**Supporting Information for**

# Diameter Dependence of Water Filling in Lithographically Segmented Isolated Carbon Nanotubes


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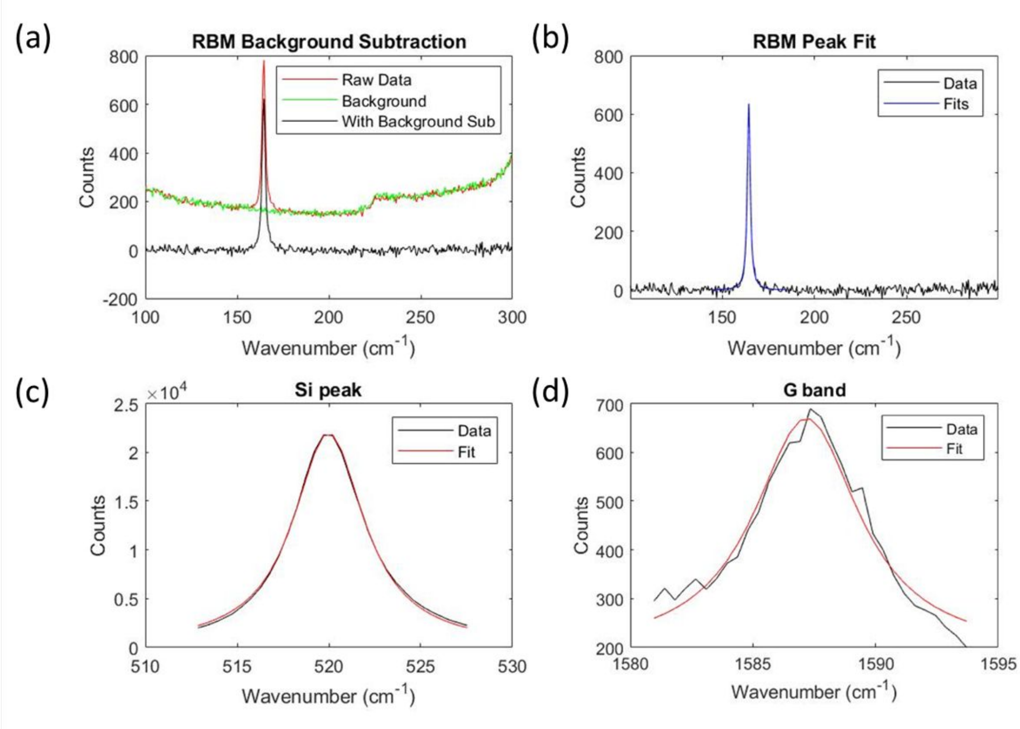


**Figure S1.** Raman data analysis. A sample Raman spectrum for one 1.51 nm CNT is analyzed. (a) The silicon background multiplied by a constant coefficient and subtracted. (b) After subtraction of a Si background signal, the RBM peak is fit to a single Lorentzian, and the peak position and full-width half maximum are calculated. (c) The Si fit is used to confirm instrument calibration. (d) The G band is used to locate CNTs and confirm that the same CNT is being observed repeatedly.

