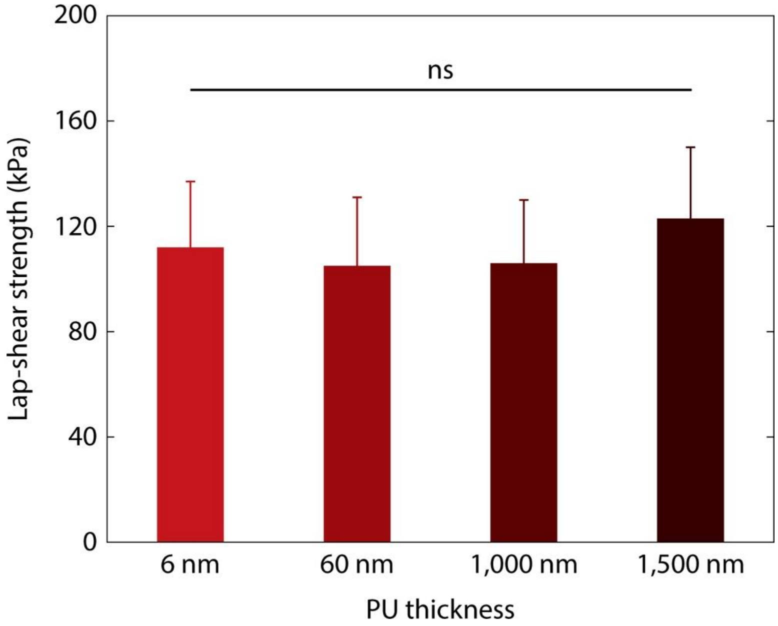

Fig. S7. PU adhesive layer thickness effect on lap-shear strength for amine-functionalized glass substrate. Conducting polymers with 10 µm thickness were used for all experiments. Values represent mean and the error bars represent 95% CI of the measured values (*n* = 5). Statistical significance and *P* values are determined by one-way ANOVA and Tukey's multiple comparison test. ns, not significant.

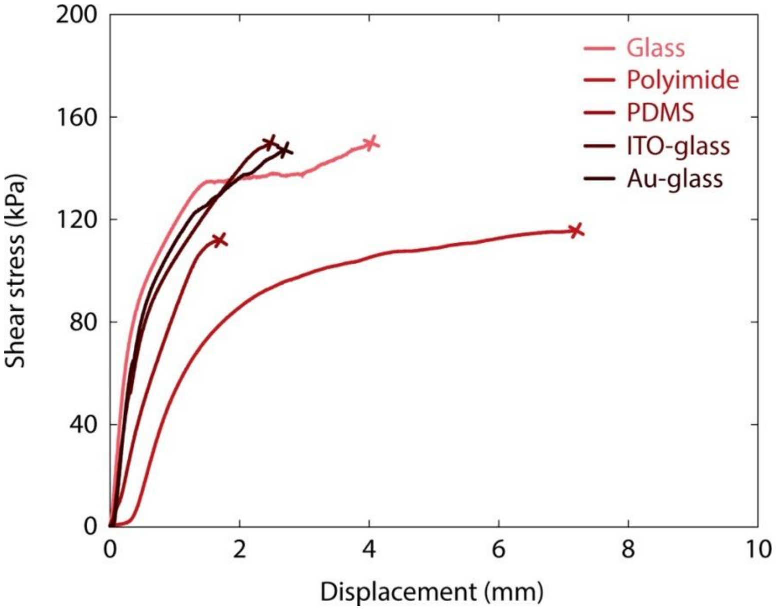

Fig. S8. Lap-shear test curves for diverse substrates. Conducting polymers with 10 µm thickness and PU adhesive layers with 60 nm thickness were used for all experiments.

