Electronic supplementary information

Electromechanical Coupling of Isotropic Fibrous Networks with Tailored Auxetic Behavior Induced by Water-Printing under Tension

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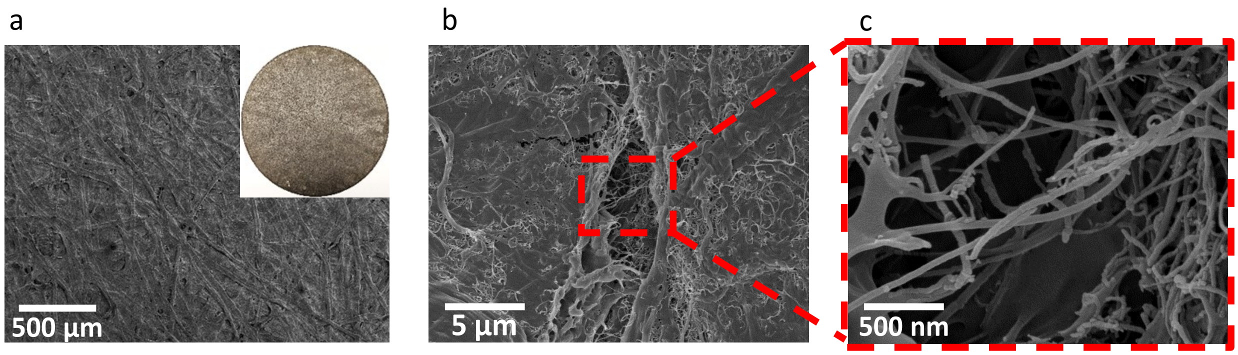

Fig. S1 (a-c) Representative SEM images of 10 CNT wt %-CPC at increasing magnification to examine the overall morphology and distribution of CNT-OH throughout the cellulose matrix. An image of the pristine CPC is shown as an inset in a.

