Electronic Supplementary Information

Direct Ink Writing of High Conductive PEDOT:PSS Dispersion with the Engineered Conformation and Electronic Structure for Printed Electronic Circuits

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EDOT conversion for the prepared dispersions

The gravimetric method was employed to determine EDOT conversion. A definit amount of the dispersion (a) was sampled at the end of the polymerization process. The sample was first dried at room temperature for 12 hours and then for 2 hours in a vacuum oven at 60°C. The resulting solid was washed three times with THF to remove any unreacted EDOT monomer. After extra drying the vacuum oven, the solid was weighed (b). The conversion (X) was calculated using Eq (S1):

$$X(\%) = \frac{b - [a \times (W_{PSS} + W_{APS})]}{a \times W_{EDOT}} \times 100 \quad \text{(S1)}$$

where W_{APS}, W_{EDOT}, and W_{PSS} represent the weight percentages of APS, EDOT, and PSS, respectively. W_{APS} varied in each polymerization recipe, while W_{EDOT} and W_{PSS} were constant at 0.375% and 0.925%, respectively. Conversion for each sample has been given in Table S1.

Table S1. EDOT Conversion for the prepared dispersions under different conditions

Sample	*X* (%)
$P_{1.5}$-10	96.9
$P_{1.5}$-25	96.4
$P_{1.5}$-40	95.9
$P_{2.25}$-10	98.1
$P_{2.25}$-25	97.8
$P_{2.25}$-40	97.3
P_3-10	98.6
P_3-25	98.55
P_3-40	98.4

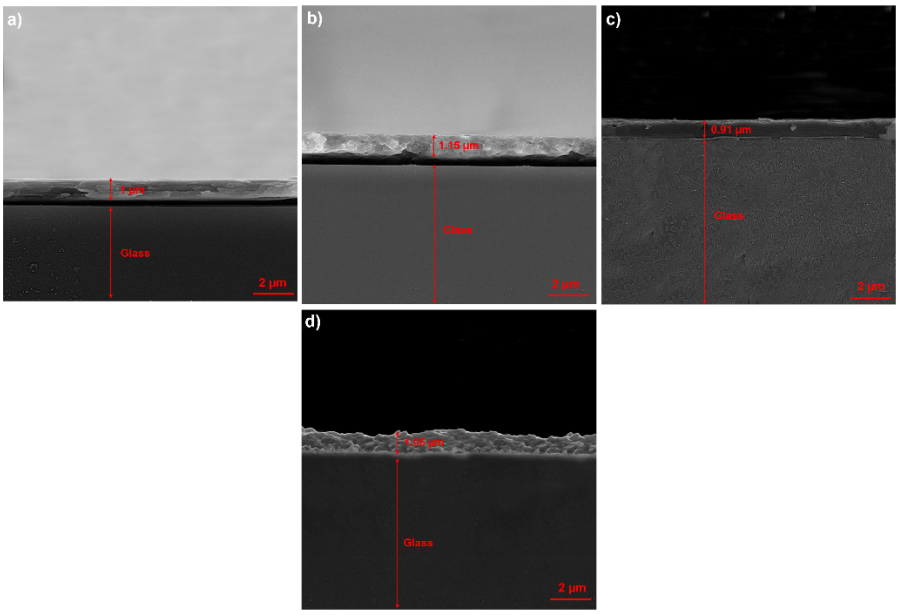

Fig. S1. FE-SEM image of cross-section of a) $P_{2.25}$-10, b) $P_{2.25}$-25, c) $P_{1.5}$-40 and d) $P_{2.25}$-40 films deposited on the treated glass

