Supplementary Information

Insights into controlling bacterial cellulose nanofiber film properties through balancing thermodynamic interactions and colloidal dynamics.

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This Supplementary Information contains:

- Supplementary Tables S1-S3
- Supplementary Figures S1-S11

Supplementary Tables

Table S1. Hansen Parameters calculations. Hansen solubility parameters of IPA-water mixtures varying with volume fraction of water (φ_{water}), with polar, dispersive and hydrogen bonding parameters. $\delta_{Solvent,CNF}$ is the solubility parameter difference between CNF and the solvents, normalized chi is obtained by dividing $\delta_{Solvent,CNF}$ with $\delta_{water,CNF}$. The solvents chosen for experimentation are marked in green.

	φ_{water}	D ($MPa^{0.5}$)	P ($MPa^{0.5}$)	H ($MPa^{0.5}$)	$\delta_{Solvent,CNF}$ ($MPa^{0.5}$)	$\chi_{normalized}$ ($\delta_{Solvent,CNF}/\delta_{WaterCNF}$)
1	0	15.8	6.1	16.4	4.75	0.18
2	0.075	15.7775	6.8425	18.3425	4.46	0.15
3	0.15	15.755	7.585	20.285	5.09	0.19
4	0.3	15.71	9.07	24.17	7.99	0.30
5	0.5	15.65	11.05	29.35	12.97	0.5
6	0.75	15.575	13.525	35.825	19.63	0.74
7	0.85	15.545	14.515	38.415	22.34	0.86
8	1	15.5	16	42.3	26.43	1.00

Table S2. Guinier fitting parameters of SAXS data for the three solvents. Intensity of the scattered X-ray at 0 scattering angle (I_0), radius of gyration (R_g), other fitting parameters ($(qRg)_{max}$ and $(qRg)_{max}$ and fitting quality factor R^2).

Sample	I_0 (au)	$(qR_g)_{max}$	$(qR_g)_{min}$	R_g (nm)	R^2
100% DI	6.82	2.53	1.16	30.5	0.87
50%/50% IPA/DI	29.73	2.17	1.35	37.06	0.95
92.5%/7.5% IPA/DI	16.57	2.40	1.36	41.01	0.96

Table S3. BIFT fitting parameters of SAXS data for the three solvents. Intensity of the scattered X-ray at 0 scattering angle (I_0), radius of gyration (R_g), maximum dimension or the largest distance between any two points within a particle (D_{max}), and fitting quality factor (R^2).

Sample	$I_{0\ (}$au)	R_g (nm)	D_{max} (nm)	R^2
100% DI	7.29	32.41	89.85	0.99
50%/50% IPA/DI	30.05	38.15	113.83	0.99
92.5%/7.5% IPA/DI	16.01	41.6	121.54	0.99

Supplementary Figures

Figure S1. Electrokinetic Studies. (a-b) Conduometric titration of CNF dispersions in DI water for surface charge measurements and linear fitting to determine surface charge neutralization of CNF. **(c)** Zeta potential variation and pH values versus normalized chi (without the addition of any base or acid).

