Supporting Information

Spin Coating Photoactive Photosystem I-PEDOT:PSS Composite Films

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Film Composition

Table S1 shows the various weight percentages, concentrations, and weight ratios, of PSI and PEDOT:PSS in solution prior to deposition and film fabrication.

Table S1. Composition of solutions to be spin coated with the solute being PSI + PEDOT:PSS.

PSI wt% in solute (PSI+ PEDOT:PSS)	C_{PSI} (mg/mL)	$C_{PEDOT:PSS}$ (mg/mL)	Ratio PSI to PEDOT:PSS
0	0	11.0	0
1	0.10	10.4	0.010
4	0.37	8.9	0.042
5	0.45	8.5	0.053
10	0.76	6.8	0.11
20	1.2	4.6	0.25
30	1.4	3.3	0.43
40	1.6	2.0	0.81
50	1.7	1.7	1.0
70	1.8	0.80	2.3
100	2	0	PSI Only

Profilometry

Spin coating membrane proteins from aqueous media is challenging, and spatially inconsistent films are expected to form from such solutions. Figure S1 shows the variations in thickness for a PSI film spin coated from an aqueous solution without PEDOT:PSS at 2000 rpm. The thickness

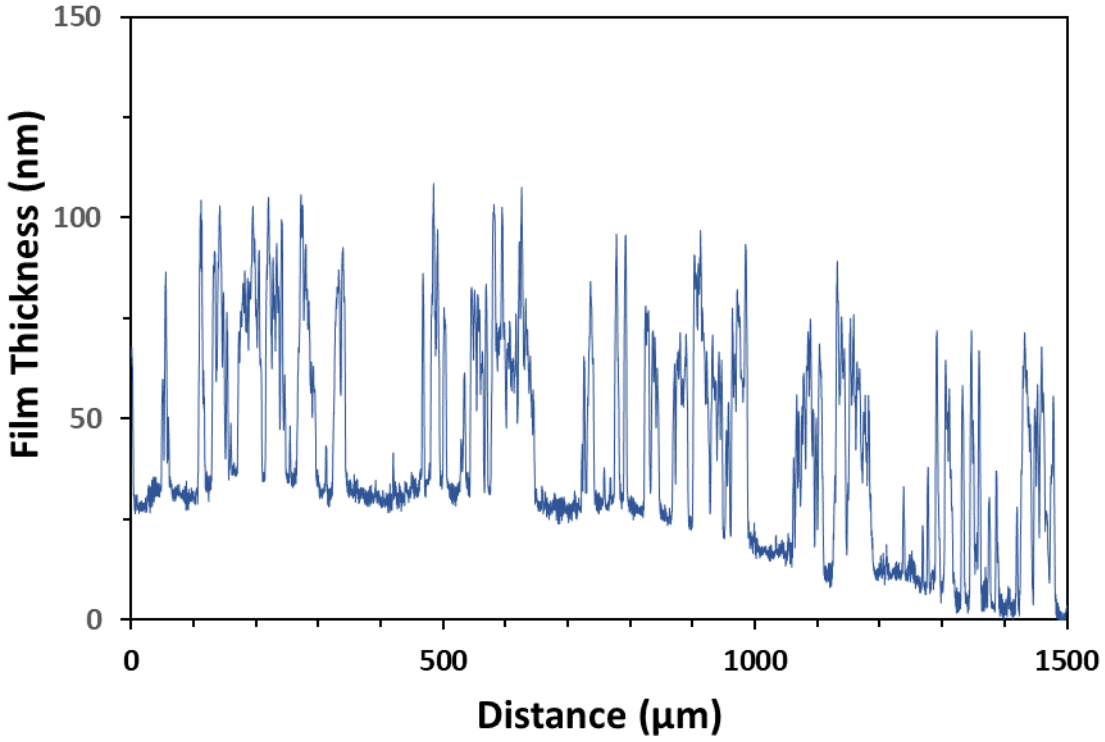

Figure S1. Profilometry data of a pure PSI film spin-coated film at 2,000 rpm on AET-modified gold. The film is inconsistent and varies from 0-100 nm across the scanned distance.

fluctuates between 0 and 100 nm with the average being ~50 nm. The addition of PEDOT:PSS results in much smoother films with lower roughness and better reproducibility.

