Supporting Information

S1: Determination of CNT spacing for OH-functionalized CNTs

The density of the OH groups considered in the MM simulations of hydroxyl-functionalized CNTs was chosen by comparing the TEM measurements of the spacing between DWNTs in the bundles (3.4 ± 0.6 Å) with MM simulations of the spacing between tube as a function of OH density (Figure S1). From this comparison, an OH density of 2-4% was chosen.

![Distance vs. OH Coverage](image)

Figure S1: Computed CNT-CNT distance (in Å) as a function of hydroxyl group coverage for a pair of (27,0) tubes, 6 nm in length. The (27,0) tubes contained (11,11) inner tubes. The experimental HRTEM images suggest the spacing between tubes is 3.4 ± 0.6 Å.

S2: Fixed versus tethered ends

There are several options for constraining the positions of tube ends during the pullout of the central tube from a seven-tube bundle. Fixing the rings of carbon atoms at tube ends is one option. This approach results in concerns related to an over-constraining of the system. In Tinker, it is also possible to tether the tube ends to fixed atoms. We used Kr atoms 50.0 Å from the ends of the center of each tube with a flat-well harmonic with a force of 500 kcal/ Å² for this purpose. This approach results in tubes

![MM3 computation of the central tube sliding force in a 6nm, 7 x CNT(27,0) bundle.](image)
that are less constrained than those in the experimental bundles. Nevertheless, this latter approach is probably more realistic. As shown in Figure S2, the main difference in using the tethers versus the fixed rings is that a force plateau was reached at a much shorter distance in the tethered case. Overall, the same average sliding force was found.

**S3: DFTB validation**

To probe the quality of the MM3 estimates of shear forces between CNTs, we have calculated these forces using the DFTB electronic structure method, and compared with the results of MM3 calculations for the same structural model. The DFTB simulations included empirical dispersion corrections. In this case, the structure studied was smaller than those considered above due to the much larger computational effort associated with the quantum mechanical calculations. Table S1 presents results for the peak force for pairs of bare tubes, and is analogous to the first peak for the tube pair in Figure 5 B. The MM3 peak force is similar to the DFTB-D peak force, suggesting that the MM3 result is reasonable. The DFTB-D result indicates that MM3 may be underestimating the force, but not by enough to change any conclusions.

**Table S1.** Comparison of peak forces for DFTB-D and MM3 results. The CNT pairs are 1 nm in length, and the modeling follows the same protocol as that used to create Figure 5.

<table>
<thead>
<tr>
<th>Model (CNT structure)</th>
<th>Maximum force (DFTB-D)</th>
<th>Maximum force (MM3)</th>
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<tbody>
<tr>
<td>Bare (5,5)</td>
<td>0.25 nN</td>
<td>0.16 nN</td>
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**S4: FT-IR and Raman difference plot for pristine vs. heat treated mats**

Figure S3 shows the difference plot for the FT-IR spectra of pristine and heat treated mats. Differences in the Raman spectra of pristine and heat treated mats showed no discernible variation in the ratio of D and G bands which was below the noise limit of the experimental data.

![Figure S3: Plot of the difference between FT-IR spectra of pristine and heat-treated mats.](image-url)