

Supporting Information

Ultra-High Vacuum-Assisted Control of Metal Nanoparticles for Horizontally-Aligned Single-Walled Carbon Nanotubes with Extraordinary Uniform Diameters

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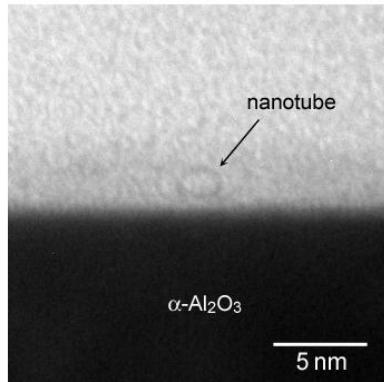
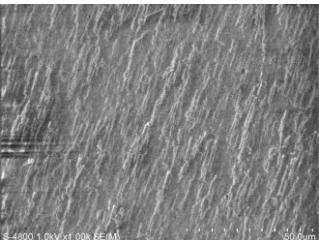
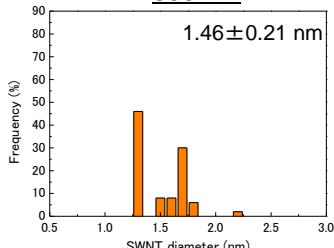


Figure SI-1. Cross-sectional TEM image of aligned nanotube on sapphire surface. The nanotube was grown without applying the UHV annealing. The sample was sliced normal to the nanotube's orientation with a FIB. The image shows that the nanotube is single-walled.

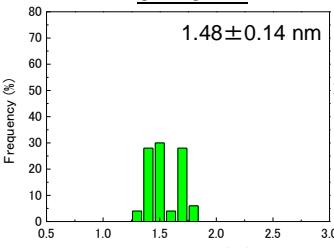
Non-annealed



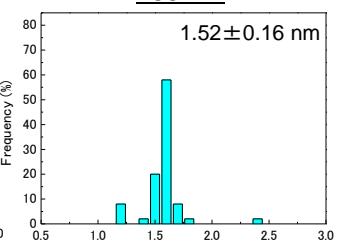
633 nm



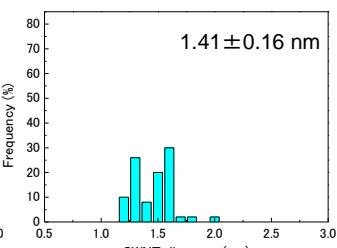
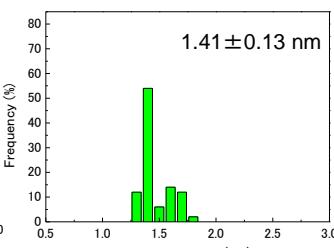
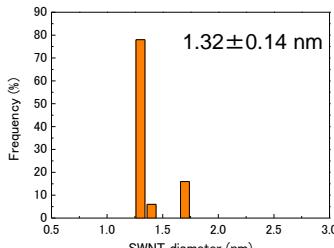
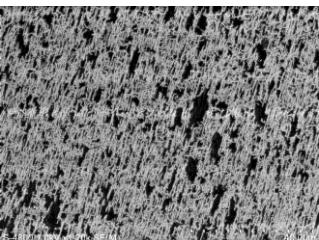
514.5 nm



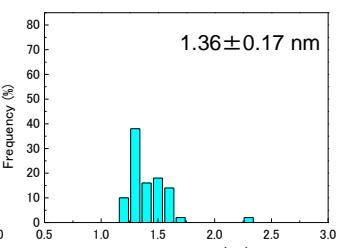
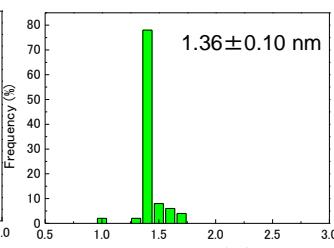
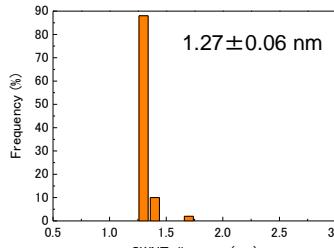
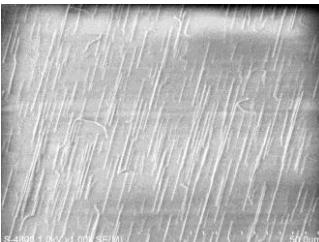
488 nm