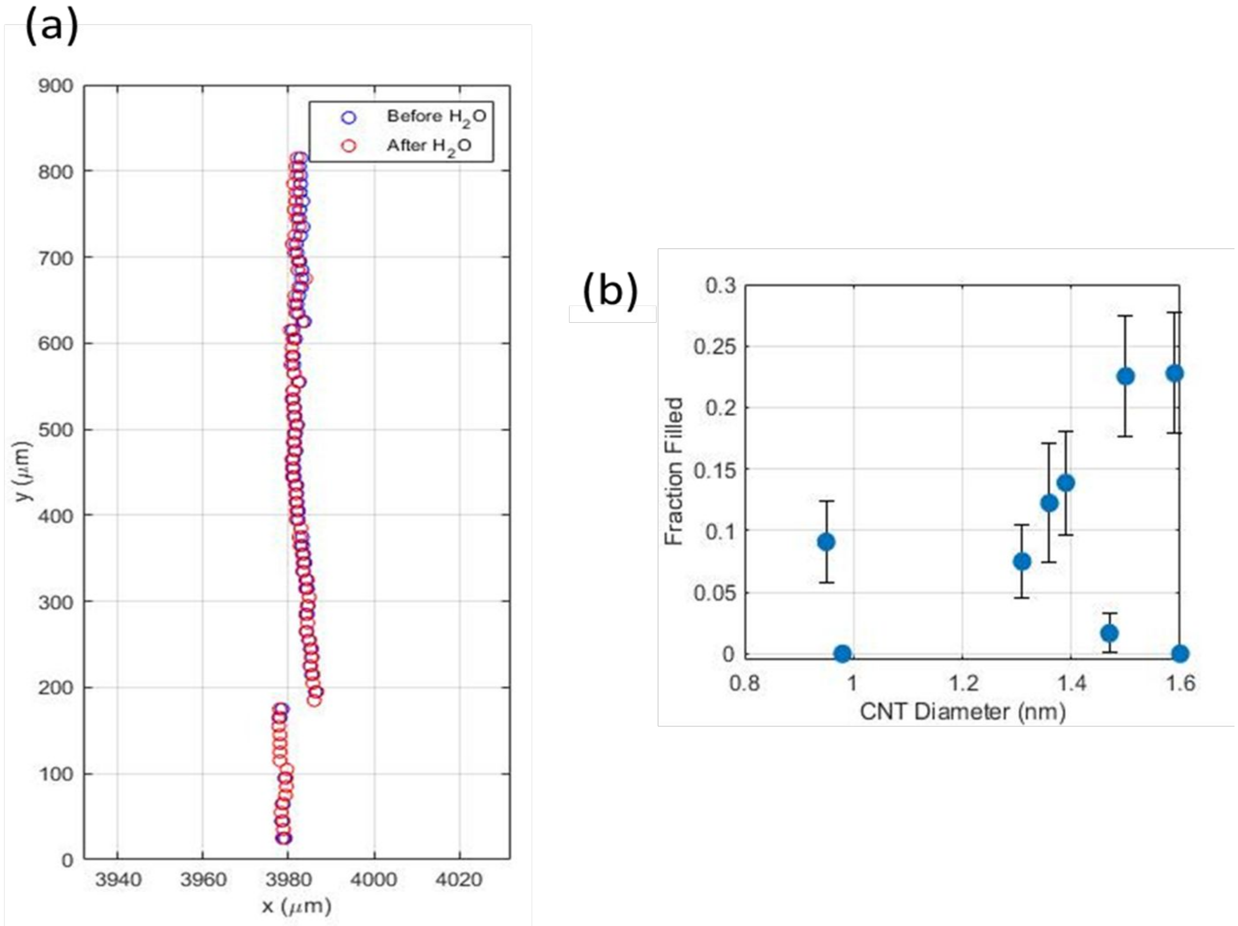


**Figure S2.** Statistics of filling from lithographically patterned CNTs. (a) Raman xy maps determine the location of etched, segmented CNTs on the silicon substrate before and after water immersion, and confirm that the same substrate-bound CNT is being observed before and after water immersion. (b) Fraction of 6 µm CNT segments filled *versus* diameter for 9 CNT chiralities, ranging from 0.95 to 1.59 nm in diameter. Each point represents observations of between 49 to 80 distinct CNT segments of the same chirality and diameter, with $\Delta\omega_{RBM}$=0.8 cm$^{-1}$ used as the threshold for fluid filling.

