SUPPORTING INFORMATION

Bio-Based PEDOT: Nanocellulose Hybrids as Efficient Hole-Transport Layers for Photoelectrochemical Devices

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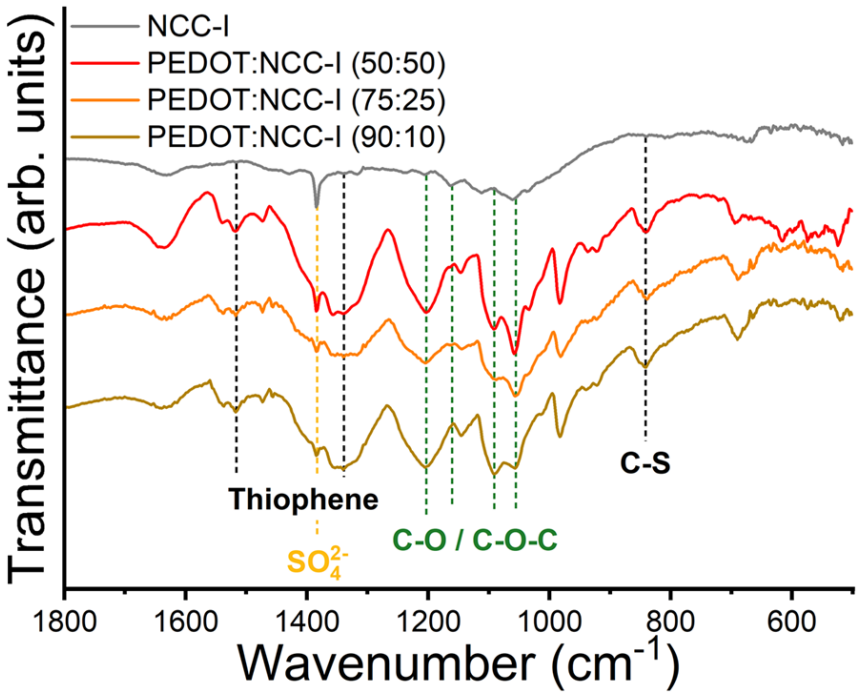

Fig. S1. FT-IR of all compositions studied of PEDOT:NCC-I 90:10, 75:25 and 50:50 using KBr pellets and diluted 1mg of sample in 1000 mg of salt.

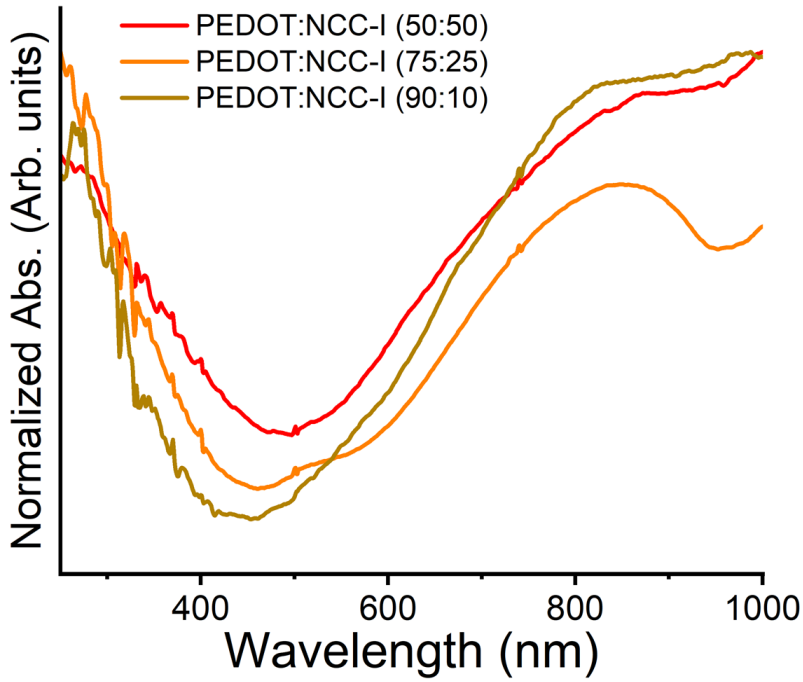

Fig. S2. UV-vis of the different PEDOT:NCC-I dispersions obtained after dialysis diluted at 25 v/v% in MiliQ water.

