

Supporting Information

Inkjet-Printed PEDOT:PSS-based Stretchable Conductor for Wearable Health Monitoring Device Applications

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S1. UV-Vis spectra of inkjet-printed PEDOT:PSS thin film

Figure S1 presents the UV-vis spectra of inkjet-printed PEDOT:PSS thin film with 5, 10 and 20 print passes. The spectra show the transmittance decreases gradually for increasing number of printing passes. The result agree with the photo in Figure 2b where the PEDOT:PSS pattern with 20 print passes is the least transparent.

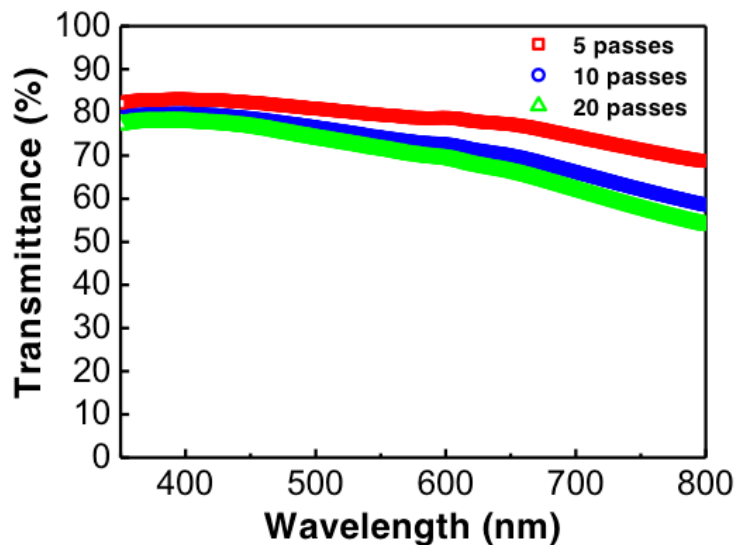


Figure S1. UV-Vis spectra of printed PEDOT:PSS thin film with different numbers of printing passes.

S2. Electrical characterization of the printed PEDOT:PSS/PEO thin film over time

Figure S2 shows the sheet resistance of inkjet-printed PEDOT:PSS thin film with 5 wt% EG and various amount of PEO measured again after 1.5 months. Due to the hygroscopic nature of PSS that absorbs moisture from surrounding environment, the sheet resistance of the PEDOT:PSS/PEO thin film gradually increases over time. The red trace was the data measured right after the thin film was fabricated and the blue trace shows the sheet resistance of the same sample after 1.5 months. The sheet resistance increased from 58 Ω/\square to 91 Ω/\square , 84 Ω/\square to 131 Ω/\square , and 205 Ω/\square to 317 Ω/\square , for printed thin films of pristine PEDOT:PSS, and PEDOT:PSS with 50 wt% and 66 wt% of PEO, respectively.

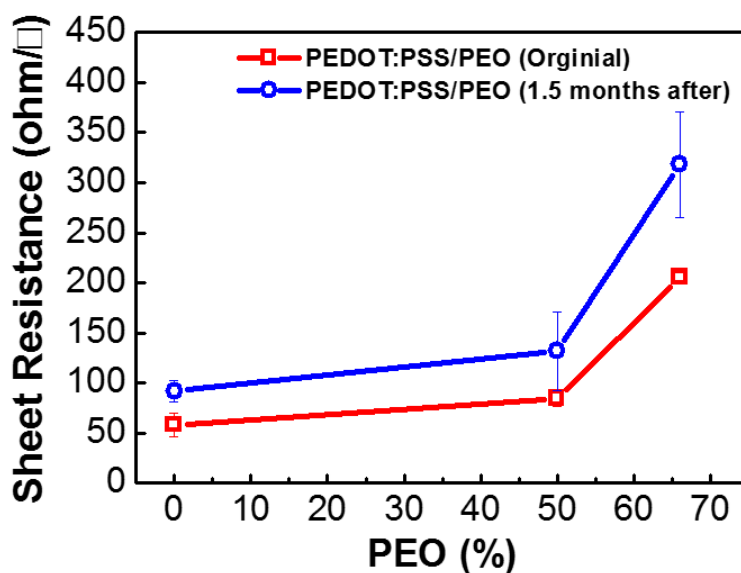


Figure S2. Comparison of sheet resistance of inkjet-printed PEDOT:PSS thin film with 5wt% EG and various amount of PEO measured again after 1.5 months.

