Supporting Information for

Role of Arginine in Mediating Protein-Carbon Nanotube Interactions

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Partial charges

The following tables contain information on the partial charges used for Arg^+ and Nva molecules used in the simulations. The net charge on Arg^+ is +1 and on Nva is 0. The partial charges on Gdm^+ molecules are already reported in Godawat et al.[1]

Table 1: Atomic partial charges for Arg^+ and Nva residues employed in our simulations

	atom	quantity	q, e
Arg^+	N	1	0.12745
	H	3	0.20526
	C_α	1	-0.02534
	H_α	1	0.12115
	C_β	1	0.00874
	H_β	2	0.01954
	C_γ	1	0.02054
	H_γ	2	0.02784
	C_δ	1	0.09045
	H_δ	2	0.04964
	N_ε	1	-0.56808
	H_ε	1	0.35617
	C_ζ	1	0.82511
	N_η	2	-0.87241
	H_η	4	0.44638
	C	1	0.85271
	O	2	-0.82971
Nva	N	1	0.10662
	H	3	0.18442
	C_α	1	-0.04618
	H_α	1	0.10032
	C_β	1	-0.01208
	H_β	2	-0.00128
	C_γ	1	-0.00028
	H_γ	2	0.00702
	C_δ	1	0.06962
	H_δ	3	0.02882
	C	1	0.83182
	O	2	-0.85048

Dependence on CNT size/curvature

Figure 1 shows the density profiles of Arg^+ and Gdm^+ solutes near (9,9), (12,12), and (15,15) CNTs. These results are consistent with the ones shown in the main manuscript for the (6,6)

and (18,18) CNTs, that is, on the exterior of the CNT, the highest peak is the Gdm^+ ion followed by the Arg^+ peak. The interior for the (9,9) and (12,12) CNTs show Arg^+ having a higher peak compared to the Gdm^+, while the (15,15) shows the same trend as for the (18,18) CNT. This discrepancy is likely due to the low number of molecules on the interior of the (9,9) and (12,12) CNTs, in conjunction with the small normalization volumes in the density calculations.
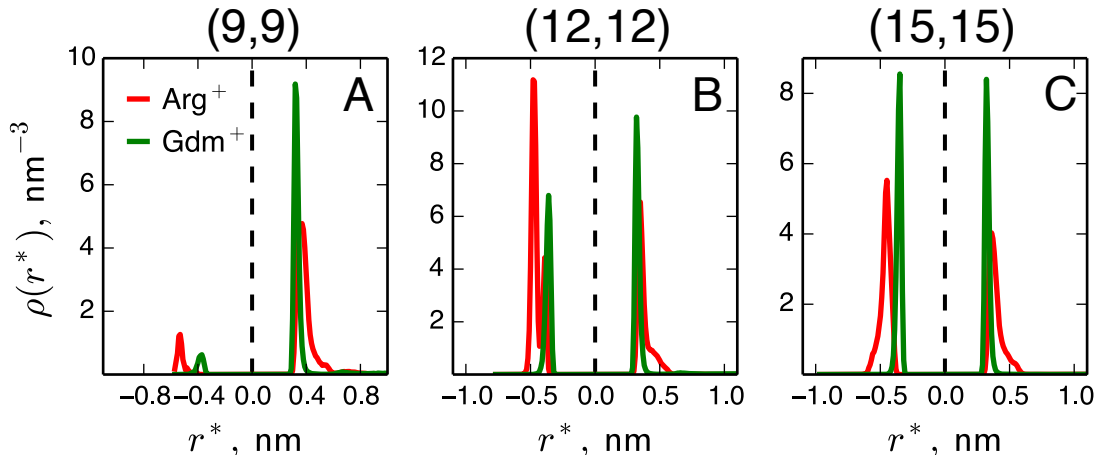

Figure 1: Density profile of Arg^+ and Gdm^+ for a (A) (9,9), (B) (12,12), and (C) (15,15) CNT using AMBER94 parameters for the CNT. The dashed line is the CNT surface.

Dependence on CNT parameters

Werder et al.[2] showed that the original AMBER parameters do not reproduce the correct contact angle when a water droplet is placed on a graphene sheet. To achieve an experimental contact angle of 86° for water on graphene, the authors tuned the non-bonded parameters between the carbon atoms in the graphene and the water atoms. Various water models were studied, including the TIP3P model for water, which is used by the AMBER force field.[2] We used the modified parameters from Werder et al. for the CNT[2] to determine if trends shown for Arg^+, Gdm^+ and Nva in the main text still hold. To achieve the mixed Lennard-Jones parameters for carbon-oxygen σ and ε according to the mixing rules, carbon-carbon