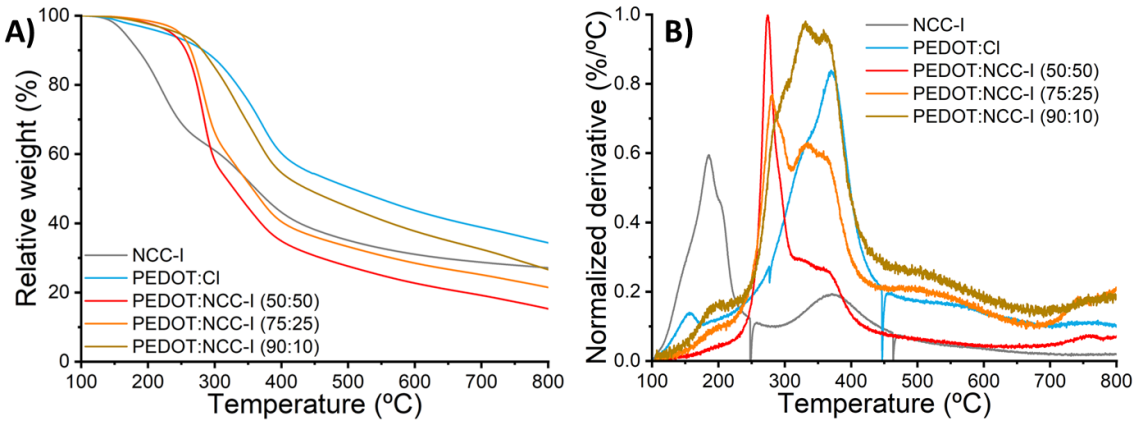

Fig. S3. A) TGA plot and its derivative of relative weight/ºC obtained under nitrogen of PEDOT:Cl as control and PEDOT:NCC-I 90:10, 75:25 and 50:50

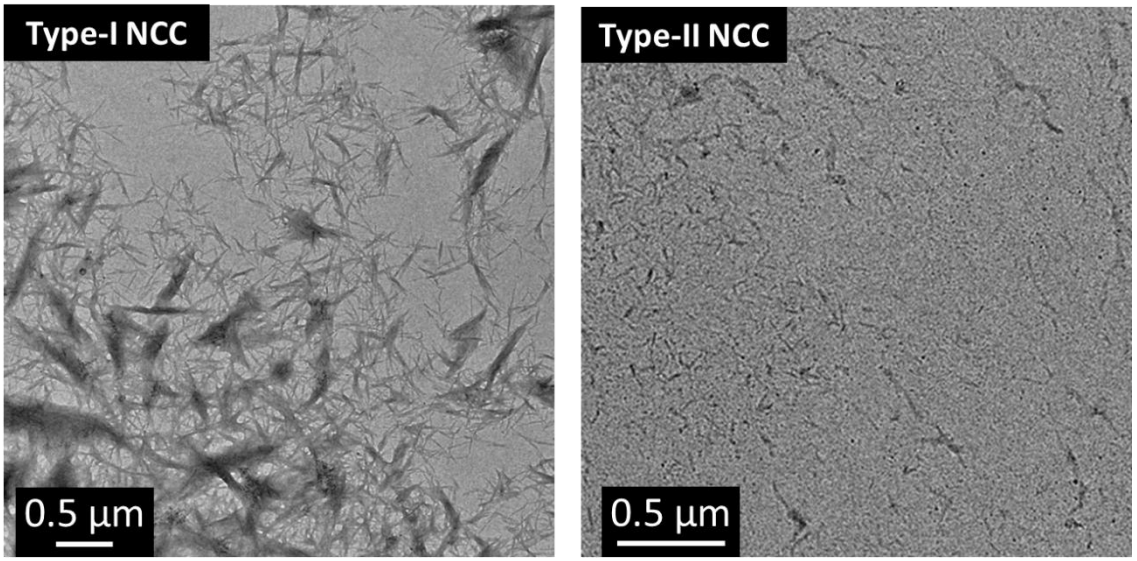

Fig. S4. TEM images for NCC type-I (left) and NCC type-II (right).

