

## Electronic Supplementary Information

### Generation of open ended, worm like and graphene like structures from spherical carbon layered materials

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#### 1.0 Experimental

##### 1.1 Addition of polyvinylpyrrolidone (PVP) to polydispersed SiO<sub>2</sub> spheres and synthesis of hollow carbon nanostructures

The polydispersed silica spheres (0.5 g) were weighed and dispersed in 40 mL of ethanol and stirred for twenty minutes. Polyvinylpyrrolidone (PVP) solution was prepared by dispersing 0.5 g of PVP in 20 mL of distilled water and sonicating for ten minutes. The PVP solution was added to the silica suspension and stirred for 1 h and 12 hours respectively. The solutions were centrifuged at 4500 rpm for 20 minutes and the products dried at 80 °C for 12 hours to give SiO<sub>2</sub>@PVP 1 h and SiO<sub>2</sub>@PVP 12 h respectively. In separate reactors, the two SiO<sub>2</sub>@PVP samples (0.06 g) were uniformly spread onto a quartz boat which was placed in the center of a quartz tube. The furnace was heated to 900 °C at 10 °C min<sup>-1</sup> under an Ar atmosphere (Ar, 200 sccm). Once the desired temperature was reached, Ar (200 sccm) was bubbled through toluene for 1 h. After this, the gas flow was stopped and the system was left to cool down to room temperature under an inert atmosphere (Ar, 200 sccm). The quartz boat was then removed from the reactor and the silica was removed with a 10

% HF solution (24 hours) and after thorough washing with distilled water the product was dried at 80 °C for 12 hours to give the two desired hollow carbon nanostructures.

### **1.2 Fabrication of a ternary blend active layer based organic solar cell**

The devices were fabricated on top of ITO-coated substrates. The ITO coated glass was cleaned in an ultrasonic bath by sequential treatments with deionized water, acetone, and ethanol and an ethanol/water mixture for 30 minutes before solvent removal by a stream of nitrogen gas. A hole transport layer, PEDOT:PSS (poly(3,4-ethylenedioxylenethiophene):poly(styrenesulfonic acid)) was then spin coated onto the ITO coated substrate at 2000 rpm for 30 seconds. The resulting film was thermally treated on a hot plate at 70 °C for 15 minutes. Two sets of P3HT and PCBM samples (10 mg each) were separately dissolved in 0.5 mL of chlorobenzene and stirred in air for 3 hours at room temperature. The hollow carbon nanostructure solutions were prepared by dissolving 1 mg of wormlike hollow carbon structures and broken hollow carbon structures separately in 0.5 mL of chlorobenzene.

The active layer was prepared by mixing the solutions of P3HT:PCBM:hollow carbon nanostructures in (1:1:0.1) weight ratio to form two different blend solutions and stirred for 2 hours. The active layer was then spin coated at 2000 rpm for 30 seconds on top of the PEDOT:PSS thin film and the cathode was deposited by thermal evaporation of aluminium under  $8.0 \times 10^{-6}$  mbar. Optical absorption measurements of the films made from the blends were carried out using a Cary 500 UV–visible Spectrometer. The surface roughness and topographic images of the films were studied using a Veeco Di3100 AFM in tapping mode installed with a  $\text{Si}_3\text{N}_4$  tip. The current density–voltage (J–V) characteristics of the films were obtained using a HP 4141B source measure unit under  $100 \text{ mW/cm}^2$  illumination (AM 1.5G). All the measurements were carried out at room temperature under standard conditions.

## 2.0 Physisorption analysis of the wormlike hollow carbon nanostructures

The surface area and porosity data were measured of the wormlike hollow carbon nanostructure sample by using a Micromeritics Tristar 3000 instrument. The sample was degassed at 250 °C in N<sub>2</sub> for 12 h. The specific surface area was calculated by the BET method from N<sub>2</sub> adsorption data and the BJH pore size distributions were calculated by analysing the desorption branches of the N<sub>2</sub> isotherms.

