Supporting Information

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Sample #	T (°C)	Ferrocene (mg mL ⁻¹)	Thiophene (vol.%)	Product	Remark
1	1000	6.7	0	SD-SWCNTs	Discontinuous
2	1000	6.7	1	MWCNTs	Continuous
3	1100	6.7	1	SWCNTs + MWCNTs	Continuous
4	1100	13.3	1	SWCNTs + MWCNTs	Continuous
5	1100	20.0	1	SWCNTs + MWCNTs	Continuous
6	1100	10.0	5	MWCNTs	Continuous
7	1100	20.0	5	MWCNTs	Continuous
8	1150	10.0	0.1	SD-SWCNTs	Continuous
9	1150	10.0	1	LD-SWCNTs	Continuous

Table S1. Experimental conditions and corresponding products.



Figure S1. Experimental setup used for the present synthesis of carbon nanotubes. Note that when no thiophene was added, singe wall carbon nanotubes could be collected only from the internal wall of ²⁵ quartz tube in the low temperature region in the form of thin film. But when thiophene was included, carbon nanotubes in the form of powder could be collected from the sample collector connected with the quartz tube.



Figure S2. Thin film (a) and powder (b) morphologies of the samples produced without and with addition of thiophene, respectively.



Figure S3. TEM images of the as-grown samples synthesized at 1000°C. (a,b) SD-SWCNTs without addition of thiophene (sample 1); (c) MWCNTs with addition of 1% thiophene (sample 2). (d) TEM images of the as-grown sample synthesized at 1100°C with addition of 1% thiophene (sample 4).

- ⁴⁵ Sample 1 was prepared when the ferrocene concentration in alcohol was 6.7 mg mL⁻¹ without addition of thiophene. It can be seen from Figure S3a that this sample consists of a fiber-like substance, and the fibers are tangled with each other to form a network. In addition, the sample contains metal particles which distribute uniformly on the fibers with only slight aggregation. The structure of sample 1 was further studied by HRTEM. Figure S3b reveals that this sample is actually made up of SWCNTs
- ⁵⁰ with diameters about 1.5-2 nm. The individual tubes apparently tend to form "ropes" or bundles. Covered on the surface of the bundle is a trace amount of amorphous carbon. However, sample 2, which was prepared under similar conditions as sample 1 but with addition of 1% thiophene, shows the presence of multi-walled CNTs (MWCNTs) with few SWCNTs (Fig. S3c). The diameter of MWCNTs is about 10-15 nm, and the wall is generally thin with several graphitic layers.
- Figure S3d presents the typical TEM images of the as-grown samples synthesized at 1100°C with the addition of 1% thiophene. Although experiments were conducted with different concentrations of ferrocene (from 6.7 to 20 mg mL⁻¹), no significant difference was found among samples 3, 4, and 5. Generally, these samples consist of both MWCNTs and SWCNTs (Fig. S3d). The MWCNTs appear to be similar to those found in sample 2 prepared at 1000°C, but the SWCNTs have much larger
- ⁶⁰ diameters than those seen in sample 1. While both SWCNTs and MWCNTs were found in samples 3, 4, and 5 with the addition of 1% thiophene, only MWCNTs were observed in samples 6 and 7 with the addition of 5% thiophene. But in this case, the MWCNTs had an external diameter of ~30 nm. The quantity of sample produced per unit time with the addition of 5% thiophene was found to be larger than that with 1% addition.









Figure S6. Raman spectra of SD- and LD-SWCNTs at high frequencies



Figure S7. Histograms of diameter distribution of Pt particle on SD-SWCNTs (a) and LD-SWCNTs (b).



Figure S8. Dependence of tube diameter on thiophene addition.