

Supporting Information

Shear-Aligned Tunicate Cellulose Nanocrystals Reinforced Hydrogels with Mechano-Thermo-Chromic Properties

*Junmei Wang,^{‡a} Qiaoyun Cheng,^{‡a, b} Shengyao Feng,^c Lina Zhang,^{*a} and Chunyu Chang^{*a}*

^a College of Chemistry and Molecular Sciences, Hubei Engineering Center of Natural Polymer-based Medical Materials, Wuhan University, Wuhan 430072, China

^b Research Center for Sugarcane Industry Engineering Technology of Light Industry Institute of Bioengineering, Guangdong Academy of Science, Guangzhou 510316, China

^c National Synchrotron Radiation Lab and College of Nuclear Science and Technology, University of Science and Technology of China, Hefei 230026, China

[‡] Authors contributed equally to this work.

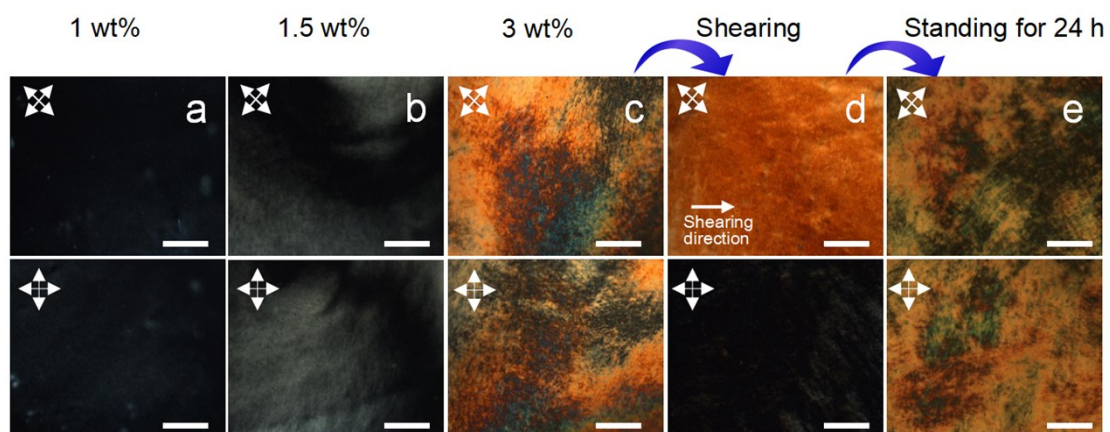


Figure S1. Polarized optical microscope (POM) images of TCNC aqueous suspensions with various concentration observed by placing samples with angles of 45° and $0/90^\circ$, respectively, the bar is $500\ \mu\text{m}$. (a) 1 wt%, (b) 1.5 wt%, (c) 3 wt%, (d) TCNC suspension (3 wt%) after shearing, and (e) the shear-oriented TCNC suspension stored for 24 h.

