

## Electronic Supplementary Information

# One-step synthesis of carbon nanosheets converted from a polycyclic compound and their direct use as transparent electrodes of ITO-free organic solar cells

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## Experimental Methods

**Synthesis of PIM-1** A mixture of  $K_2CO_3$ , 3,3,3',3'-tetramethyl-1,1'-spirobisindane-5,5',6,6'-tetrol (3.40 g, 10 mmol), and 2,3,5,6-tetrafluorophthalonitrile (2.00 g, 10 mmol) in dry DMF (dimethylformamide) (80 ml) was stirred at 65 °C for 72 hr. Upon cooling, the mixture was added to water, and the crude product was collected by filtration. Repeated reprecipitations from methanol gave 4.26 g (92.4% yield) of yellow polymer.  $^1H$  NMR( $CDCl_3$ ) 1.16-1.45(12H, m), 2.04-2.46(4H, m), 6.41(2H, s), 6.80(2H, s).

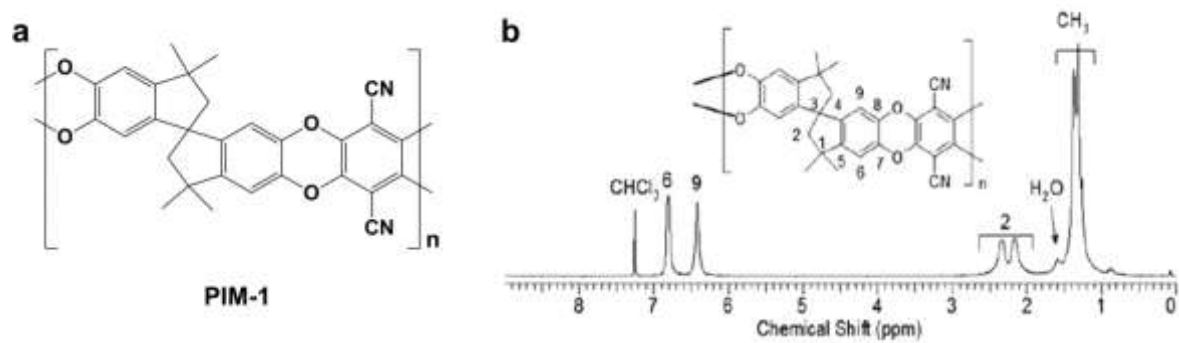
Molecular weight and molecular weight distributions were measured by GPC using PLgel 3  $\mu$ m MIXED-E columns and THF as the eluent with polystyrene standards. ( $M_n = 34,000$  g  $mol^{-1}$ , PDI = 2.4)

**Synthesis and characterization of CNSs** PIM-1 was dissolved in chloroform at 35 °C for 30 min. The solutions ranging from 0.1 to 0.6 wt% were spin-coated onto cleaned quartz substrates. The spin-coated films were carbonized up to 1200 °C with a temperature increase rate of 5 °C/min under a  $H_2/Ar$  mixture.

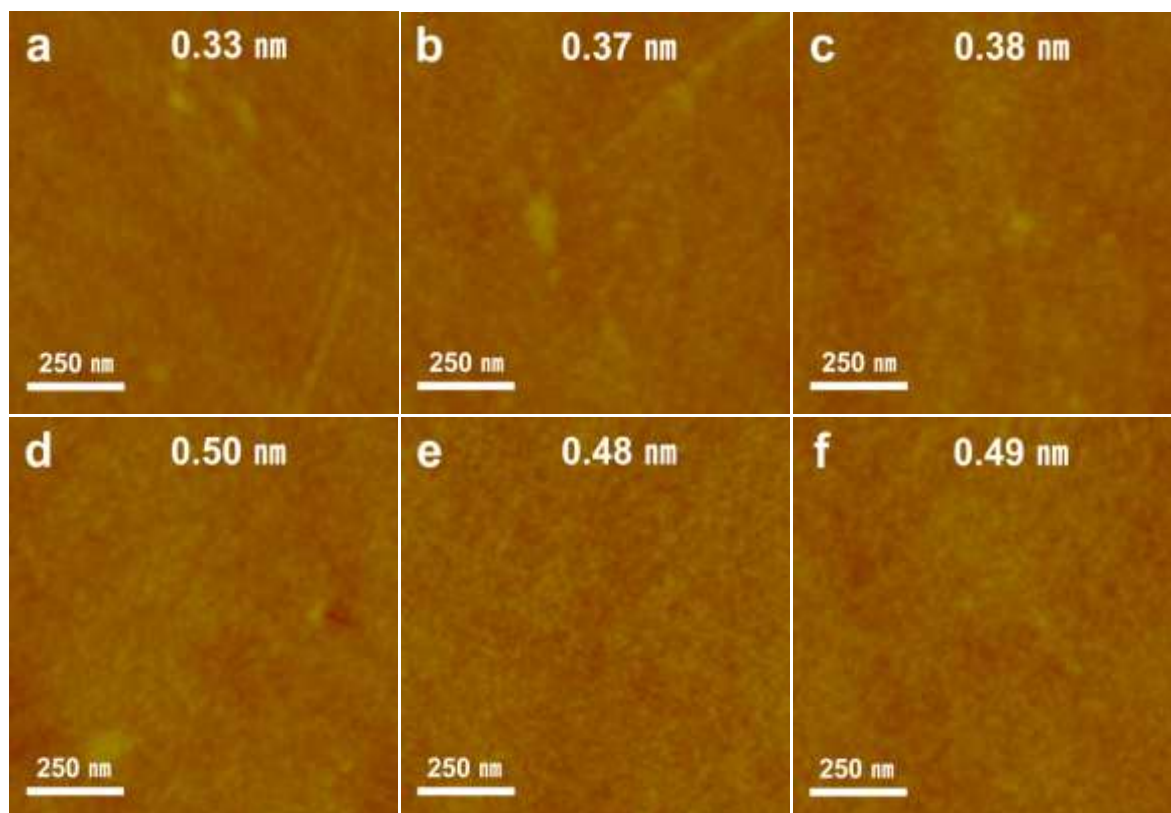
The thickness and roughness of the CNSs were analyzed by atomic force microscopy (AFM, Dimension 3100, Veeco) in tapping mode. The sheet resistance and transmittance of CNSs were measured using a four-point probe (FPP-RS8, Dasol Eng.) and ultraviolet-visible spectroscopy (UV-VIS, V-570, Jasco), respectively. The degree of the structure development of CNSs was confirmed by Raman spectroscopy (Horiba) with a 514.5 nm laser beam. The components of the CNSs were examined using x-ray photoelectron spectroscopy (XPS, PHI 5000 VersaProbe, Ulvac-PHI). The structure properties of the CNSs were investigated by transmittance electron microscopy (TEM, Tecnai G2 F20, FEI).