FTIR Spectra

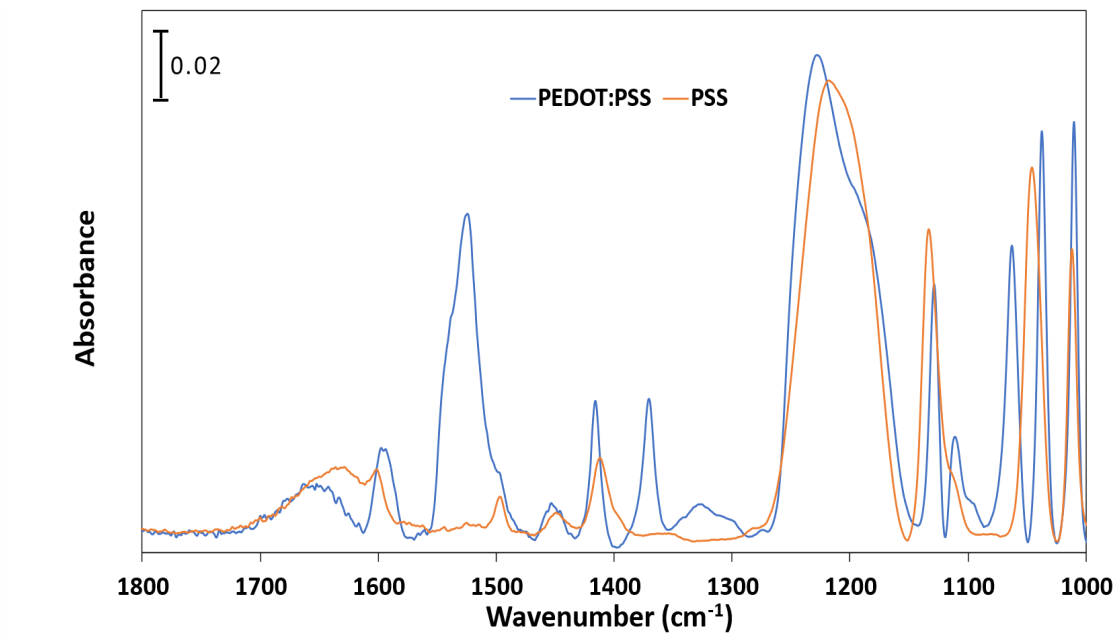

Figure S2. FTIR/ATR spectroscopy of PSS and PEDOT:PSS. Different peaks and peak shifts help attribute absorbances to one component vs the other.

To identify distinct IR absorbance peaks for PEDOT:PSS and PSS, the IR spectra of both components were plotted as shown in Figure S2, by separately spin coating a PEDOT:PSS stock solution at 1500 rpm and drop casting a 0.1 M sodium Na PSS solution from aqueous solution on a gold substrate. The most prominent peaks associated with PEDOT are the inter-ring stretching at 1526 cm^{-1} and the C-C or C=C quinoidal structure of thiophene at 1328 cm^{-1}. The peaks assigned to PSS at around 1010 and 1040 cm^{-1} are due to the S=O stretching of sulfonate bonds. All other peaks have contributions from both PSS and PEDOT.

Multiple Depositions

Figure S3 shows thicknesses of PSI-PEDOT:PSS films prepared from multiple spin coating depositions. Between each step, the film was left to dry for ~30 min in the fume hood until the

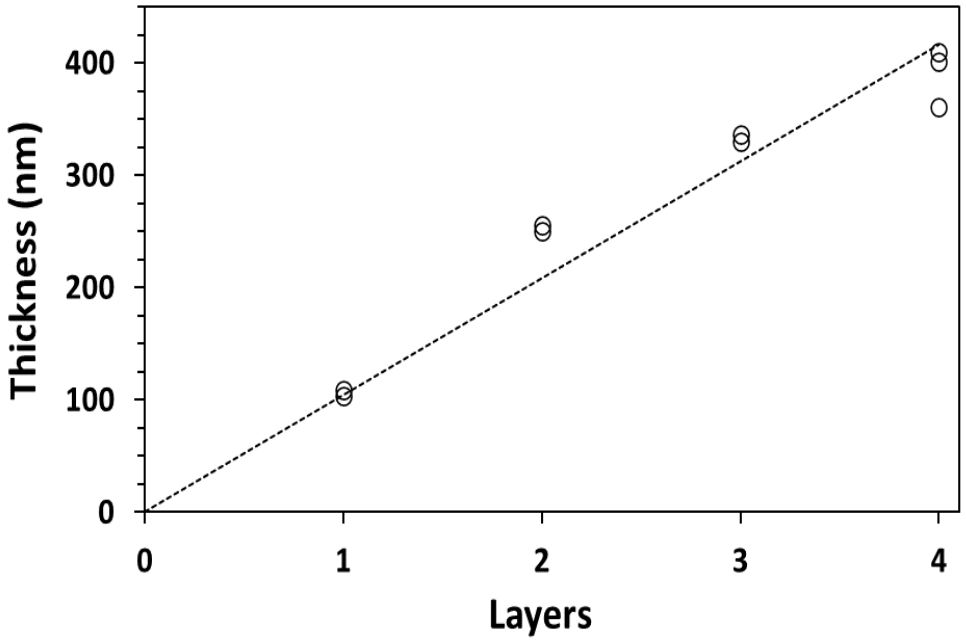

Figure S3. Film thickness after different numbers of layers (at 10% PSI in solution) deposited via spin coating 500 μL per deposition at 1600 rpm. The dashed line is fitted to the data points as a guide to the eye for the general trend.