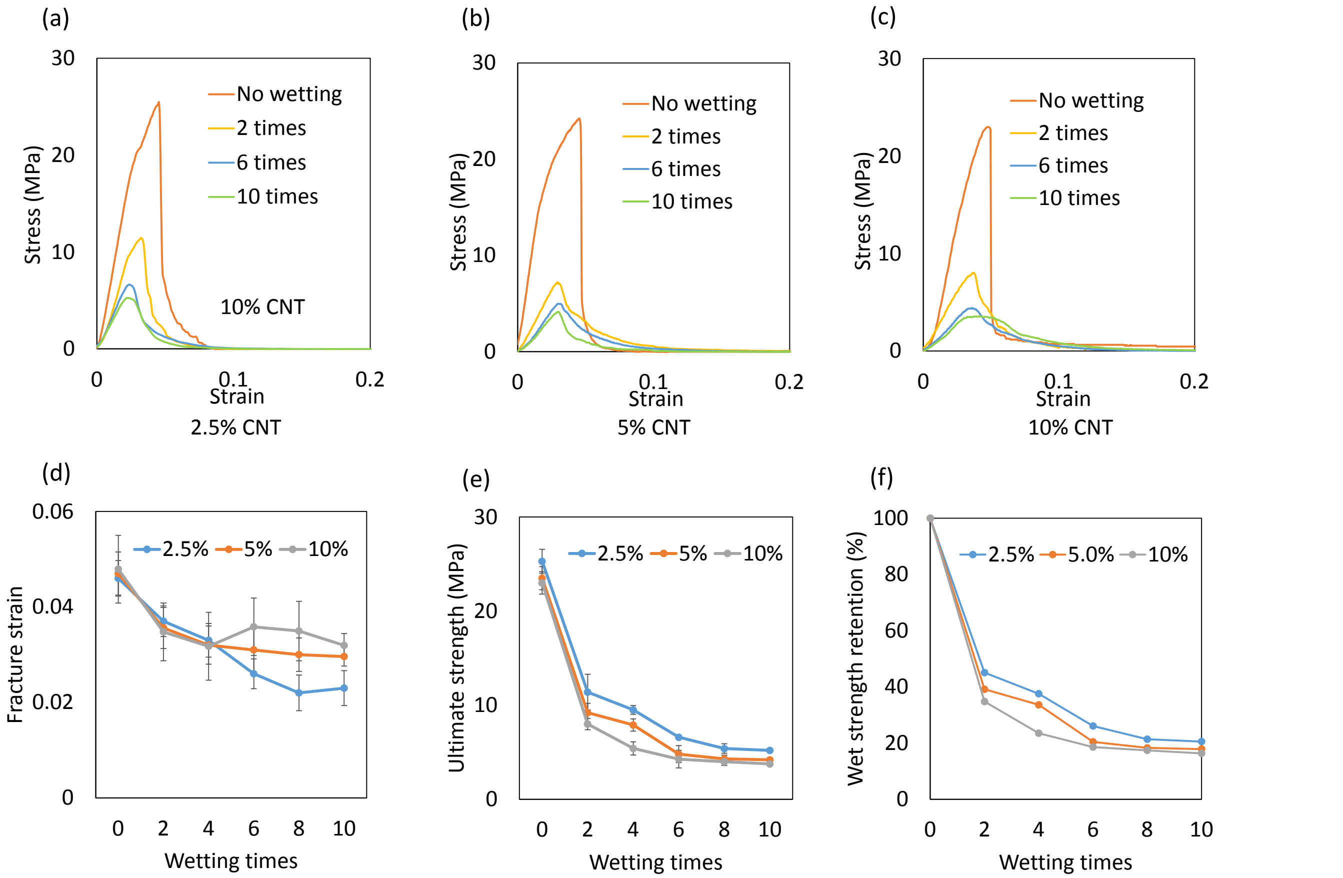

Fig. S2 Mechanical properties of CNT-cellulose composites by wet-straining method. (a-c) Stress-strain curves of CNT-cellulose composites with 2.5, 5 and 10-CNT wt % of after being applied wetting for 0, 2, 6 and 10 times. (d-e) Fracture strain and ultimate strength summary for CNT-cellulose composites with 2.5, 5 and 10-CNT wt % after being applied wetting for 0, 2, 4, 6, 8 and 10 times. (f) Wet strength retention of CNT-cellulose composites with 2.5, 5 and 10-CNT wt % after being applied wetting for 0, 2, 4, 6, 8 and 10 times.

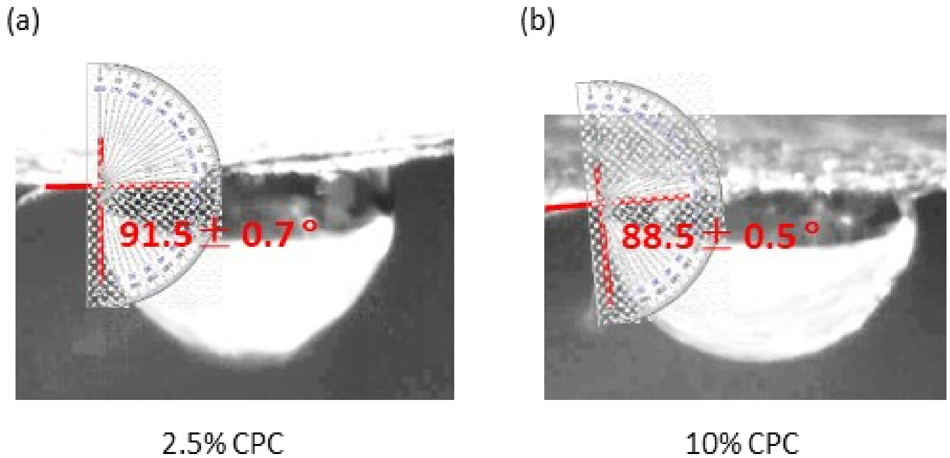

Fig. S3 Contact angles of (a) CPC with 2.5 wt %, and (b) CPC with 10 wt %.