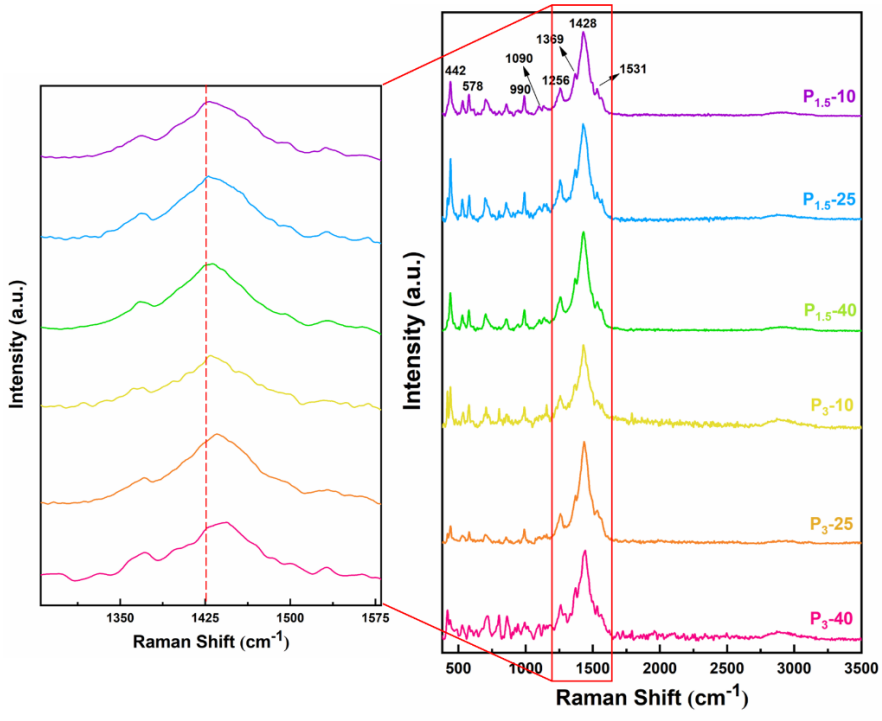

Fig. S2. Raman spectra of $P_{1.5}$-10, $P_{1.5}$-25, $P_{1.5}$-40, P_3-10, P_3-25, and P_3-40 and the corresponding expanded spectra in the region of 1260-1580 cm^{-1}

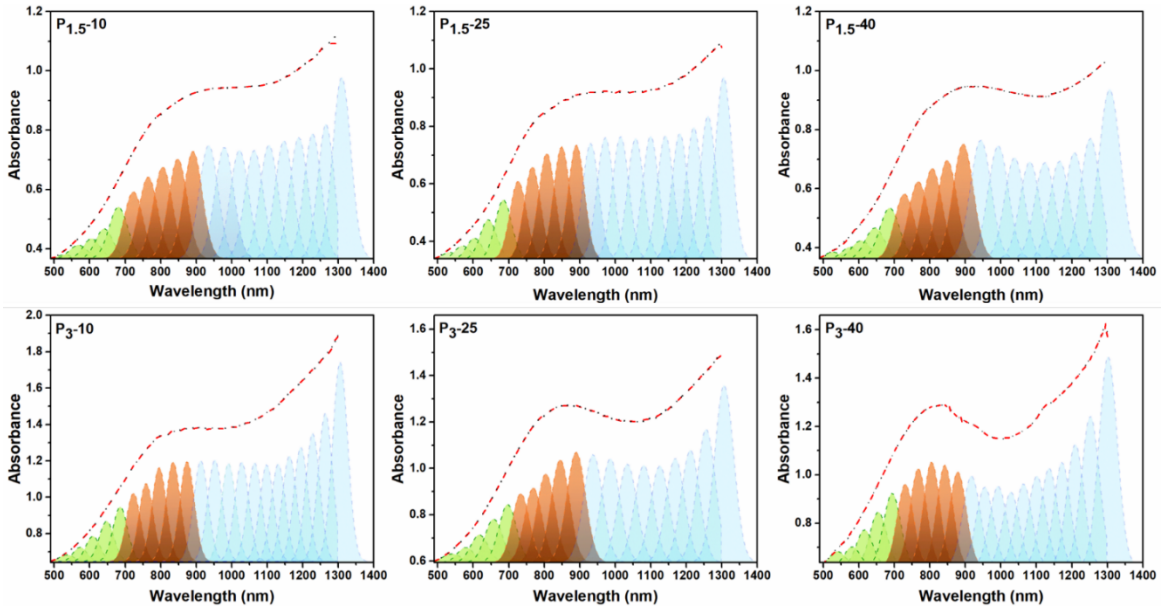

Fig. S3. Deconvoluted UV-Vis-NIR spectra of diluted $P_{1.5}$-10, $P_{1.5}$-25, $P_{1.5}$-40, P_3-10, P_3-25, and P_3-40 samples. Green, brown and blue areas represent neutral, polaron and bipolaron oxidation state, respectively.