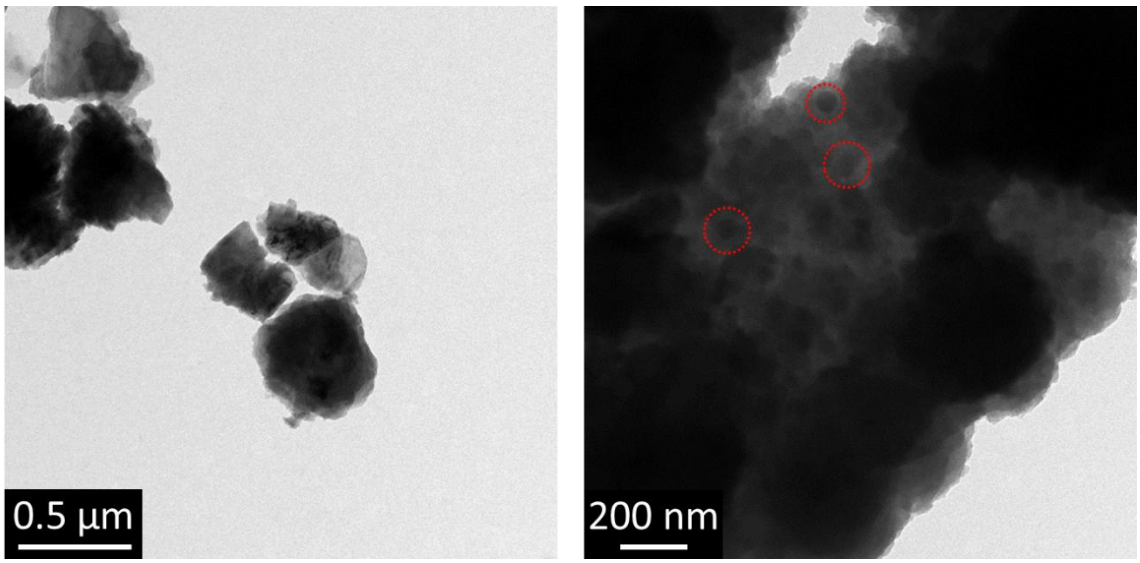

Fig. S5. TEM images for PEDOT:Cl control. Higher magnification image (right) shows the presence of small particles within big aggregated particles.

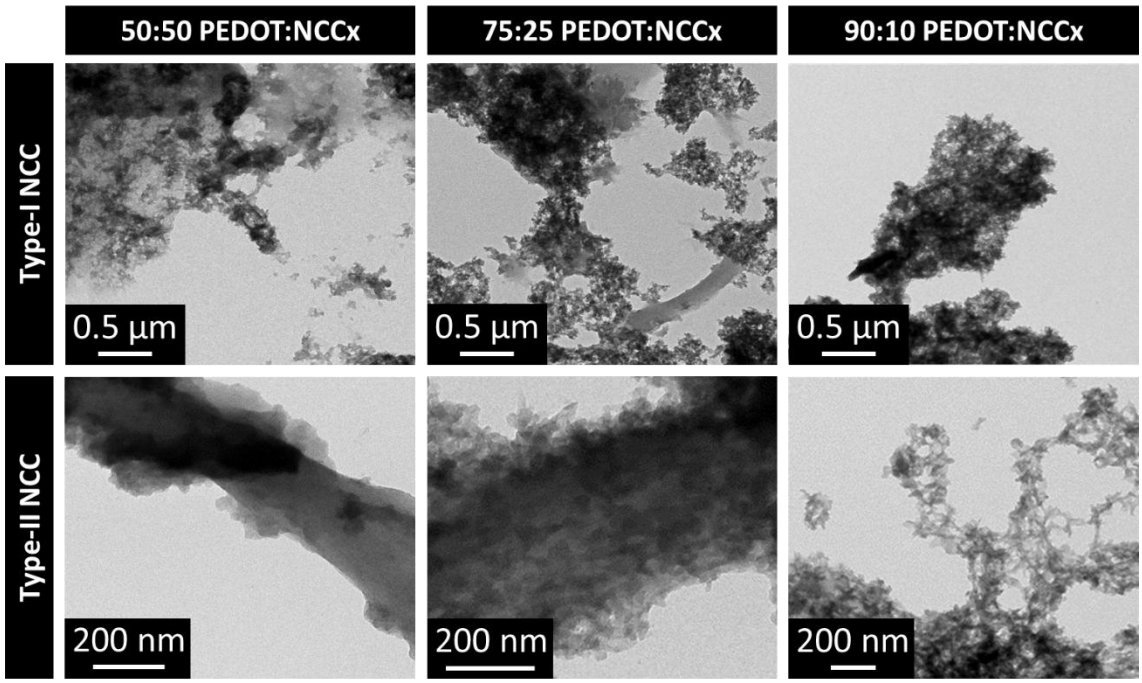

Fig. S6. TEM images for PEDOT:NCCx 50:50, 75:25 and 90:10 composites using type-I and type-II nanocellulose.