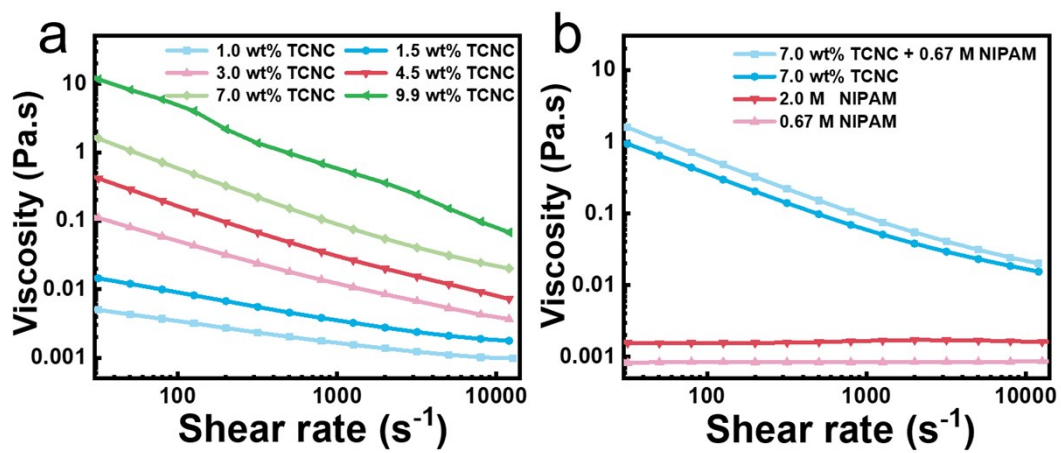


Figure S2. Viscosity of TCNC suspensions with different concentration as a function of shear rate (a), and viscosity of TCNC suspension (7 wt%), TCNC/0.67 M NIPAM mixture, and NIPAM solutions as a function of shear rate (b) at 25 °C.

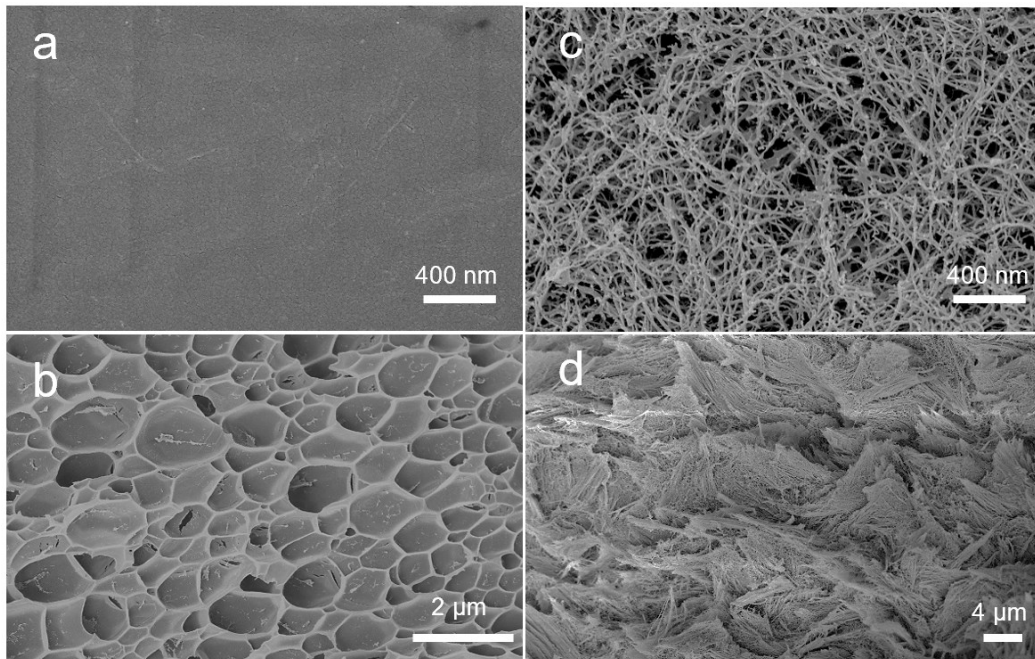


Figure S3. SEM images of surface (a, c) and cross-section (b, d) of PNIPAM hydrogel (a, b) and random TCNC/PNIPAM hydrogel (c, d).

