Supplementary Information

Assembly of cellulose nanocrystals in a levitating drop probed by time-resolved small angle X-ray scattering

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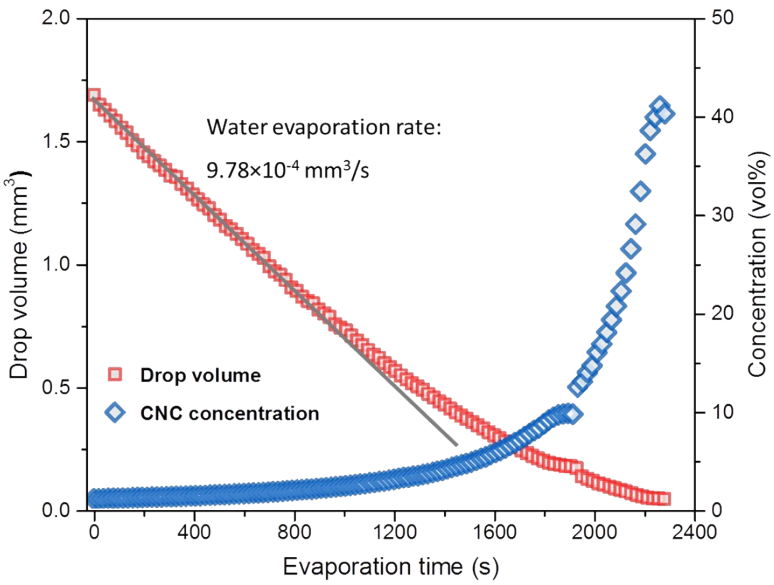

Fig. S1 Change of the CNC drop volume and concentration as a function of evaporation time. The grey line denotes a constant water evaporation rate (9.78×10^{-4} mm^3/s) of the drop until 1000 s.

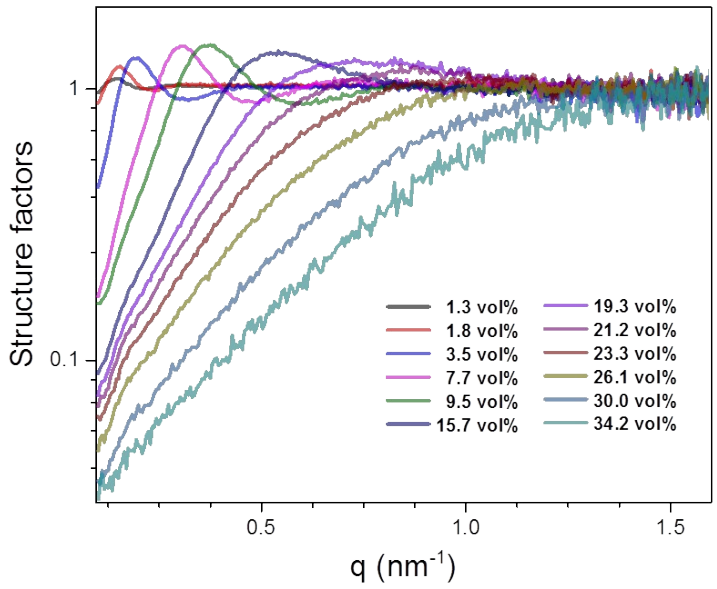

Fig. S2 Change of the structure factor as a function of the CNC concentration.