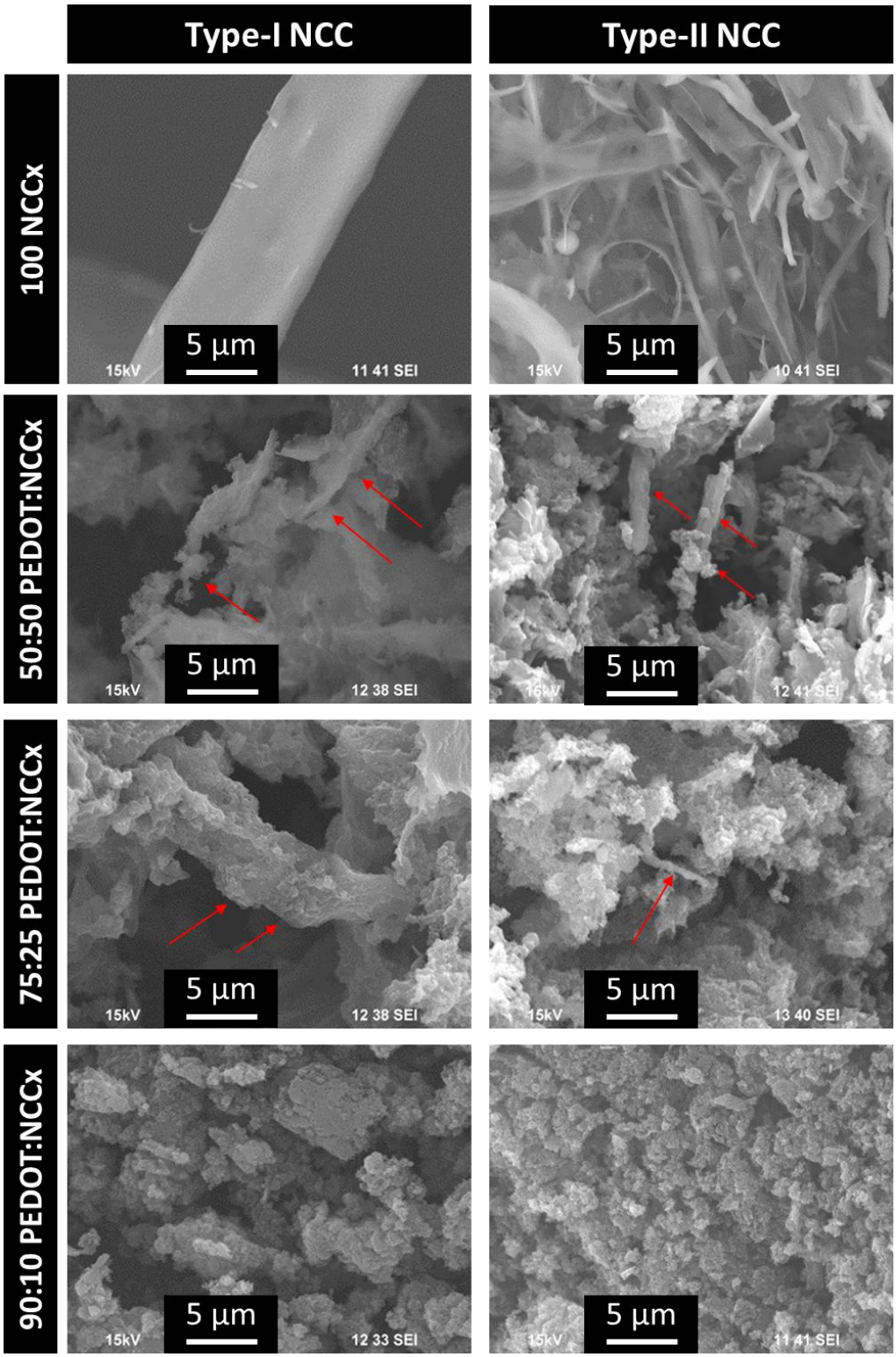

Fig. S7. SEM images of type-I and type-II nanocellulose along with the 50:50, 75:25 and 90:10 PEDOT: NCCx hybrids using both types. Red arrows show the morphological structure of the nanocellulose wrapped by PEDOT.

