Electronic Supplementary Information

Facile diameter control of vertically aligned, narrow single-walled carbon nanotubes

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Fig. S1. (a) Negligible effect of H₂ on the nanotube diameter distribution during the CVD process and (b) optimized CVD process for the continuous and pulsed growths of SWNT forests. For the ultrathin Fe catalysts used in this study, the growth condition was chosen without H₂ addition. We did not observe a significant difference in the nanotube diameter distribution between CNTs grown from the pure C₂H₂ condition and the C₂H₂/H₂ condition. We interpret that C₂H₂ is a very reactive gas, a source of C as well as H₂, negating somehow the need of H₂. However, growth rate continued to decrease with H₂ vol%. This decrease in the growth rate hints that a large concentration of surface adsorbed hydrogen may inhibit the generation and/or adsorption of carbon building blocks. In addition, prolonged H₂ exposure could not only reduce the iron oxide but also promote the agglomeration of the Fe catalyst layer and the catalyst coarsening.

![Diagram showing continuous and pulsed growth processes](image-url)
**Fig. S2.** HR-TEM images of SWNTs grown from ultrathin Fe catalysts with various thicknesses: (a) 0.1 nm, (b) 0.15 nm, (c) 0.2 nm, (d) 0.3 nm, and (e) 0.4 nm. The nanotubes were grown at 750 °C and 70-80 mbar using Ar/C$_2$H$_2$ (8000:5 sccm) for 10 min and sampled for comparison. Plot of SWNT mean diameter as a function of the Fe layer thickness is shown in (f). Additional Raman spectra using 532-nm excitation are provided in (g).
Nominal Fe thickness, $t_{CNT}$ (nm)

$$d_{CNT} (\text{nm}) = 4.72 \times t_{CNT} (\text{nm}) + 0.65$$

$R^2 = 0.99$

Raman Shift ($\text{cm}^{-1}$), $\lambda = 532 \text{ nm}$
Fig. S3. (a) Time dependent growth kinetic curves represented by the heights (h) of SWNT forests grown from the catalysts of various Fe thicknesses: 0.1 (red), 0.2 (orange), 0.3 (green) and 0.4 nm (blue) (b, c) Monomolecular fitting parameters for the growth curves, $h = A(1 - e^{-t/\tau})$, where $A$ (black) and $\tau$ (blue) are asymptotic height and time constant, respectively. Lines are drawn for visual guidance.
**Fig. S4.** TEM diameter histograms of SWNTs grown from the ultrathin Fe catalysts (>0.2 nm) for different durations. (a, b) Diameters of SWNTs grown from 0.3-nm-thick Fe catalysts on 20-nm-thick Al₂O₃ with growth time of (a) 10 min and (b) 60 min. (c, d) Diameters of SWNTs grown from 0.2-nm-thick Fe catalysts on 20-nm-thick Al₂O₃ catalysts with growth time of (c) 10 min and (d) 60 min.
Fig. S5. $P_{\text{C}_2\text{H}_2}$ dependence of average heights (growth time: 3 min) of the SWNT forests obtained from 0.2-nm-thick Fe catalysts, with an exponential fit: $h \propto \exp(-P_{\text{C}_2\text{H}_2}/P^*)$, where $P^*$ is a characteristic decay constant equal to 0.051 mbar.