

## Supporting Information

# Exploring anisotropic properties of chiral nematic cellulose nanocrystal aerogels: Outstanding directional mechanical strength and unexpected surface-dependent thermal conductivity

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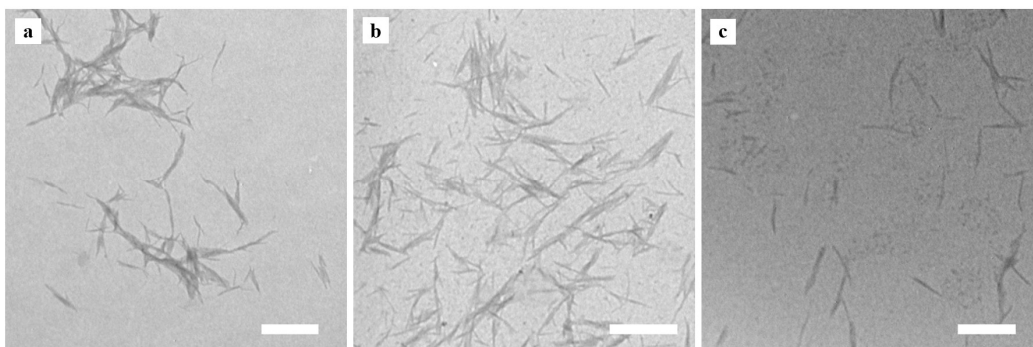
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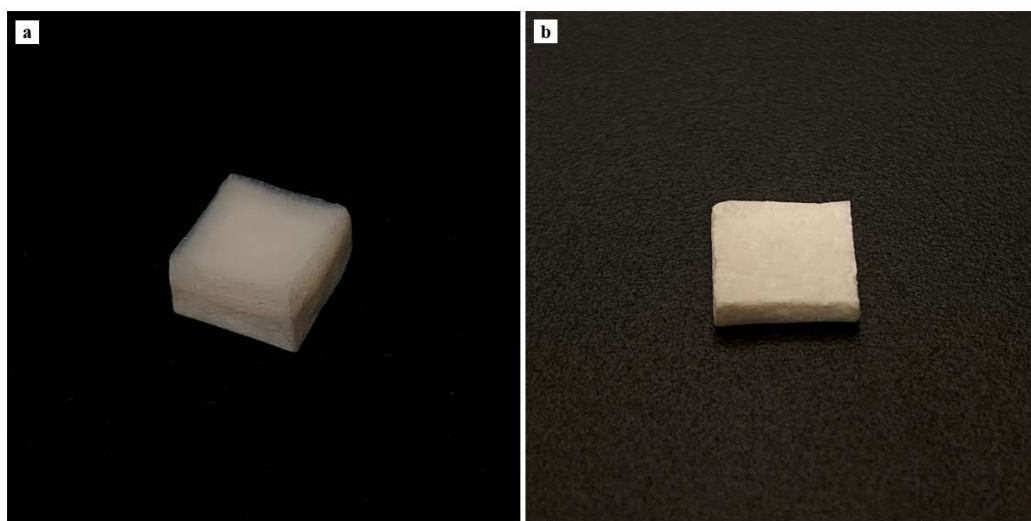
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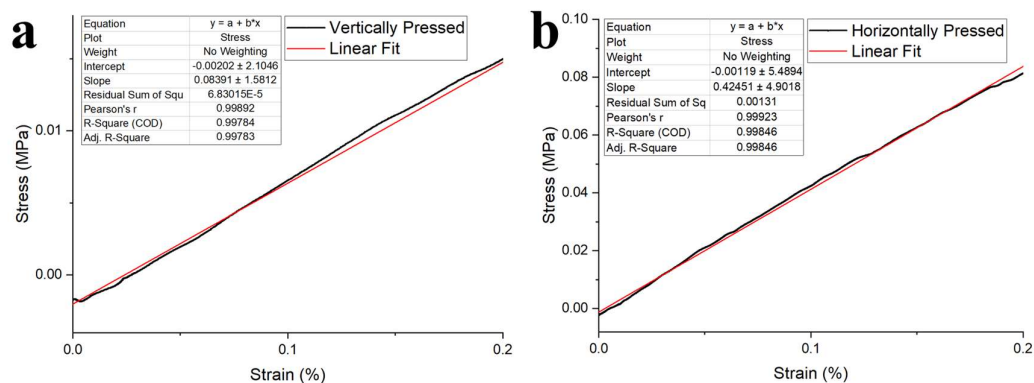
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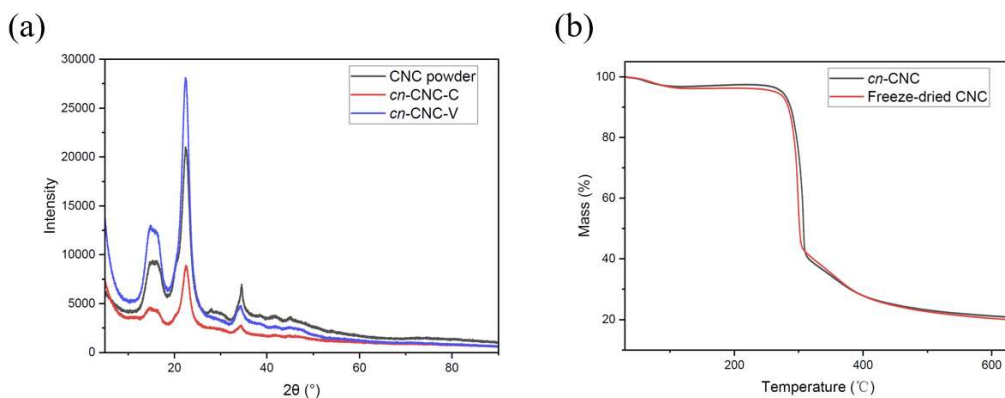
**Figure S1.** Transmission electron micrographs of CNCs from (a) the uniform CNC suspension, (b) the anisotropic phase of CNC suspension, and (c) the isotropic phase of CNC suspension. ImageJ (NIH, <http://imagej.nih.gov/ij/>) was used to manually measure CNC lengths and widths. Scale bar represents 500 nm for (a) and (b), 300 nm for (c).