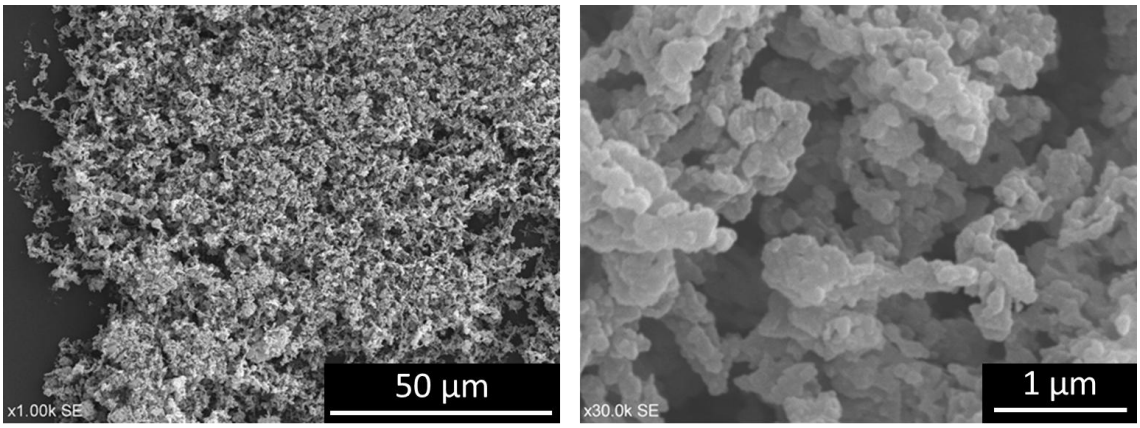

Fig. S8. Low (left) and high-magnification (right) SEM micrographs of the as-deposited P3HT nanoparticles on FTO.

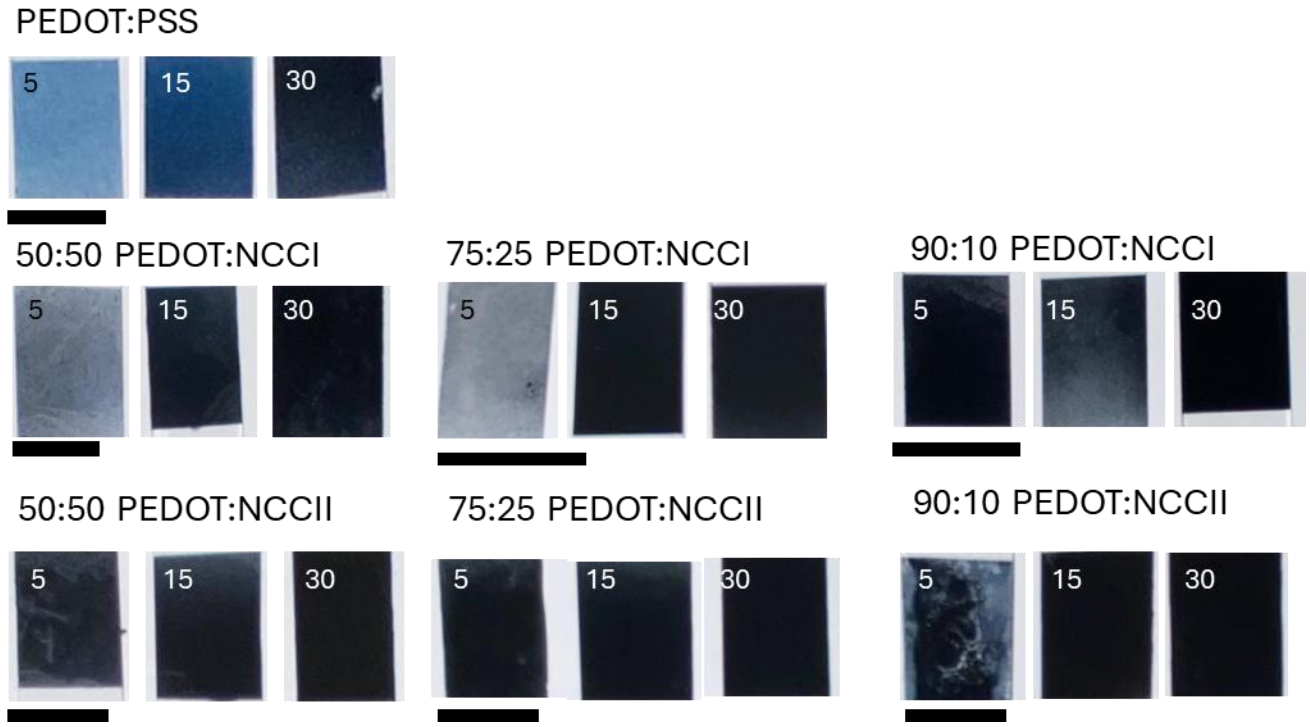

Fig. S9. Photographs of spray-coated films based on 50:50, 75:25 and 90:10 PEDOT:NCCI and PEDOT:NCCII hybrid, showing the film continuity and surface appearance as a function of the number of deposited layers 5, 15, and 30 layers (scale bar= 1cm).

