Electronic Supplementary Information for

Improved hole transport in inverted perovskite solar cells with carbon nanorings

Experimental section

Synthesis of carbon nanorings (CNRs)

Fructose (80 mg, 99%) and Glucose (80 mg, 99%) were dissolved in water (26 g, distilled) in a 50 mL glass vial with a PET cap. The vial was closed, hand-shaken and sonicated at 40 °C for 30 min. A 0.05 g amount of Ferrocene and PAN powder were placed in a graphite crucible enclosed within a graphite susceptor, and heated up to the reaction temperature using an induction furnace with a flow of Ar (1000 sccm) and H_2 (100 sccm). H_2 was allowed to bubble through the vial. The temperature of the susceptor was controlled to be heated to 1000 °C. After growth for 15 min, the H_2 flow was stopped and the chamber was cooled down to room temperature. During the cooling process, the system was purged with Ar to prevent a backflow of air from the exhaust line.

The product was purified by treatment with 10 wt % HCl, then suspended in 100 mL of concentrated sulfuric acid. Ultrasonication was applied for 30 minutes using an ultrasonic bath cleaner (USK-4R) to facilitate dispersion. Subsequently, 17 g of potassium permanganate ($KMnO_4$) was gradually added to the suspension. The reaction mixture was stirred at 0 °C for 30 minutes, followed by heating to 40 °C and maintaining this temperature for an additional hour. During cooling, 100 mL of distilled water was slowly introduced into the mixture within an ice water bath. As the temperature dropped to approximately room temperature (about 2 hours), 400 mL of distilled water and 20 mL of 30 wt % hydrogen peroxide (H_2O_2) were added to quench the reaction. The resulting solution was separated by centrifugation at 4800 rpm (using a Hitachi Himac CR-GII centrifuge). The remaining slurry was washed with 3 M HCl three times and with acetone five times, each wash performed via centrifugation. The purified product was then freeze-dried using an EVELLA FD-1000 lyophilizer. Following oxidation and washing, the product was exfoliated under ultrasonic agitation for 1 hour to achieve delamination.

The exfoliated product was further washed three times by replacing the liquid with distilled water and HCl to allow soluble impurities to diffuse out over at least 2 hours. After washing, the sample was freeze-dried to remove residual liquids.

Precursor preparation

461 mg PbI_2, 159 mg Methylammonium iodide (MAI), and 78 mg dimethyl sulfoxide (DMSO) (molar ratio 1:1:1) are dissolved in 600 mg dimethylformamide (DMF). The solution was stirred overnight. Prior to device fabrication, the precursor solution was filtered twice with a 0.22 µm PTFE filter. $PC_{61}BM$ solution (20 mg mL^{-1}) was prepared by dissolving PCBM in CBZ and stirred at room temperature over 2 h. CNR solution (0.01 mg mL^{-1}) was used. Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS, PVP AI 4083) and henyl-C61-butyric acid methyl ester (PCBM, Nano-C, 99.5%) were used as HTL and ETL, respectively.

Device fabrication

The perovskite solar cells were fabricated on indium tin oxide (ITO)-coated glass substrates with the configuration of ITO/PEDOT:PSS/$CH_3NH_3PbI_3$/PCBM/Ag or ITO/CNRs/PEDOT:PSS/ $CH_3NH_3PbI_3$/PCBM/Ag. ITO substrates were cleaned by sequential ultrasonic treatment in acetone, methyl alcohol, isopropyl alcohol, and deionized water for 15 min each and then dried with a nitrogen stream. Then the precleaned ITO substrates were UV−ozone treated for 10 min. In order to form dense random networks of CNRs, poly(L-lysine) (PLL) solution (Sigma Aldrich, 0.1% (w/v) in H_2O) was spin-coated onto the ITO substrate for 20 min. After soft blowing with nitrogen, CNRs solution was drop-cast on the substrates for 20 min, and the preparation was finished with soft blowing using nitrogen. Then, PEDOT:PSS solution (filtered with 0.45 μm PTFE-H) was spin-coated onto the ITO or ITO/CNRs substrates at 4000 rpm for 30 s and then dried at 100 °C for 10 min in air. Afterward, perovskite precursors (filtered with 0.45 μm PVDF) were spin-coated onto the substrates at 5000 rpm for 50 s after being accelerated up to 5000 rpm for 5 s in a glovebox; 0.5 mL of anti-solvent (CBZ) was dropped at 5–6 s during the main spin-coating duration. The resulting substrates were then annealed at 100 °C for 10 min on hot plates in air. Perovskite precursor solution was spin-coated on the ITO/PEDOT:PSS or ITO /PEDOT:PSS/CNRs layer at 4000 rpm for 30 s. Subsequently, the sample was annealed at 100 ^{o}C for 10 min. Afterward, the PCBM solution was spin-coated onto the perovskite films at 4000 rpm at 30 s. Finally, a 100 nm thick Ag were deposited on the substrates through a shadow mask. The effective cell area was defined as 4 mm^2.