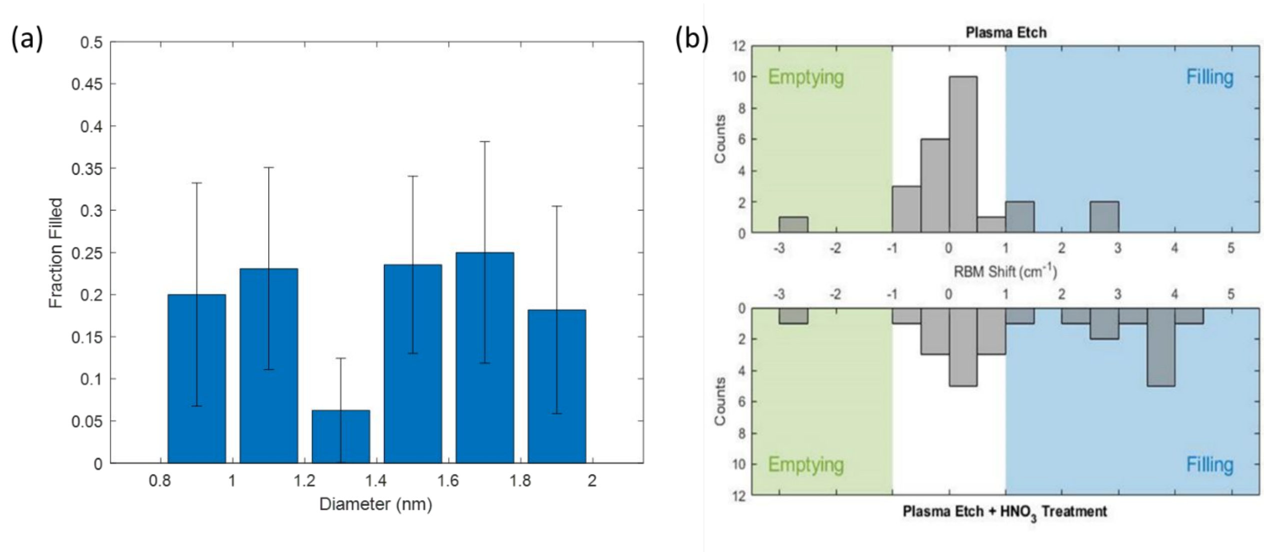


**Figure S3**. (a) The fraction of CNTs that fill with water, as indicated by an RBM shift above 0.8 $cm^{-1}$, is shown *versus* diameter from 0.8 nm to 2.0 nm. Variations in fraction filled with diameter are not statistically significant. (b) Effect of nitric acid treatment on water filling of carbon nanotubes. Each data point represents paired observations of the same location on the same chirality, substrate-bound CNT. On top, Raman spectra are taken before and after 1 hr $H_2O$ immersion for CNTs subject to oxygen plasma etching to open CNT ends (6.8 W, 3 min). On bottom, Raman spectra are taken before and after 1 hr $H_2O$ immersion for CNTs subjected to oxygen plasma etching plus supplemental immersion in nitric acid (2.6 M, 1 hr) and subsequent drying. CNTs are more likely to fill with water after immersion in nitric acid than they are without nitric acid treatment, as shown by the difference in $\Delta\omega_{RBM}$ distributions.

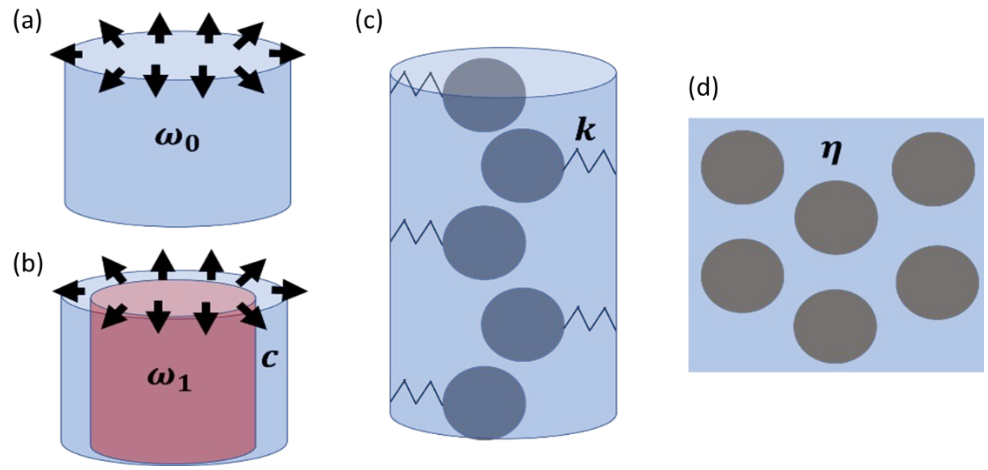


**Figure S4.** (a-b) In a quasi-continuum model, the change in CNT RBM frequency $\Delta\omega_{RBM} = \omega_1 - \omega_0$ can be related to the area-normalized spring constant c applied by a rigid fluid shell on the elastic shell of the CNT. The spring constant c in this model is the product of (b) the molecular spring constant k, and (c) the fluid areal density η. In this model, η alone is assumed to vary with CNT diameter.