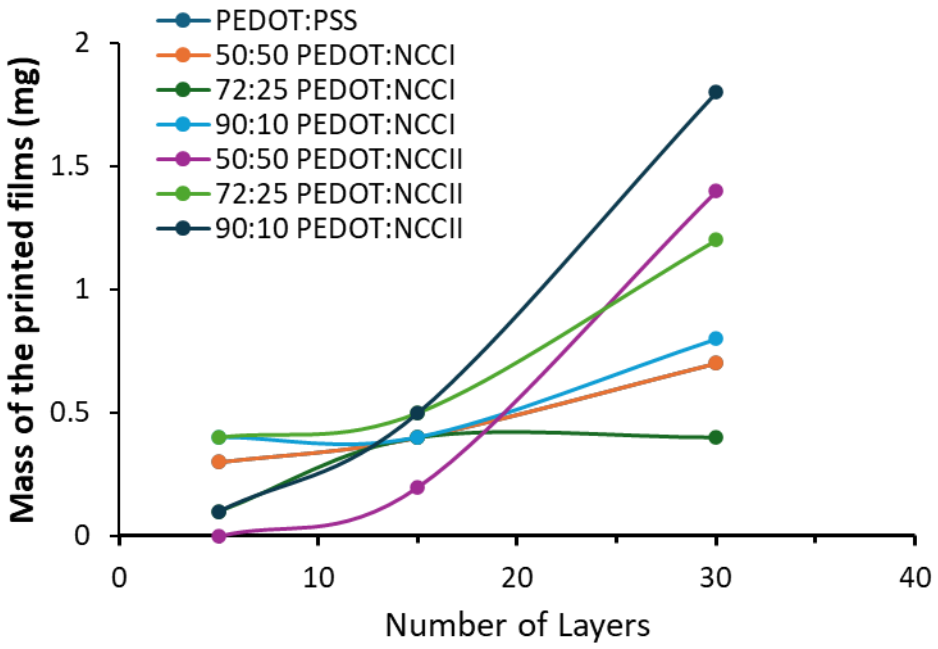

Fig. S10. Mass weight of the different of spray-coated films based on 50:50, 75:25 and 90:10 PEDOT:NCCI and PEDOT:NCCII hybrid as a function of the number of deposited layers (5, 15, and 30 layers).

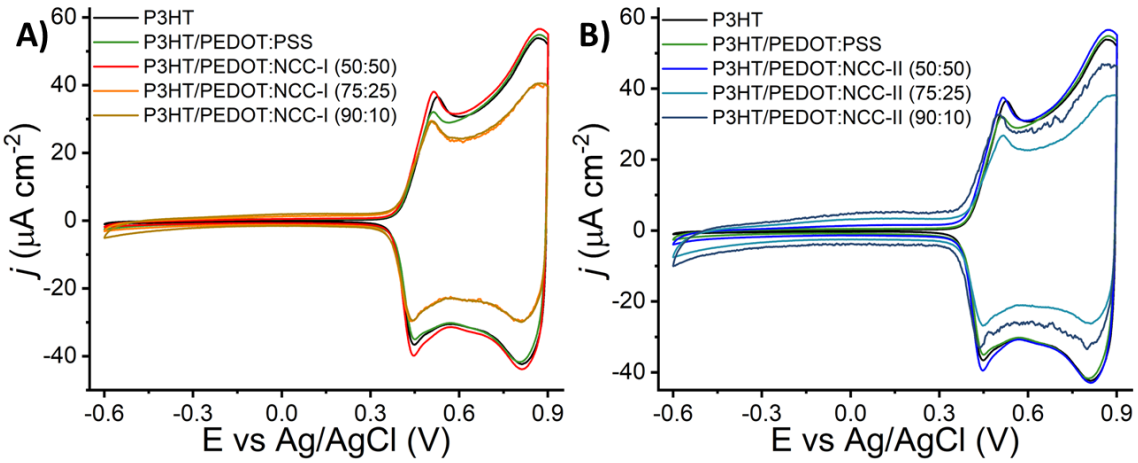

Fig. S11. Cyclic voltammetry of P3HT, P3HT/PEDOT:PSS control and P3HT/PEDOT:NCCx (50:50) hybrid films with both NCC A) type I or B) type-II.