next deposition step occurred. Under these conditions, the spin coating of a 10% PSI solution leads to successively thicker films up to 4 depositions. If the drying step was omitted, the dispensed aqueous solution on the substrate attached to the spin coater's rotating disk dissolved the top layers of the film rather than adding to its thickness, resulting in inconsistencies in the final thickness.

Turnover Number Calculations

TN are calculated as described in Eq 2 in the main text.

For spin coated films, the following calculations and assumptions are shown for a film deposited from a 1 %wt PSI solution. All PSI-PEDOT:PSS films analyzed for TN had a thickness of ~150 nm, an assumed density of 1 g/ cm^3, and a geometric area of 1 cm^2. For the spin coated composite films, the mass loading of PSI in the film is assumed to be the same as that in solution

Film volume=1.5×10^{-5} cm^3 and a mass m=1.5×10^{-5} g

1% PSI: 1% of the film is PSI by weight (assumed based on solution composition)

so, the mass of PSI=1.5×10^{-7} g PSI.

Using 500 kDa as the molecular weight, the number of PSI moles = 3×10^{-13} mol

Areal concentration of PSI: $C_{PSI} = 3 \times 10^{-13}$ mol/cm^2

PCA current density: 140 nA/cm^2 = 1.4×10^{-7} A/cm^2

$$TN = \frac{1.4 * 10^{-7} \frac{\mathrm{A}}{\mathrm{cm}^2}}{3.0 * 10^{-13} \frac{\mathrm{mol}}{\mathrm{cm}^2} * 96485 \frac{\mathrm{C}}{\mathrm{mol}}} = 4.8 \frac{mol_{e^-}}{s * mol_{PSI}}$$

For drop casted films, we are using known volumes of solutions with known concentrations. Here the calculation is shown for a 70%wt PSI film:

70 % PSI: used 25 µL of a solution at ~ 1.8 mg/mL PSI on 0.27 cm^2 of AET gold

So, mass of PSI = 4.5×10^{-5} g PSI

Using 500 kDa as the molecular weight, the number of PSI moles = 9.0×10^{-11} mol PSI

Areal concentration of PSI: $C_{PSI} = 3.3 \times 10^{-10}$ mol/cm^2

$$TN = \frac{7.3 * 10^{-7} \frac{\mathrm{A}}{\mathrm{cm}^2}}{3.3 * 10^{-10} \frac{\mathrm{mol}}{\mathrm{cm}^2} * 96485 \frac{\mathrm{C}}{\mathrm{mol}}} = 0.023 \frac{mol_{e^-}}{s * mol_{PSI}}$$

Multi-Angle Dynamic Light Scattering (MADLS)

MADLS is a zeta sizing technique that provides size distributions based on percentage volume and percentage intensity. For polydisperse systems, percentage intensity could be misleading as the larger sized particles will always reflect more light, causing their representation to be most prominent in the sample. Figure S4 shows the percentage intensity of a PSI-PEDOT:PSS solution containing 10% PSI. Comparing Figure S4 to its percentage volume companion in Figure 5 of the manuscript shows that the presence of the larger particles is amplified in this representation.