S3. Electrical Properties of PEDOT:PSS/PEO thin film when stretched along the transverse direction

Figure S3 shows the relative change in resistance plotted as a function of tensile strain for PEDOT:PSS thin film with 5 wt% EG and various amount of PEO when stretched along the transverse direction. In this experiment, the printed pattern is a straight line with $\sim 450\ \mu\text{m}$ width and $\sim 1.5\ \text{cm}$ length. For the results presented in Figure 4f, when stretched along the longitudinal direction by 50%, the devices experience 41% and 18% increase in sheet resistance for samples with 50 wt% and 66 wt% of PEO additives, respectively. When stretched along the transverse direction, the samples exhibit 61% and 32% increase in sheet resistance for samples with 50 wt% and 66 wt% of PEO additives, respectively. The increase in sheet resistance may be because the PEDOT grains tend to be stacked along the printing direction (longitudinal direction) during printing. As a result, the stretching in the transverse direction may have more impact on the grains sliding than stretching along the printing direction.

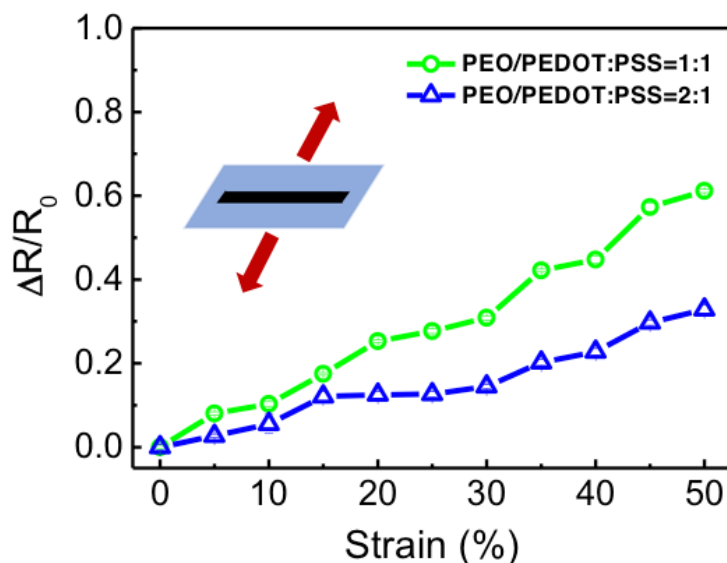


Figure S3. Relatively change in resistance as a function of tensile strain when the sample is stretched along the transverse direction.

S4. Rheological properties of the PEDOT:PSS-based conducting polymer ink

Figure S4 shows the viscosity as a function of shear rate for the PEDOT:PSS-based conducting polymer inks with various types of additives. The viscosity was measured (Rheometer, TA Instruments AR G2) at the shear rate from 1 to 1000 s^{-1} . The results show typical non-Newtonian behavior of the PEDOT:PSS solution with decreasing viscosity for increasing shear rates.^{1,2} For the pristine PEDOT:PSS ink, the viscosity is 0.1863 Pa-s at a shear rate 1 s^{-1} and the viscosity drop to 0.02327 Pa-s at a shear rate 1000 s^{-1} . The PEDOT:PSS solution mixed with 5wt% of EG, DMSO DMF and glycerol all exhibit similar behavior compare with pristine PEDOT:PSS ink. Moreover, the PEDOT:PSS mixed with 5 wt% EG and 50 wt% PEO (PEO/PEDOT:PSS = 1:1) or 5 wt% EG and 66 wt% PEO (PEO/PEDOT:PSS = 2:1) exhibit higher viscosity compared to the one without PEO under all shear rates tested. The result may be attributed to the high molecular weight of PEO. At a shear rate of 1000 s^{-1} , the viscosity of the PEO/PEDOT:PSS mixture exhibit a fairly low viscosity of 0.041 and 0.039 Pa-s for 50 wt% of PEO and 66 wt% of PEO, respectively. Our ink exhibit similar rheological behavior compared to the viscosity of PEDOT:PSS solution reported previously in the literature.^{3,4}

Typically, the inkjet printer has a shear rate greater than 1000 s^{-1} and the printability of the ink is defined by the Ohnesorge number whose value typically lies between 0.1 to 10.⁵⁻⁸ In supporting information Table S2, we have systematically measured and provided the detailed information about the ink properties including the ink density, surface tension and the calculated Ohnesorge number. The inkjet printer used in this experiment (The GIX Microplotter, Sonoplot Inc.) is capable of printing inks with viscosity up to 0.450 Pa-s and the calculated Ohnesorge number for all types of PEDOT:PSS solution fall in the range between 0.1 to 10. Therefore, all types of PEDOT:PSS solution in this work are printable by inkjet printing process.