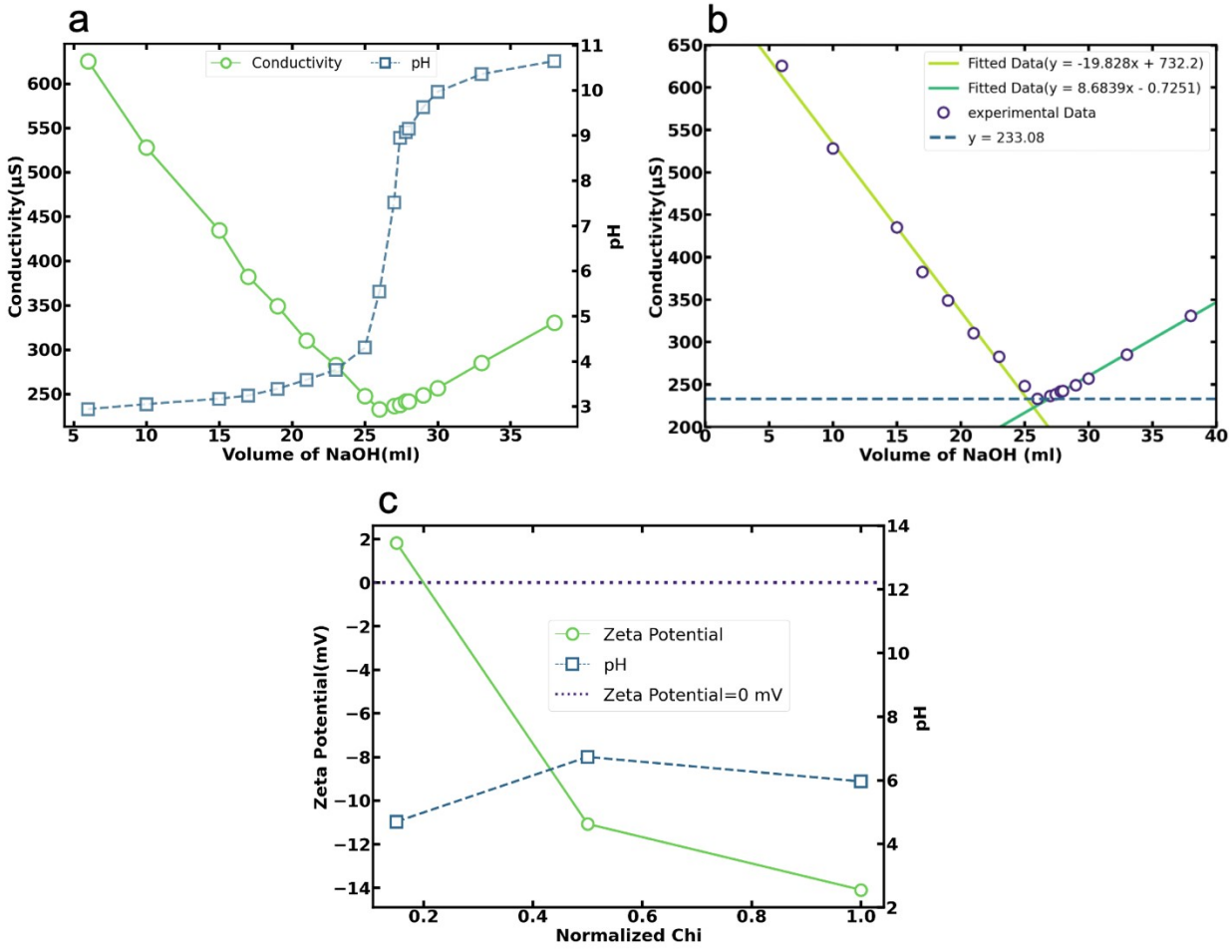


Figure S2. Hansen Parameters space. Fig 1: Polar, dispersive and hyderogen bonding parameters plotted for the different solvents and cellulose for visual representation of $\delta_{Solvent,CNF}$.