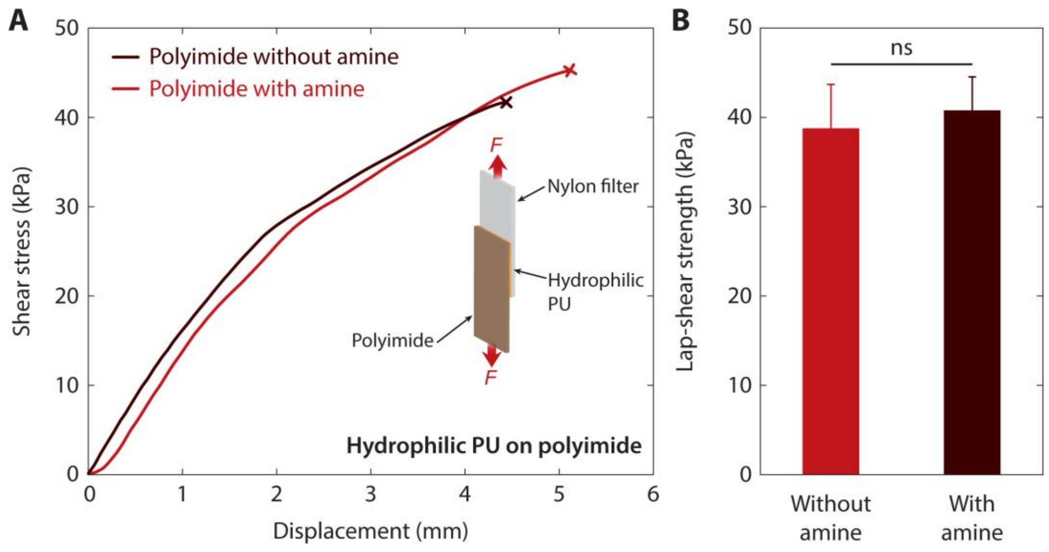

Fig. S9. Adhesion of hydrophilic PU to polyimide. (**A**) Displacement vs. shear stress curves from lab-shear tests of hydrophilic PU on polyimide substrates with and without primary amine functionalization. (**B**) Lap-shear strength of hydrophilic PU on polyimide substrates with and without primary amine functionalization. Hydrophilic PU layers with 10 μm thickness were used for all experiments. Values in **B** represent mean and the error bars represent 95% CI of the measured values (*n* = 5). *P* values are determined by Student's *t* test. ns, not significant.

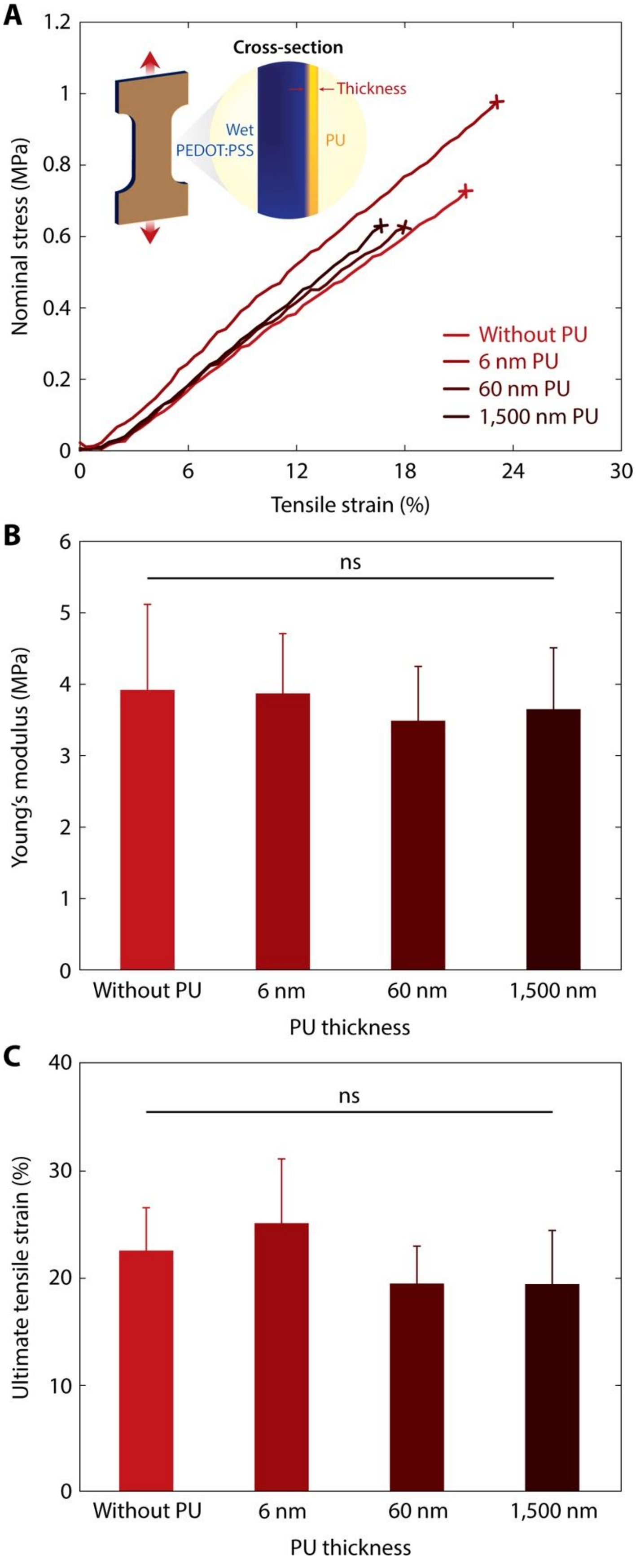
A
Cross-section
Thickness
Wet
PEDOT:PSS
PU
Nominal stress (MPa)
1.2
1
0.8
0.6
0.4
0.2
0
0
6
12
18
24
30
Tensile strain (%)
Without PU
6 nm PU
60 nm PU
1,500 nm PU
B
ns
Young's modulus (MPa)
6
5
4
3
2
1
0
Without PU
6 nm
60 nm
1,500 nm
PU thickness
C
ns
Ultimate tensile strain (%)
40
30
20
10
0
Without PU
6 nm
60 nm
1,500 nm
PU thickness