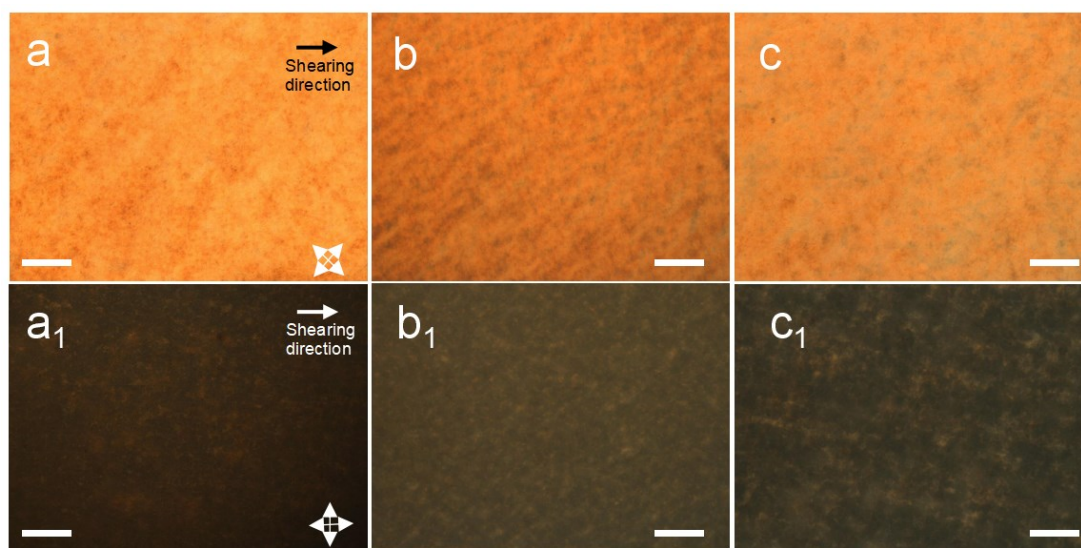


Figure S4. POM images of 7 wt% TCNC/0.67 M NIPAM mixture after shearing (a, a₁), hydrogel (b, b₁) and swollen hydrogel (c, c₁). The shear direction oriented at 45° and 0/90° with respect to the polarization axis of either polarizers. The bar is 500 μm.

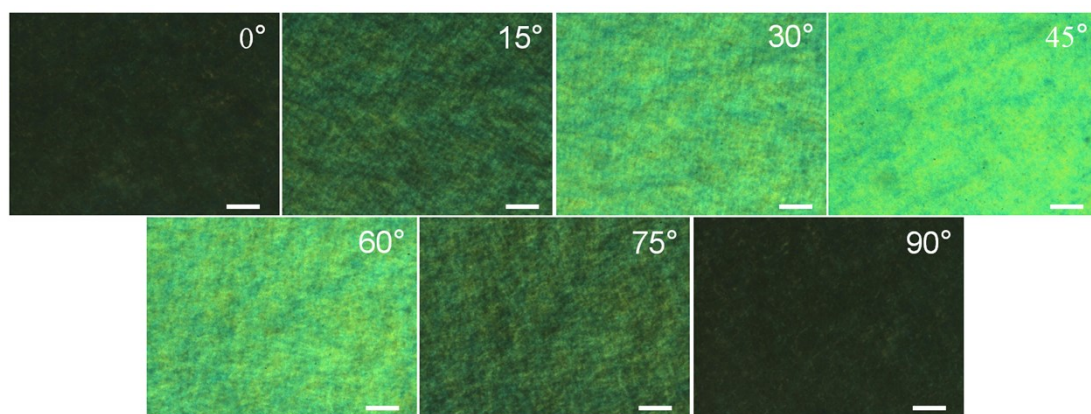


Figure S5. Transmission images of **OH-9** at different angle. The scale bar is 150 μm .

