Electronic Supplementary Information (ESI)

Dramatically enhancing the yield of carbon nanotubes by simply adding oxygen-containing molecules in solid-state synthesis

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EXPERIMENTAL SECTION

Materials and Instrumentation.

Organometallic compound **M** was synthesized in our previous work. All other reagents were used as received without further purification. Transmission electron microscopy (TEM) was performed on a JEM-2010HT or JEM-2010FEF microscope at an accelerating voltage of 200 kV. TEM samples were prepared by drying a droplet of the suspension on a TEM copper grid with a carbon film. Scanning electron microscopy (SEM) was performed on a ZEISS-SIGMA scanning electron microscope. Energy-dispersive X-ray spectroscopy (EDS) was taken on the SEM. The X-ray diffraction (XRD) analyses were performed on a Bruker D8 Advanced X-ray diffractometer with CuKα radiation (λ=1.5418 Å). Raman spectroscopy was performed on a RM-1000 Confocal Raman Microspectroscopy (Renishaw Company) using 514.5 nm laser excitation.

Pyrolysis Process: Powders of the organometallic precursor M and different oxygen-containing molecules were put together and mixed uniformly. Then, the mixed powders were placed in quartz tubes, respectively. These tubes were sealed under high vacuum and placed into the furnace, and underwent the same heating programs. They were first heated slowly to 180 ^{o}C (1.5 ^{o}C min^{-1}), held for two hours to ensure the complete decomposition of the Co complexes, then to a higher temperature (700 ^{o}C, 2.2 ^{o}C min^{-1}), held for eight hours. After slowly cooled to room temperature, the obtained products were characterized by TEM, SEM, EDS and powder X-ray diffraction. Because the production of gas in a sealed space, so it need to control the amount of the additive feedstrocks. In this work, the utilized quartz tubes are 12 cm in length with an internal diameter of 1 cm, for safety, the total mass of feedstrocks should better less than 120 mg.

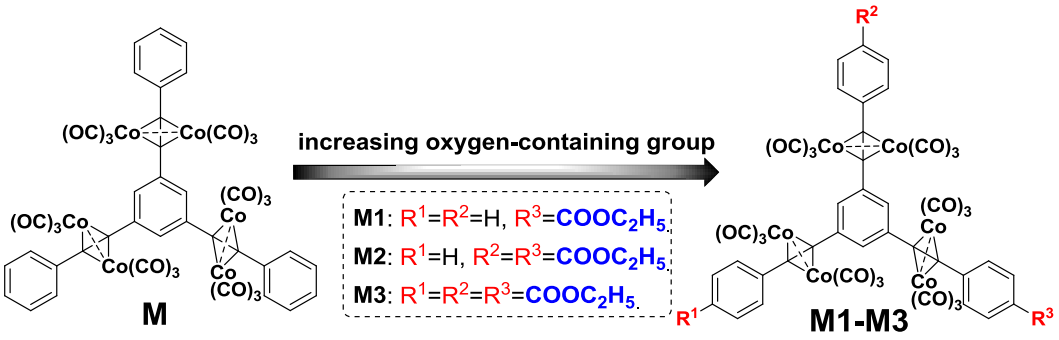

Chart S1. Chemical structures of the organometallic compounds M and M1-M3.