Characterization:

The scanning electron microscopy (SEM) image is measured by using S-4800 scanning electron microscope at an acceleration voltage of 2 kV. The X-ray diffraction (XRD) patterns were taken on a Rigaku Ultima IV diffractometer using Cu Ka radiation. Raman spectra were recorded with a Renishaw RM-1000 Micro Raman Spectrometer. Investigations of chemical compositions were performed using X-ray photoelectron spectroscopy (XPS, Physical Electronics PHI 5600). The UV-Vis absorption spectra were measured using a UV-1700 spectrometer. The PL spectra were recorded on a fluorescent spectrophotometer with a 150W Xe lamp as an excitation source at room temperature. For time-resolved PL (TRPL) measurements, a tunable Ti:sapphire femtosecond pulsed laser was used as the excitation light source with an excitation wavelength of 400 nm (3.8 MHz, 55uW). Electrochemical impedance spectroscopy was conducted using a CHI 760E electrochemical workstation. The current density-voltage (J-V) characteristics were recorded under illumination and dark using a Keithley 2400 source meter under AM1.5 simulated solar light in air. The illumination intensity was calibrated using a single-crystal Si photovoltaic cell. The incident photon-to-electron conversion efficiency (IPCE) was performed with a Newport Oriel IPCE measurement kit.

Table S1 The detailed photovoltaic parameters including Jsc, Voc, FF and PCE of PSCs with different proportions of CNRs.

Concentration (mg/mL)	**V_{OC}**	**J_{SC}**	**FF**	**PCE**
0	1.04	23.68	78.97	21.87
0.05	1.042	24.32	79.24	22.35
0.1	1.05	25.12	83.64	23.16
0.25	1.052	24.11	80.56	22.03
0.5	1.043	23.98	77.82	20.96

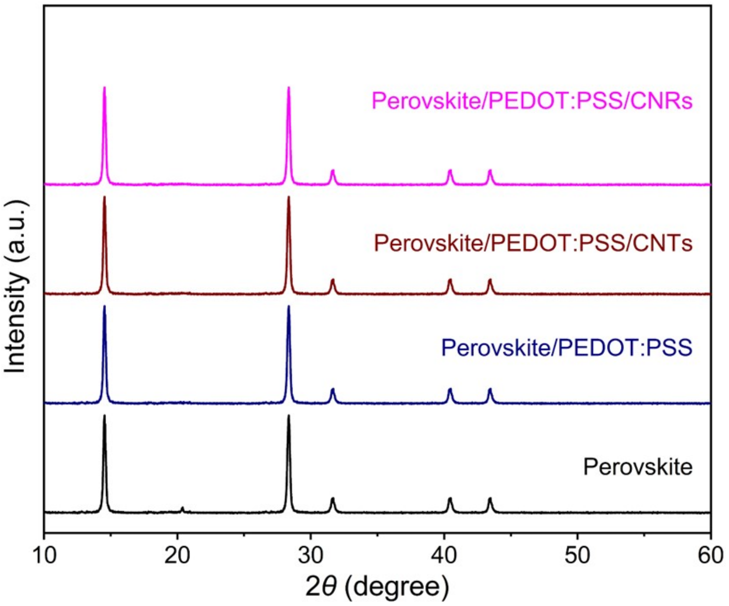

Figure S1. XRD spectra of perovskite/PEDOT:PSS films with and without CNRs.