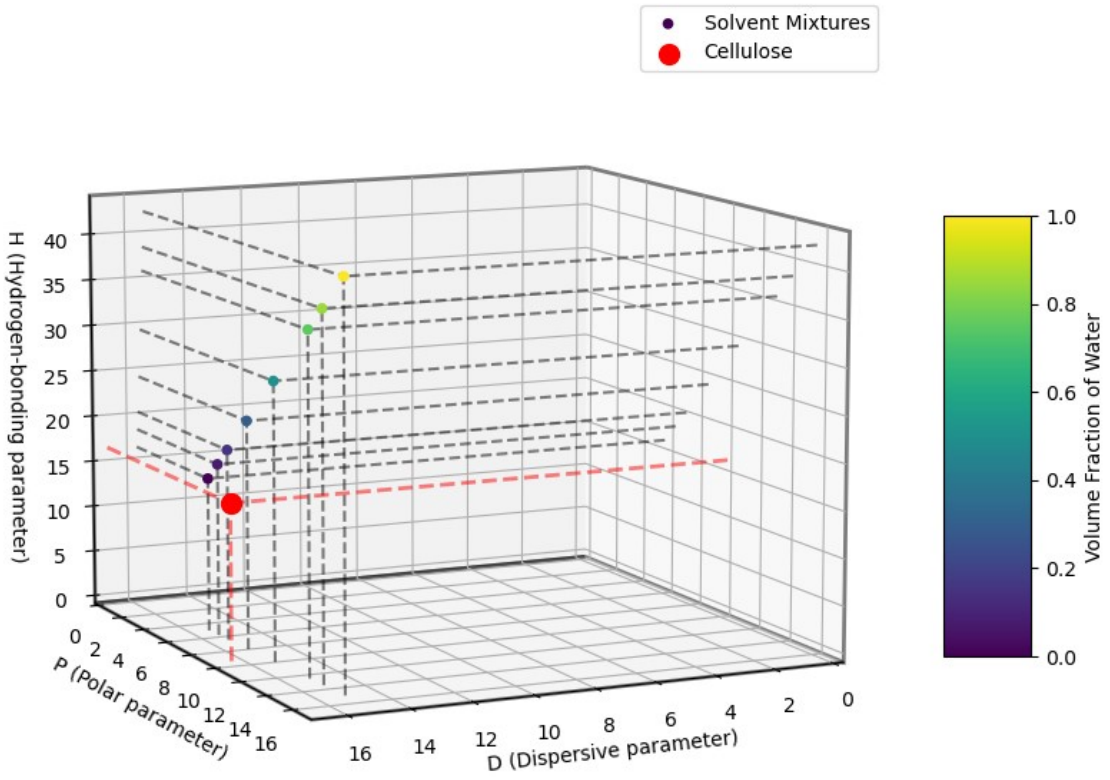
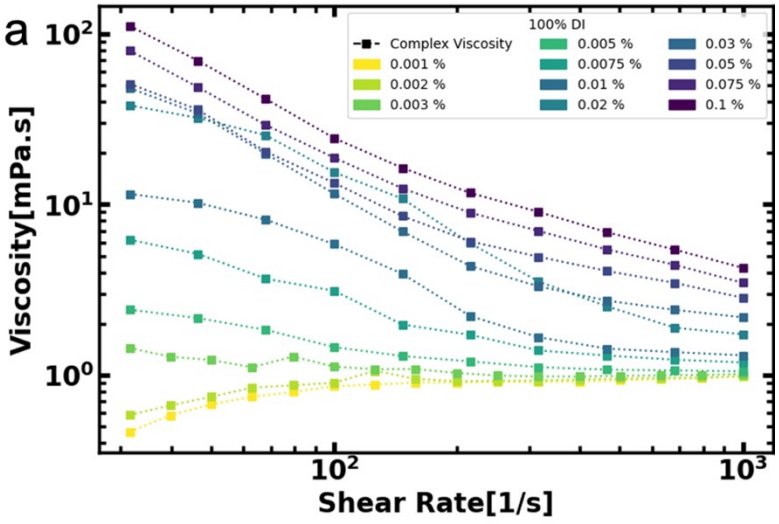
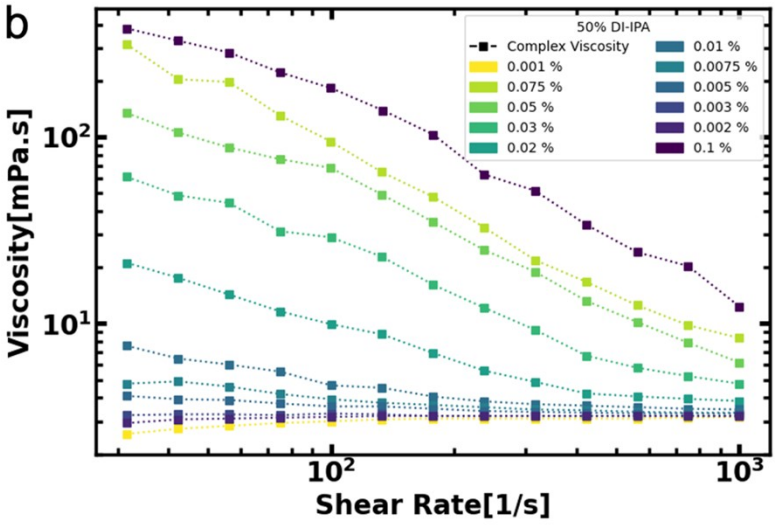


Figure S3. Rheological characterization. Viscosity against shear rate plots for (a) 100 % water, (b) 50 % water /50% IPA and (c) 7.5% water \ 92.5% IPA dispersions from 0.001 wt. % to 0.1 wt.%.


a
100% DI
Complex Viscosity
0.001 %
0.002 %
0.003 %
0.005 %
0.0075 %
0.01 %
0.02 %
0.03 %
0.05 %
0.075 %
0.1 %
Viscosity[mPa.s]
Shear Rate[1/s]


b
50% DI-IPA
Complex Viscosity
0.001 %
0.075 %
0.05 %
0.03 %
0.02 %
0.01 %
0.0075 %
0.005 %
0.003 %
0.002 %
0.1 %
Viscosity[mPa.s]
Shear Rate[1/s]

