Supporting Information for

Structural Stability and Binding Strength of a Designed Peptide-Carbon Nanotube Hybrid

Daniel Roxbury,[1] Shao-Qing Zhang,[3,4] Jeetain Mittal,[1] William F. DeGrado,[3] Anand Jagota[1,2]*

[1]Department of Chemical Engineering

[2]Bioengineering Program, Lehigh University

[3]Department of Physics and Astronomy, University of Pennsylvania

[4]Department of Pharmaceutical Chemistry, University of California, San Francisco

[*]*corresponding author*: D331 Iacocca Hall, Lehigh University, Bethlehem PA 18017 (USA)

(1-610 758 4396); anj6@lehigh.edu)

S1. Circular Dichroism of HexCoil-Ala-SWCNT Samples with Added Surfactant

In this study, we make use of several small surfactant molecules in an attempt to displace HexCoil-Ala peptide from the surface of a preexisting SWCNT suspension. A valid concern would be that the surfactant and peptide might interact, influencing the reaction to halt. One can imagine that the surfactant, with amphiphilic properties, could bind to the likewise amphiphilic peptide on the SWCNT. Further, if this were to happen, would the secondary alpha-helical structure of the peptide be disrupted? To examine these scenarios, we performed far-UV circular dichroism (CD) spectroscopy on samples of HexCoil-Ala and HexCoil-Ala-SWCNT. To these samples, we have added a small surfactant, sodium dodecyl sulfate (SDS), chosen for its low

absorbance in the UV region. This surfactant is similar to SDBS, with the exception of a missing benzyl group (which causes high UV absorbance, and could not be used for CD). As shown in Figure S1, HexCoil-Ala has two negative CD peaks at 208 and 225 nm, indicating the presence of alpha-helical structure. These peaks were not significantly affected by whether or not HexCoil-Ala was wrapping a SWCNT. In addition, SDS also had negligible influence on the two dominant peaks. This suggests that the surfactant is not disrupting the secondary structure of the peptide.

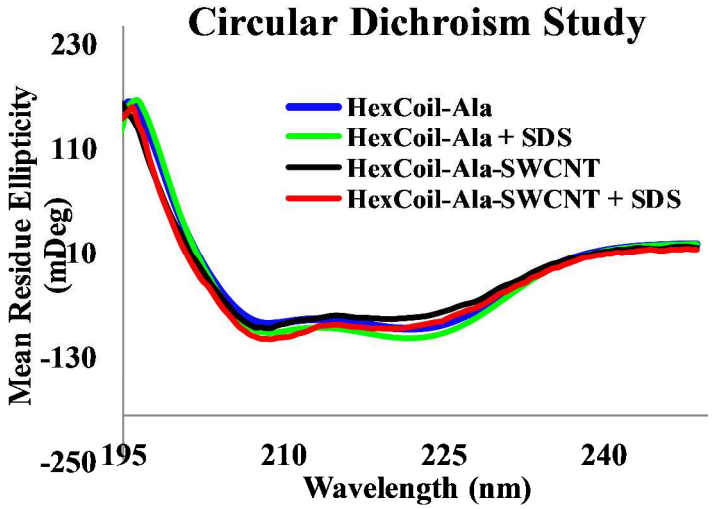

Figure S1. Spectral data from a circular dichroism experiment on HexCoil-Ala and HexCoil-Ala-SWCNT samples illustrating the negligible effect of surfactant, SDS.

S2. Two-Dimensional Fluorescence Maps

Scanning excitation/emission fluorescence maps can be very useful tools for determining the quality of a dispersed SWCNT sample. Here we present fluorescence maps from the 'ending' samples of HexCoil-Ala-SWCNT and $(TAT)_4T$-SWCNT after 10 minutes of incubation with SDBS at 60 ˚C (Figure S2). In conjunction with the absorbance scans in Figure 1 of the main text, we can make two conclusions from these fluorescence maps. We can first clearly say