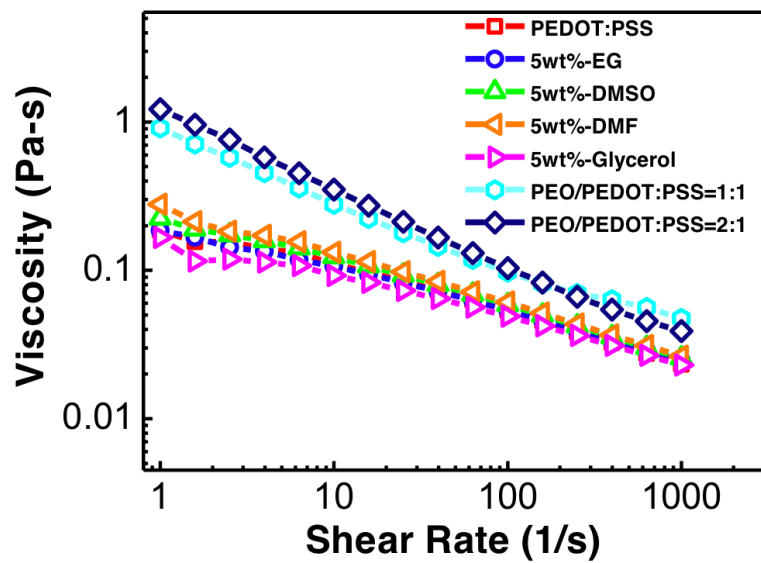


Figure S4. Measured viscosity as a function of shear rate for the PEDOT:PSS-based conducting polymer inks with various types of additives.

Table S1. Comparison of PEDOT:PSS based conductor with various kinds of treatment and fabrication methods

Materials	Fabrication Method	Stretchability	Conductivity / Sheet resistance
PEDOT:PSS with H ₂ SO ₄ post-treatment ⁹	spin-cast	N/A	4380 S cm ⁻¹
PEDOT:PSS hydrogel with H ₂ SO ₄ treatment ¹⁰	blade cast	N/A	880 S cm ⁻¹
H ₂ SO ₄ /DMSO-treated PEDOT:PSS ¹¹	spin coating with layer-by-layer (LBL) process	N/A	3026 S cm ⁻¹
PEDOT:PSS doped with benzenesulfonic acid ¹²	spin coating	N/A	1996 S cm ⁻¹
PEDOT:PSS with H ₂ SO ₄ treatment ¹³	spin-cast	N/A	2938 S cm ⁻¹
PEDOT:PSS with PEG and EG ¹⁴	spin coating	~ 25%	101 S cm ⁻¹
PEDOT:PSS with PVA and EG ¹⁴	spin coating	47%	172 S cm ⁻¹
PEDOT:PSS with PDMS EG and Triton-X 100 ¹⁵	molding	82%	20 Ω/□
PEDOT:PSS with poly(acrylic acid) (PAA) ¹⁶	spin coating	40%	125 S cm ⁻¹
PEDOT:PSS with PU ¹⁷	drop cast	700%	79 S cm ⁻¹

Table S2. Summary of ink formulations and measured properties

Ink Composition	Density (kg/m ³)	Surface Tension (J/m ²)	Nozzle Diameter (m)	Viscosity (Pa-s) at shear rate = 1000 (1/s)	Ohnesorge Nunmer
PEDOT:PSS	1020.00	0.0725	0.0004	0.02327	0.135
PEDOT:PSS + 5wt% EG	1040.00	0.0706	0.0004	0.02456	0.143
PEDOT:PSS + 5wt% DMSO	1065.33	0.0687	0.0004	0.0256	0.149
PEDOT:PSS + 5wt% DMF	1002.67	0.0653	0.0004	0.02636	0.162
PEDOT:PSS + 5wt% Glycerol	1091.67	0.0760	0.0004	0.02294	0.125
PEO/PEDOT:PSS = 1:1	1012.00	0.0540	0.0004	0.04736	0.320
PEO/PEDOT:PSS = 2:1	1030.38	0.0543	0.0004	0.03894	0.260

Supplementary References

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