***Fabrication of OSCs*** To form the buffer layer, poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS, Clevios PH1000) was spin-coated on the CNSs and was then dried at 120 °C for 10 min. To fabricate the active layer, a blended solution with 50 mg of poly(3-hexylthiophene) (P3HT, Rieke Metals) and 50 mg of [6,6]-phenyl-C<sub>61</sub> butyric acid methyl ester (PCBM, Nano-C) in 2 ml of 1,2-dichlorobenzene was spin-coated in a N<sub>2</sub> glove box. The solvent-annealing treatment was performed for 120 min. Then, thermal annealing was carried out at 110 °C for 10 min. Lastly, Ca (20 nm)/Al (100 nm) metal cathodes were thermally evaporated in a vacuum at 10<sup>-6</sup> torr.

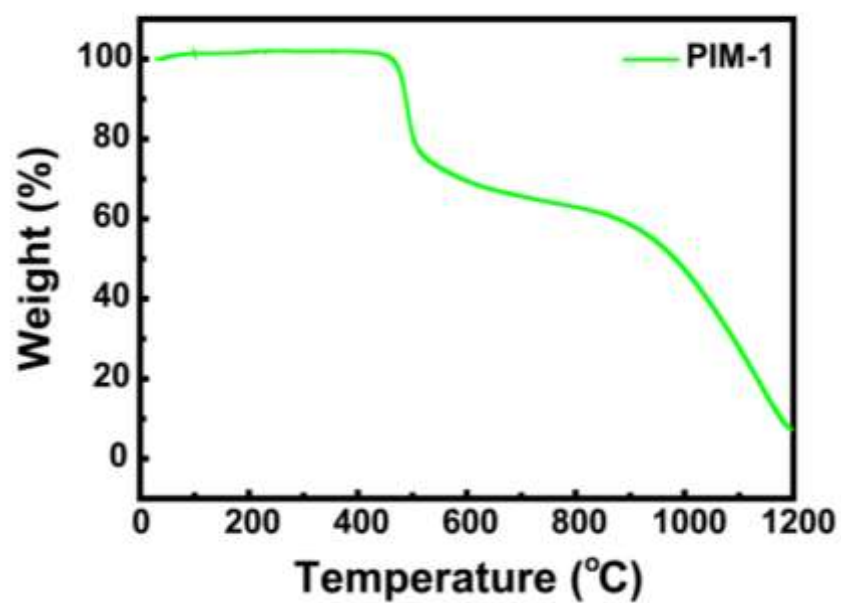
The current density-voltage (J-V) characteristics of the cells were measured using a Keithley 1200 instrument under AM 1.5 G conditions under 100 mWcm<sup>-2</sup>. For a more accurate measurement, the light intensity was calibrated using a reference Si solar cell certified by the International System of Units (SI) (SRC-1000-TC-KG5-N, VLSI Standards, Inc).