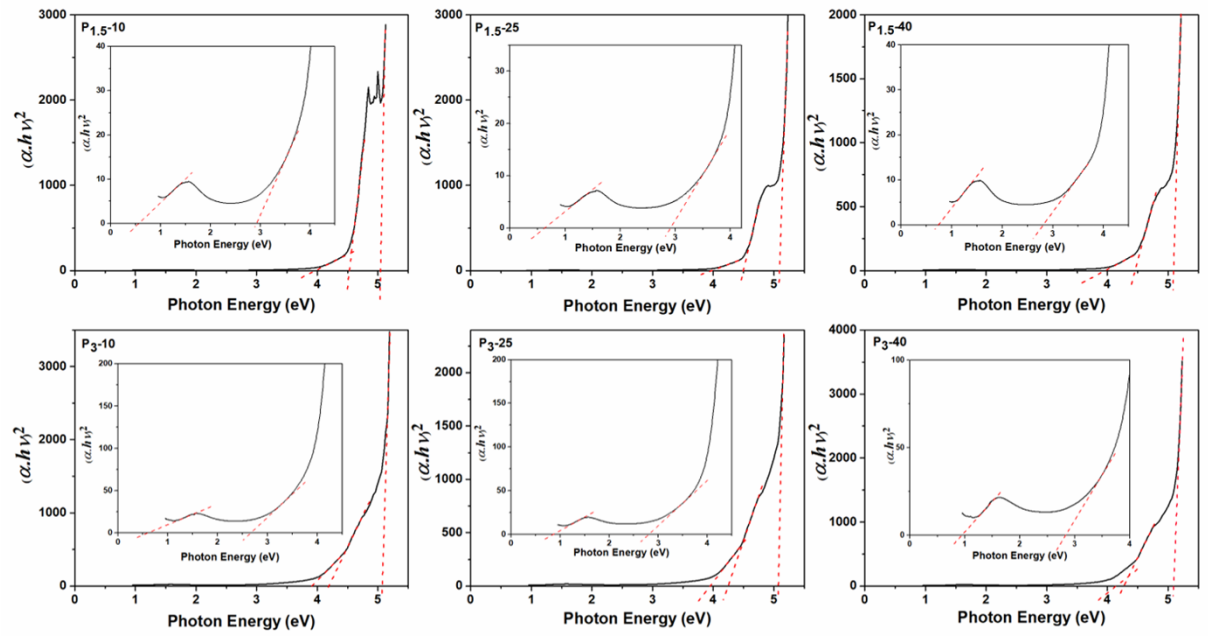

Fig. S4. Tauc plots of $P_{1.5}$-10, $P_{1.5}$-25, $P_{1.5}$-40, P_3-10, P_3-25, and P_3-40 samples derived from the corresponding UV-Vis-NIR spectra. Insets show the expanded regions.

Sample	E_g-1	E_g-2	E_g-3	E_g-4	E_g-5
$P_{1.5}$-10	5.03	4.51	3.97	2.92	0.56
$P_{1.5}$-25	5.09	4.5	3.97	2.93	0.62
$P_{1.5}$-40	5.09	4.46	3.99	2.82	0.74
$P_{2.25}$-10	5	4.21	3.91	2.7	0.176
$P_{2.25}$-25	4.99	4.23	3.93	2.84	0.51
$P_{2.25}$-40	5.02	4.23	3.92	2.83	0.82
P_3-10	5.07	4.2	3.91	2.69	0.59
P_3-25	5.09	4.24	3.95	2.81	0.86
P_3-40	5.09	4.27	3.94	2.82	0.95

Table S2. Band gap energies (eV) of PEDOT:PSS samples derived from the corresponding Tauc plots

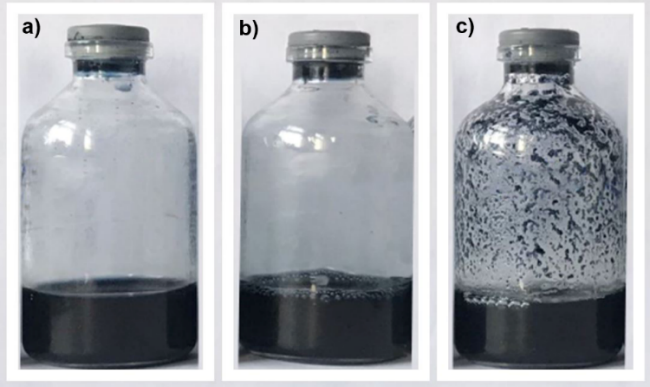

Fig. S5. Colloidal stability investigation of a) $P_{2.25}$-10, b) $P_{2.25}$-25 and c) $P_{2.25}$-40 dispersions after one month. Pictures were taken after vigerous shaking.