Fig S9 indicates the Nitrogen adsorption-desorption isotherm of the open ended wormlike hollow carbon nanostructures that can be classified as a type (III) adsorption isotherm according to the International Union of Pure and Applied Chemistry (IUPAC) nomenclature. The measured isotherm exhibited a H3 hysteresis loop at a relative pressure of  $P/P_0 = 0.8 - 1.0$  indicating assemblage of slit shaped pores or platelike particles.<sup>1</sup> Consequently, the material exhibited a BET surface area of 98 m<sup>2</sup>/g and a pore volume of 0.64 cm<sup>3</sup>/g. Fig. S9 inset shows the pore size distribution calculated by the Barrett-Joyner- Halenda (BJH) method with a sharp peak observed at 23.4 nm; characteristic of mesopores and a broad pore size distribution in the macroporous region (50 to 100 nm). This indicated the coexistence of hierarchical mesopores and macropores derived from the hollow interconnected network and interparticle voids in the sample.<sup>2, 3</sup> Indeed, the presence of non-spherical hollow cavities in carbon nanomaterials have been reported to reduce the charge transport pathway in electrochemical devices.<sup>4</sup>

## 2.1 Energy Dispersive X-ray Spectroscopy (EDS) analysis

The energy dispersive X-ray spectroscopy (EDS) analysis was done to check the elemental composition of the synthesized carbon nanostructures. The EDS spectrum of the wormlike hollow carbon nanostructure shows a high carbon peak and a negligible oxygen peak (Fig. S10). This confirmed the complete removal of silica during etching.

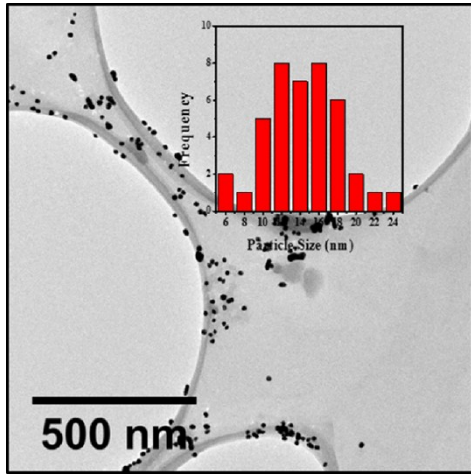


Fig. S1 TEM image of synthesized gold nanoparticles.

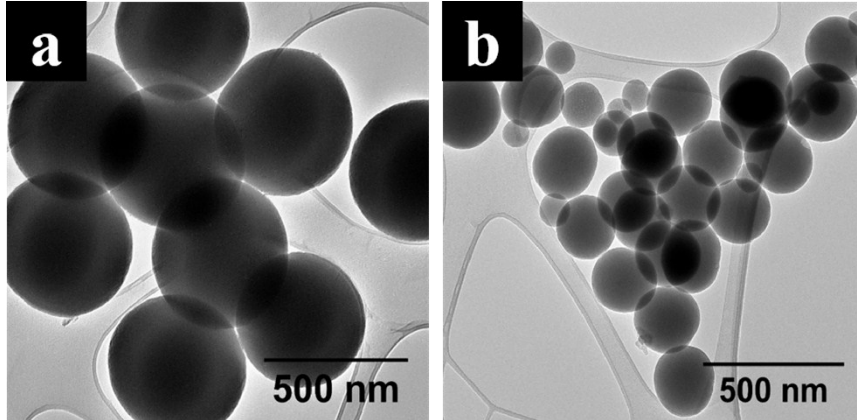


Fig. S2 TEM images of (a) monodispersed SiO<sub>2</sub> spheres and (b) polydispersed SiO<sub>2</sub> spheres.