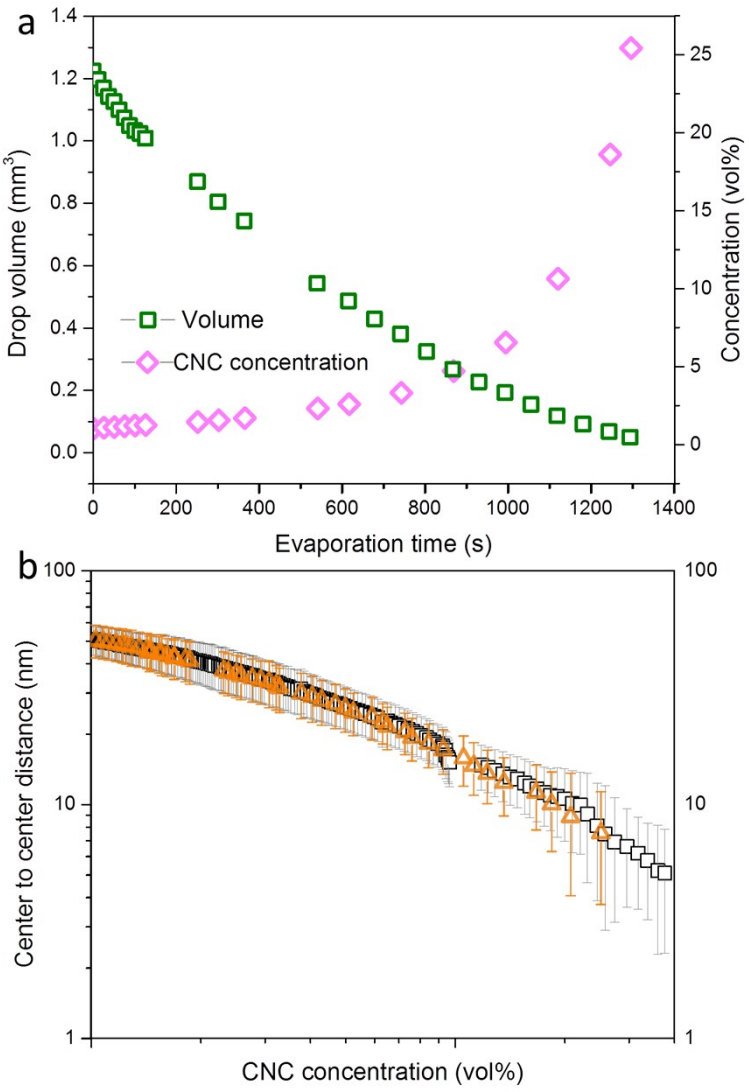

Fig. S3 Additional time-resolved SAXS study on a levitating drop of CNC aqueous dispersion. (a) Change of the CNC drop volume and concentration as a function of the evaporation time. (b) Center-to-center separation distance between CNC particles with an increasing CNC concentration. Grey squares refer to the data of Fig. 2 in manuscript.

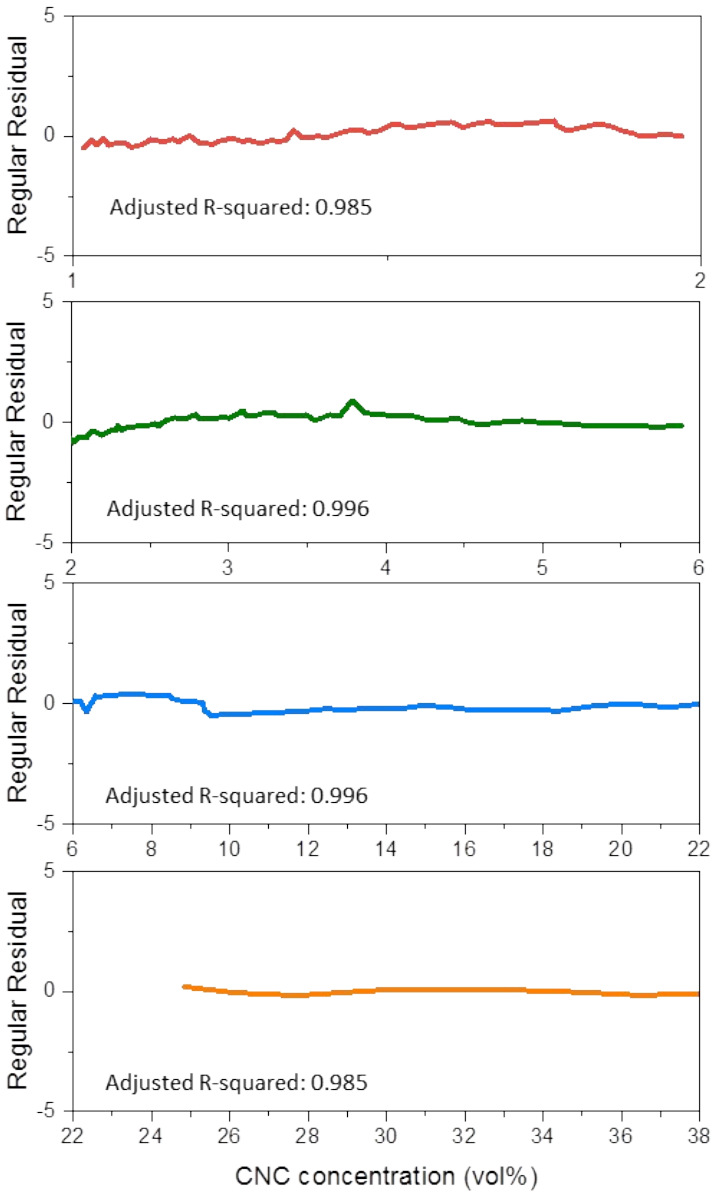

Fig. S4 The normalized residual for the power law fitting of CNC separation distance with concentrations.

