Supplementary Information

Revealing the Impact of Catalyst Phase Transition on Carbon Nanotube Growth by *In Situ* Raman Spectroscopy

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Keywords

* Present Address: Honda Research Institute, Columbus, OH 43212 USA
Carbon nanotubes, CVD growth, phase transition, Raman spectroscopy, *In situ*

**Figure S1.** Plots of catalyst lifetimes vs. growth temperatures corresponding to Fig. 3a in the manuscript. The above plots have been plotted on linear scales. (a) The data in Regions I and II have been fitted with exponential curves. (b) The combined data points have been fit to a single exponential.

The temperature vs. catalyst lifetime plots shown above in Fig. S1 have been plotted on linear scales unlike the corresponding semi-log plot in Fig. 3a. The data points in Regions I and II in Fig. S1a have been fitted to single exponential curves. Note, however, that the fit in Region II corresponds only to the data above the dashed transition region (i.e. > 1080 °C). The fits to the data in Fig. S1a show that the lifetimes decrease exponentially with increasing temperatures in both Regions I and II in spite of the scatter in the data. This can also be seen in Fig. S1b where the combined data points from Regions I and II have been fit to a single exponential. The poor fit of the single exponential in Fig. S2b furthers the argument to fit the datasets to separate curves.
Figure S2. Plots of initial growth rates ($\nu$) against growth temperatures for (a) Fe, and (b) Ni. The phase transition region corresponding to the modified Fe-C phase diagram is shown by the dashed lines in (a).
Figure S3. Histograms of initial particle size distributions prior to SWNT growth, (a), and after SWNT growth, (b), from Fe (bottom panel) and Ni (top panel) catalysts. SEM images of Fe nanoparticles before, (c), and after, (d), growth, respectively.
Figure S4. (a) SEM image and (b) corresponding Raman spectrum collected from an individual SWNT grown from Ni catalyst. A single peak in the low-frequency RBM region and a narrow G band identify the individual SWNT.
Figure S5. Normalized in situ growth curves and corresponding SEM images from two different experiments conducted at low (820 °C), and high (950 °C) temperatures, resulting in single and multiple SWNTs, respectively.

Figure S4 shows an example of growth curves from two different experiments conducted at low and high temperatures, which resulted in single and multiple SWNTs, respectively. Both growth curves have been normalized to unity and superimposed upon each other. Also included are the corresponding SEM images from the two experiments that show evidence for single and multiple SWNT growth in the two experiments. From the shape of the growth curves in Fig. S4, it can be seen that the experiment at the higher growth temperature has a similar lifetime as the growth at lower temperature, although only a single exponential has been used to fit both curves. This example shows that we did not observe (within our experimental noise levels) nucleation and termination of individual SWNTs within an ensemble at various times within the growth experiment. Moreover, the single exponential growth curve from multiple SWNTs suggests that the measured lifetime is an average value from all the SWNTs.