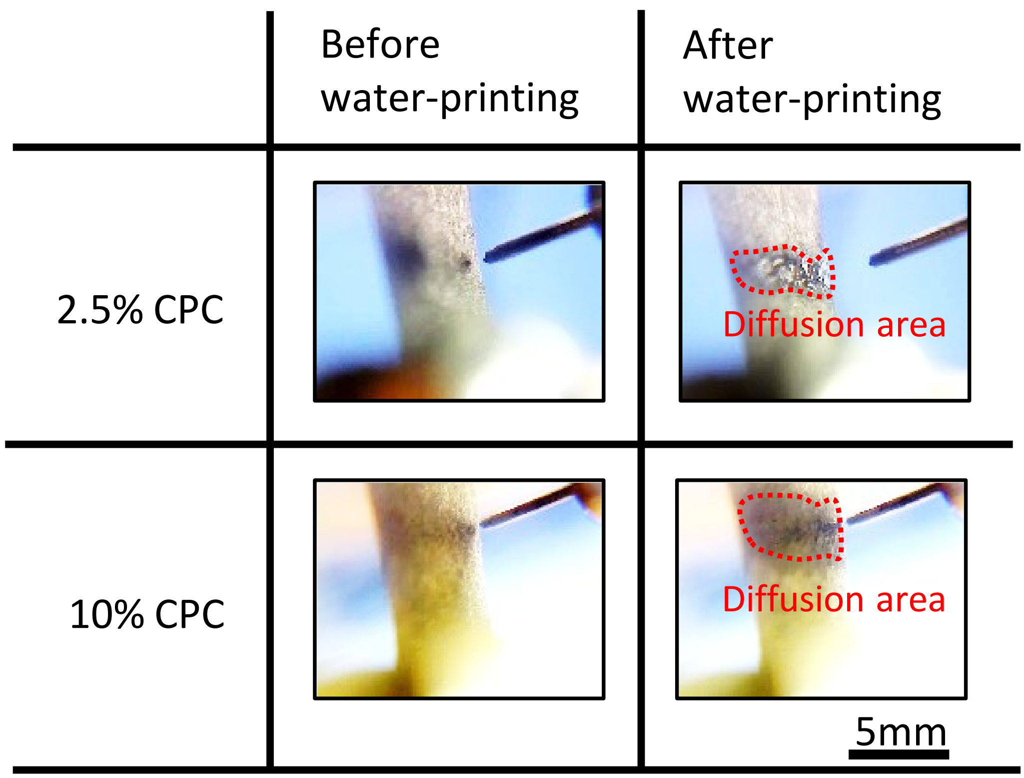

Fig. S4 Pictures of water-printing regions on CPC with 2.5 and 10 CNT wt %.