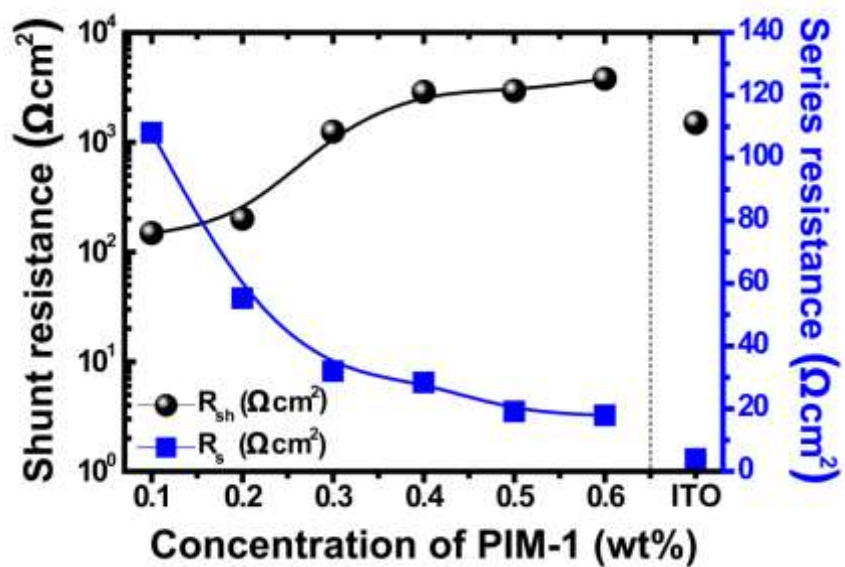
**Figure S1.** (a) The PIM-1 structure used as a CNS precursor. (b) Chemical structure and <sup>1</sup>H NMR spectra of 2-5 wt% PIM-1.



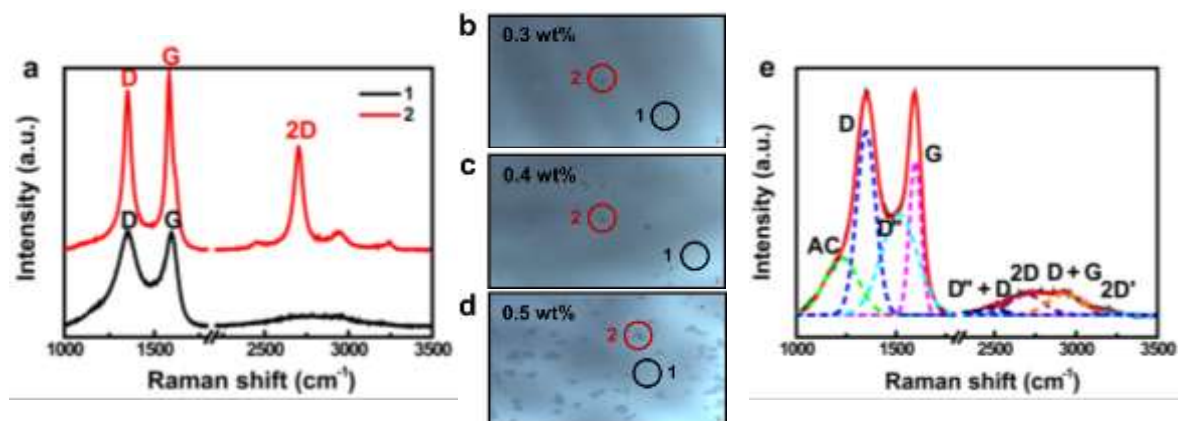
**Figure S2.** AFM images of CNS derived PIM-1 polymers: (a) 0.1, (b) 0.2, (c) 0.3, (d) 0.4, (e) 0.5, and (f) 0.6 wt% PIM-1 solutions.



**Figure S3.** TGA analysis of PIM-1 under N<sub>2</sub> gas flow.



**Figure S4.** The shunt resistance and series resistance of CNSs as a function of PIM-1 concentration.



**Figure S5.** (a) Two representative Raman spectra of 0.3 wt%-converted CNSs. (b-d) The Raman optical images of 0.3, 0.4, and 0.5 wt%-converted CNSs. The spectrum at the position of the red circle indicates that the graphitic structure of CNSs is improved compared to the others. The area of the improved structure increases with increasing the PIM-1 concentration. (e) The deconvoluted Raman spectrum (bottom curve in Figure S5-a) of a CNS derived from a 0.3 wt% PIM-1 solution.



**Table S1.** The atomic concentration for the CNS as a function of PIM-1 concentration.

| Polymer contents<br>[wt%] | Atomic concentration [%] |   |       |       |
|---------------------------|--------------------------|---|-------|-------|
|                           | C                        | N | O     | Si    |
| 0.1                       | 72.28                    | 0 | 16.48 | 11.24 |
| 0.2                       | 93.19                    | 0 | 3.67  | 3.14  |
| 0.3                       | 96.60                    | 0 | 1.90  | 1.50  |
| 0.4                       | 98.50                    | 0 | 1.04  | 0.46  |
| 0.5                       | 98.56                    | 0 | 1.05  | 0.39  |
| 0.6                       | 97.98                    | 0 | 1.60  | 0.42  |