Supporting Information

One-Step Solid-State Thermolysis of a Metal-organic Framework: A Simple and Facile Route to Large-Scale of Multi-walled Carbon Nanotubes

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Experimental Section

Synthesis. In a typical synthesis, the samples were obtained by the following method. Powders of the precursor [Ni₃(btc)₂·12H₂O]ₙ (about 0.3 g) were placed in a ceramic boat (with inside diameter of 0.6 cm and length of 5 cm). First, the ceramic boats were placed the middle of a quartz tube (with inside diameter of 3 cm and length of 40 cm) along the axis of the tube with one end of the tube closed. Then the tube was heated to different temperature with the heating ratio of 10 °C·min⁻¹ in a horizontal tube furnace (in Figure S0) over the different course of time. After slow cooling to room temperature, pale products appeared in the left of the quartz-boat, and fiber-like black powder, which was obtained without further purity, came forth in the right and wall of the quartz-boat.

Figure S0. The sketch map of the tube furnace used for thermolysis.

Characterization. Raman spectra (Raman) were measured on a France JY HR-800 confocal laser micro Raman spectrometer. Powder X-ray diffraction (XRD) patterns were collected in a Shimadzu XRD-6000 (operating at 40 kV and 30 mA) with graphite-monochromatized Cu Kα radiation (wavelength λ = 1.5147 Å). Field emission scanning electron microscopy (FE-SEM) studies were conducted with a well-aligned LEO1530VP SEM (Carl Zeiss INC.) operating at 200 kV and with an Oxford INCA energy dispersive X-ray analysis (EDX) to fulfill element microanalysis. Transmission electron microscopy (TEM) studies were operated on a JEOL 2010 TEM using at an accelerating voltage of 200 kV. Thermogravimetric (TG) analysis was displayed on PerKinElmer Pyris 1 TGA under atmospheric pressure with N₂ as the carrier gas. Thermogravimetry-mass spectrometry (TGA-MS) was performed on a NETZSCH STA-499C-Thermal Star 300 thermal analyzer.
**Figure S1.** XRD patterns of the Ni-MOF.
Figure S2. (a) TGA and (b) DTG curves of the Ni-MOF in N₂ flow with the heating rate of 10 °C·min⁻¹.
Figure S3. Photography of (a) the Ni-MOF and (b) as-synthesized products by thermolysis of the Ni-MOF at 500 °C for 20 h in the one-end closed tube furnace.

(a) the Ni-MOF:

(b) Products after thermolysis:
**Figure S4.** XRD patterns of three different products with color changing from argent to pale and black and from left to right (a) - (c) at the place in the ceramic boat where the Ni-MOF was placed at 400 °C for 20 h.
Figure S5. XRD patterns of the synthesized MWCNTs by thermolysis of the Ni-MOF in the one-end closed horizontal tube furnace at 500 °C for (a) 5 h, (b) 10 h, (c) 15 h, and (d) 20 h.
Figure S6. EDX analysis of as-synthesized MCNTs by thermolysis of the Ni-MOF in the one-end closed horizontal tube furnace at 500 °C for (a) 5 h, (b) 10 h, and (c) 20 h.
**Figure S7.** FE-SEM image and EDX of the remained Ni particles after thermolysis of the Ni-MOF in the one-end closed horizontal tube furnace at 500 °C for 20 h.
Reference
