## What is Below the Support Layer Affects Carbon Nanotube Growth: an Iron Catalyst Reservoir Yields Taller Nanotube Carpets

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## Supporting Information:





**Figure 1** AFM measurements of samples annealed for 10 minutes at 790°C with and without reservoir. We can observe the large particles on the sample with the Fe reservoir.



**Figure 2** Raman spectroscopy measurements taken samples with and without Fe reservoir after 5 min anneal at 790°C. The G/D ratios are comparable

5 min anneal at 790 degrees- with "Fe reservoir"



5 min anneal at 790 degrees- without "Fe reservoir"



**Figure 3** (a) and 2(b) show top-view HRSEM images taken for both Fe reservoir and the non-Fe reservoir samples, respectively, after a 5 minute annealing period, at 790<sup>°</sup> (the magnifications are 50,000, 100,000 and 200,000 respectively).

## With Fe "reservoir"



## Without Fe "reservoir"



**Figure 5** HRTEM images showing the crystalline CNT structure for samples with and without the Fe reservoir for annealing durations between 2-30 min. We can see that the CNT diameters are smaller for the sample with the Fe reservoir compared with the non-reservoir sample. This result correlates with the annealing experiments and support the mechanism described in the text.



**Figure 6** SEM images showing the CNT carpets for samples with and without the Fe reservoir with 5 min anneal for 15 min growth durations at  $790^{\circ}$ . (1) Fe 10 nm/Al<sub>2</sub>O<sub>3</sub> 30 nm/Fe 1.2 nm (2) Fe 10 nm/Al<sub>2</sub>O<sub>3</sub> 10nm/Fe 1.2 nm (3) Fe 10 nm/Al<sub>2</sub>O<sub>3</sub> 5nm/Fe 1.2 nm (4) Fe 10 nm/Al<sub>2</sub>O<sub>3</sub> 3nm/Fe 1.2 nm (5) Al<sub>2</sub>O<sub>3</sub> 3nm/Fe 1.2 nm



**Figure 7** SEM images showing the result of a 5 minute anneal followed by a 20 second growth (at 790 °C). The sample without reservoir (left) exhibited only micron-long entangled CNTs while the sample with reservoir (right) exhibited a 15  $\mu$ m-tall carpet of vertically aligned CNTs.