**Figure S2.** *cn*-CNC-C sample (a) before and (b) after slicing off the top part.



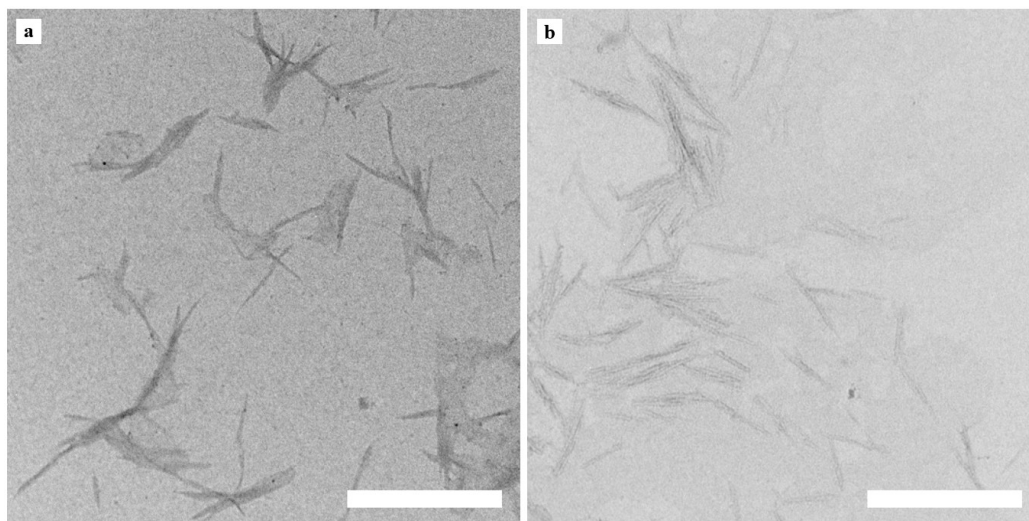
**Figure S3.** Linear regression analysis for representative stress-strain curves achieved from (a) vertical and (b) horizontal compression tests.



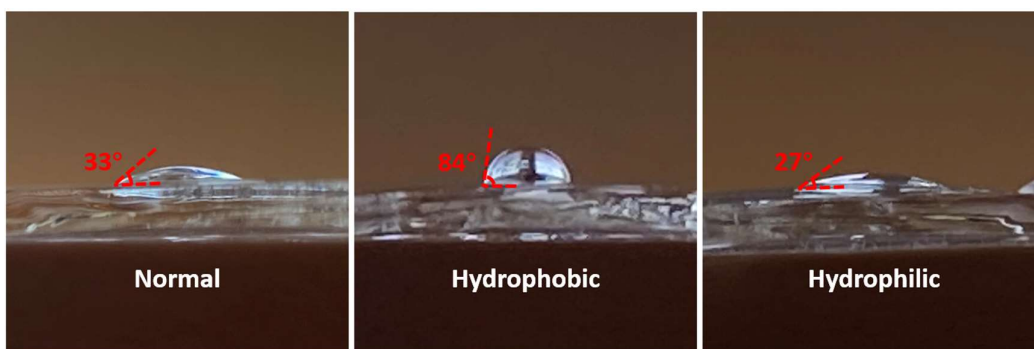
**Figure S4.** (a) Powder X-ray diffractograms of *cn*-CNC aerogels and freeze-dried CNC powder. Both display peaks characteristic of crystalline cellulose. (b) TGA curves of *cn*-CNC aerogel and freeze-dried CNC (nitrogen atmosphere; 10 °C/min). Both of them show similar thermal stability.