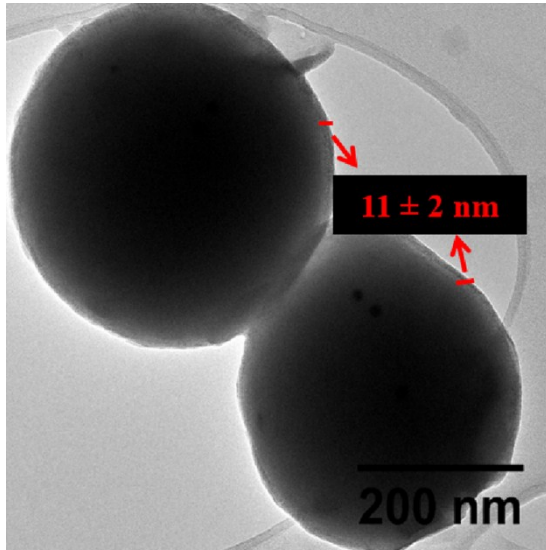


Fig. S3 TEM image of Au@SiO<sub>2</sub>C@C1 after 1 h carbonization time.

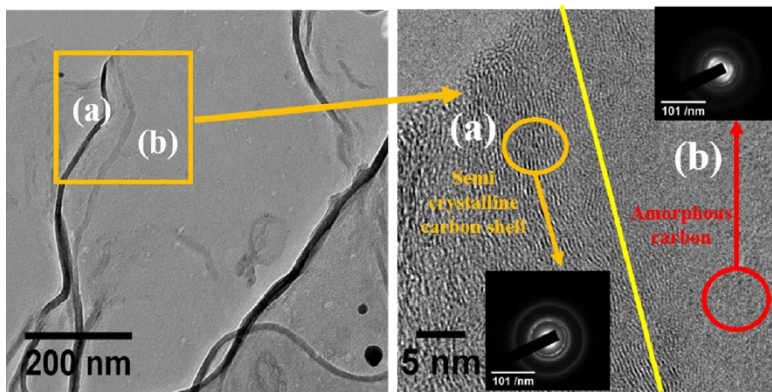


Fig. S4 HRTEM images of Au@HCSC1; (a) carbon shell which is semicrystalline as shown by the presence of diffuse rings with tiny spots (see SAED pattern) and (b) the amorphous carbon.

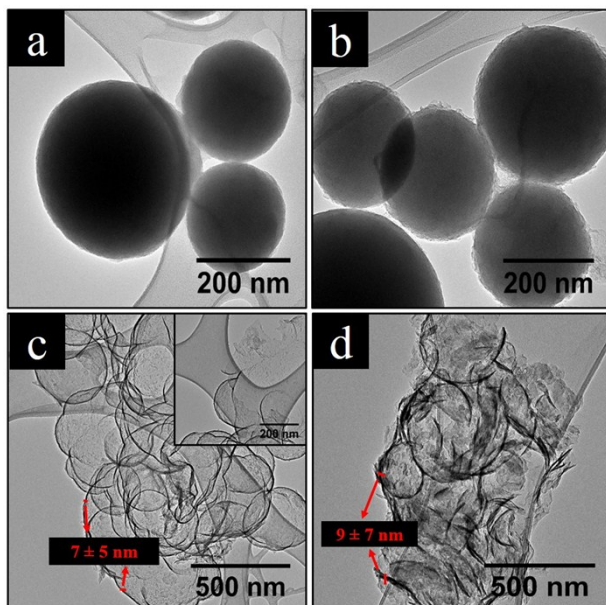


Fig. S5 TEM images of polydispersed SiO<sub>2</sub> spheres mixed with PVP (a) 1 h and (b) 12 h stirring times and the hollow carbon nanostructures obtained from (c) SiO<sub>2</sub>@PVP 1 h and (d) SiO<sub>2</sub>@PVP 12 h after 1 h carbonization time and SiO<sub>2</sub> removal respectively.

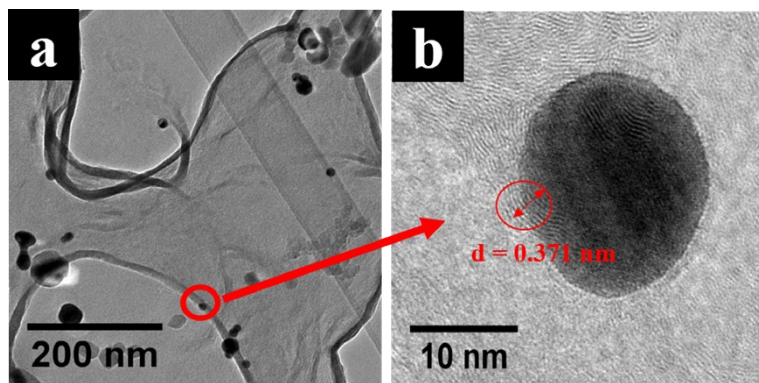


Fig. S6 HRTEM images of Au embedded in Au@HCSC1.