Fig. S10. Mechanical properties of wet PEDOT:PSS with varying PU adhesive layer thickness. (**A**) Nominal stress vs. tensile strain curves for wet PEDOT:PSS with the varying PU adhesive layer thickness. (**B**) Young's moduli of wet PEDOT:PSS with the varying PU adhesive layer thickness. (**C**) Ultimate tensile strain of wet PEDOT:PSS with the varying PU adhesive layer thickness. Values in **B**,**C** represent mean and the error bars represent 95% CI of the measured values ($n = 5$). Conducting polymers with 100 µm thickness were used for all experiments. Statistical significance and P values are determined by one-way ANOVA and Tukey's multiple comparison test. ns, not significant.

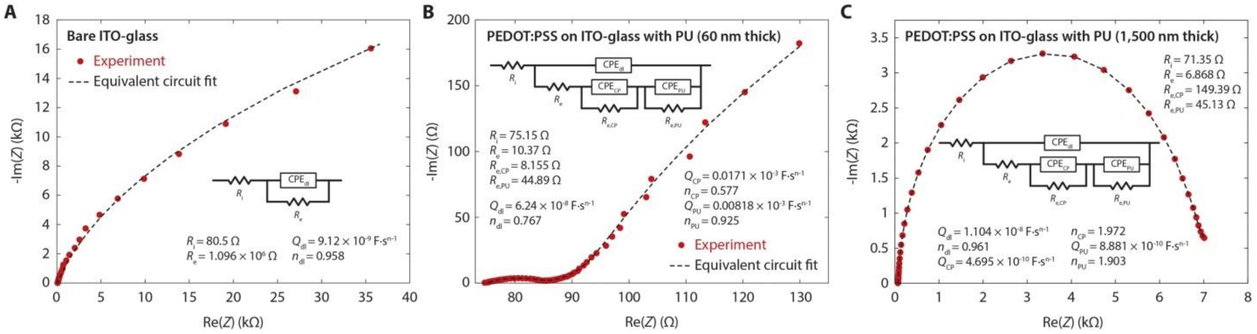

Fig. S11. Nyquist plots for EIS measurements of adhesive interface by varying PU adhesive layer thickness. (**A-C**) Nyquist plot obtained from the EIS measurement of a bare ITO-glass electrode (A), a wet PEDOT:PSS (10 µm thickness) on an amine-functionalized ITO-glass electrode with the PU adhesive layer (60 nm thickness) (B), and a wet PEDOT:PSS (10 µm thickness) on an amine-functionalized ITO-glass electrode with the PU adhesive layer (1,500 nm thickness) (C) overlaid with the plot predicted from the corresponding equivalent circuit models. In the equivalent circuit models, R_i represents ionic resistance for the electrolyte, R_e represents electronic resistance of the ITO-glass electrode, $R_{e,CP}$ represents electronic resistance of the wet conducting polymer, $R_{e,PU}$ represents electronic resistance of the PU adhesive layer, CPE$_{dl}$ represents the double-layer capacitive phase element (CPE) for the ITO-glass electrode, CPE$_{CP}$ represents CPE of the wet conducting polymer, and CPE$_{PU}$ represents CPE of the PU adhesive layer. CPE is used to account inhomogeneous or imperfect capacitance, and are represented by the parameters Q and n where Q represents the peudocapacitance value and n represents the deviation from ideal capacitive behavior. The true capacitance C can be calculated from these parameters by using the relationship $C = Q\omega_{max}^{n-1}$, where ω_{max} is the frequency at which the imaginary component reaches a maximum.

