Supporting Information

High Temperature Selective Growth of Single-Walled Carbon Nanotubes with a Narrow Chirality Distribution from CoPt Bimetallic Catalyst

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S1. Catalyst preparation.

We prepared silica-supported Co and CoPt catalysts using an impregnation method. For the preparation of Co catalyst, cobalt (II) acetate tetrahydrate ($Co(CH_3COO)_2 \cdot 4H_2O$, 0.228 g) was first added into ethanol (20 ml) and sonicated for 10 min. Porous silica supports (1 g, 7 nm in average particle size and BET surface area of 390 ± 40 m^2/g, Sigma Aldrich) were then added to the above solution to make the silica-supported Co catalyst. For the preparation of CoPt bimetallic catalyst, $Co(CH_3COO)_2 \cdot 4H_2O$ (0.120 g) was first added into ethanol (15 ml) and sonicated for 10 min. Then, ethanol solution of hexachloroplatinic (IV) acid hexahydrate ($H_2PtCl_6 \cdot 6H_2O$, 0.097 mol/L, 5 ml) was added into the above solution and the mixture was sonicated for another 10 min. After that, silica supports (0.5 g) were added to make the silica-supported CoPt bimetallic catalysts with Co:Pt molar ratio of 1:1. For both catalysts, the silica supports together with the metal solution were further sonicated for 1 h and baked at 80 °C for 12 h to evaporate the ethanol. Finally, the obtained pink (for Co) and dark green (for CoPt) blocks were ground in a mortar for 15 min to obtain fine powders, which were used as catalysts for subsequent single-walled carbon nanotube (SWCNT) growth.

S2. SWCNT growth.

For the growth of SWCNTs, ~30 mg of catalyst powders were put into a quartz boat, which was then placed at the center of a horizontal chemical vapor deposition (CVD) tubular furnace with a 25-mm-diameter quartz tube. The furnace temperature was raised to 800 °C (or 850 °C) under the protection of Ar (200 sccm) flow. The catalyst was then reduced at the growth temperature for 4 min under Ar (500 sccm) and H_2 (200 sccm) flow. After that, H_2 was turned off and Ar was switched to bubble ethanol into the CVD furnace with a flow rate of 200 sccm. The ethanol was kept in a container at a constant temperature of 75 °C and the SWCNT growth was performed at atmospheric pressure. After SWCNT growth for 20 min, the furnace was cooled down under Ar (500 sccm) and H_2 (200 sccm) flow.

S3. Materials characterization.

The catalyst powders were characterized using X-ray diffraction (XRD, Rigaku diffractometer with CuKα radiation). The as-grown SWCNTs were systematically investigated using multi-laser resonant Raman spectroscopy (Horiba Jobin Yvon

HR800 with 532 nm, 633 nm, and 785 nm lasers, and the laser power below 2 mW) and transmission electron microscopy (TEM, Tecnai G^2 F30, 300 kV). The silica supports were removed using a 30% HF acid before scanning electron microscopy (SEM, Nova Nano SEM 430, 15 kV) observations.

For optical absorption and emission measurements, ~20 mg of as grown SWCNTs together with the silica supports were added into 5 ml D_2O containing 2% (wt: v) sodium cholate hydrate (SC) as surfactant. The mixture was tip sonicated for 2 h with a corn-horn sonicator (900 W, with 20% power output and a tip radius of 3 mm, SCIENTZ-II D). After sonication, the resulting black suspension was centrifugated at 16,000 rpm (18,140 g) for 2 h (Fulgor GL-20B). The temperature was kept below 20 °C during the sonication and centrifugation processes. The upper 70% of the supernatant was carefully decanted and used for further measurements. UV-vis-NIR absorption measurements were carried out in a 1 cm path-length quartz cell using a Varian Cary 5000 spectrophotometer in the range of 200-1900 nm with D_2O containing 2% (wt: v) SC as reference. The same suspension was further used for photoluminescence excitation/emission (PLE) measurements on a Jobin-Yvon Nanolog-3 spectrofluorometer. The excitation wavelength scanned from 500 to 770 nm and the emission spectra was collected from 900 to 1600 nm with 5 nm steps for both samples. The silica supports were not removed because they precipitate during the centrifugation process and consequently have no effect on the spectroscopic measurements.