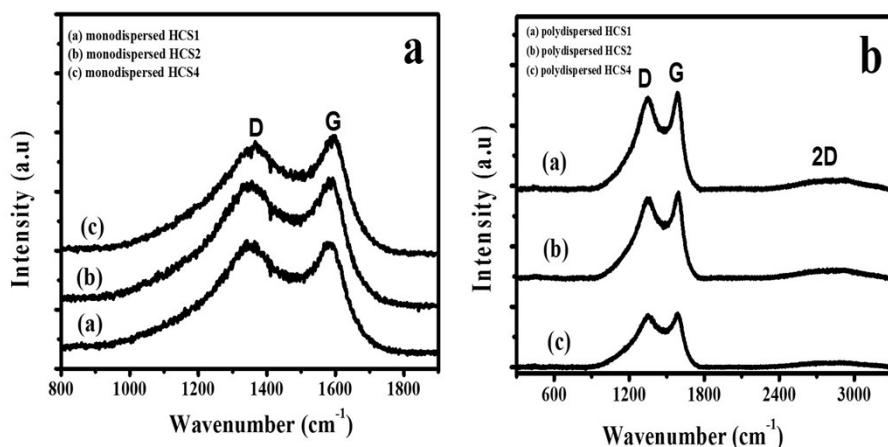


Fig. S7. Raman spectra of HCSs obtained from (a) monodispersed SiO<sub>2</sub> and (b) polydispersed SiO<sub>2</sub> spheres after (1 h, 2 h, 4 h) carbonization times and SiO<sub>2</sub> removal respectively.

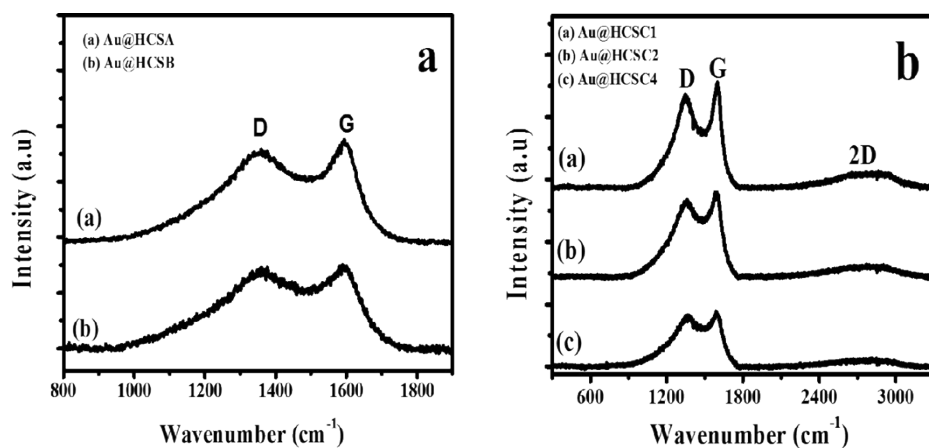


Fig. S8 Raman spectra of Au@HCSs obtained from (a) monodispersed Au@SiO<sub>2</sub> spheres after 1 h carbonization time and SiO<sub>2</sub> removal and (b) polydispersed Au@SiO<sub>2</sub> spheres after (1 h, 2 h, 4 h) carbonization times and SiO<sub>2</sub> removal respectively.