that SWCNT emission signals from $(TAT)_4T$ are blue-shifted relative to HexCoil-Ala with.(6,5) emission signal peaks at 980 and 992 nm, respectively. Fluorescence emission in these maps correlates directly with the peak position observed in Figure 1. Therefore, the data confirm that in large measure HexCoil-Ala remains on the SWCNT after an attempted SDBS exchange. The second conclusion that we can draw from the data is that HexCoil-Ala preferentially disperses (6,5)-SWCNT relative to $(TAT)_4T$. Note the intensities of the (8,3) and (7,5)-SWCNTs in both of the maps when normalizing the data by the peak of the (6,5)-SWCNT. The (7,5) peak intensities are 0.46 and 0.83 for HexCoil-Ala and $(TAT)_4T$ dispersions of SWCNT, respectively, showing that HexCoil-Ala preferentially disperses the (6,5)-SWCNT compared to $(TAT)_4T$.

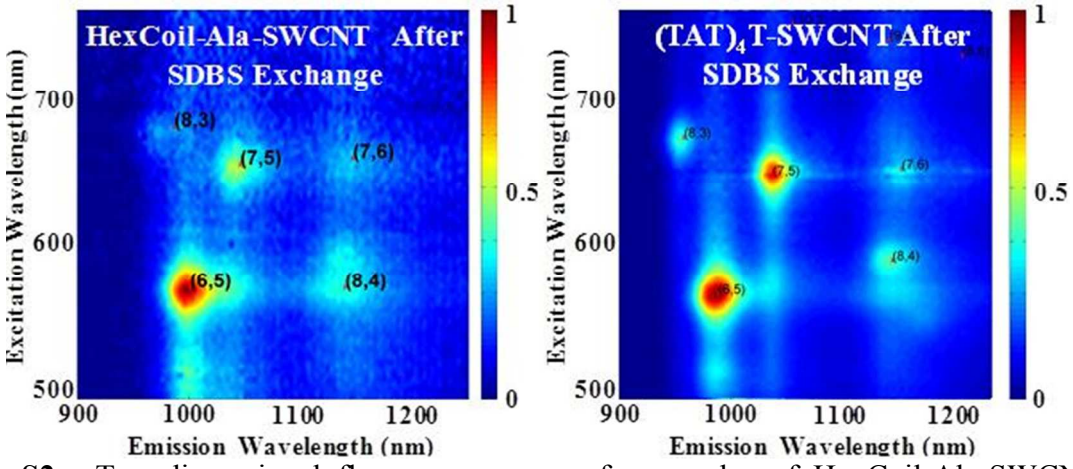

Figure S2. Two-dimensional fluorescence maps for samples of HexCoil-Ala-SWCNT and $(TAT)_4T$-SWCNT after SDBS exchange. Blue-shifted emission wavelengths are seen in the $(TAT)_4T$ sample, suggesting the removal of DNA from the surface of the SWCNTs. Additionally, HexCoil-Ala intensities of SWCNTs other than (6,5) are much lower than in the $(TAT)_4T$ dispersion.

S3. Convergence in Simulated Structure Analysis

When examining simulations of complex systems, it is useful to estimate the degree of convergence along appropriate reaction coordinates. In the case of simulations of HexCoil-Ala on the (6,5)-SWCNT, we have examined the progressive change in helical fraction for all six strands. Average helicity versus trajectory for all configurations is shown in Figure S3. Except for an initial drop in the average helicity for each strand, the configuration remains relatively stable over the course of the simulated trajectory.