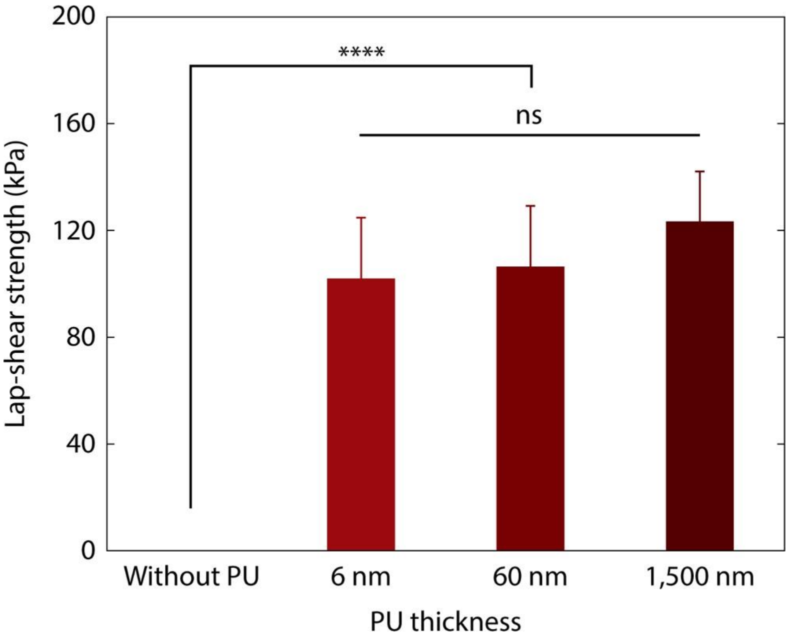

Fig. S12. PU adhesive layer thickness effect on lap-shear strength for amine-functionalized ITO-glass substrates. Conducting polymers with 50 µm thickness were used for all experiments. Values represent mean and the error bars represent 95% CI of the measured values (*n* = 5). Statistical significance and *P* values are determined by one-way ANOVA and Tukey's multiple comparison test. ****$P \leq 0.0001$; ns, not significant.