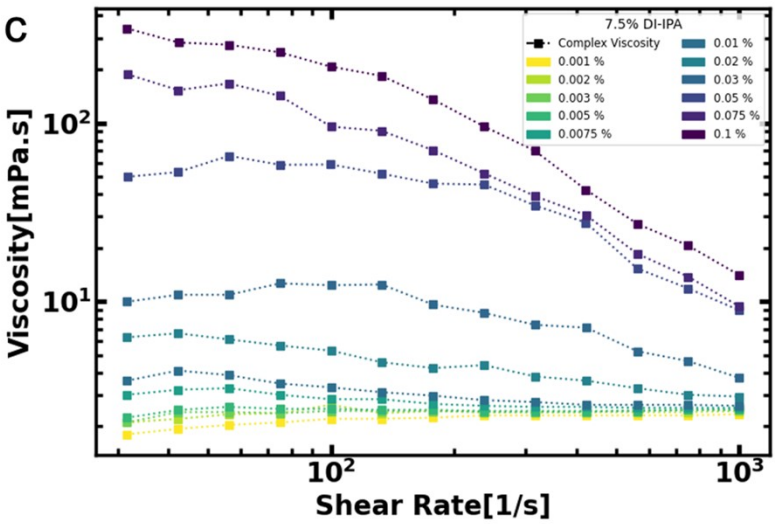

c
7.5% DI-IPA
Complex Viscosity
0.001 %
0.002 %
0.003 %
0.005 %
0.0075 %
0.01 %
0.02 %
0.03 %
0.05 %
0.075 %
0.1 %
Viscosity[mPa.s]
Shear Rate[1/s]

Figure S4. Rheological study of thermodynamic behavior. Plot comparing relative viscosity against concentration for the three dispersions.