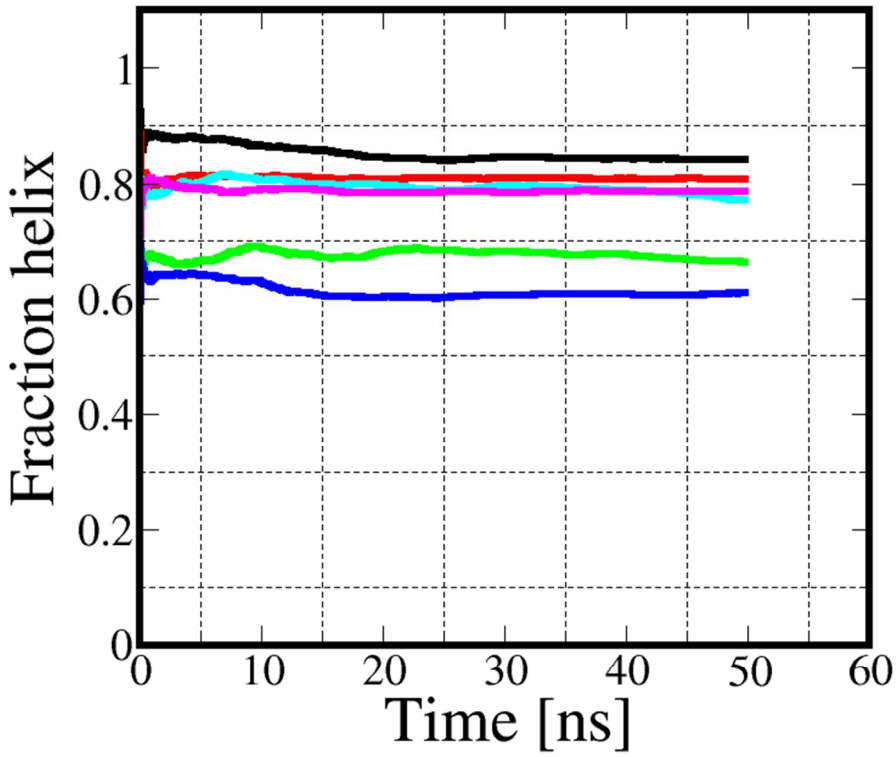

Figure S3. Average helicity vs. time for six strand configurations of HexCoil-Ala simulated on a (6,5)-SWCNT. Convergence is determined by a steady value of helicity, generally after an asymptotic decay.

S4. Characteristic Analysis of the Simulations

When the peptide hexamer binds to SWCNT, its configuration remains stable in simulations. It indicates a sizable affinity between the hexamer and SWCNT. The mean Cα RMSD of the structures in Figure S4a is 1.79 Å. The configuration of the hexamer is different from the designed model by its irregularity. Chain F protrudes from the assembly and has much less contact with SWNCT than the other chains. The local structure of Chain E, F and G resembles that of the tetramer crystal structure. However, the substantial binding between Chains E and G and SWCNT makes a poor alignment in Figure S4b. The mean Cα RMSD on the middle ten residues is 3.20 Å. The stable binding between the hexamer and SWCNT can be easily visualized by compact packing of the peptides on the hydrophobic surface of SWCNT.