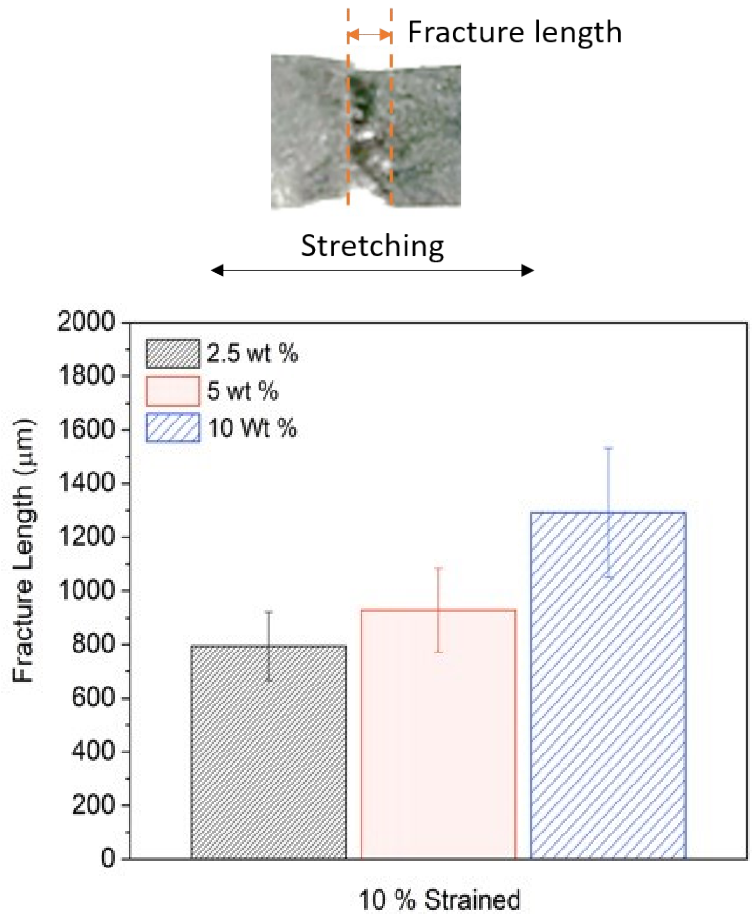

Fig. S5 Fracture lengths of CPC with 2.5, 5, and 10 CNT wt %.

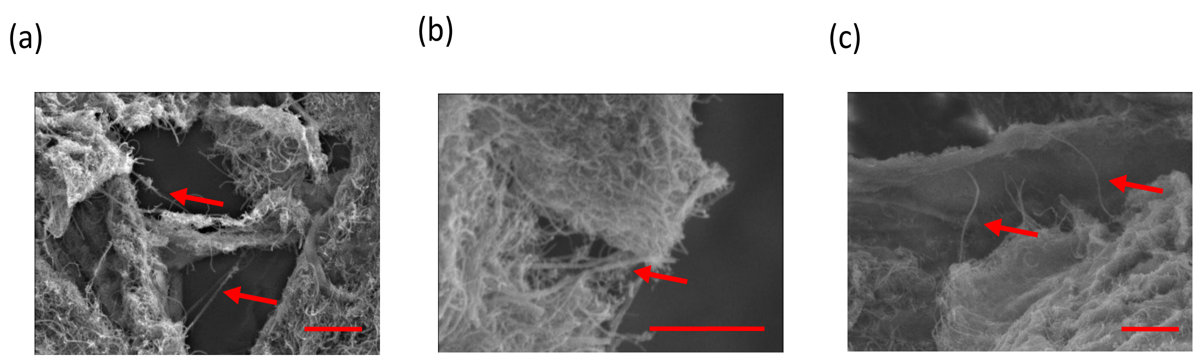

Fig. S6 SEM images of CNT spanning cellulose fibers. The scale bar is 2 μm.