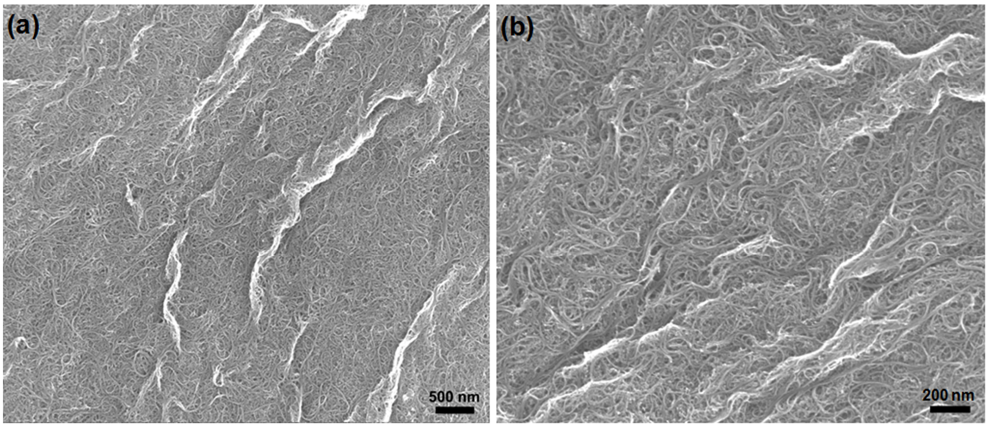

Figure S1. SEM images of the SWCNTs grown from CoPt catalyst at 800 °C. Before SEM imaging, the silica supports were removed by etching in a 30% HF solution.

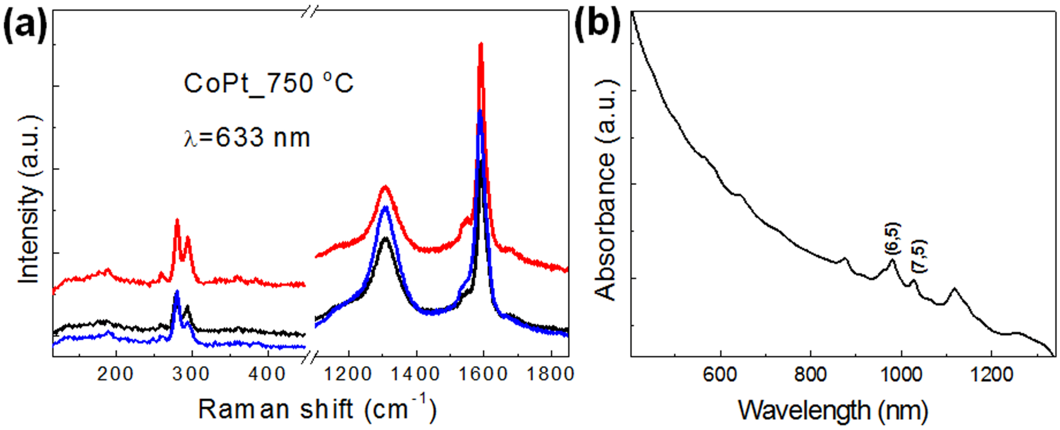

Figure S2. (a) Raman and (b) optical absorption spectra of SWCNTs grown from CoPt catalyst at 750 °C under atmospheric pressure. The Raman spectra in (a) were taken at different sample positions. The high I_D/I_G in Raman spectra and high absorption background in optical absorption spectra indicate the poor quality of the as grown SWCNTs.

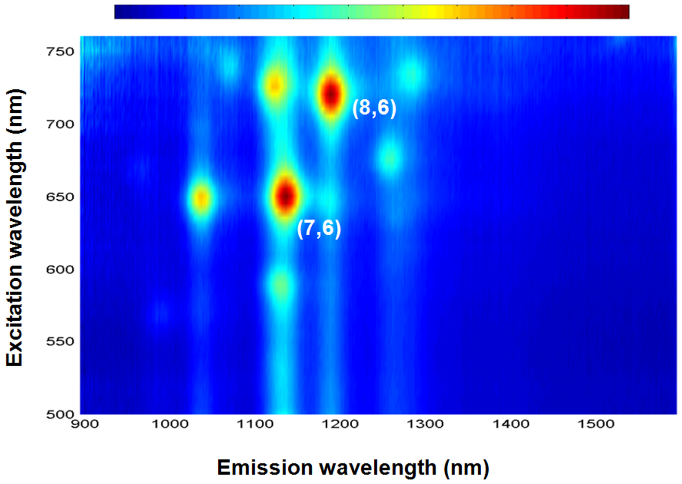

Figure S3. A photoluminescence spectrum of SWCNTs grown from Co catalyst at 800 °C.