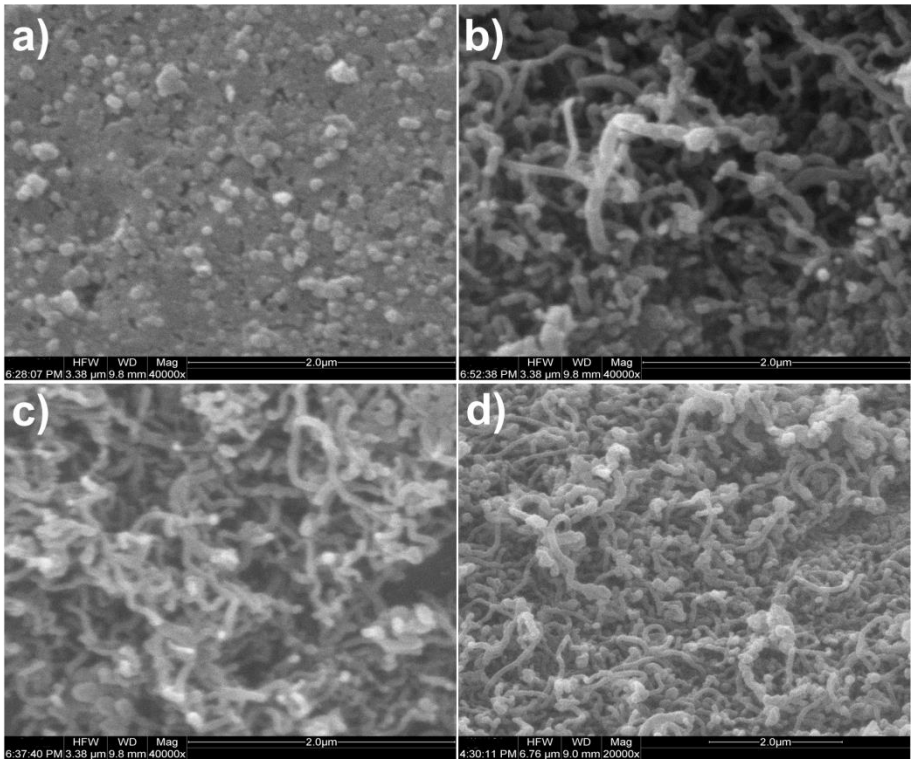

Figure S1. SEM (a, b, c, d) images of the materials obtained through thermolysis of compounds M (a), M1 (b), M2 (c) and M3 (d).