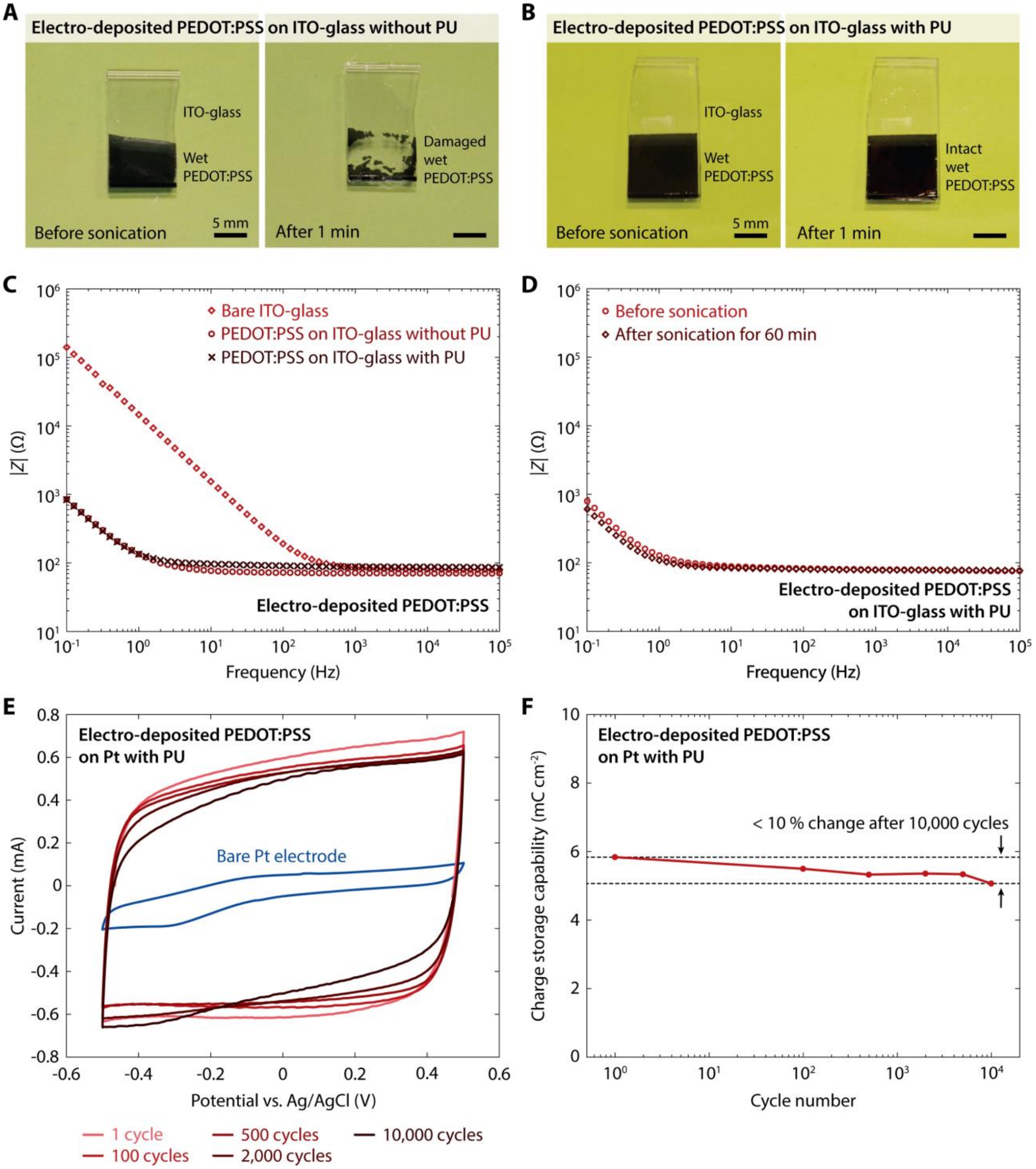
A
Electro-deposited PEDOT:PSS on ITO-glass without PU
ITO-glass
Wet PEDOT:PSS
Before sonication
5 mm
Damaged wet PEDOT:PSS
After 1 min
B
Electro-deposited PEDOT:PSS on ITO-glass with PU
ITO-glass
Wet PEDOT:PSS
Before sonication
5 mm
Intact wet PEDOT:PSS
After 1 min
C
Bare ITO-glass
PEDOT:PSS on ITO-glass without PU
PEDOT:PSS on ITO-glass with PU
Electro-deposited PEDOT:PSS
|Z| (Ω)
Frequency (Hz)
D
Before sonication
After sonication for 60 min
Electro-deposited PEDOT:PSS on ITO-glass with PU
|Z| (Ω)
Frequency (Hz)
E
Electro-deposited PEDOT:PSS on Pt with PU
Bare Pt electrode
Current (mA)
Potential vs. Ag/AgCl (V)
1 cycle
100 cycles
500 cycles
2,000 cycles
10,000 cycles
F
Electro-deposited PEDOT:PSS on Pt with PU
< 10 % change after 10,000 cycles
Charge storage capability (mC cm⁻²)
Cycle number

Fig. S13. Adhesion stability of electrodeposited wet conducting polymer. (**A**) Images of an electro-deposited wet PEDOT:PSS on a ITO-glass substrate without the PU adhesive layer before and after ultrasonication for 1 min. Photo Credit: Hyunwoo Yuk, MIT. (**B**) Images of an electro-deposited wet PEDOT:PSS on an amine-functionalized ITO-glass substrate with the PU adhesive layer before and after ultrasonication for 1 min. Photo Credit: Hyunwoo Yuk, MIT. (**C**) EIS curves for a ITO-glass electrode without the PU adhesive layer, an electro-deposited wet PEDOT:PSS on a ITO-glass electrode without the PU adhesive layer, and an electro-deposited wet PEDOT:PSS on an amine-functionalized ITO-glass electrode with the PU adhesive layer. (**D**) EIS curves for an electro-deposited wet PEDOT:PSS on an amine-functionalized ITO-glass electrode with the PU adhesive layer before and after ultrasonication for 60 min in PBS. (**E**) Long-term CV curves for an electro-deposited wet PEDOT:PSS on an amine-functionalized Pt electrode with the PU adhesive layer in PBS. (**F**) Measured CSC vs. CV cycle number for an electro-deposited wet PEDOT:PSS on an amine-functionalized Pt electrode with the PU adhesive layer in PBS. Conducting polymers with 500 nm thickness and PU adhesive layers with 60 nm thickness were used for all experiments.

