Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Electronic Supplementary Information: Packing Morphology of Wavy Nanofiber Arrays

Itai Y. Stein,[a,*] **and Brian L. Wardle**[b]

[a] *Department of Mechanical Engineering, Massachusetts Institute of Technology, 77 Massachusetts Ave, Cambridge, MA 02139, USA. E-mail: iys@mit.edu*
[b] *Department of Aeronautics and Astronautics, Massachusetts Institute of Technology, 77 Massachusetts Ave, Cambridge, MA 02139, USA.*

S1 Two Dimensional Coordinations and Resulting Constitutive Triangles

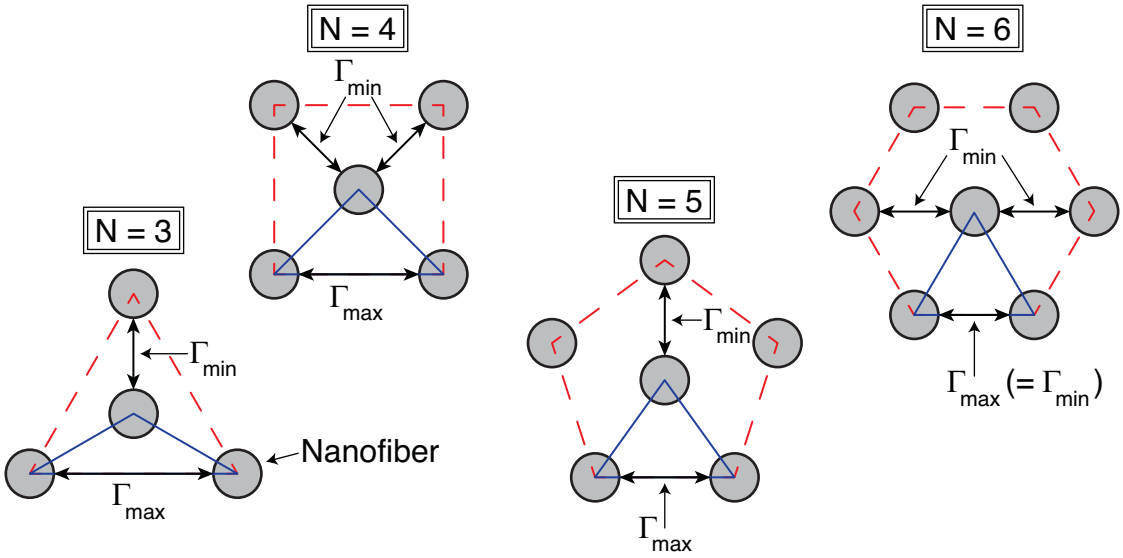

Fig. S1 Illustration of the two dimensional nanofiber (NF) coordination number (N), and the effective constitutive triangles that comprise each coordination.[1,2] The average of the minimum (Γ_{min}) and maximum (Γ_{max}) inter-NF spacings, defined as Γ, is used in the main text as an easily accessible measure of the morphology of nanofiber arrays.

S2 Waviness Correction for Square and Hexagonal Packing

Scaling of the waviness correction (Ω) with the waviness ratio (w) for square packing ($\rightarrow \Omega_{\square}(w)$) with coefficient of determination $\mathbb{R}^2 > 0.997$:

$$\Omega_{\square}(w) = \begin{cases} 4(w)^{1.6} + 1, & w < 0.05 \\ -0.0012(w)^{-1.2} + 1.076, & 0.05 \leq w \leq 0.125 \\ -0.0057(w)^{-0.47} + 1.076, & 0.125 < w \leq 0.3 \end{cases} \tag{S1}$$

Scaling of Ω with w for hexagonal packing ($\rightarrow \Omega_{\hexagon}(w)$) with $\mathbb{R}^2 > 0.991$:

$$\Omega_{\hexagon}(w) = \begin{cases} 2.5(w)^{1.9} + 1, & w \leq 0.1 \\ -0.00143(w)^{-1.56} + 1.082, & 0.1 < w \leq 0.3 \end{cases} \tag{S2}$$

Table S1 Waviness correction for NFs exhibiting square packing, $\Omega_{\square}(w)$, and hexagonal packing, $\Omega_{\hexagon}(w)$, as a function of the waviness ratio, w, as presented in Fig. 3 in the main text.

w []	$\Omega_{\square}(w)$ []	$\Omega_{\hexagon}(w)$ []
0.01	1.002	1.000
0.025	1.011	1.002
0.05	1.033	1.008
0.075	1.049	1.018
0.1	1.058	1.031
0.125	1.061	1.043
0.15	1.062	1.054
0.175	1.063	1.061
0.2	1.064	1.066
0.25	1.065	1.070
0.3	1.066	1.071