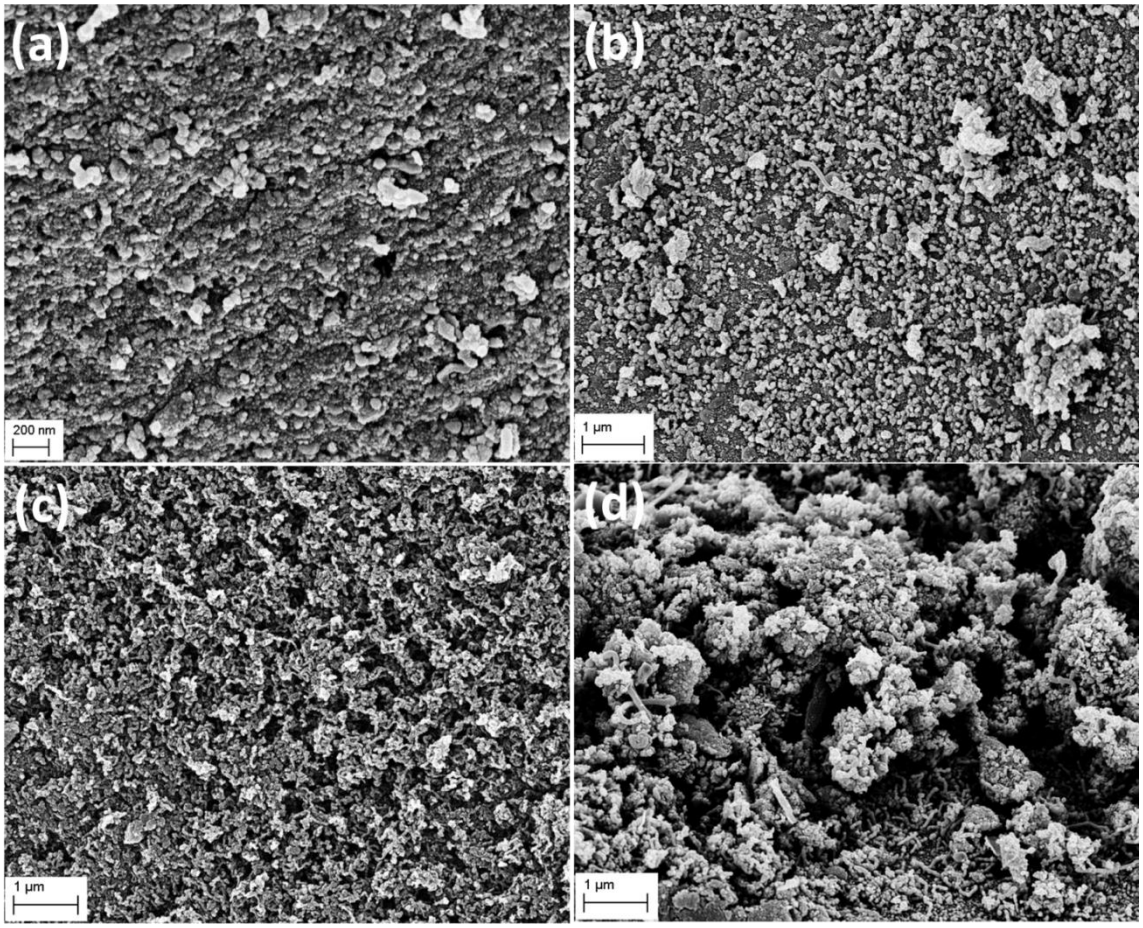

Figure S2. SEM images of the materials obtained through thermolysis of **M** with different amount of **A1**. The molar ratio of oxygen to **M** was 3:1 (b), 6:1 (c) and 12:1 (d). Pyrolytic produce obtained from **M** without addition of **A1** was shown as a contrast (a).

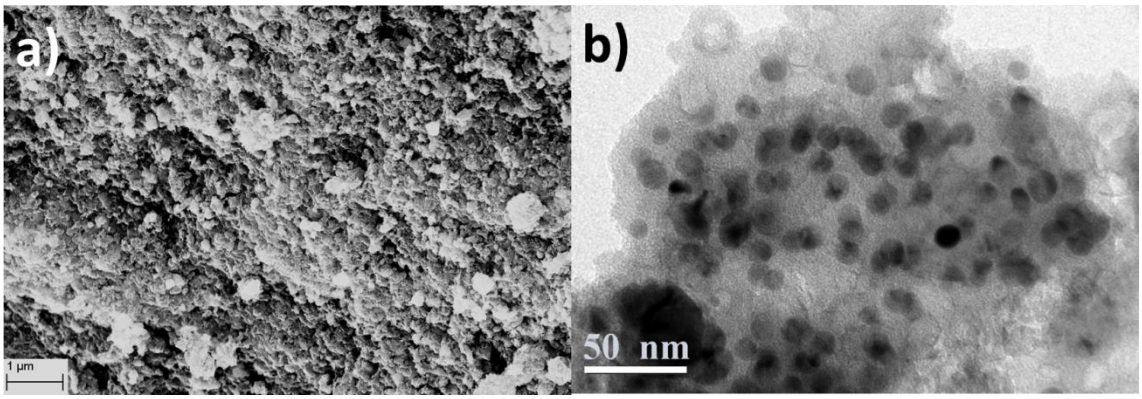

Figure S3. SEM (a) and TEM (b) images of the materials obtained through thermolysis of **M** with the addition of **D**.