parameters of $\sigma = 0.32$ nm and $\varepsilon = 0.24$ kJ/mol were used. Figure 2 shows the results from all of the CNT sizes studied with the parameters from Werder et al.[2]
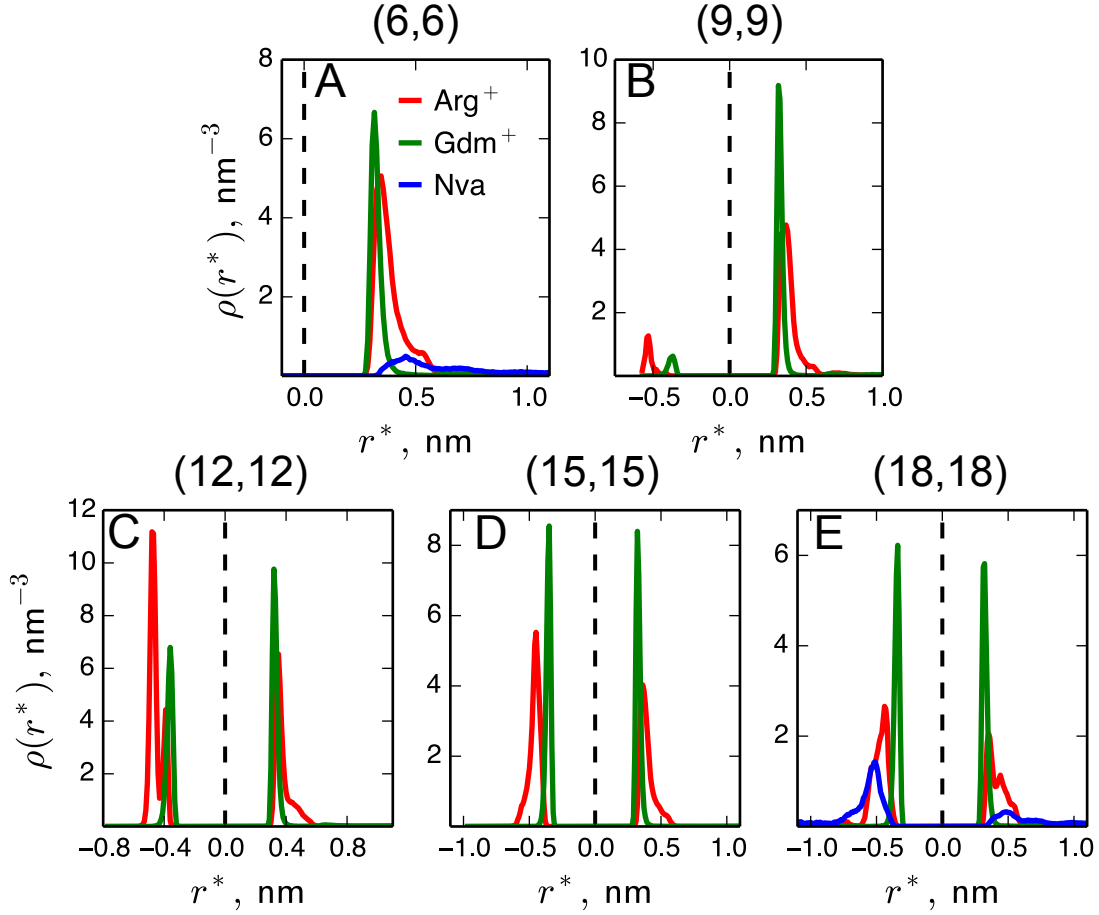

Figure 2: Density profile of Arg$^+$, Gdm$^+$, and Nva with CNT parameters from Werder et al.[2] for a (A) (6,6), (B) (9,9), (C) (12,12), (D) (15,15), and (E) (18,18) CNT. The dashed line is the CNT surface.

The results show that density profiles from the main text (Figure 5) and Figure 1 compared to Figure 2 are qualitatively similar in regards to the peak height on the exterior of the CNT. The density profile of the interior also shows similar trends compared to the AMBER results from the main text and Figure 1. These calculations suggest that the use of the AMBER force field as opposed to the Werder et al. parameters does not change the main conclusions of our manuscript that Arg$^+$ mediates the lysozyme-CNT interaction or that the Gdm$^+$ is the key contributor to Arg$^+$-CNT interactions.

References

(1) Godawat, R.; Jamadagni, S. N.; Garde, S. Unfolding of Hydrophobic Polymers in Guanidinium Chloride Solutions. *J. Phys. Chem. B* **2010**, *114*, 2246–2254.

(2) Werder, T.; Walther, J. H.; Jaffe, R. L.; Halicioglu, T.; Koumoutsakos, P. On the Water–Carbon Interaction of Use in Molecule Dynamics Simulations of Graphite and Carbon Nanotubes. *J. Phys. Chem. B* **2003**, *107*, 1345–1352.