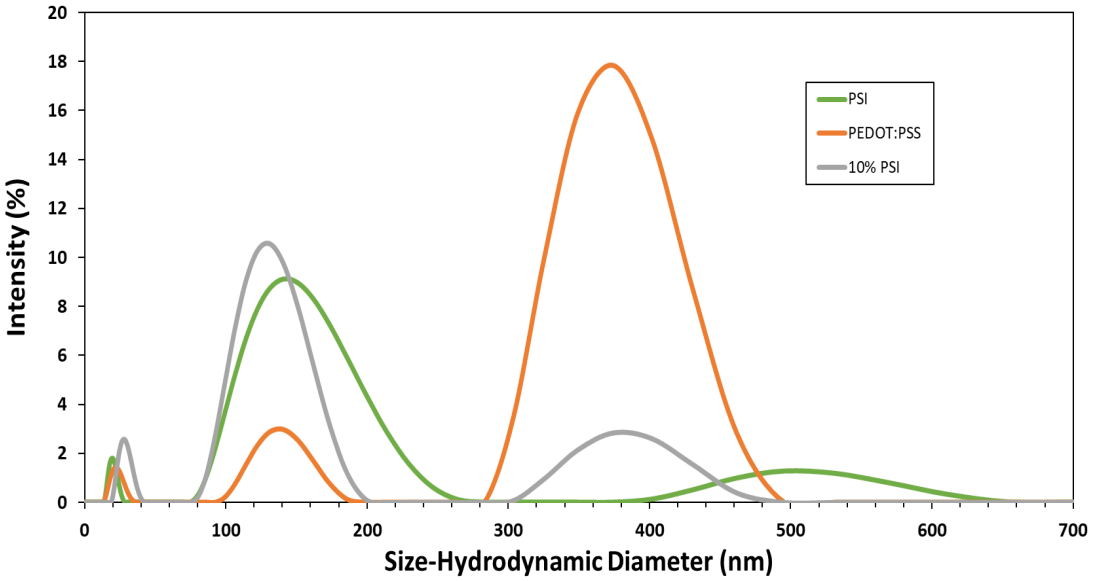

Figure S4. Size distribution by % intensity of PSI, PEDOT:PSS, and a 10% PSI solution showing the hydrodynamic diameter of the particles.

UV-Vis

UV-Vis spectroscopy was used to determine the percent transmittance (%T) for composite films spin coated from a 10 wt% PSI solution onto glass substrates at different speeds, with uncoated glass serving as the baseline. The results in Figure S5 show that even for a film as thick as ~865 nm, light can penetrate the film and reach the deep-seated layers within the film. In accordance with Beer Lambert's Law, the data are best fitted to a linear trend, where the %T for the two distinct chlorophyll *a* characteristic absorbance wavelengths (440 nm and 675 nm) are plotted versus the film thickness. Light passing through the film at the conditions herein is not a limiting factor to its photoactive properties.

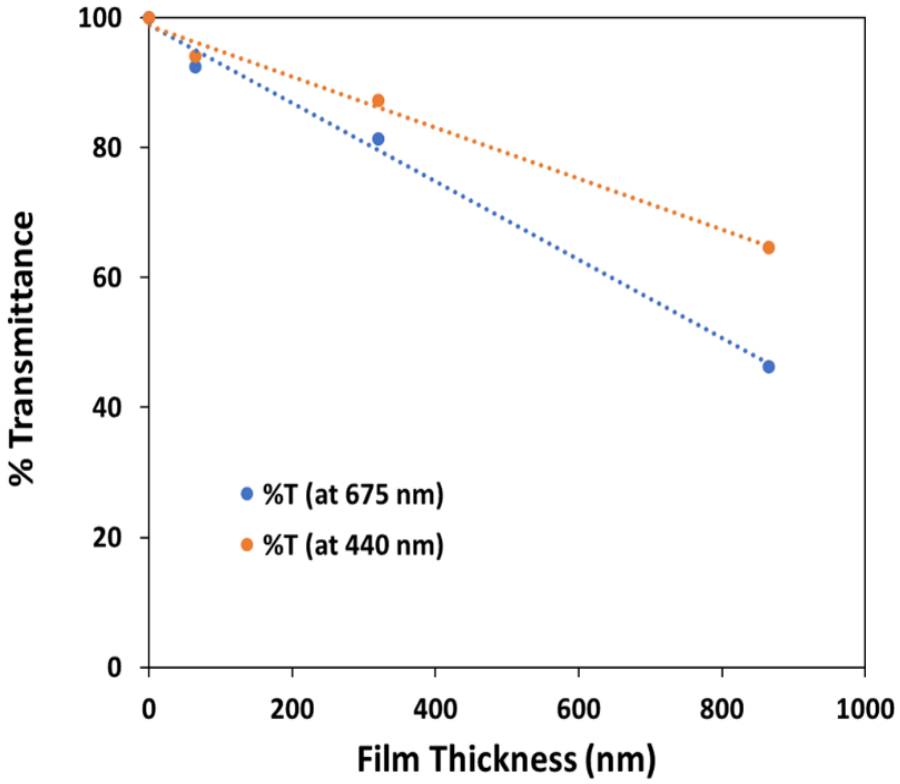

Figure S5. UV-Vis transmittance of light through 10 %wt PSI composite films with varying thicknesses on glass substrates. The blue dotted line is a linear fit for data collected at 675 nm, and the orange dotted line is that for data collected at 440 nm.