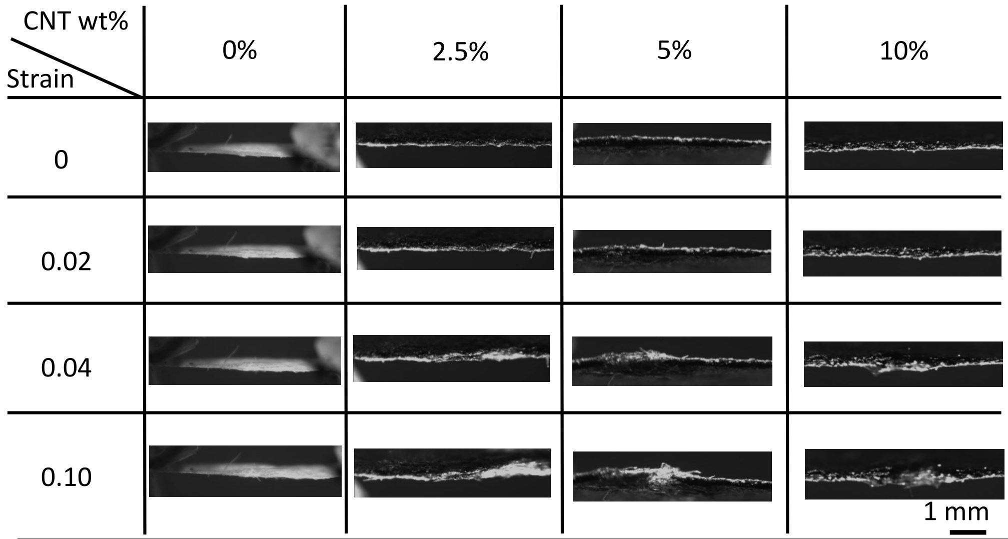

Fig. S7 Optical microscope images of profiles of pure paper and CPC with 2.5, 5, and 10 CNT wt %, showing the thickness at the strain of 0, 0.02, 0.04, and 0.10.

Simulation of the stress distribution of water-printed specimens under tension

The stress distribution of water-printed specimens under tension was studied by COMSOL. The solid mechanics models were built (Fig. S8a-c). The dimension of the specimen is 10 mm× 5 mm, with a 0.3 mm× 5 mm water-printed region in the middle, and two 0.1 mm× 5 mm transition regions between the water-printed and non-water-printed region. The dimensions of the transition regions were determined by the measurement of the shades on water-printed specimens. Regarding the actual printing-stretching, three scenarios were discussed with the auxetic behavior of paper, and the effect of water-printing alone, and the auxetic behavior of paper alone.

