Supplementary Information

Controlled synthesis of a large fraction of metallic single-walled carbon nanotube and semiconducting carbon nanowire networks

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Figure S1. **The synthesis of high-density SWCNT networks occurred only in a narrow process parameter space**. When the growth temperature is reduced to 850 °C (a) or the pressure is kept at 1 **Torr** (b), no SWCNTs were observed. On the other hand, when 40 sccm H_2 were used in both the pretreatment and growth stages, a lower density of nanotubes was obtained (c); or when 200 sccm H_2 and a trace amount of H_2O were added in the CVD processes, a lower density and shorter nanotubes were observed. **Moreover, SWCNTs with some impurities were produced when the thickness of Fe was 1 nm (e) and 1.8 nm (f); while MWCNTs and amorphous carbon were produced when the thickness of Fe was 3 nm (g). If the Fe/SiO_2 catalyst was used, a lower density and long nanotubes were observed (h).** Nanotubes in **(c) – (h)** were also bound to the surface, in contrast to the "**bridge**" morphology (*i.e.*, free-standing) observed in Fig. 1 in the context.

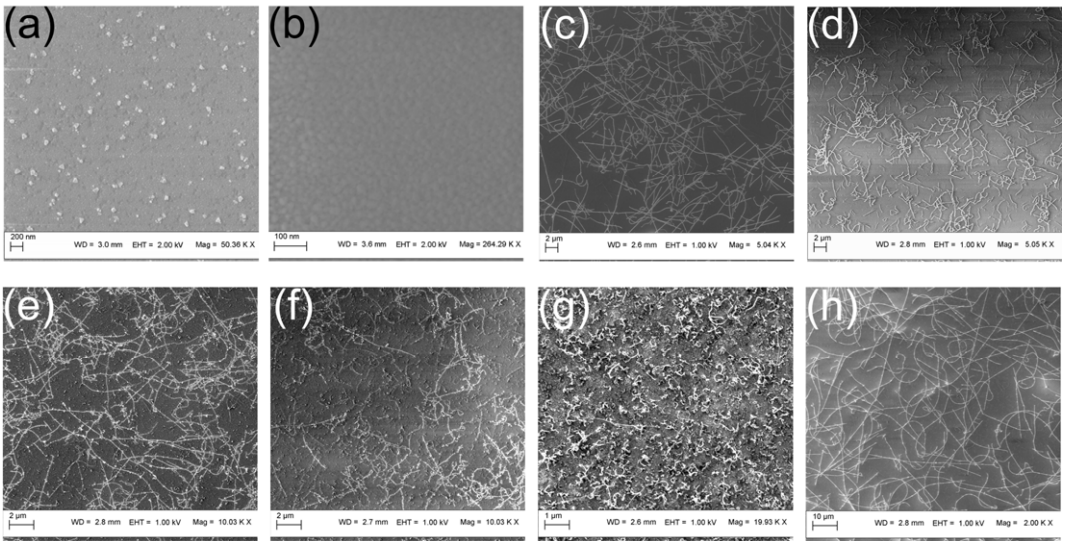


Figure S2. Resonance Raman for SWCNTs with different diameters. To choose the correct laser excitation in resonance Raman, information on the diameter distribution of the SWCNTs has to be known. For HiPco, its diameter is 1.0±0.2 nm. In this range, 1.96 eV excitation can be in resonant with both metallic and semiconducting nanotubes by assuming a resonant window of ±0.1 eV (a).[1] However, for CoMoCAT nanotubes which were enriched with >50% (6,5) indexed and >90% semiconducting nanotubes, a higher energy of laser excitation at 2.54 eV has to be chosen in order to be resonant with the E_{11}^{M} region of the metallic nanotubes (b). The energy of E_{11}^{S} in most cases is well below the Raman laser excitation energy.[2] The quantification of metallic and semiconducting contributions is therefore analyzed through the relative intensity between E_{11}^{M} and E_{22}^{S}.[3] The Kataura plot is from S. Maruyama of The University of Tokyo.