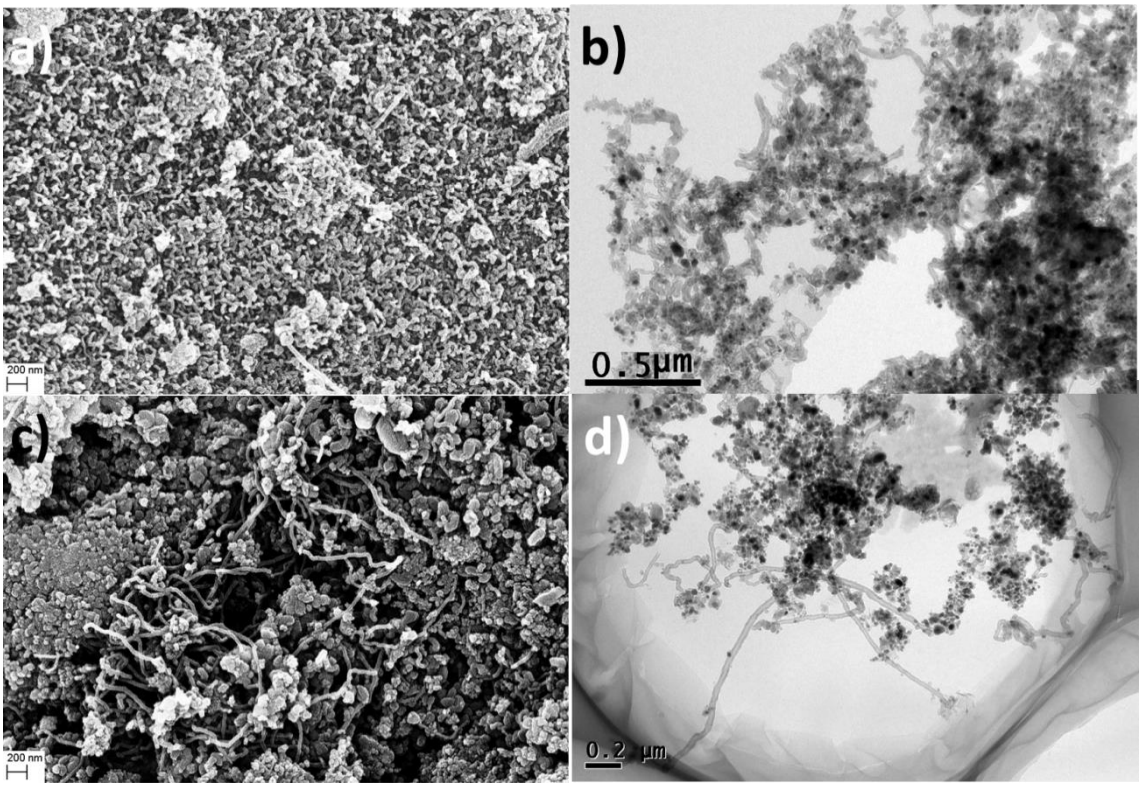

Figure S4. SEM (a, c) and TEM (b, d) images of the materials obtained through thermolysis of **M** with the addition of **C1** and **C2**. (a, b) for **M-C1**, (c, d) for **M-C2**.