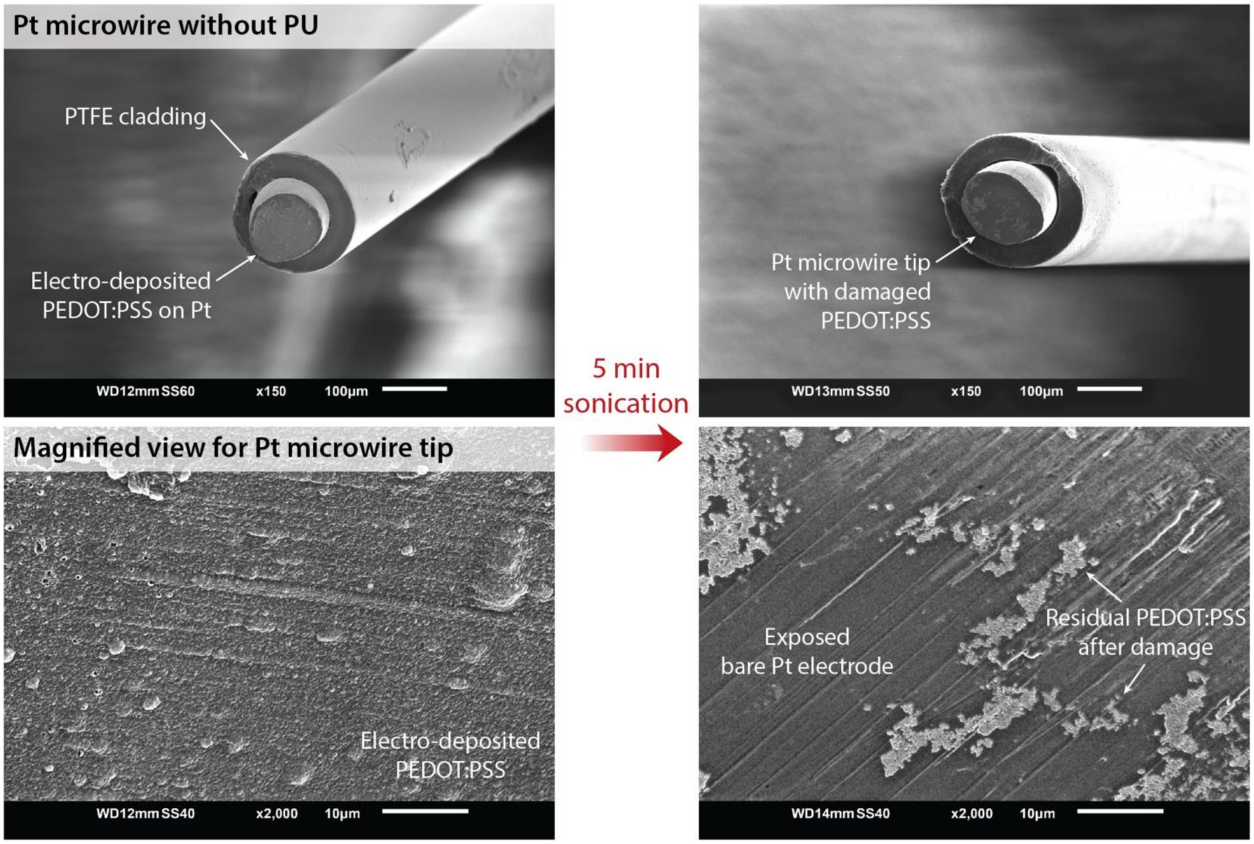

Fig. S14. SEM images of electrodeposited PEDOT:PSS on Pt microwire electrode without PU adhesive layer. Electro-deposited PEDOT:PSS on a Pt microwire electrode without the PU adhesive layer exhibits significant damage and delamination from the Pt electrode after ultrasonication for 5 min in PBS. Conducting polymers with 500 nm thickness and PU adhesive layers with 60 nm thickness were used for all experiments.