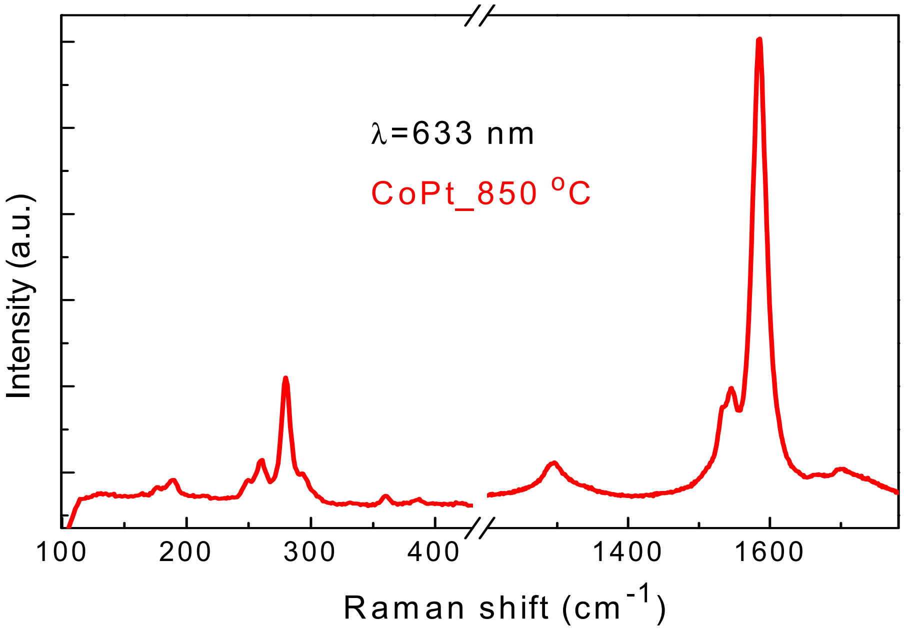

Figure S4. A typical Raman spectrum of the SWCNTs grown from CoPt catalyst at 850 °C. The excitation laser is 633 nm.

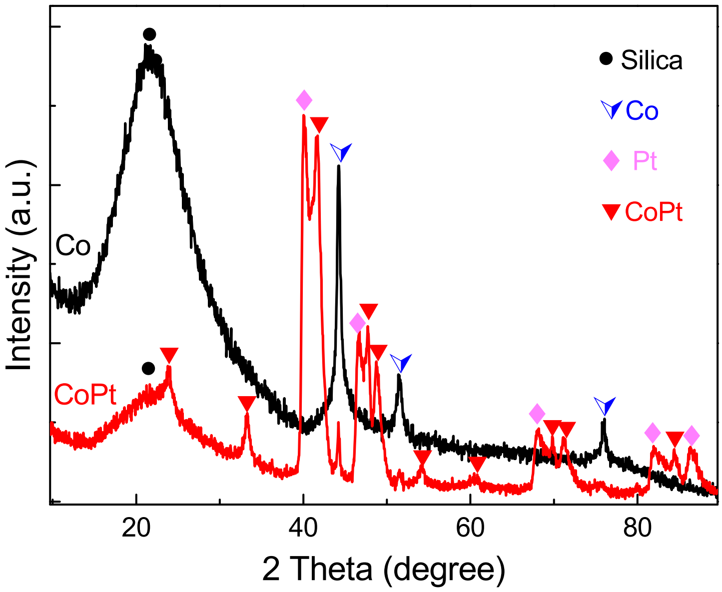

Figure S5. XRD patterns of the Co and CoPt catalysts after pretreatment while just before ethanol introduction. The diffraction peaks denoted by blue, pink, and red symbols can be assigned to monometallic Co (PDF number 15-0806), monometallic Pt (PDF number 65-2868), and CoPt alloy (PDF number 43-1358), respectively.