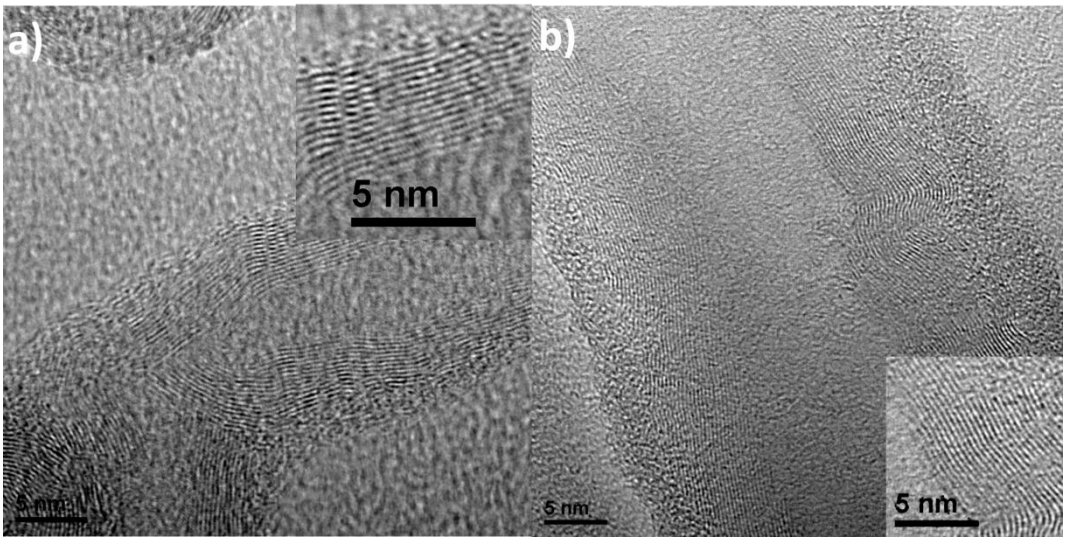

Figure S5. HRTEM images of the materials of **M-C1** (a) and **M-C2** (b)**.**

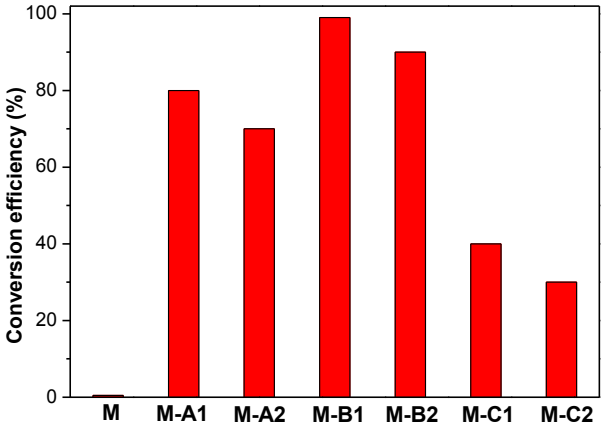

Figure S6. Conversion efficiency of carbon feedstocks to carbon nanotubes for **M** without/with different oxygen-containing molecules. Conversion efficiency of carbon means the conversion rates of carbon in feedstocks (deducted 'CO' from the $[Co_2(CO)_6]$ units in organometallic precursor) to the CNTs. The value was evaluated by calculating their mass recovery of the crude carbon material and comparing their SEM and TEM images.

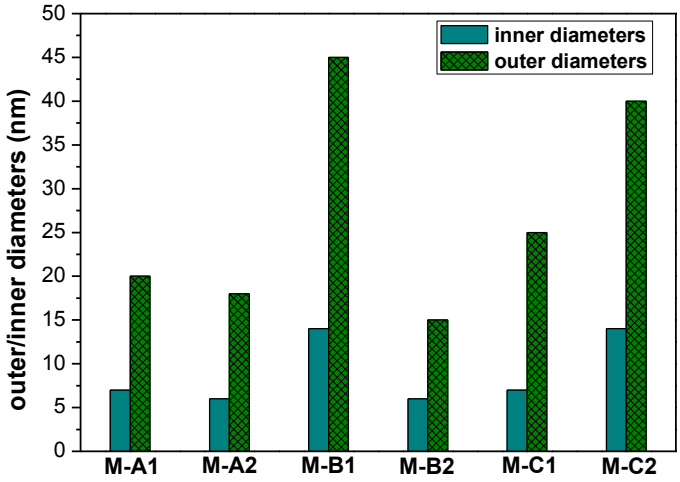

Figure S7. Outer and inner diameters of the obtained CNTs.

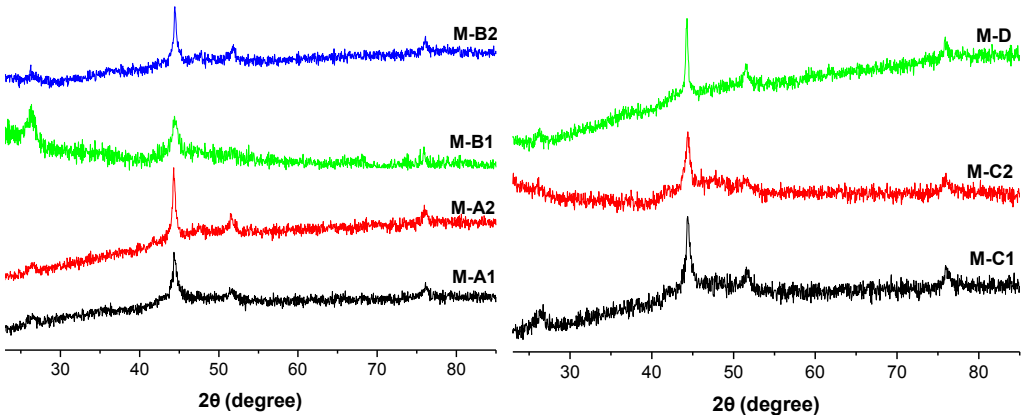

Figure S8. XRD diffractograms of the materials obtained through thermolysis of **M** with the addition of other compounds.