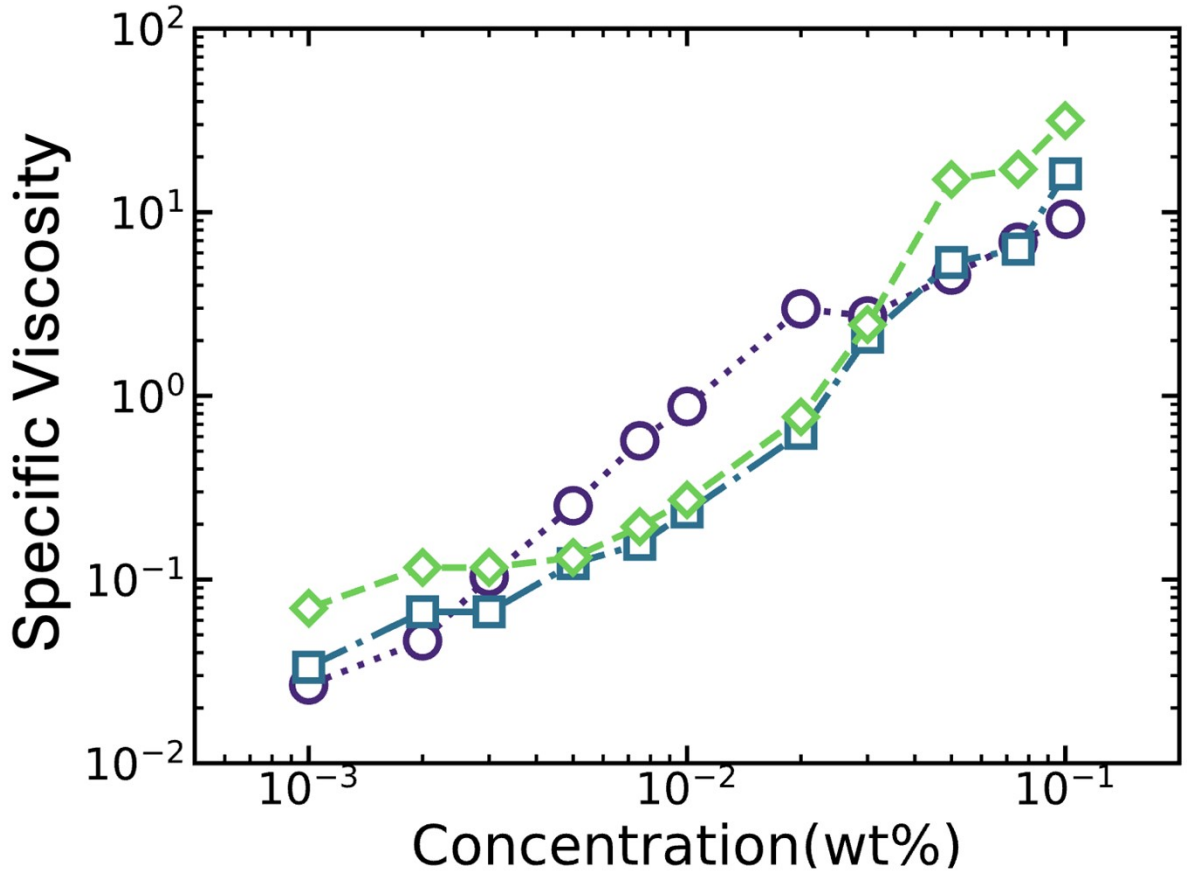


Figure S5. SAXS analysis. Guinier Fitting (left) and Residual Plots of the Guinier Fit (right) for (a) 100 % water, (b) 50 % water /50% IPA and (c) 7.5% water \ 92.5% IPA dispersions.