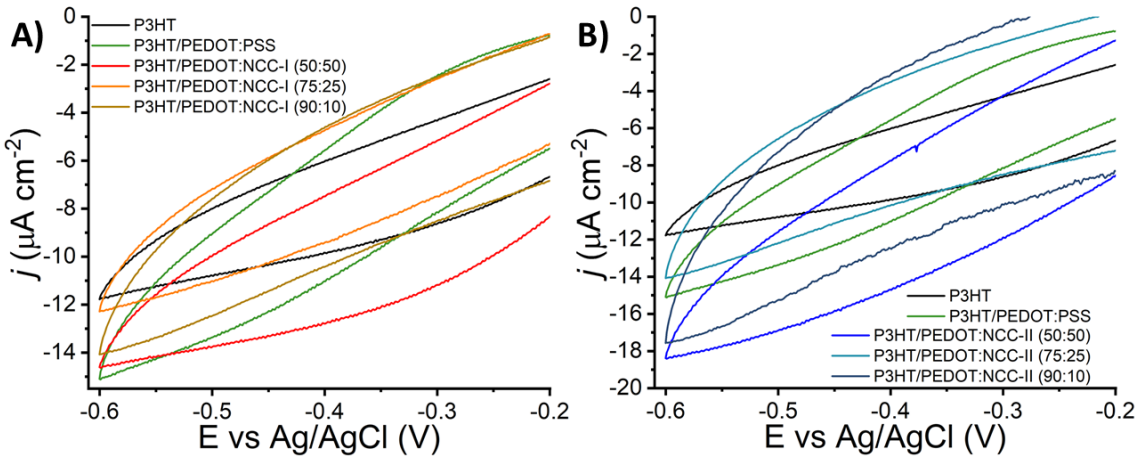

Fig. S12. Zoomed-in CV under illumination of P3HT, P3HT/PEDOT:PSS control and P3HT/PEDOT:NCCx (50:50) hybrid films with both NCC A) type I or B) type-II, showing the maximum cathodic photocurrent achieved for each hybrid composition.