S3 Waviness Scaling with Carbon Nanotube Volume Fraction

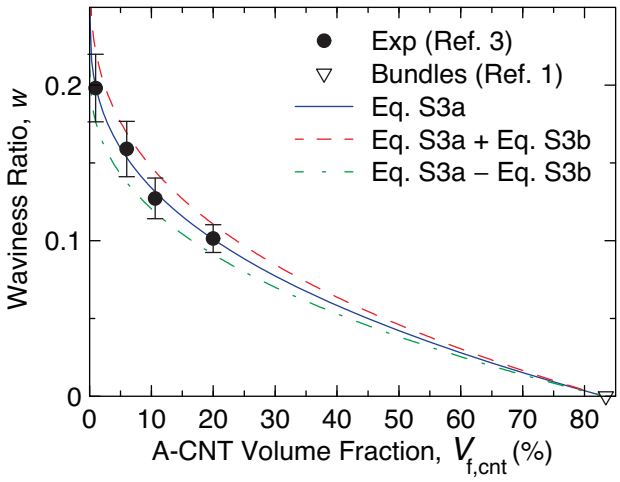

Fig. S2 Scaling of the waviness ratio (w) with the A-CNT volume fraction ($V_{\mathrm{f,cnt}}$).[3] The plot shows that w can be reduced by $\sim 50\%$ by increasing V_{f} from ~ 1 vol. % CNTs to ~ 20 vol. % CNTs, and that the scaling of w with $V_{\mathrm{f,cnt}}$ can be modeled using a combination of power laws for both the mean values (eqn (S3a)) and variances (eqn (S3b)).[3]

The scaling of the waviness ratio (w) as a function of the aligned carbon nanotube (A-CNT) volume fraction ($V_{\mathrm{f,cnt}}$) was studied experimentally in a recent study (see Fig. S2).[3] Using the theoretical maximum $V_{\mathrm{f,cnt}}$ of ~ 8 nm outer diameter CNTs ($\rightarrow V_{\mathrm{f,cnt}}^{\mathrm{max}} \approx 83.45\%$ CNTs) where $w = 0$,[1] both the mean values of w (μ_{w}) and the statistical uncertainties in μ_{w} ($\sigma_{\mathrm{w}}/\sqrt{n}$) were fit independently via power laws as follows:

$$\mu_{\mathrm{w}}(V_{\mathrm{f}}) = a_1 (V_{\mathrm{f}})^{b_1} + c_1 \tag{S3a}$$

$$\sigma_{\mathrm{w}}(V_{\mathrm{f}})/\sqrt{n} = a_2 (V_{\mathrm{f}})^{b_2} + c_2 \tag{S3b}$$

where $a_1 = -0.04967$, $b_1 = 0.3646$, and $c_1 = 0.2489$ with coefficient of determination ($\mathbb{R}^2$) of 0.9996, and $a_2 = -0.0852/\sqrt{n}$, $b_2 = 0.2037$, $c_2 = 0.2100/\sqrt{n}$, and $n = 30$ CNTs with $\mathbb{R}^2 = 0.9812$.[3]

S4 Waviness Corrected Carbon Nanotube Separation and Coordination Number

Table S2 Average aligned carbon nanotube (A-CNT) separation, Γ_{cnt}, and waviness correction, Ω_{cnt}, as a function of the A-CNT volume fraction, $V_{f,cnt}$, evaluated using the simulation described in the main text (see Fig. 4a).

$V_{f,cnt}$ [%]	Γ_{cnt} [nm]	Ω_{cnt} []
0.6	106.44	1.0685
0.7	97.81	1.0689
0.8	90.75	1.0682
0.9	85.00	1.0686
1	80.06	1.0681
1.5	63.42	1.0676
2	53.51	1.0673
3	41.72	1.0656
4	34.69	1.0639
5	29.89	1.0618
6	26.36	1.0600
7	23.62	1.0584
8	21.41	1.0564
9	19.58	1.0544
10	18.03	1.0522
11	16.71	1.0503
12	15.55	1.0480
13	14.53	1.0461
14	13.63	1.0443
15	12.81	1.0422
16	12.08	1.0399
17	11.41	1.0379
18	10.80	1.0355
19	10.24	1.0336
20	9.72	1.0315
21	9.25	1.0294
22	8.81	1.0275
23	8.39	1.0254

Table S3 Effective two dimensional coordination number, N, estimated for aligned carbon nanotubes (A-CNTs) as a function of the A-CNT volume fraction, $V_{\mathrm{f,cnt}}$, evaluated using previously reported experimental data (see Fig. 4b in the main text).[1]

$V_{\mathrm{f,cnt}}$ [%]	N
1	4.30 ± 0.05
6	4.70 ± 0.03
10.6	4.90 ± 0.1
20	5.30 ± 0.05

References

[1] I. Y. Stein and B. L. Wardle, Phys. Chem. Chem. Phys. **15**, 4033 (2013).

[2] I. Y. Stein, N. Lachman, M. E. Devoe, and B. L. Wardle, ACS Nano **8**, 4591 (2014).

[3] I. Y. Stein, D. J. Lewis, and B. L. Wardle, Nanoscale **7**, 19426 (2015).