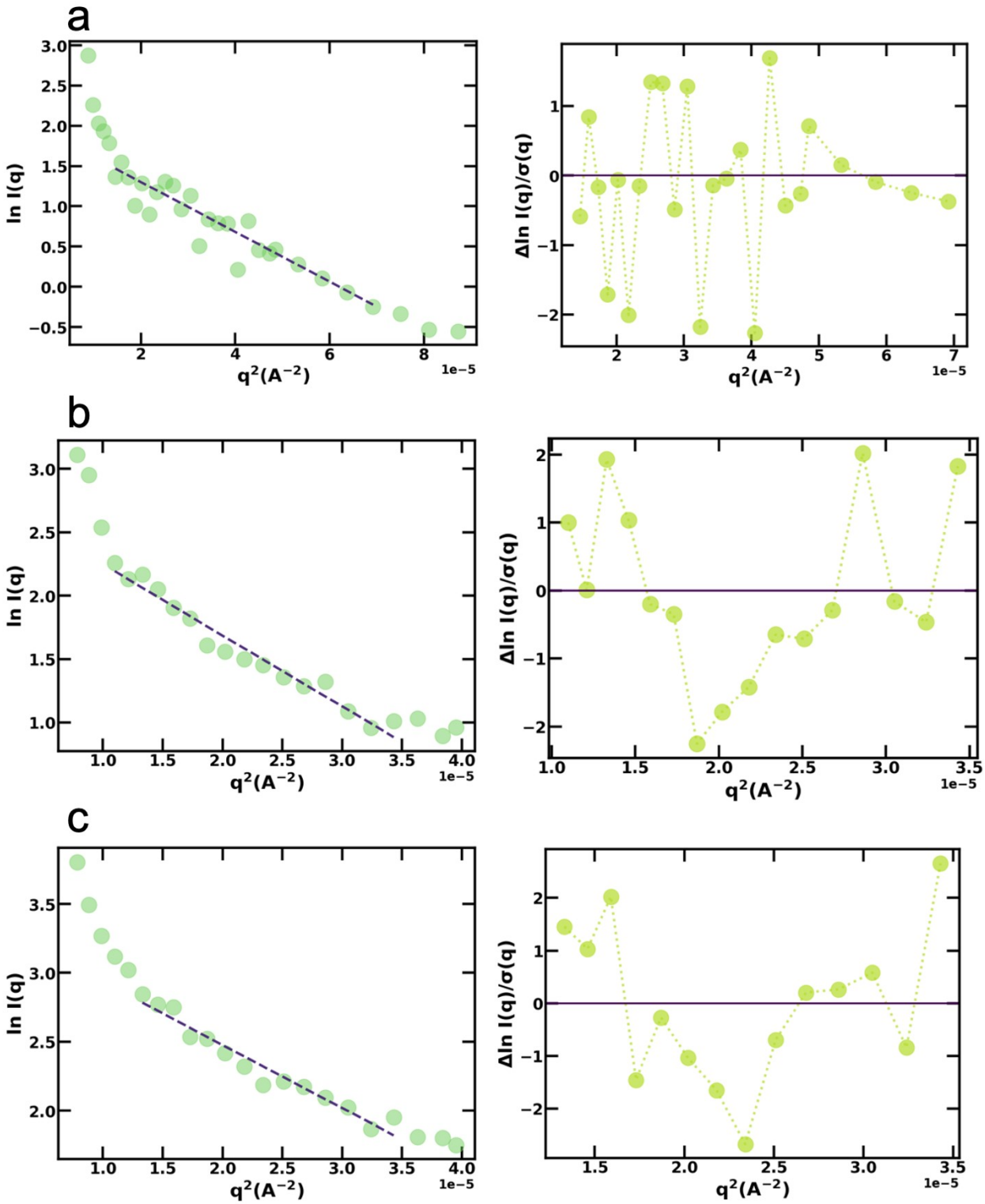


Figure S6. Mechanical testing of films. Engineering stress against engineering strain tests for (a) 100 % water, (b) 50 % water /50% IPA and (c) 7.5% water \ 92.5% IPA casted films of 0.2 wt.% CNF concnetration, along with values for Yong's modulus, ultimate tensile strength, elongation to break and toughness (n=12).
from 0.001 wt. % to 0.1 wt.%.

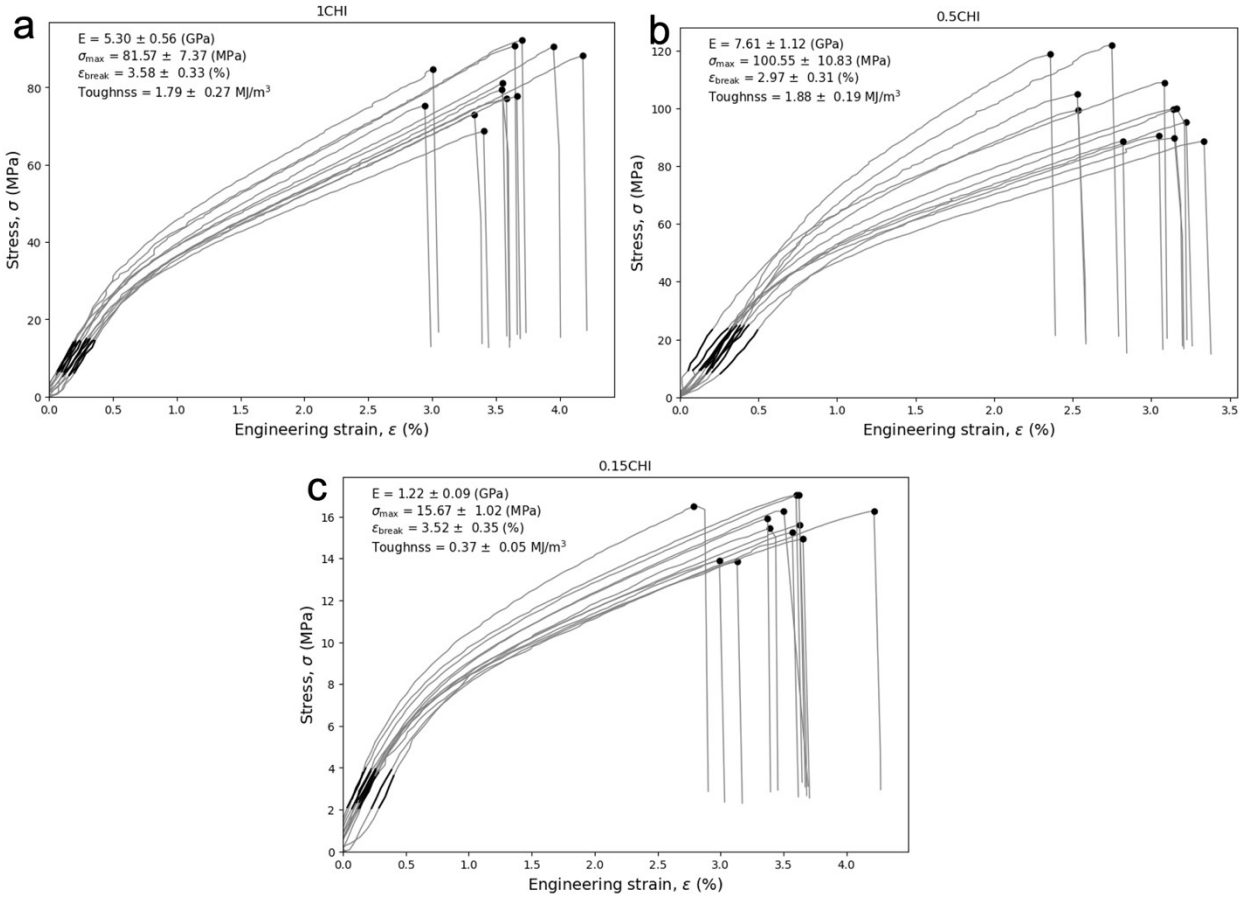


Figure S7. SEM image analysis. Detected voids from SEM images of the fractured films casted from (a) 100 % water, (b) 50 % water /50% IPA and (c) 7.5% water \ 92.5% IPA dispersions.