**Table S1.** Calculated density and porosity of *cn*-CNC-V

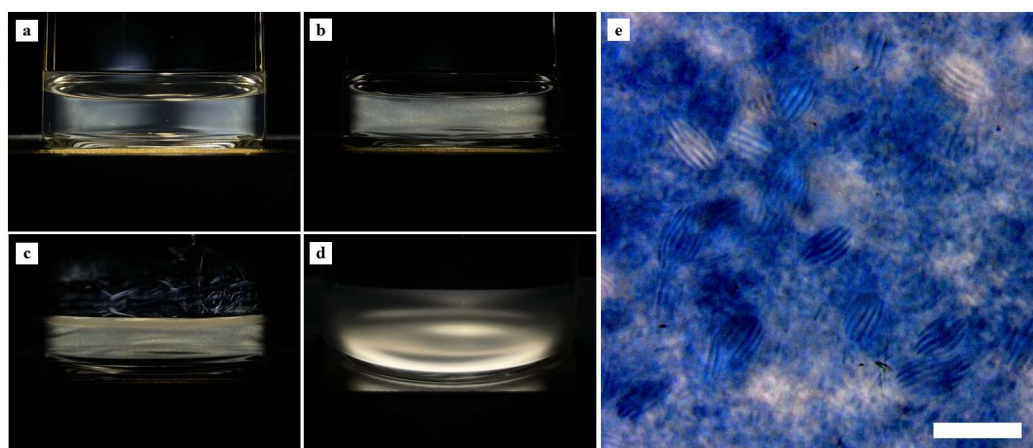
Diameter [mm]	Thickness [mm]	Weight [mg]	Density [ $\text{g cm}^{-3}$ ]	Porosity [%]
21.5	1.83	92.4	0.139	91.5



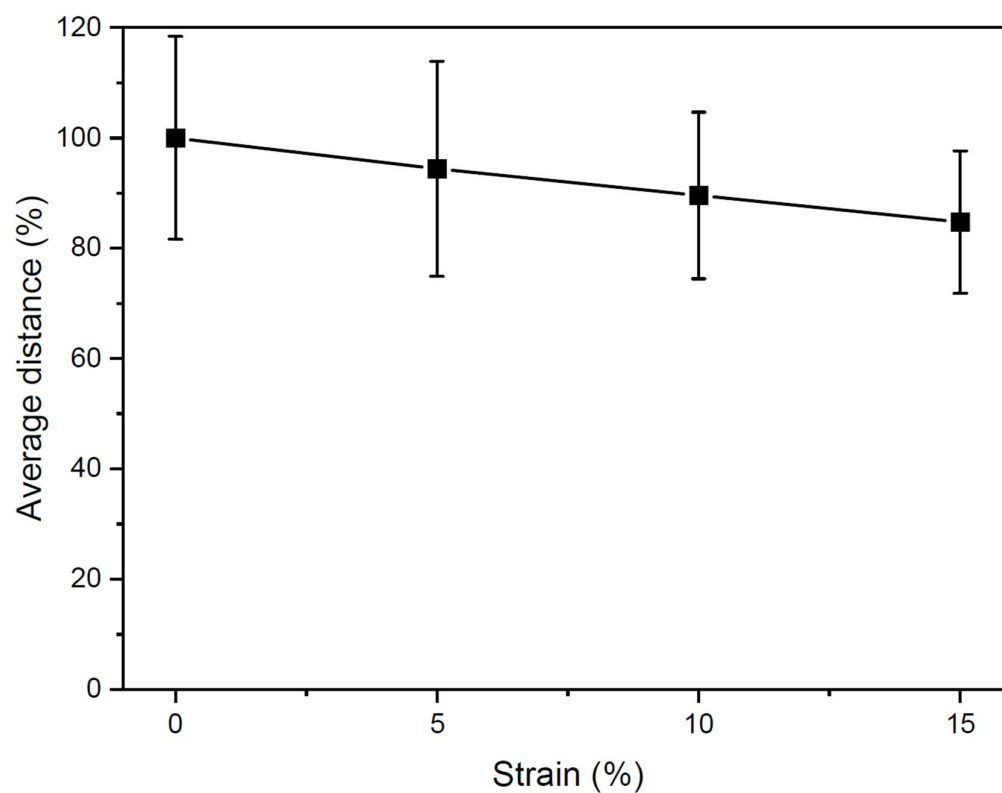
**Figure S5.** Transmission electron micrographs of CNCs from (a) the top part and (b) the bottom part of *cn*-CNC-V. ImageJ (NIH, <http://imagej.nih.gov/ij/>) was used to manually measure CNC lengths. Scale bar represents 500 nm.



**Figure S6.** Photographs of (a) normal vial, (b) hydrophobic vial and (c) hydrophilic vial with a 5  $\mu\text{L}$  droplet of water. Red dotted lines indicate water contact angles. ImageJ (NIH, <http://imagej.nih.gov/ij/>) was used for the analysis.



**Figure S7.** Time lapse study of isotropic CNC sample with equilibration. (a) After sonication, (b) after 24 h equilibration, (c) after adding ethanol, (d) after the gelation, and (e) POM micrograph of this equilibrated isotropic alcogel, showing tactoids as oval areas with fingerprint textures. Images shown in (a-c) are viewed between crossed polarizers. Scale bar represents 50  $\mu\text{m}$ .



**Figure S8.** The relationship between average distance between CNC pseudolayers (corresponding to half helical pitch) with vertical strain applied to the material. ImageJ (NIH, <http://imagej.nih.gov/ij/>) was used to manually measure the half helical pitch for the chiral nematic structure of the CNCs.