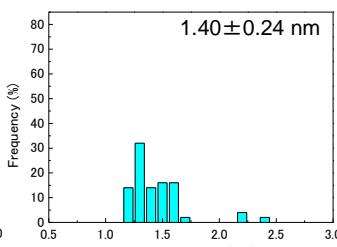
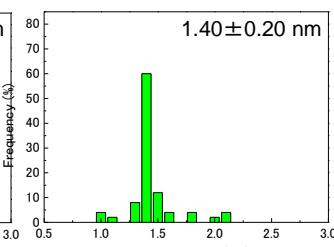
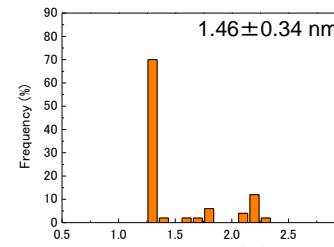
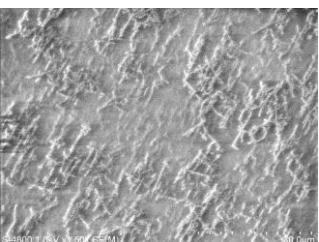
UHV anneal for 30 min



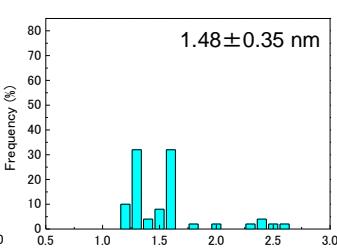
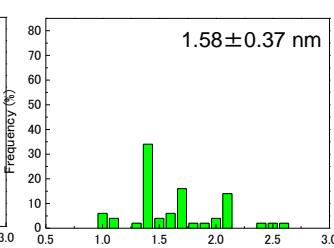
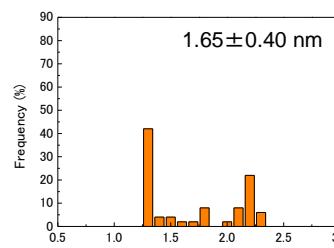
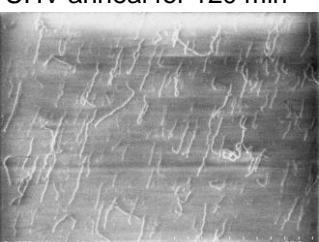
UHV anneal for 60 min