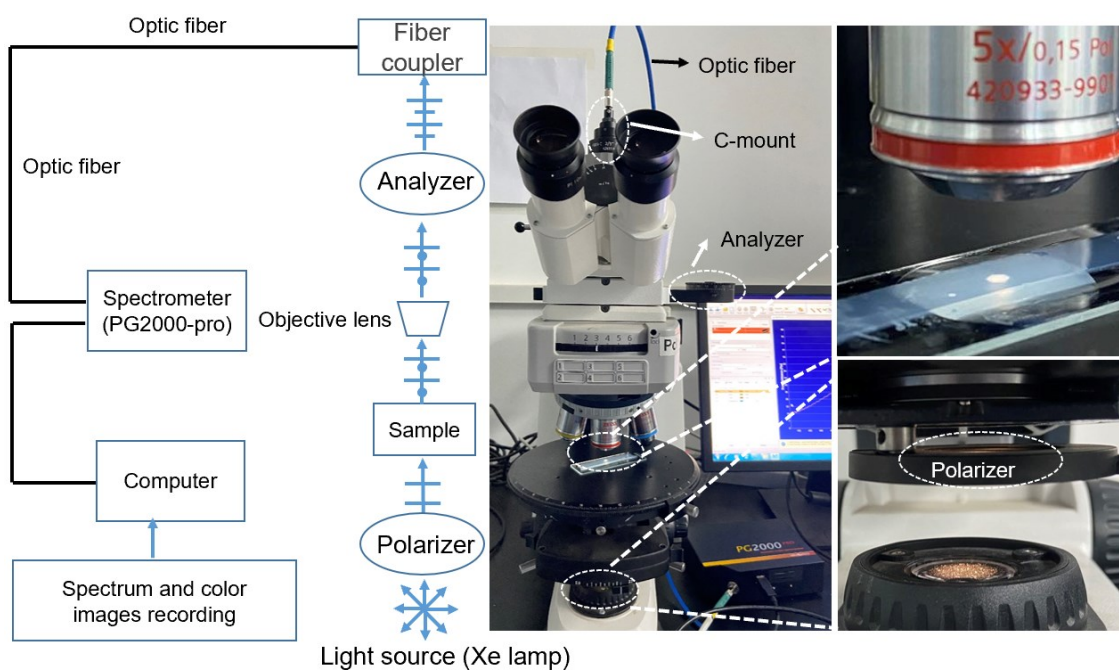


Figure S6. Schematic diagram of the optical device consisted of an optical fiber spectrometer and a polarizing optical microscope for recording spectra of TCNC nanocomposite hydrogels.

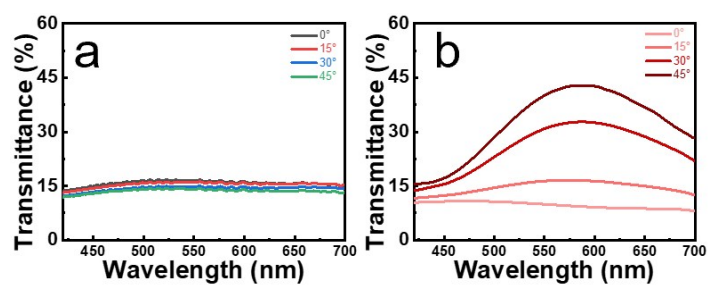


Figure S7. Transmission spectra of **RH** (a) and **OH-9** (b) measured by using the optical device (Figure S6) at different angles.

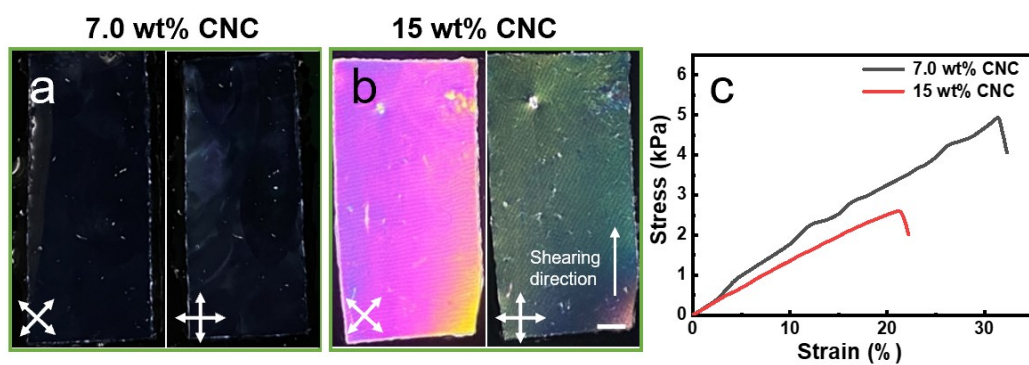


Figure S8. Photographs and tensile stress-strain curves (c) of plant-based CNC/PNIPAM hydrogel with 7 (a), 15 wt% (b) CNC. Scale bar = 2.5 mm.