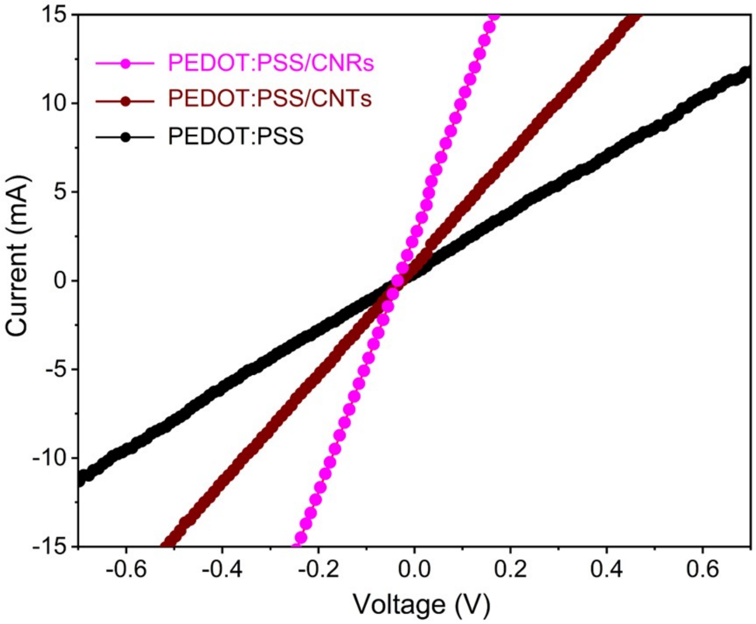

Figure S2. The current-voltage curves of PEDOT:PSS, PEDOT:PSS/CNTs and PEDOT:PSS/CNRs.

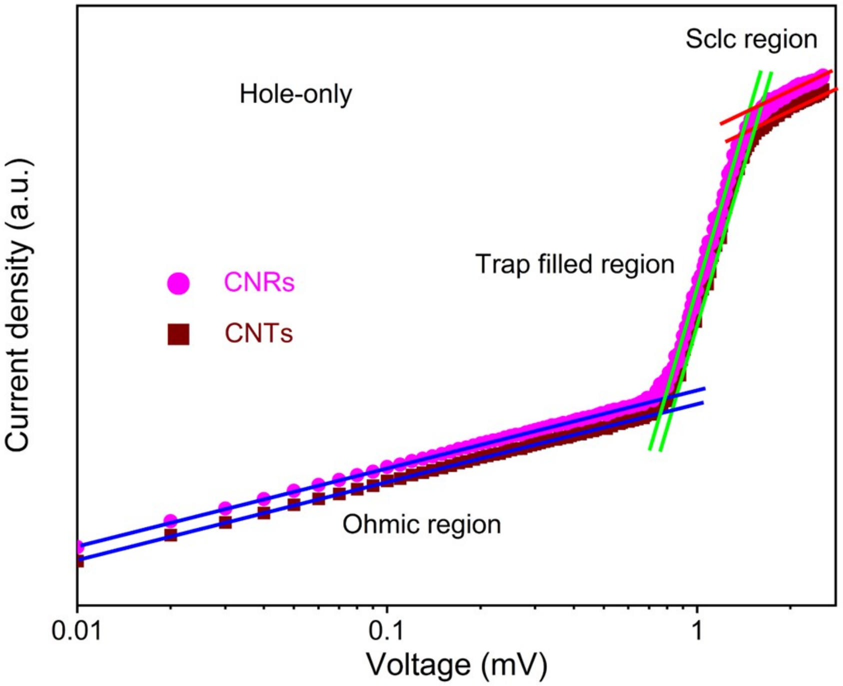

Figure S3. Space charge limited current measurements of the hole-only devices.