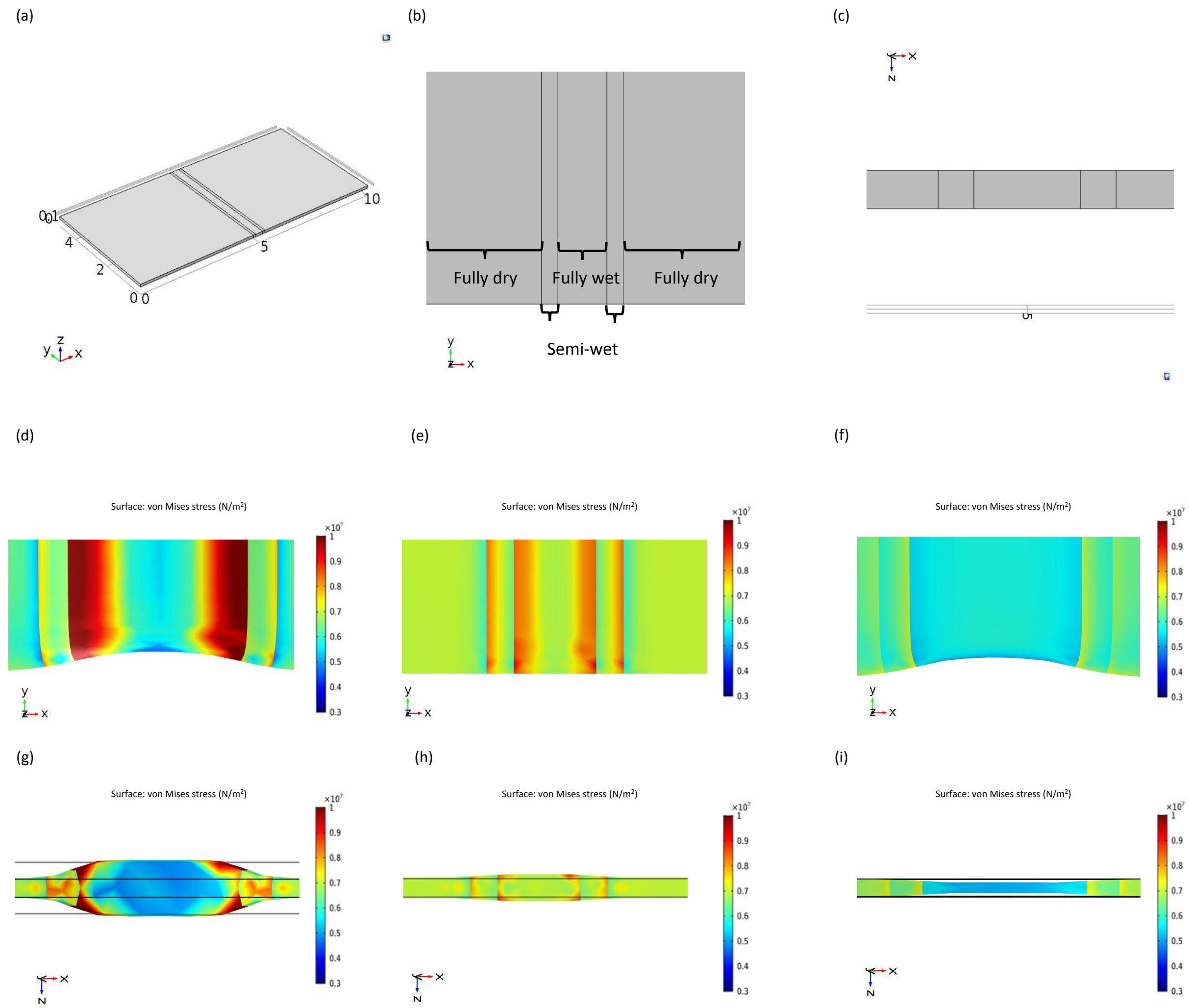

Fig. S8 COMSOL 3D models and simulation results (a) 3D view. (b) x-y planar view zoomed in dry, fully-wet, and semi-wet regions. (c) x-z planar view zoomed in a fully-wet region. (d) Stress distribution of x-y plane in scenario 1. (e) Stress distribution of x-y plane in scenario 2. (f) Stress distribution of x-y plane in scenario 3. (g) Stress distribution of x-z plane in scenario 1. (h) Stress distribution of x-z plane in scenario 2. (i) Stress distribution of x-z plane in scenario 3. (Scale factor for displacement: 50).

The parameters were as follows. The Young's modulus data were obtained from tensile tests (Fig. 2a). The Poisson's ratio was obtained from the characterization of the auxetic behavior (Fig. 2b).

<u>Scenario 1 - printing-stretching:</u>
Young's moduli for dry, semi-wet, and fully wet regions were 700, 420, and 130 Mpa
The Poisson's ratios were (0.2, 0.2, −0.2), (0.2, 0.2, −0.6), and (0.2, 0.2, −0.9).
<u>Scenario 2 - no water-printing, but with auxetic behavior:</u>
Young's moduli for dry, semi-wet, and fully wet regions were the same as 700 Mpa.
The Poisson's ratios were (0.2, 0.2, −0.2), (0.2, 0.2, −0.6), and (0.2, 0.2, −0.9)
<u>Scenario 3 - no auxetic behavior, but with water-printing:</u>
Young's moduli for dry, semi-wet, and fully wet regions were 700, 420, and 130 Mpa.
The Poisson's ratios were the same as (0.2, 0.2, 0.2).
The Young's moduli and Poisson's ratios of dry and fully-wet regions were obtained from the experiment. The data for semi-wet regions were the average values of dry and fully-wet regions. Each scenario was applied to build the 3D models.