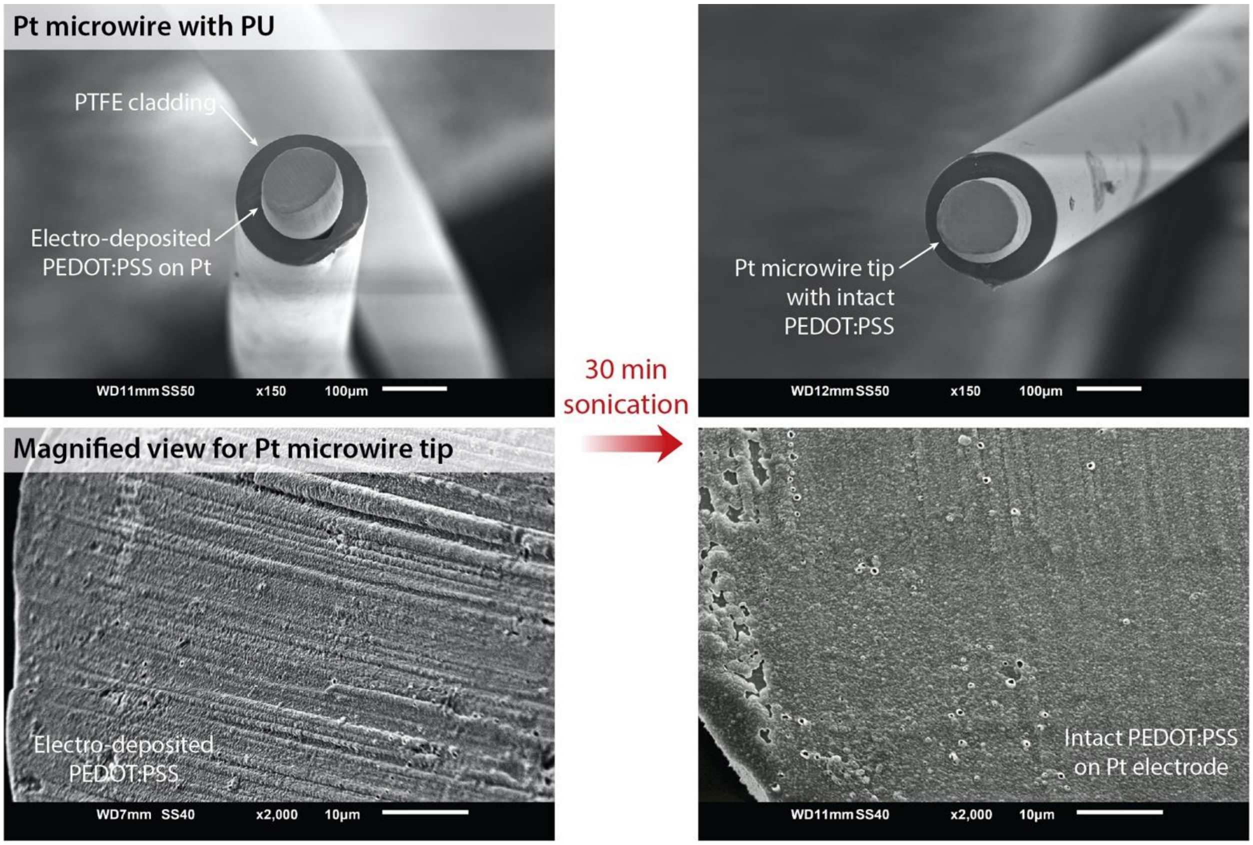

Fig. S15. SEM images of electrodeposited PEDOT:PSS on amine-functionalized Pt microwire electrode with PU adhesive layer. Electro-deposited PEDOT:PSS on an amine-functionalized Pt microwire electrode with the PU adhesive layer remains intact on the Pt electrode after ultrasonication for 30 min in PBS. Conducting polymers with 500 nm thickness and PU adhesive layers with 60 nm thickness were used for all experiments.