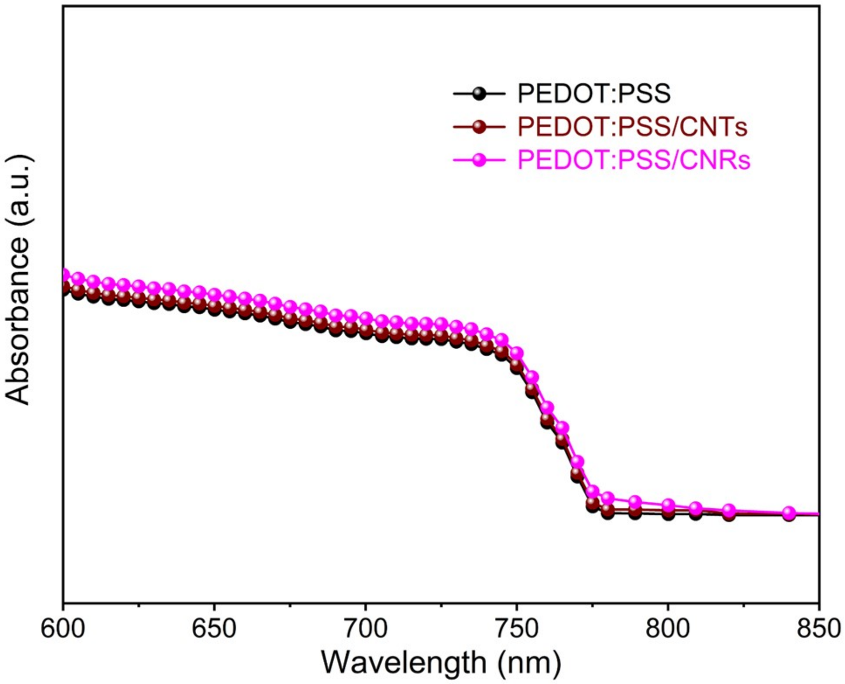

Figure S4. UV-vis spectroscopy of the PEDOT:PSS, PEDOT:PSS/CNTs and PEDOT:PSS/CNRs coated perovskite films.

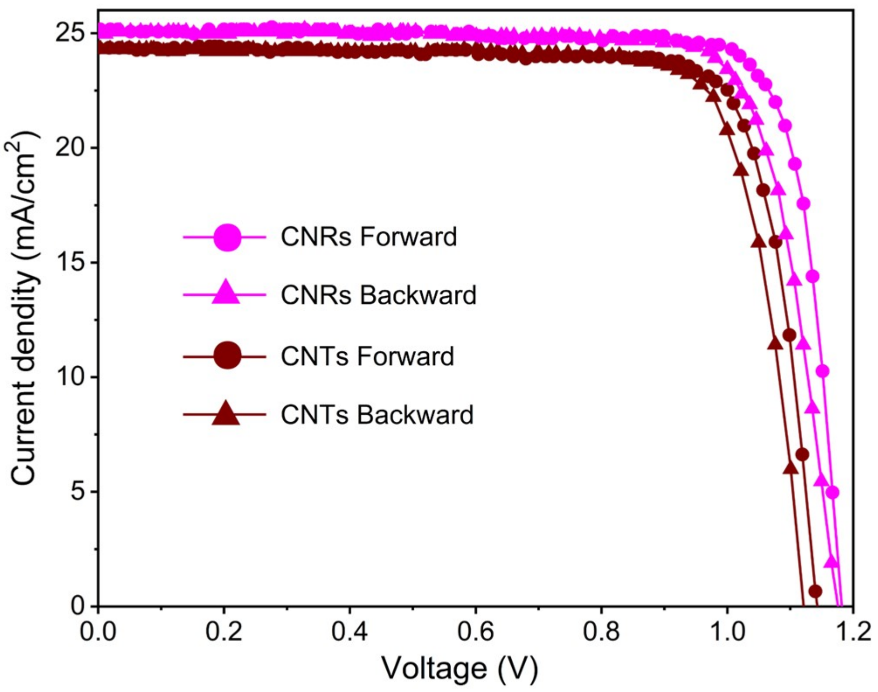

Figure S5. J-V curves extracted from forward and backward sweeping.