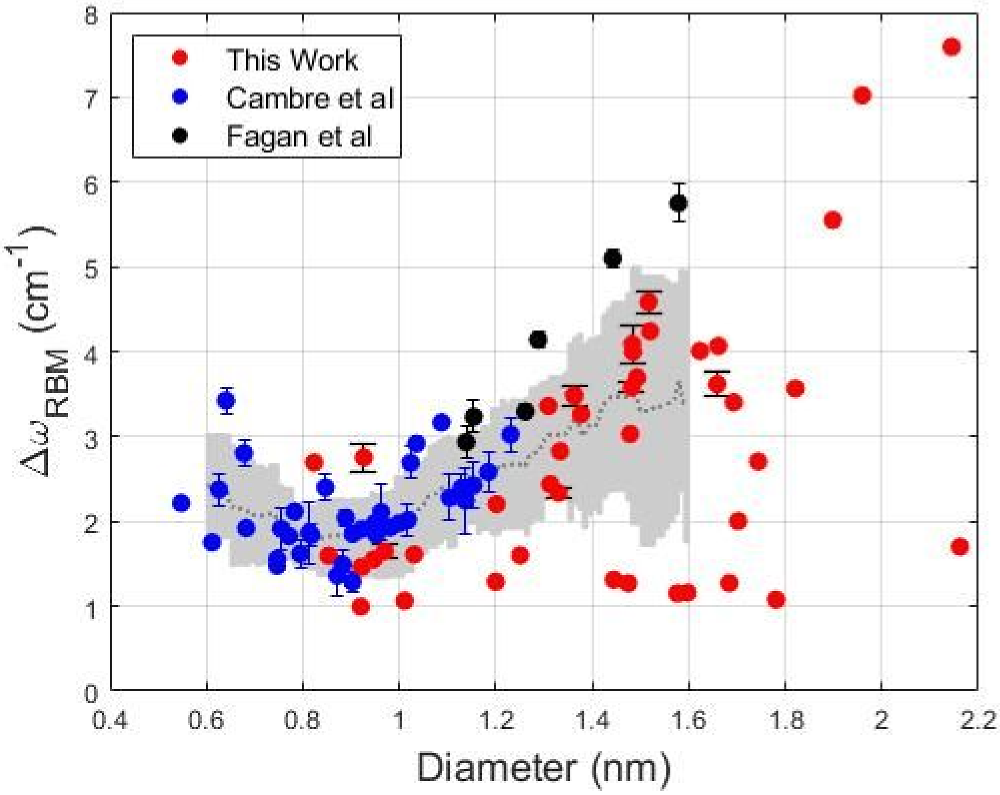


**Figure S5.** RBM shift is shown *versus* diameter in CNTs that are presumed to have filled or emptied with comparison to solution-phase work by Cambré *et al*.[30] and Fagan *et al*.[31] Individual paired observations do not have confidence intervals, while repeated measurements on the same chirality CNT are shown with 95% confidence intervals. A moving average and standard deviation of $\Delta\omega_{RBM}$ is shown in grey.

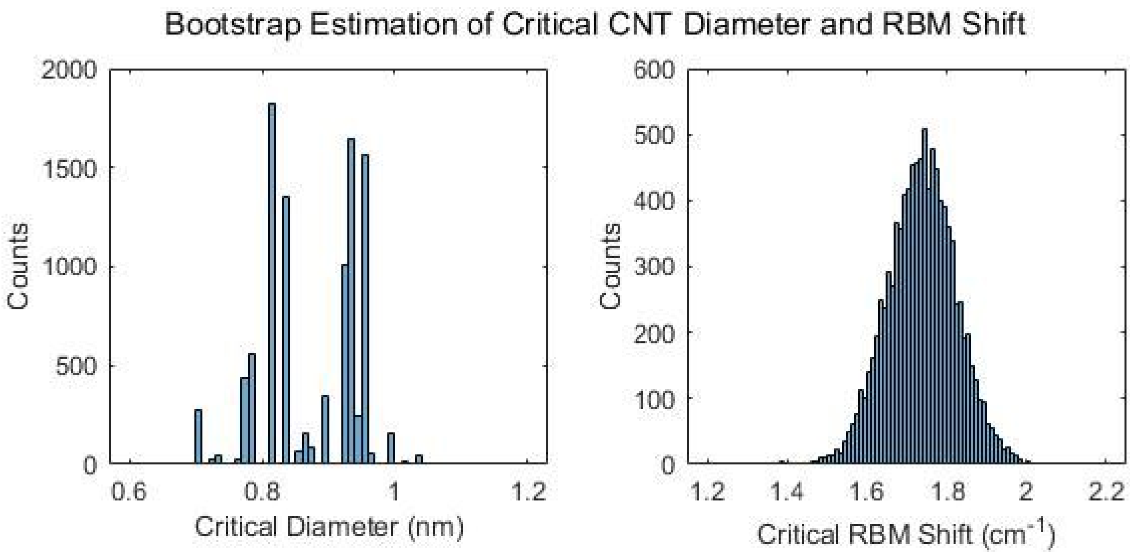


**Figure S6.** Bootstrap estimation of standard error of the mean in critical CNT diameter (±0.08 nm) and critical RBM shift (±0.09 $cm^{-1}$). Standard errors of the mean were estimated by analysis of 10,000 bootstrapped samples.