UHV anneal for 90 min



UHV anneal for 120 min



UHV anneal for 180 min

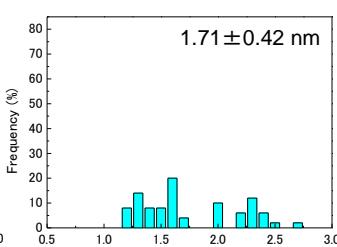
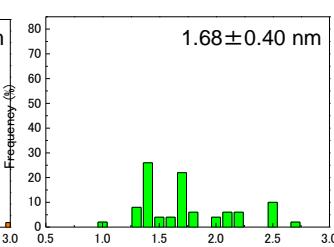
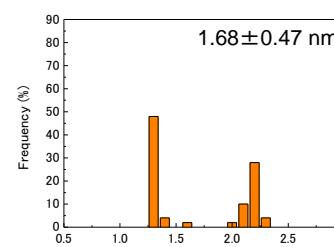
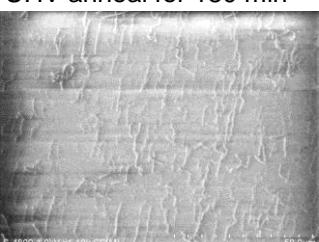
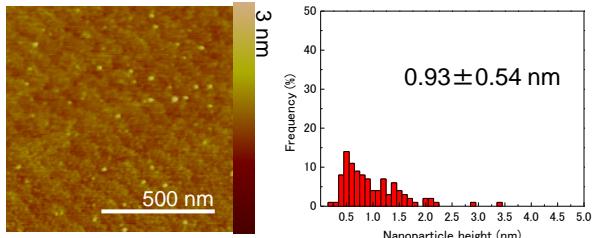
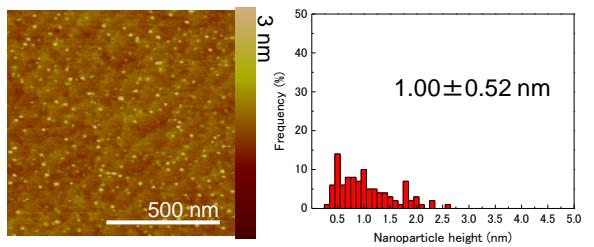
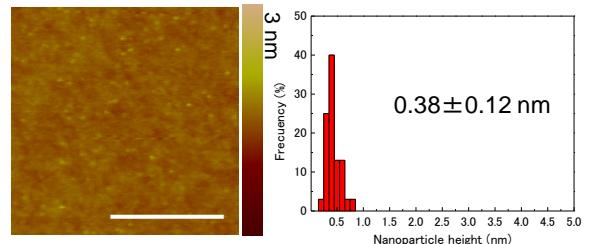
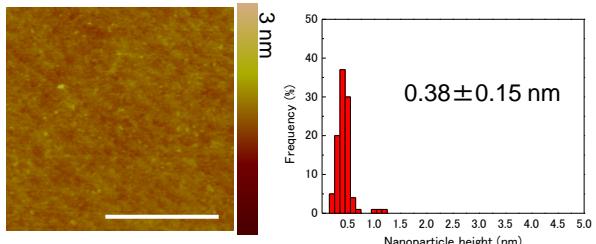


Figure SI-2. SEM images and histograms of diameter distribution of nanotubes grown with Fe metal nanoparticles treated under different annealing conditions. $N = 50$ for each wavelength.

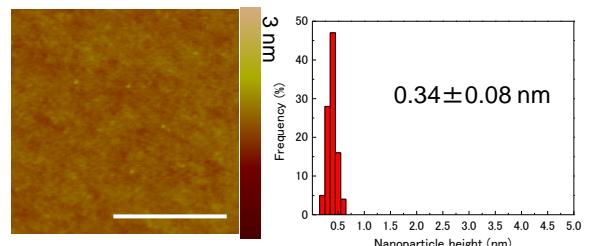
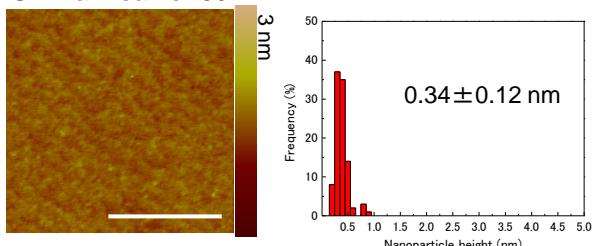
Non-annealed

After H₂ treatment at 900 °C

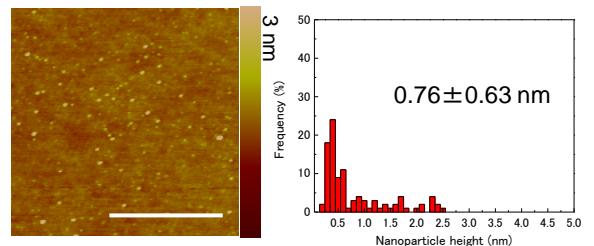
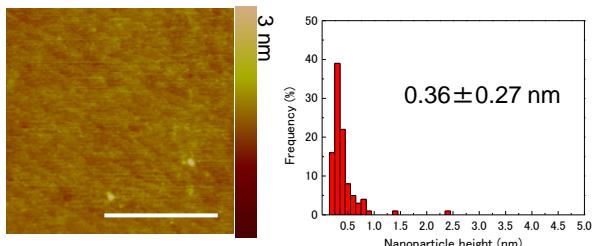
UHV anneal for 30min