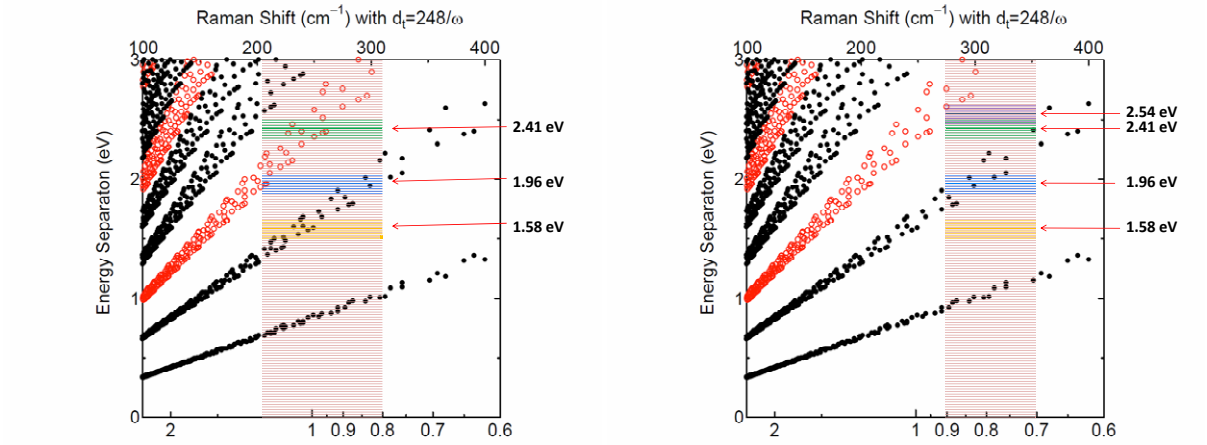


Figure S3. Raman G-band analysis. Enlarged Raman G-band fittings for this work (a), HiPco (b) and CoMoCAT (c) nanotubes. One Lorentzian line shape and one BWF line shape were resolved in (a) whereas several Lorentzian line shapes were resolved in (b) and (c). The asymmetric BWF line shape is described by:[4] $I(\omega) = I_0 * (1 + (\omega - \omega_{BWF})/q/W)^2 / (1 + ((\omega - \omega_{BWF})/W)^2)$, where ω_{BWF} is the BWF peak frequency at maximum intensity I_0, $1/q$ is a measure of the interaction of the phonon with a continuum of states, and W is the peak width. The fitting in (a) yielded a value of $1/q$ = -0.17, showing the significance of the coupling of phonons with the electronic continuum. The smaller separation between the lower-frequency BWF line shape and the higher-frequency G^+-band (at ~1590 cm^{-1}) in (a) also indicated that the diameter of the nanotubes in this work was larger according to the nanotube curvature effect.[5]