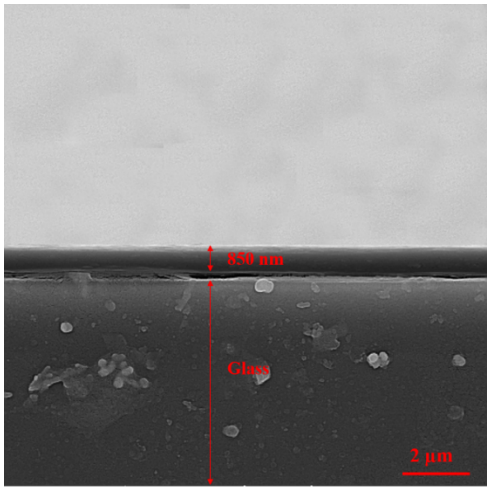

Fig. S6. FE-SEM image of $P_{2.25}$-10-D film deposited on the glass after rinsing with methanol

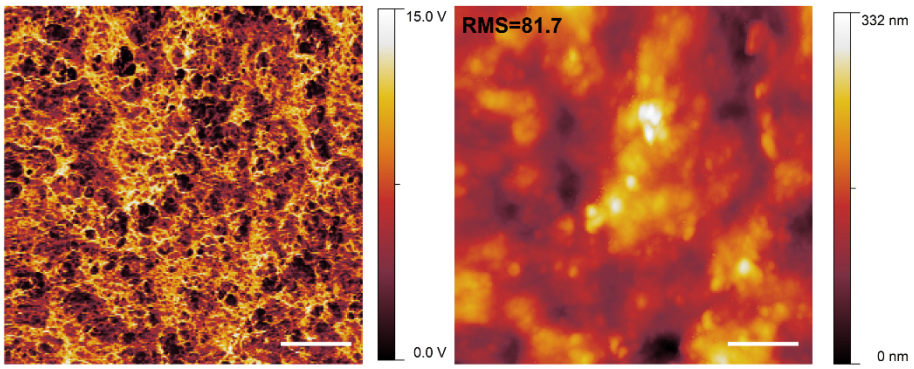

Fig. S7. Phase (left) and height (right) AFM images of $P_{2.25}$-10-D film deposited on the glass after rinsing with methanol. The scale bar in the images is 5 µm.

Table S3. Contact angles and surface energies of untreated and treated PET by corona discharge

Material	Contact angle (water)	Contact angle (diiodomethane)	Polar component ($mN.m^{-1}$)	Dispersive component ($mN.m^{-1}$)	Overall surface energy ($mN.m^{-1}$)
Untreated PET	57.4	35.1	12.99	41.99	54.97
Treated PET	27.4	19.1	25.90	48.03	73.93

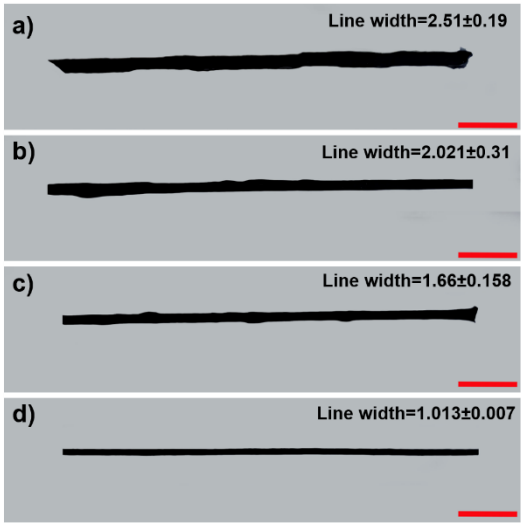

Fig. S8. Digital images of the printed lines at varying printing speeds: a) 500; b) 1000; c) 2000 and d) 4000 $mm.min^{-1}$. The nozzle-to-substrate distance was set as 1 mm and the ink was printed

using the nozzle with inner diameter of 0.7 mm at a constant pump speed of 1 rpm. All scale bars represent 1 cm.

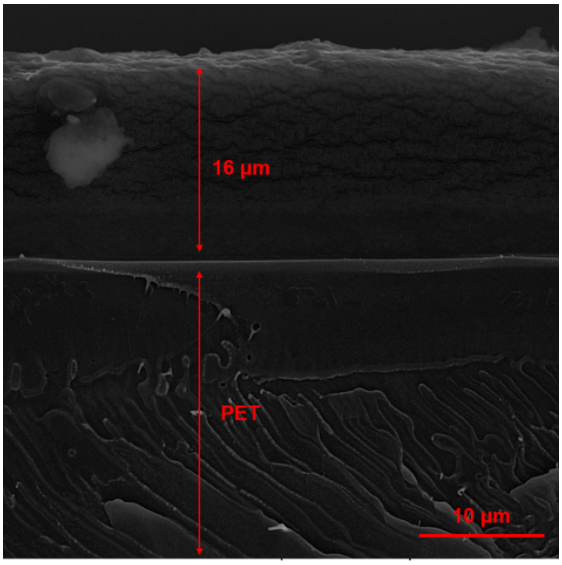

Fig. S9. FE-SEM image of cross-section of the three-layered printed circuit on PET as a substrate after rinsing with methanol and drying at 60 °C