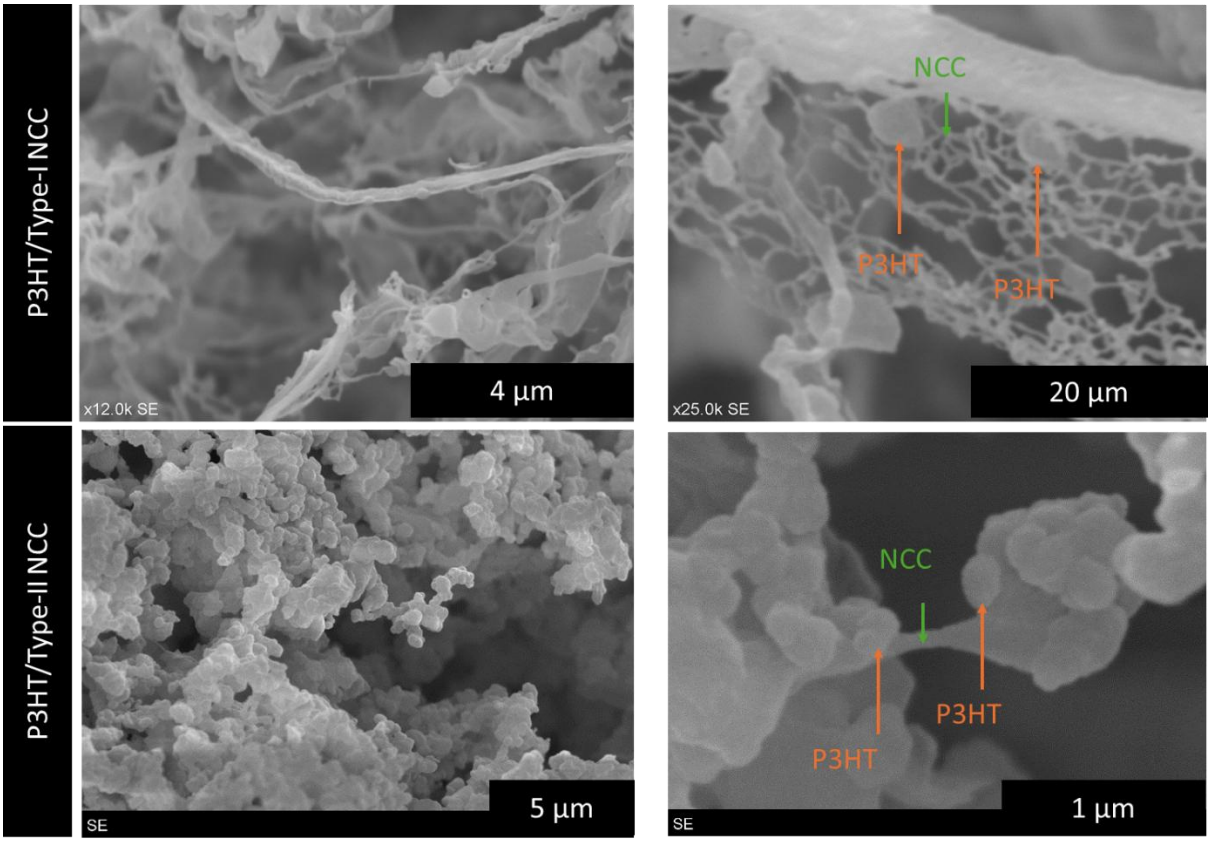

Fig. S13. Scanning electron microscopy (SEM) images of P3HT blended with NCC-I and NCC-II.

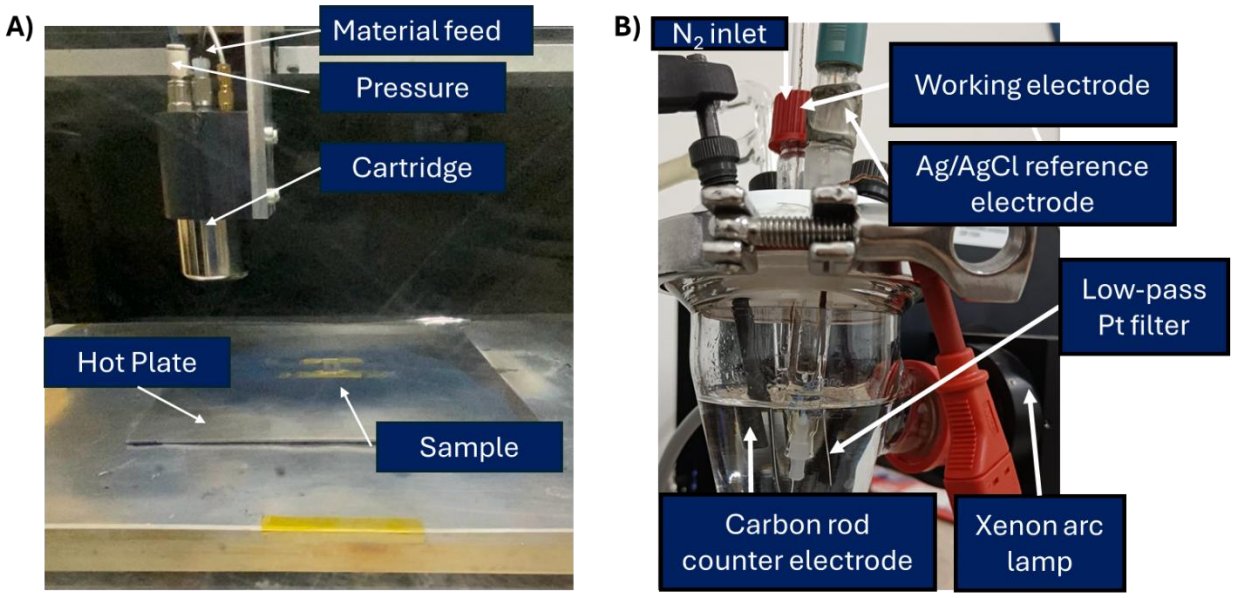

Fig. S14. A) Photograph of the spray-coating setup. The ink is delivered under controlled pressure through a spray cartridge and deposited onto the substrate positioned on a hot plate maintained at 100 °C, enabling rapid solvent evaporation during film formation. B) Photograph of the photoelectrochemical setup. The working electrode consists of spray-coated P3HT/PEDOT:PSS or P3HT/PEDOT:NCC hybrid architectures. Illumination is provided by a Xenon lamp.

		Resistance (Ω)	
Type	**5 spray-coated layers**	**15 spray-coated layers**	**30 spray-coated layers**
PEDOT:PSS	5345	6279	1472
50:50 PEDOT:NCCI	N.A.	15482.07	17737.63
75:25 PEDOT:NCCI	N.A.	18646.06	23530.97
90:10 PEDOT:NCCI	8558.03	18116.60	2594.57
50:50 PEDOT:NCCII	N.A.	6178.18	15456.65
75:25 PEDOT:NCCII	15118.38	17595.59	9640.39
90:10 PEDOT:NCCII	29632.13	1292.91	2230.94

Table S1. Electrical resistance of PEDOT:NCCx hybrid films and PEDOT:PSS control samples as a function of the number of spray-coated layers (5, 15, 30), extracted from I-V measurements using a four-point probe station.