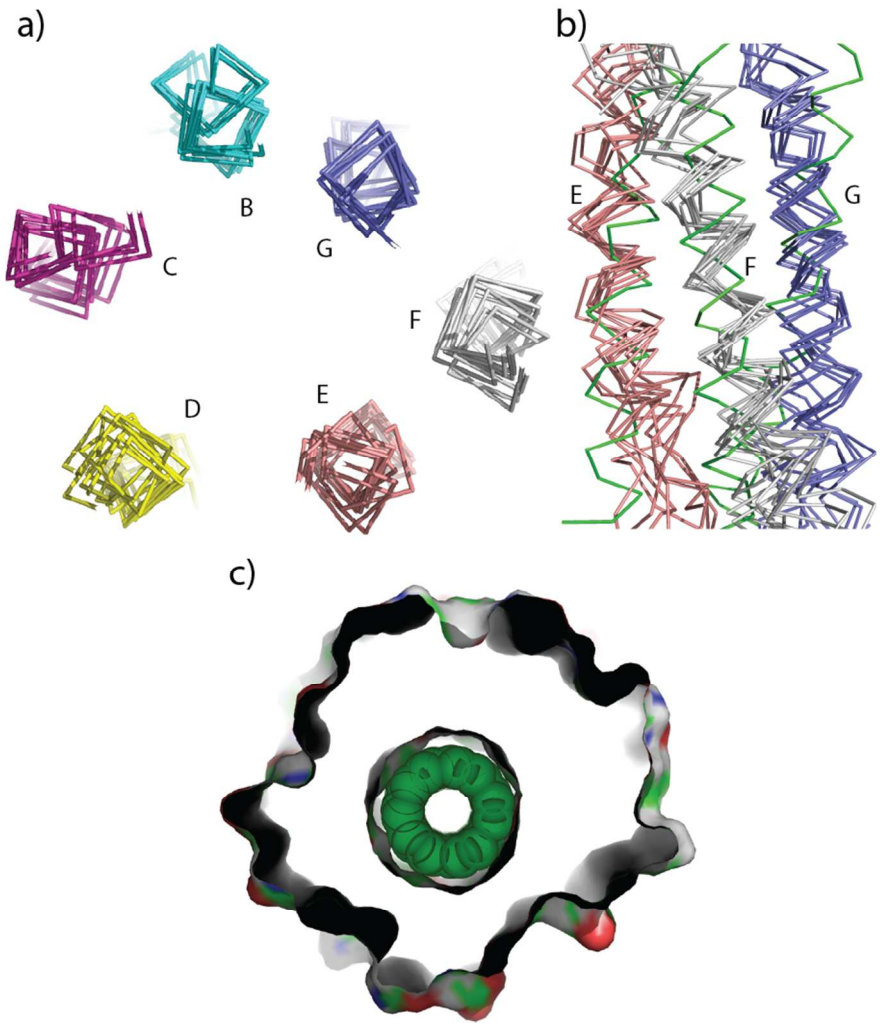

Figure S4. (a) The overlap of the middle ten residues on the configurations from 5 ns to 50 ns with an interval of 5 ns. The chain names are labeled. (b) Chains E, F and G from the structures in (a) are aligned with three consecutive chains from HexCoil-Ala tetramer crystal structure (green) (PDB ID: 3S0R). (c) The surface representation of the peptide hexamer with SWCNT in spheres.

S5. The Leu-Zipper and Ala-Coil Interfaces

The crystal structure and the designed model have two types of interface, the leucine zipper and Ala-Coil. They have a sequence motif in heptad repeats: L/AXXXXXX, where X stands for any residue. Leucine and alanine are supposed to make close interchain contacts in their specified interfaces as shown in Figure S5.

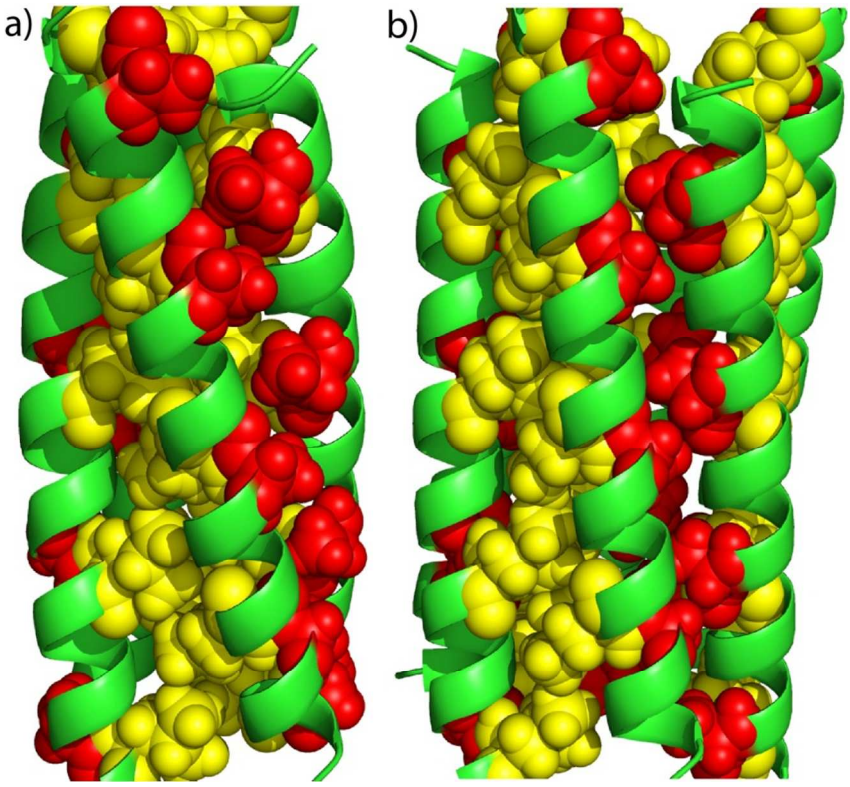

Figure S5. The packing of leucine (yellow) and alanine (red) in the tetramer crystal structure (PDB ID: 3S0R) (a) and the designed hexamer model (b).