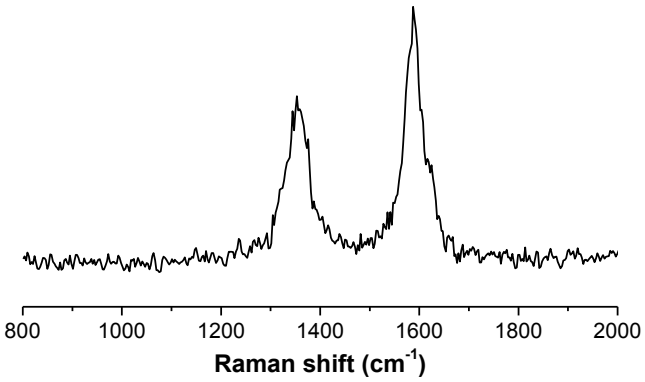

Figure S9. Raman spectrum of **M-B1**.

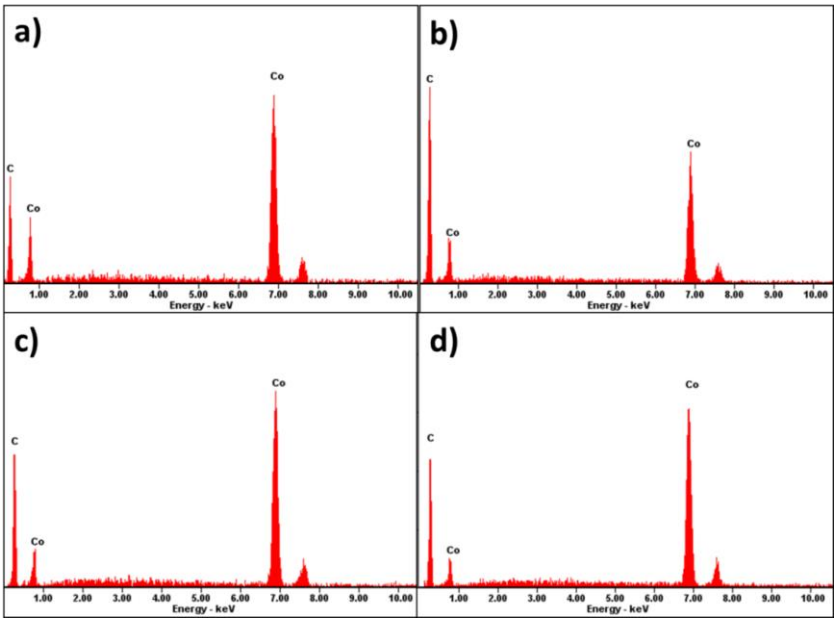

Figure S10. SEM-EDX spectra of (a) **Ms**, (b) **M-A1**, (c) **M-A2** and (d) **M-D**.

