SUPPOTING INFORMATION

Formation of C₆₀ fullerene-bonded-CNTs by radio frequency plasma

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Fig. S1 Schematic representation of carbon nano-peapod.

Description of CVD method for the synthesis of CNBs structure.

A schematic presentation of CVD method is given as follows.¹ Typically, catalyst particles were grown in situ via ferrocene vapor decomposition (Scheme S1). The precursor was vaporized by passing ambient temperature CO (with a flow rate of 300 cm³·min⁻¹) through a cartridge filled with ferrocene powder. The partial vapor pressure of ferrocene in the reactor was maintained at 0.7 Pa. The flow containing ferrocene vapor was then introduced into the high temperature zone of the ceramic tube reactor through a water-cooled probe and mixed with additional CO (100 cm³·min⁻¹). The reactor wall temperature was varied from 800 to 1150 $^{\circ}$ C. Meanwhile, controlled amounts of H₂O and CO₂ were also added together with the carbon source via a mass flow controller to alter the density of fullerenes on the CNTs. In order to introduce the H₂O vapor, a flow of a carrier gas was passed through a water saturation vessel. The amount of H_2O was varied from 0 to 405 ppm. The concentration of introduced CO₂ was varied from 0 to 12000 ppm. The carrier gas water vapor saturation conditions were monitored on-line by a Gasmet Fouriertransform infrared (FT-IR) spectrometer. And nano-buds were collected downstream of the reactor either by filtering on silver or nitrocellulose disk filters or by depositing on TEM grids via an electrostatic precipitator (ESP).



Scheme S1 Schematic presentation of the experimental setup for the preparation of CNBs.

Materials and characterization of the CNBs structure.

Both CNTs and C₆₀ fullerenes were purchased from Alfa Aesar. The materials were treated with hydrochloric acid (HCl) to remove the metal catalysts before radio frequency plasma (RF plasma) treatment. High-resolution transmission electron microscopy (HRTEM) photographs were taken on a high-resolution transmission electron microscope (HRTEM, Tecnai Model JEOL-2010) at an accelerating voltage of 200 kV. The Raman spectra were recorded at ambient temperature on a SPEX 1403 spectrometer with an argon-ion laser at excitation energy of 2.33 eV.

Detailed synthesis of CNBs.

In a typical method, CNTs was firstly mixed with C_{60} fullerenes by grinding with their mass ratio of 1:1 using a mortar. Secondly, the whole mixture was transferred to a glass flask and the HV pulsed dc power with 100 W was applied to the plasma coil until N₂ vacuum degree of the glass flask reached a relative stable state about 3.9 Pa. The whole mixture was then treated under this condition for about 1 hour. Subsequently, 20 mL carbon disulphide (CS₂) was injected into the glass flask, which was then continuously stirred for about 24 h under ambient temperature. In the end, the sample was collected and washed several times with CS_2 to remove the unreacted C_{60} . With no extra post-synthetic process, the purified sample was dried in a vacuum oven at 60 °C for 12 hours, some of which was dispersed in CS_2 and sonicated before transferring onto TEM grids. And the schematic and specific experimental setups were shown in Fig. S2 and Fig. S3, respectively.



Fig. S2 Schematic view of the experiment setup for the synthesis of CNBs.



Fig. S3 Specific experimental setup for the synthesis of CNBs.



Fig. S4 (a) and (b): HRTEM images of CNBs obtained for 30 min and 120 min,

respectively.



Fig. S5 (a) and (b): HRTEM images of CNBs obtained with C_{60} : CNTs ratios of 0.5 and



2, respectively.

Fig. S6 TEM image of CNTs after plasma treatment.





Fig. S7 TEM (a) and HRTEM (b) images of C_{60} fullerenes after plasma treatment.

Fig. S8 The OES (optical emission spectroscopy) of N_2 plasma.