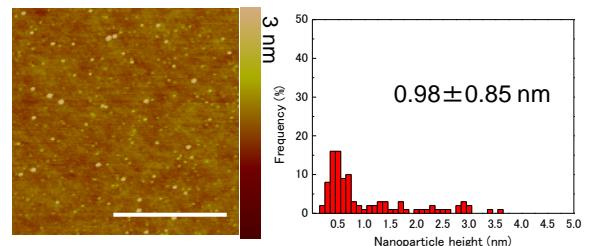
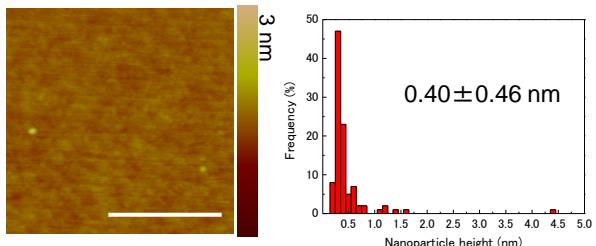
UHV anneal for 60 min



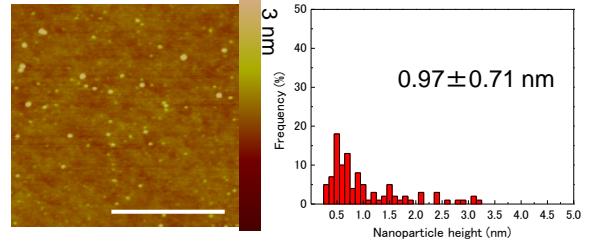
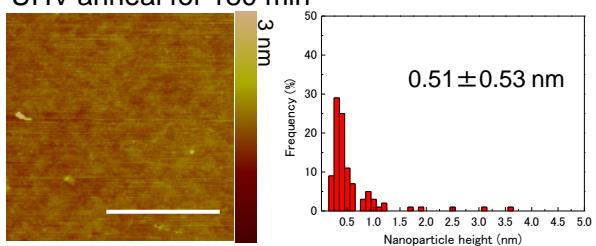
UHV anneal for 90 min



UHV anneal for 120 min



UHV anneal for 180 min

**Figure SI-3.** AFM images and histograms of Fe nanoparticle height. The Fe nanoparticles was first annealed in UHV (left), and then treated in H₂ flow at 900 °C under atmospheric pressure (right).

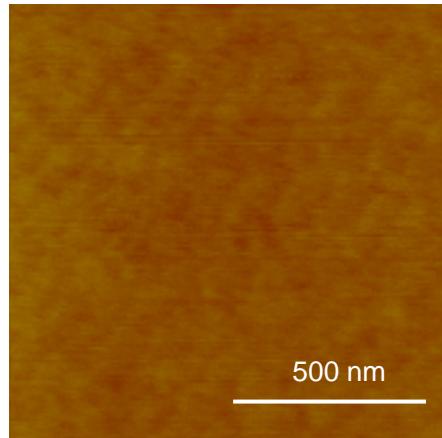


Figure SI-4. AFM images of sapphire surfaces without sputtered Fe catalyst. The AFM was measured after annealing in UHV at 900 °C for 3h. No apparent particles are observed.