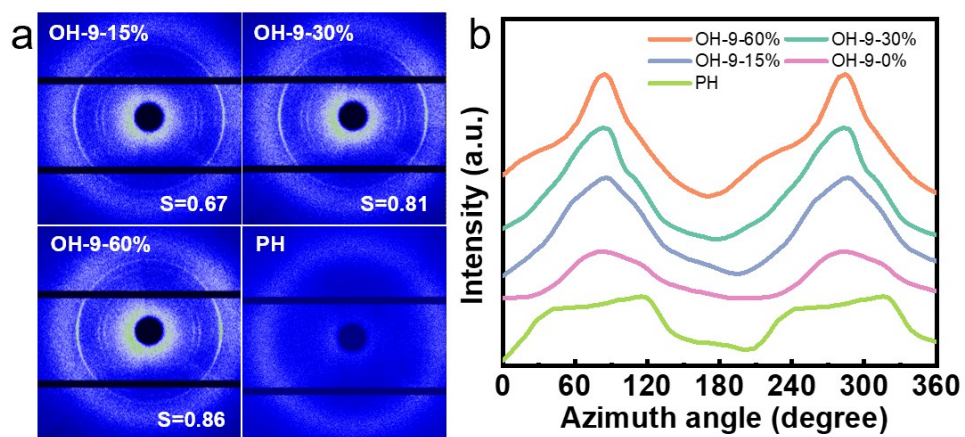


Figure S9. 2D WAXS patterns of **OH-9** with different strain (15, 30, and 60%) and **PH** (a); Azimuthal-integrated intensity distribution curves, where 0° represents the perpendicular direction (b).

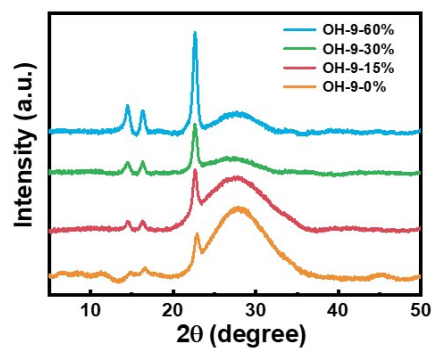


Figure S10. XRD patterns of **OH-9** at different strain.

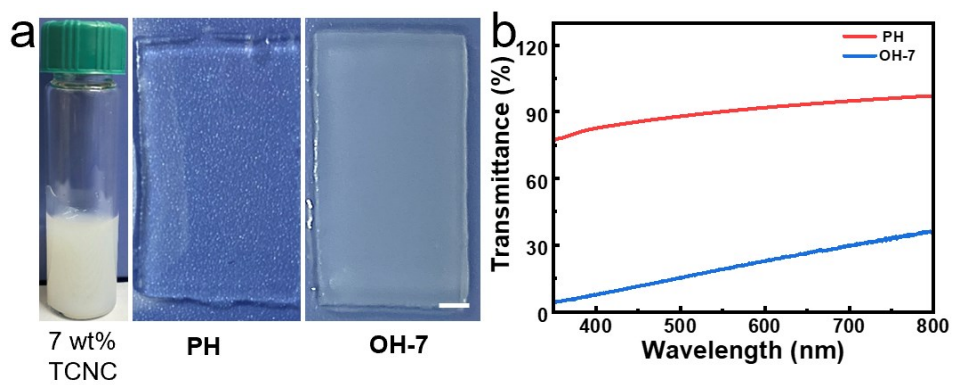


Figure S11. Photograph (a) and transmission spectra (b) of 7 wt% TCNC, **PH**, and **OH-7**.