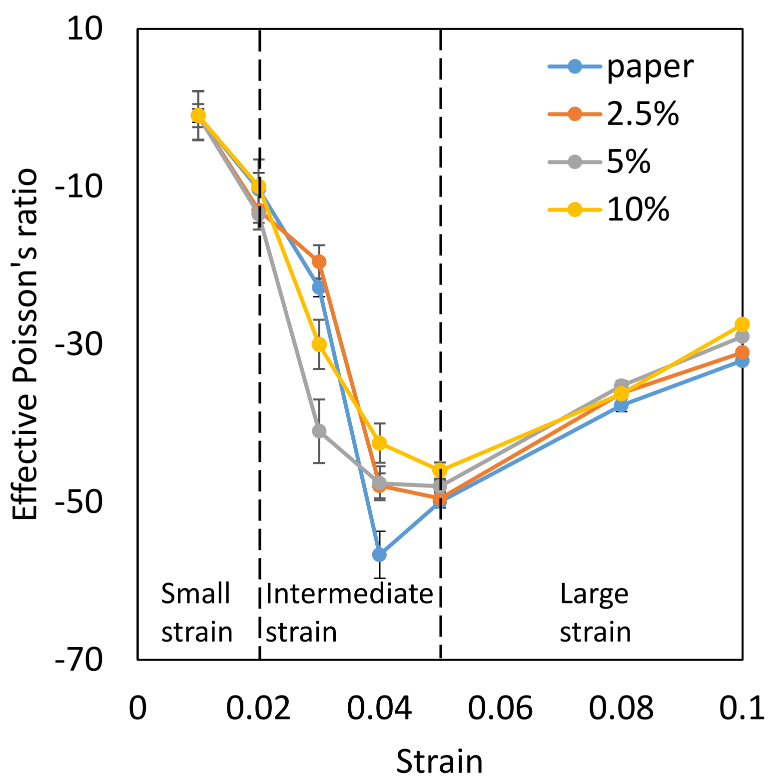

Fig. S9 Effective Poisson's ratio of pure paper and CPC with 2.5, 5, and 10 CNT wt% at strains of 0, 0.01, 0.02, 0.03, 0.04, 0.05, 0.08, and 0.10.

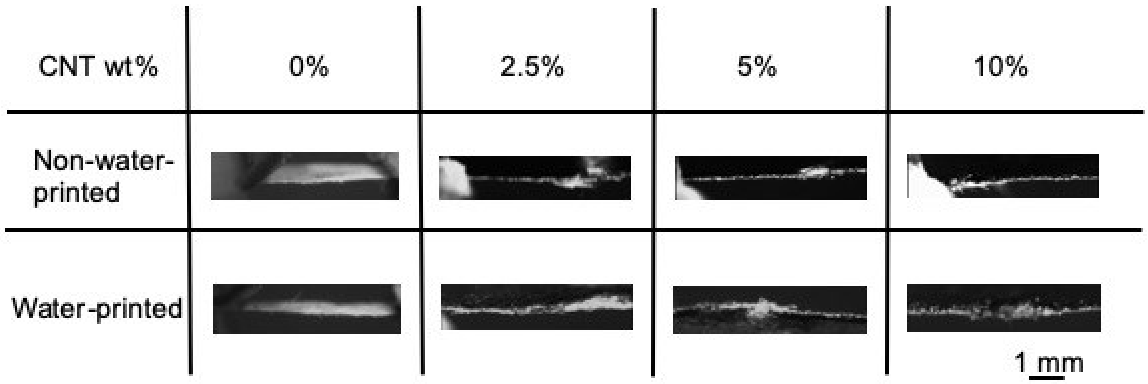

Fig. S10 Optical microscope images show the profiles of water-printed and non-water-printed pure paper and CPC with 2.5, 5, and 10 CNT wt %, at their maximum thickness.