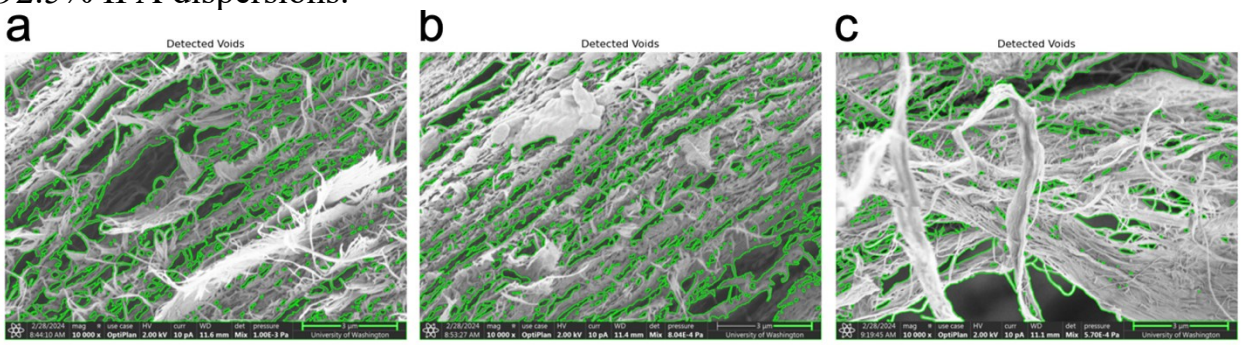


Figure S8. SEM image analysis. Surface roughness and fiber pull-out counts quantified using (a) Canny Edges and (b) Sobel Edges from SEM images of the fractured films casted from 100 % water dispersions. (c) Labeled regions from the analysis.

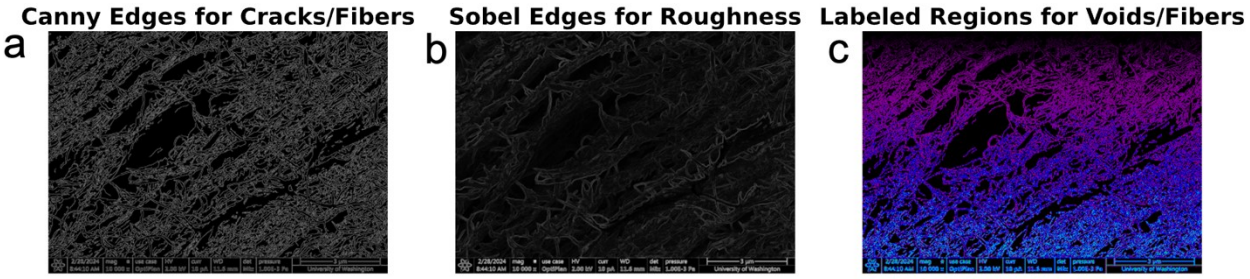


Figure S9. SEM image analysis. Surface roughness and fiber pull-out counts quantified using (a) Canny Edges and (b) Sobel Edges from SEM images of the fractured films casted from 50 % water /50% IPA dispersions. (c) Labeled regions from the analysis.

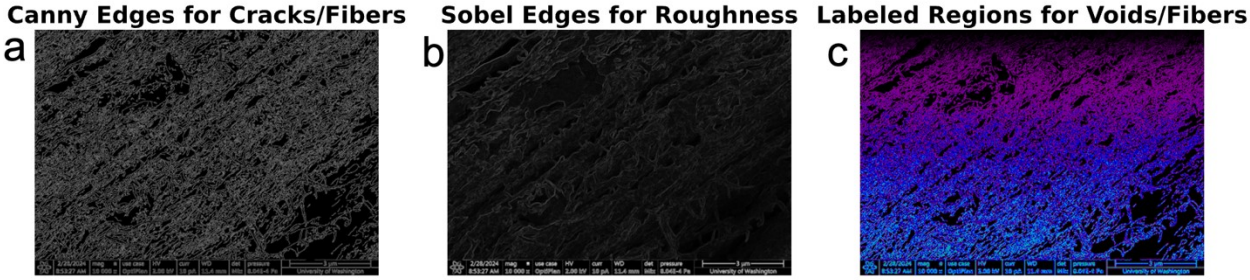


Figure S10. SEM image analysis. Surface roughness and fiber pull-out counts quantified using (a) Canny Edges and (b) Sobel Edges from SEM images of the fractured films casted from 7.5% water \ 92.5% IPA dispersions. (c) Labeled regions from the analysis.