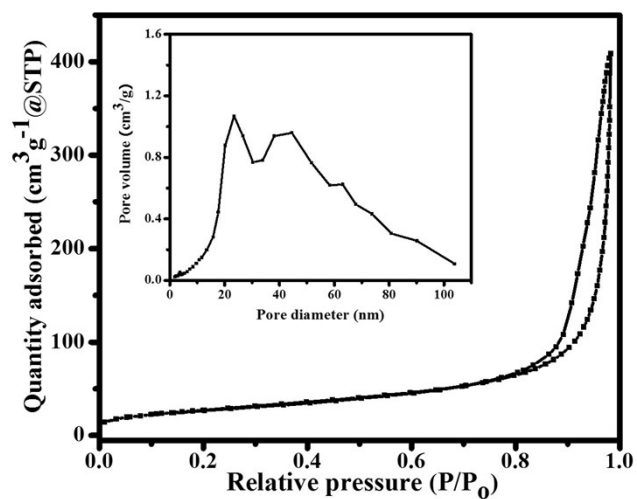


Fig. S9 Adsorption and desorption curve of the wormlike carbon nanostructure. Inset shows the pore size distribution.

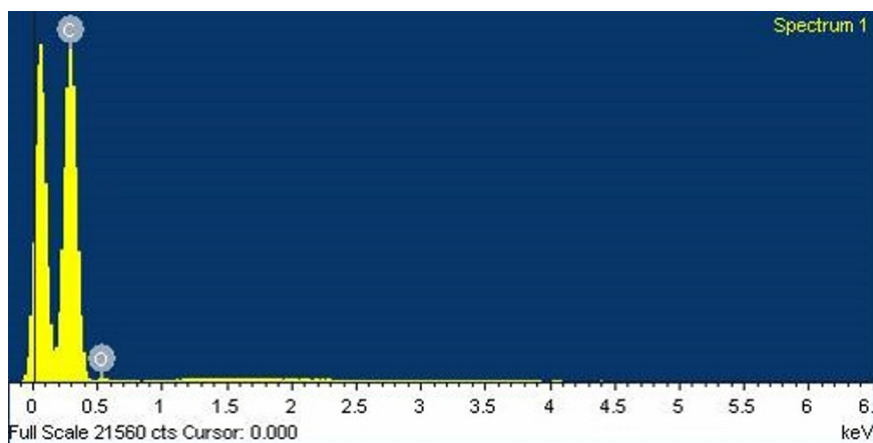


Fig. S10 EDS spectra of the obtained wormlike hollow carbon nanostructures.



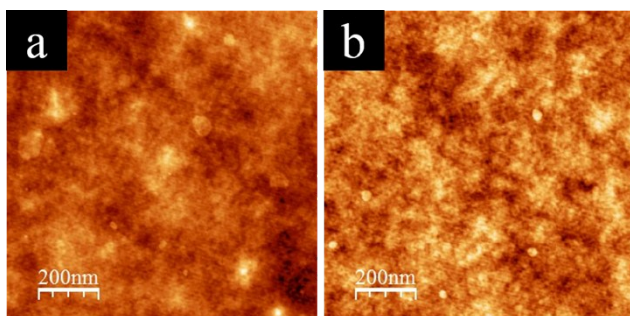


Fig. S11 AFM topographic images of (a) P3HT:PCBM:wormlike HCSs and (b) P3HT:PCBM:broken HCSs films.

Table S1  $I_D/I_G$  ratios of HCSs synthesized at different carbonization time

Description	D band position ( $\text{cm}^{-1}$ )	G band position ( $\text{cm}^{-1}$ )	$I_D/I_G$ ratio
monodispersed HCS1	1356	1592	0.97
monodispersed HCS2	1363	1589	1.00
monodispersed HCS4	1342	1575	1.01
polydispersed HCS1	1349	1583	0.95
polydispersed HCS2	1364	1593	0.93
polydispersed HCS4	1350	1587	0.97

Table S2 I<sub>D</sub>/I<sub>G</sub> ratios of Au@HCSs synthesized at different carbonization time

Description	D band position (cm <sup>-1</sup> )	G band position (cm <sup>-1</sup> )	I <sub>D</sub> /I <sub>G</sub> ratio
Au@HCSA	1359	1593	0.90
Au@HCSB	1376	1588	0.97
Au@HCSC1	1370	1590	0.91
Au@HCSC2	1363	1588	0.92
Au@HCSC4	1365	1585	1.03

## References

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