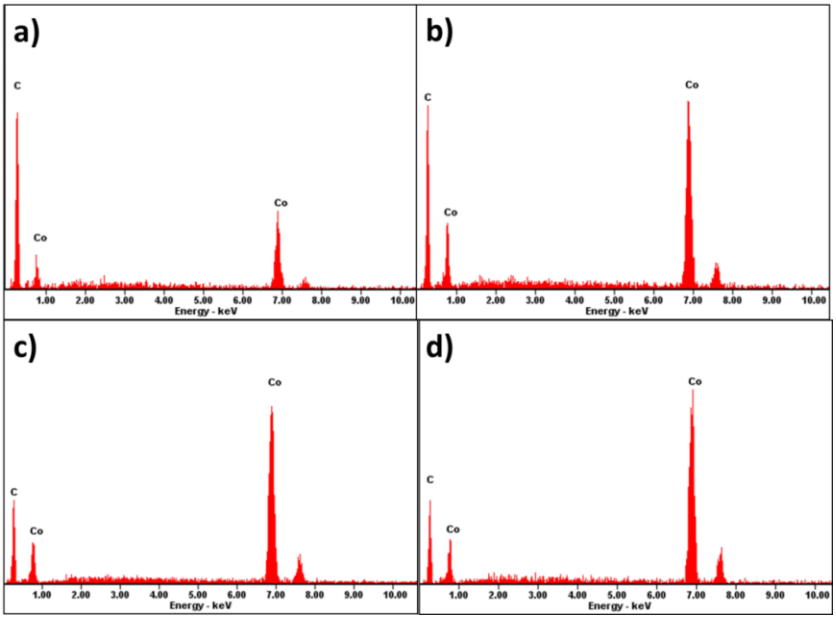

Figure S11. SEM-EDX spectra of (a) **M-B1**, (b) **M-B2**, (c) **M-C1** and (d) **M-C2**.