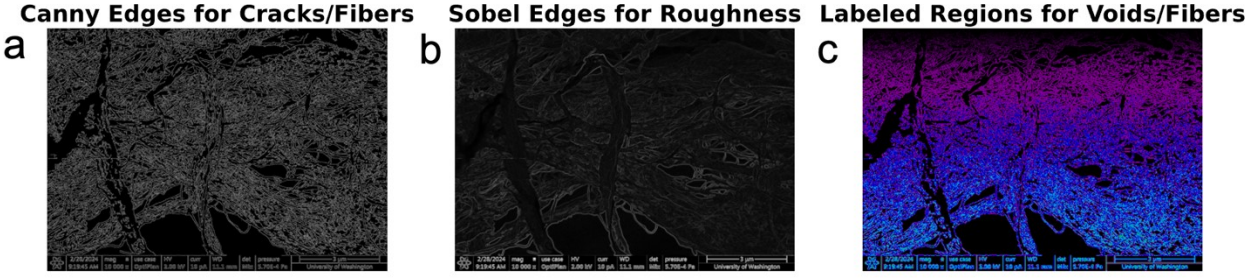


Figure S11. SEM image analysis. Normalized roughness, void and fiber pull-out count and void areas analyzed from the SEM images of the fracture surfaces of the CNF films.

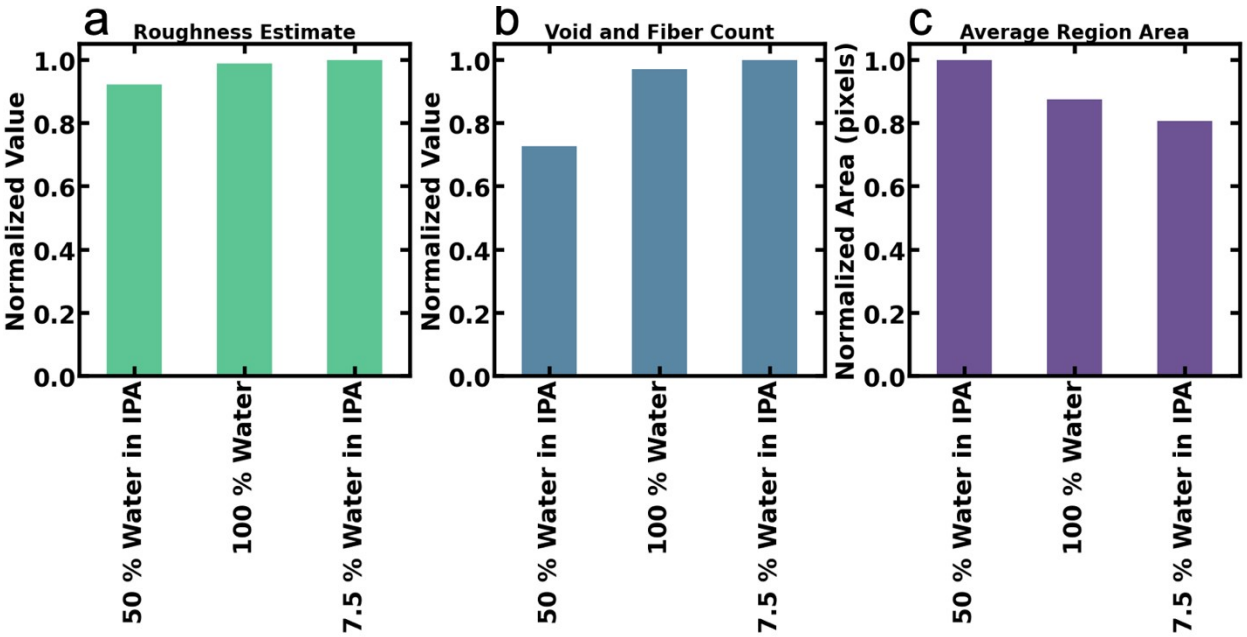


Figure S12. Colloidal Behavior of 0.02 wt% CNF in 100% DI. Three vials showing the colloidal behavior containing 0.02 wt% CNF, three samples were tried for repeatability.