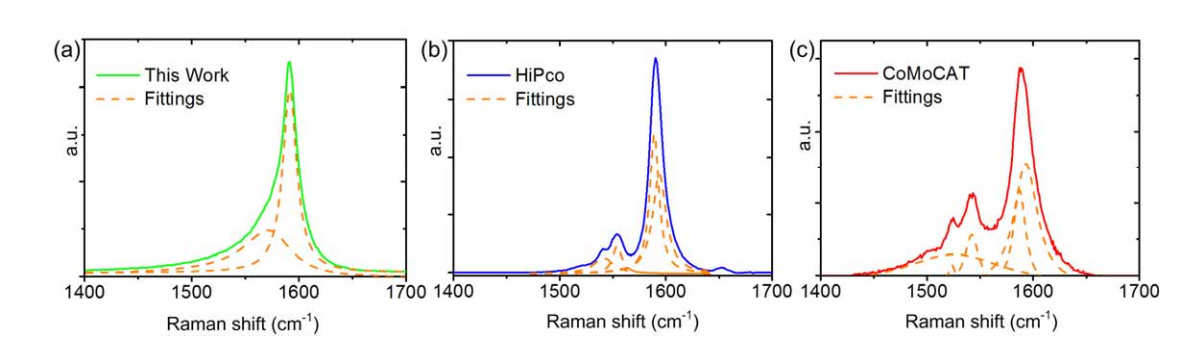


Figure S4. SWCNTs grown by other catalysts. When the Fe/Al_2O_3 catalyst was replaced by Co/SiO_2 (a), $Co/Mo/Al_2O_3$ (b), and Ni/Si (c), SWCNTs grown by the same conditions showed different density, length, and morphology. Moreover, no preferential growth of metallic nanotubes was observed. The nominal thicknesses of Co, Mo, and Ni were 1, 0.2, and 0.6 nm, respectively.

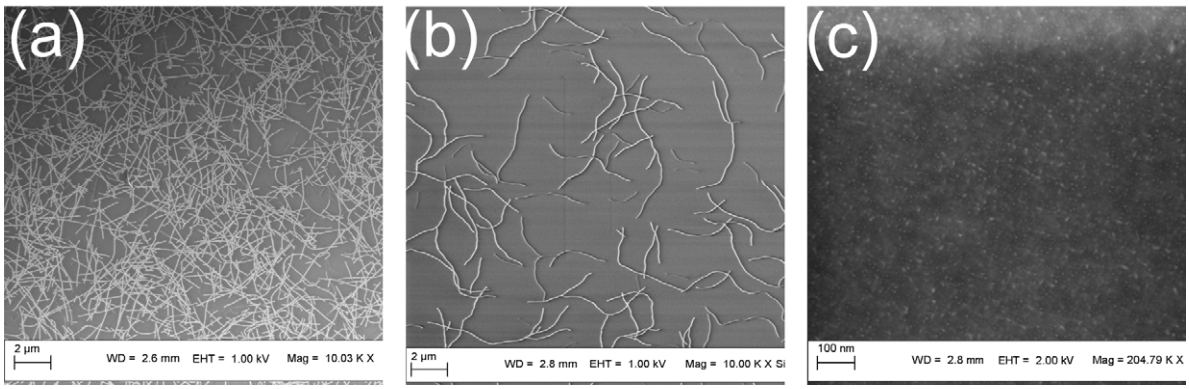
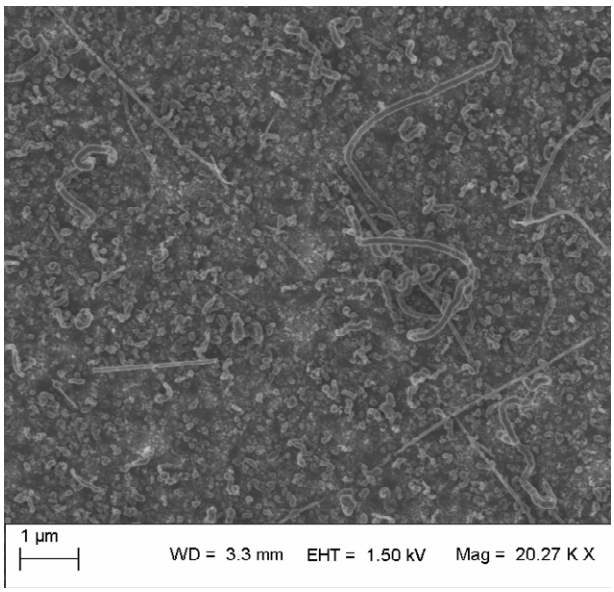


Figure S5. MWCNTs were grown if the catalysts were pretreated without H_2. When the growth temperature is kept at 1000 °C in the catalytic CVD processes of growing CNW networks, MWCNTs are resulted if no H_2 was used in the pretreatment stage. The image below also shows possible formation of amorphous carbon due to methane decomposition at high temperatures.



References

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