

## Supplementary Information:

### Confined self-assembly of cellulose nanocrystals in a shrinking droplet

Fernando Jativa<sup>a,b</sup>, Christina Schütz<sup>c,d</sup>, Lennart Bergström<sup>c</sup>, Xuehua Zhang<sup>b</sup> and Bernd Wicklein<sup>\*d#</sup>

<sup>a</sup> Department of Mechanical Engineering, University of Melbourne, Parkville, VIC 3010, Australia.

<sup>b</sup> Department of Civil, Environmental & Chemical Engineering, RMIT University, Melbourne, VIC 3001, Australia.

<sup>c</sup> Department of Materials and Environmental Chemistry, Stockholm University, 106 91 Stockholm Sweden.

<sup>d</sup> Wallenberg Wood Science Center, KTH, Teknikringen 56, 100 44 Stockholm, Sweden.

# Present affiliation: Materials Science Institute of Madrid-CSIC, Spain.

\*corresponding author: Bernd Wicklein (bernd@icmm.csic.es)

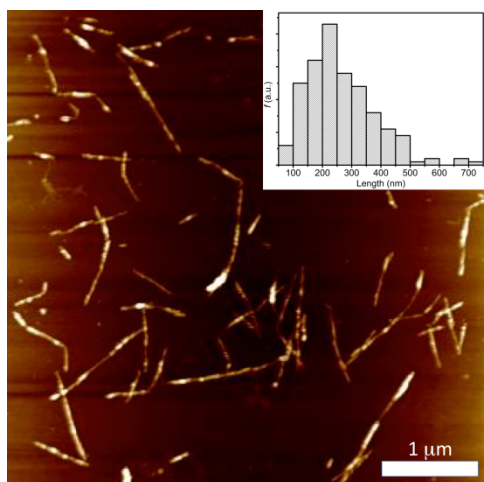


Fig. S1: AFM image obtained in air of CNCs deposited on a mica surface and corresponding histogram of the length distribution (inset).

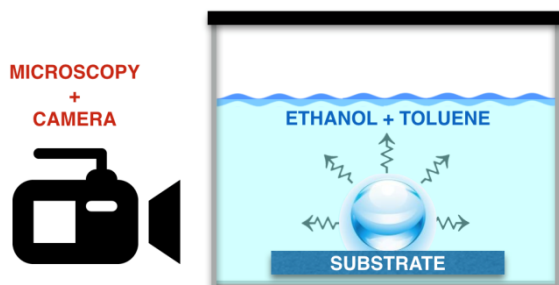


Fig. S2: Schematic of the setup used for assessing the water dissolution dynamics of aqueous CNC droplets in toluene/ethanol solutions. Side-view images of the shrinking droplet were recorded with a contact angle measurement equipment. The droplet was placed on a hydrophobized OTS-silicon substrate immersed in the binary liquid. The glass vial was sealed to prevent solvent evaporation.

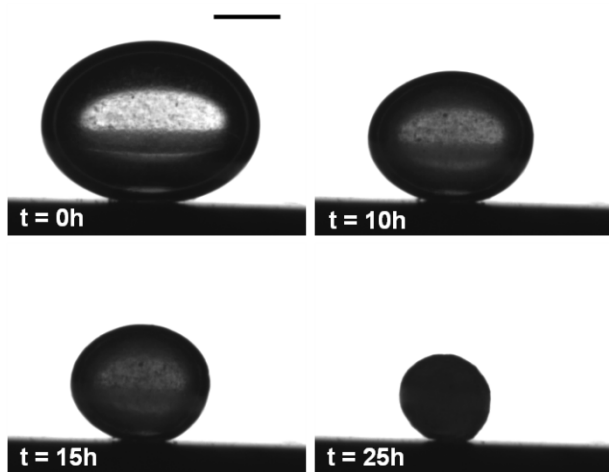


Fig. S3: Snapshots of a shrinking CNC droplet in toluene/0.5 vol% ethanol solution taken at different times. The initial droplet volume was 2  $\mu\text{L}$ . These exemplifying images show that the spherical shape of the droplet is maintained throughout the dissolution process owing to the hydrophobized substrate on which it resides. The scale bar is 600  $\mu\text{m}$ .

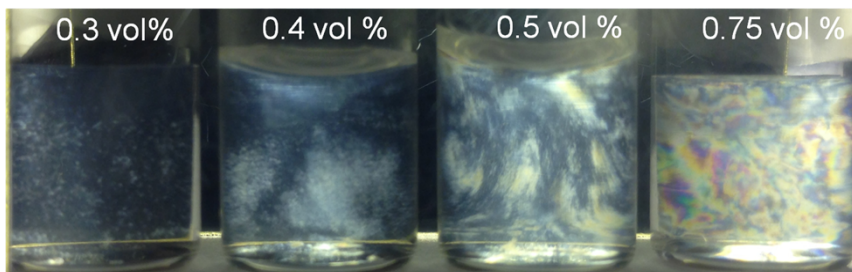


Fig. S4: Photograph showing suspensions of carboxylated CNC positioned between crossed polarizers. The image indicates the biphasic region between the isotropic and liquid crystalline phase with an onset concentration of 0.3 vol% and the 100 % liquid crystalline phase at 0.75 vol%. The image was taken 48 h after sample preparation and no phase separation occurred until then.

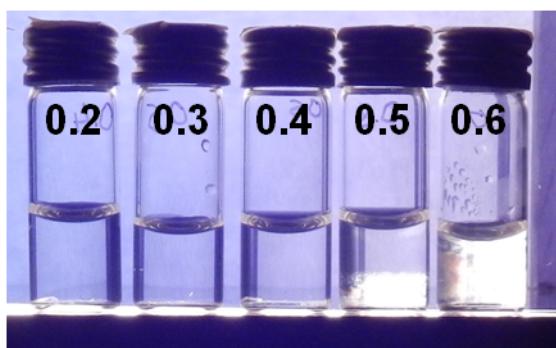


Fig. S5: Photograph showing suspensions of carboxylated CNC positioned between crossed polarizers. The image indicates a phase separation between a chiral nematic and an isotropic phase with an onset concentration of 0.3 vol% and the 100 % chiral nematic phase at 0.6 vol%. The image was taken 6 months after sample preparation. The numbers in the photograph indicate the vol% of CNC. The chiral nematic order was confirmed by laser diffraction.