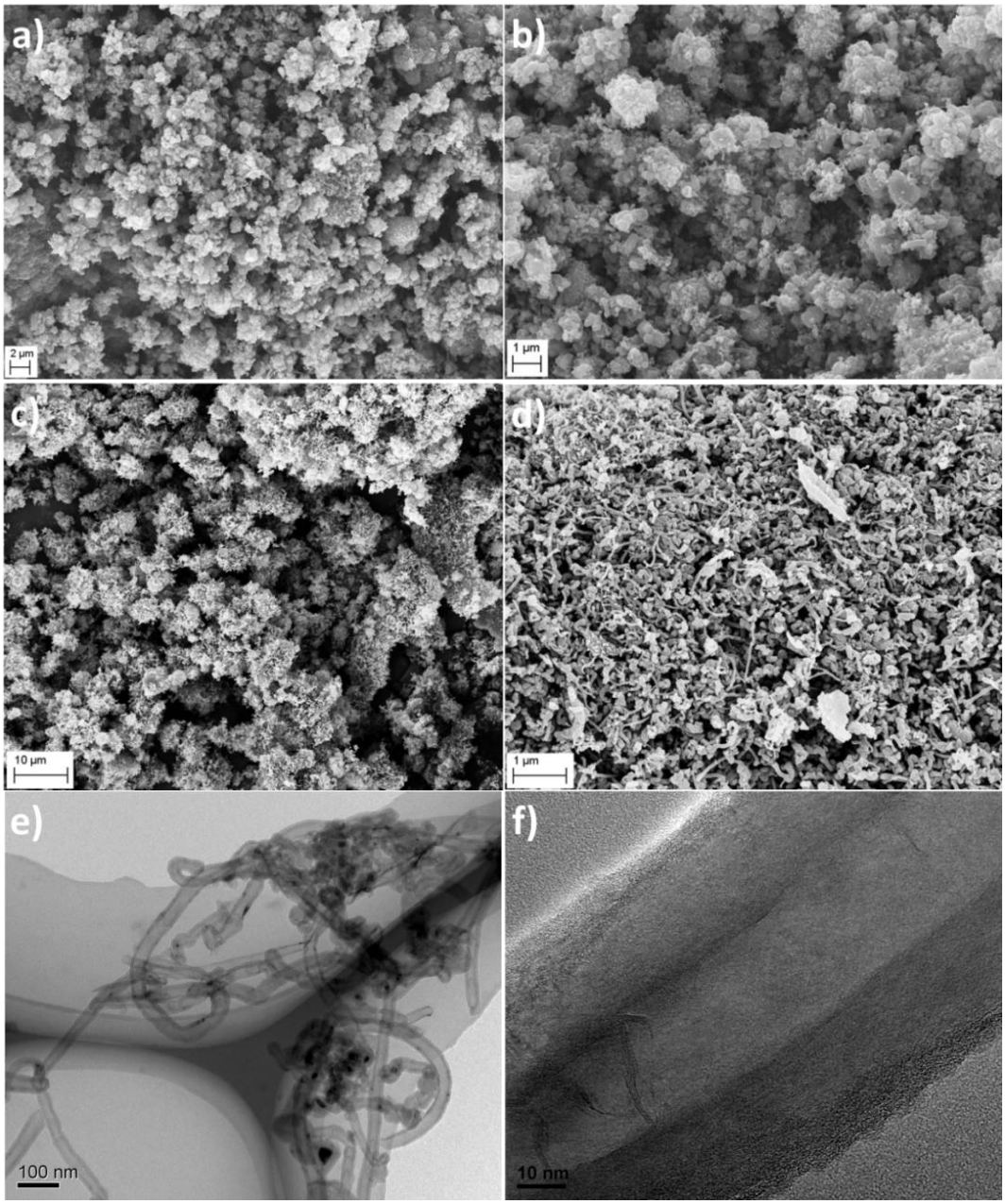

Figure S12. SEM (a-d) and TEM (e, f) images of the materials obtained through thermolysis of pure ferrocene (a, b) and ferrocene/**B1** mixture (c, d, e, f).

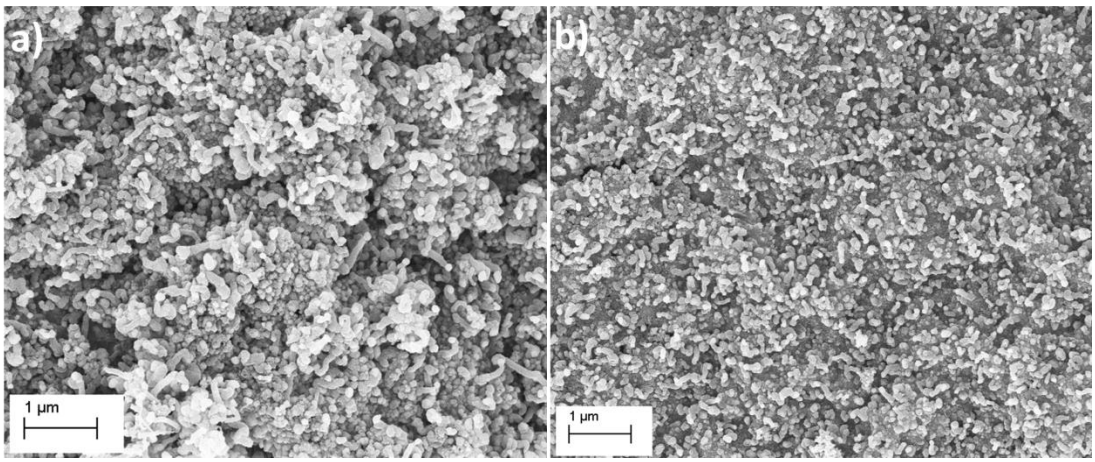

Figure S13. SEM images of the materials obtained through pyrolysis of $Co_2(CO)_8$/B1/1,3,5-tris(phenylethynyl)benzene mixture (a) and $Co_2(CO)_8$/B1/ biphenyl mixture (b).

Table S1. CNTs yield and the outer/inner diameter of the produced CNTs with different feedstocks.

Sample name	Feedstocks (mg) M : OCMs[a]	CNTs yield (%)[b]	Outer/inner diameter (nm)
Ms	40 : 0	0	----
M-A1	40 : 9.6	~80	7，20
M-A2	40 : 6.3	~70	6，18
M-B1	40 : 9.8	~100	14，45
M-B2	40 : 5.7	~90	6，15
M-C1	40 : 8.1	~40	7，25
M-C2	40 : 4.4	~30	14，40
M-D	40 : 9.6	0	----

[a] OCMs: oxygen-containing molecules; A1: phthalic anhydride, A2: maleic anhydride, B1: methyl 4-hydroxybenzoate, B2: dimethyl oxalate, C1: phthalic acid, C2: oxalic acid (D: biphenyl). [b] CO in $[Co_2(CO)_6]$ moieties in organometallic precursor M was deducted when calculated the yield of carbon nanotubes.