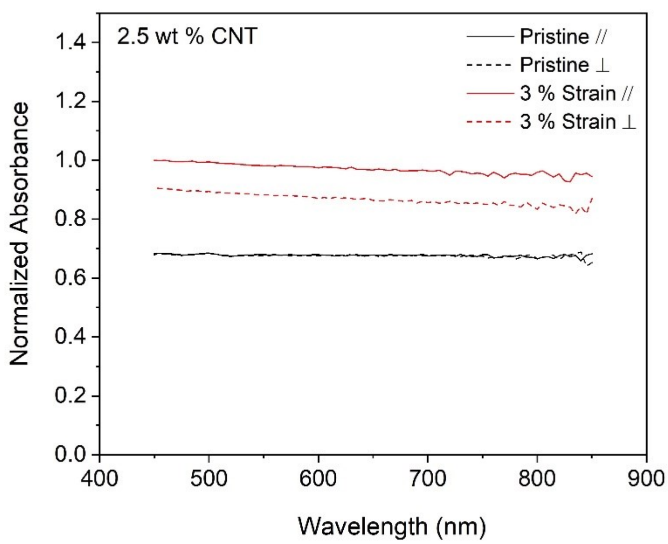

Fig. S11 Polarized absorption spectra of conductive papers containing 2.5 CNT wt % under strains at 0.0 and 0.03.

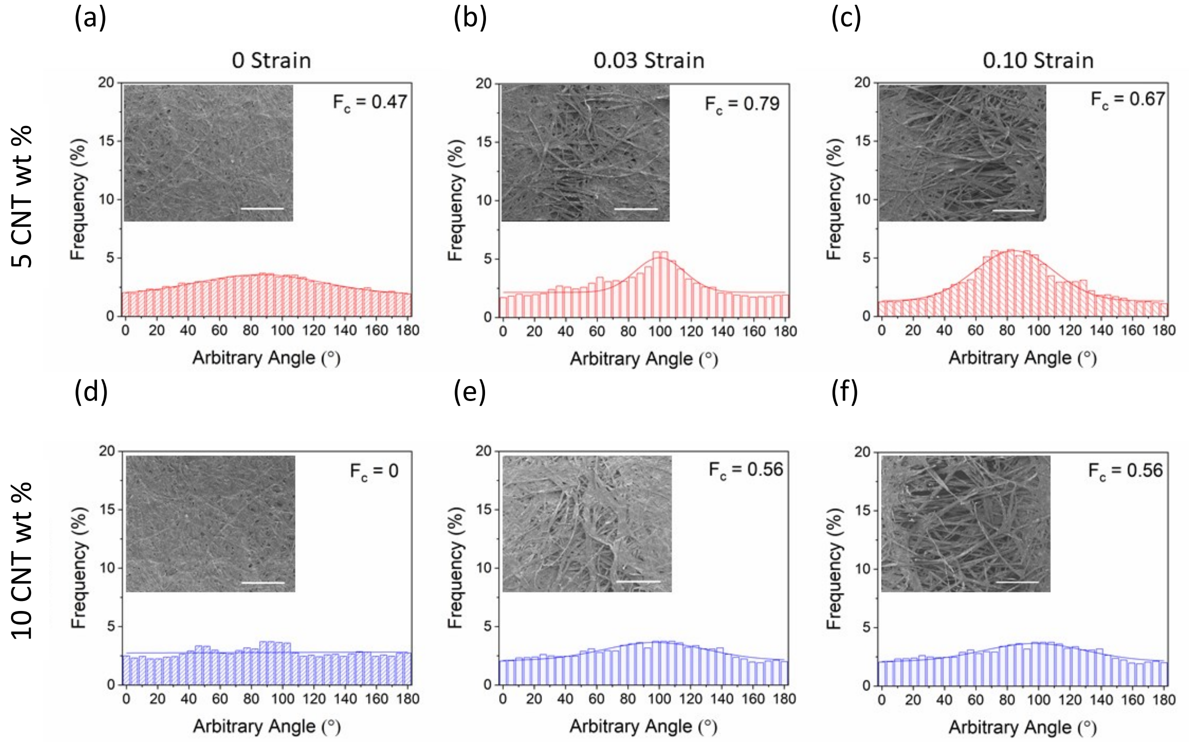

Fig. S12 SEM images of in-plane cellulose fiber orientations and corresponding orientation factors for CPC with (a-c) 5 CNT wt % at strain of 0 (a), 0.03 (b), and 0.10 (c), and (d-f) 10 CNT wt % at strains of 0 (d), 0.03 (e), and 0.10 (f).