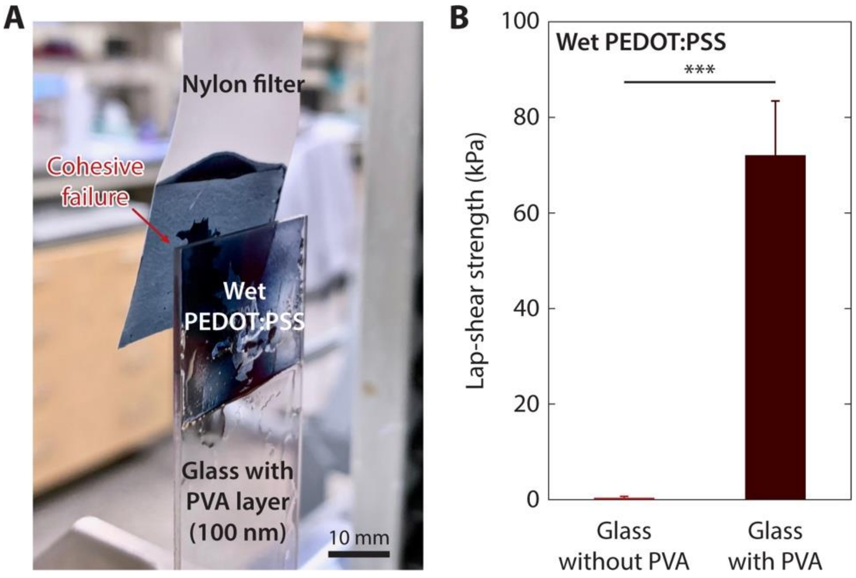

Fig. S16. Strong adhesion of wet conducting polymer by PVA adhesive layer. (**A**) Image of cohesive failure during lap-shear test for a wet PEDOT:PSS on a glass substrate with the PVA adhesive layer. Photo Credit: Hyunwoo Yuk, MIT. (**B**) Lap-shear strength of wet PEDOT:PSS on glass substrates with and without the PVA adhesive layer. Conducting polymers with 10 μm thickness and PVA adhesive layers with 100 nm thickness were used for all experiments. Values in **B** represent mean and the error bars represent 95% CI of the measured values ($n = 5$). P values are determined by Student's t test. ***$P \leq 0.001$.

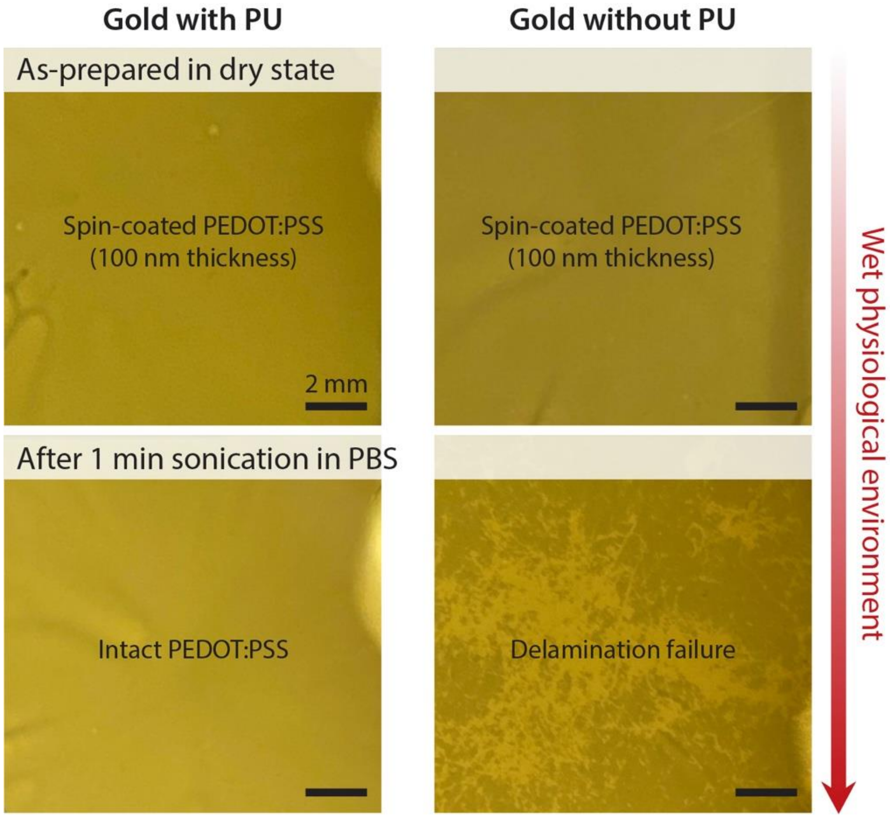

Fig. S17. Adhesion of thin spin-coated conducting polymers in wet physiological environment. Images of spin-coated PEDOT:PSS (~ 100 nm thickness) on gold substrates with and without the PU adhesive layer (60 nm thickness) after ultrasonication for 1 min in PBS. Photo Credit: Hyunwoo Yuk, MIT.

Legend for Supplementary Movie

Movie S1. Adhesion stability of wet PEDOT:PSS under cyclic bending deformations.