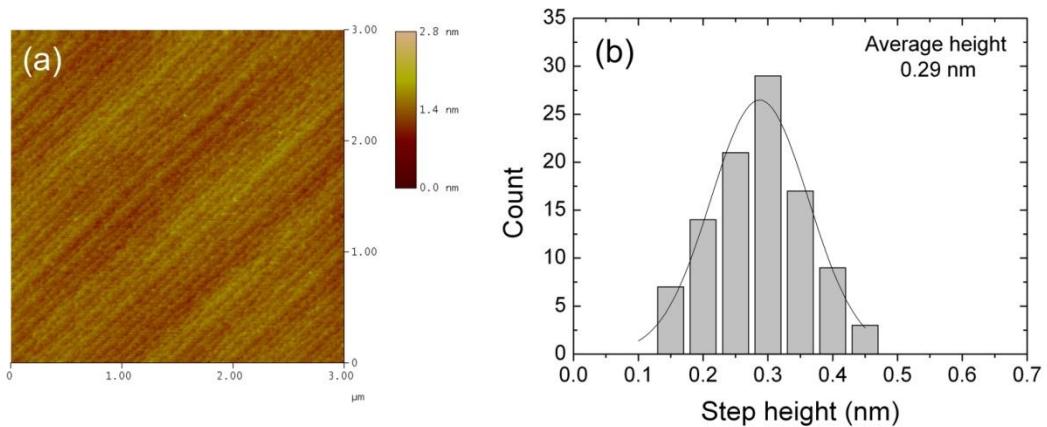


Figure SI-5. Reliability measurement for the AFM height analysis. A calibrated Si substrate with 0.31 nm step height purchased from NTT-AT was used to check the reliability of our AFM setup. (a) AFM image and (b) the measured histogram of the step height. The line is a Gaussian fitting curve. The data shows that the observed large discrepancy between the AFM height of the Fe nanoparticle and SWNT diameter (Raman analysis) is not due to error of the AFM height.

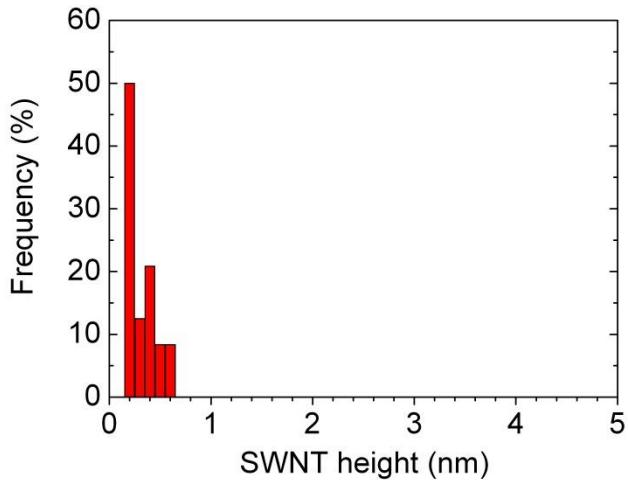


Figure SI-6. Histogram of the SWNT height measured by AFM. The sputtered Fe was annealed in UHV for 1h, and then subjected to the CVD at 900 °C. Number of counted SWNTs is 24. Comparison with Figure 1f indicates that the AFM height underestimates the actual SWNT diameter determined by the Raman spectroscopy.

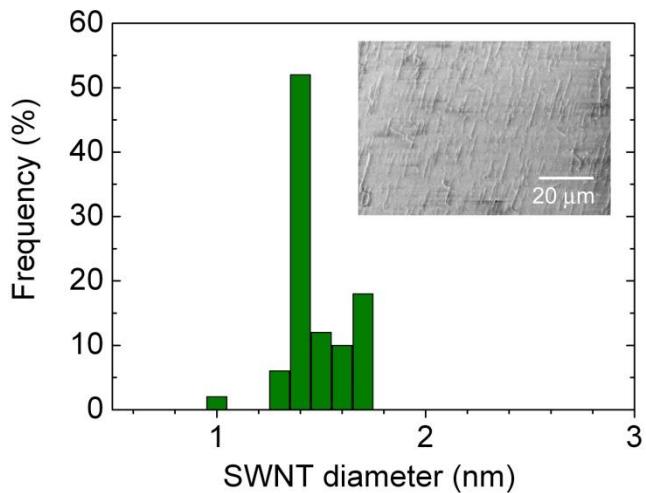


Figure SI-7. Histogram of the SWNT diameter measured by Raman RBM frequency. The sputtered Fe was annealed in low vacuum of 10^{-2} Pa for 1h, and then subjected to the CVD at 900 °C. Number of counted SWNTs is 50 and the excitation wavelength is 514.5 nm. The average diameter and the standard deviation are 1.41 and 0.13 nm, respectively. Although the diameter distribution is narrower than that of the SWNTs grown over the non-annealed catalyst, it is wider than that annealed in UHV (see Figure 1f, green). This result suggests the vacuum pressure is one of important parameters to control the nanoparticle size.