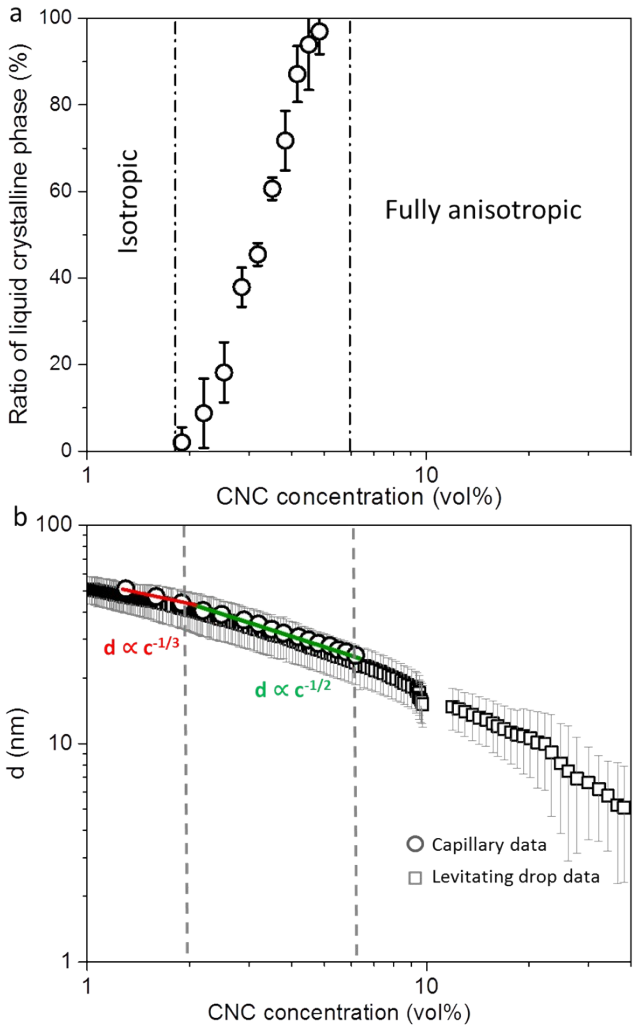

Fig. S5 Structural evolution of CNC dispersions with an increasing particle concentration. (a) Equilibrium phase diagram of CNC dispersions in capillaries.[1] (b) Change of separation distance between CNC particles in levitating drops and capillaries. The solid curves describe a power law relation ($d \propto c^{-x}$) with exponents of 1/3 (red) and 1/2 (green), respectively, for the data measured in capillaries.

Fig. S6 Photograph of CNC dispersion at a concentration of 1 vol% between crossed polarizers.

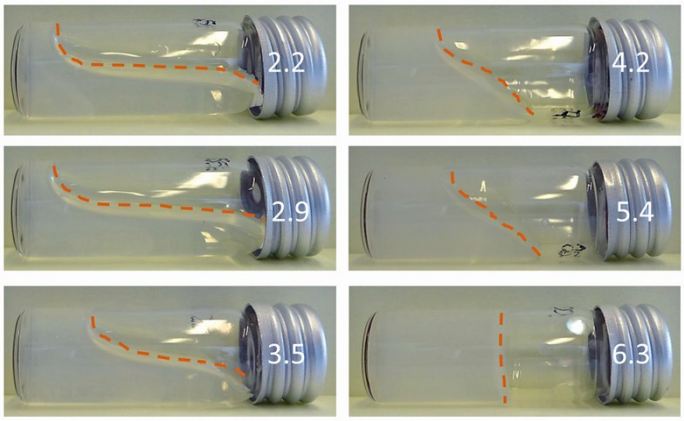

Fig. S7 Digital photographs of 90° tilted vials of CNC dispersions with different concentrations (vol%). The dispersions were allowed to equilibrate in non-tilted position overnight prior to the measurements. The yellow dash curves indicate the liquid-air interface.

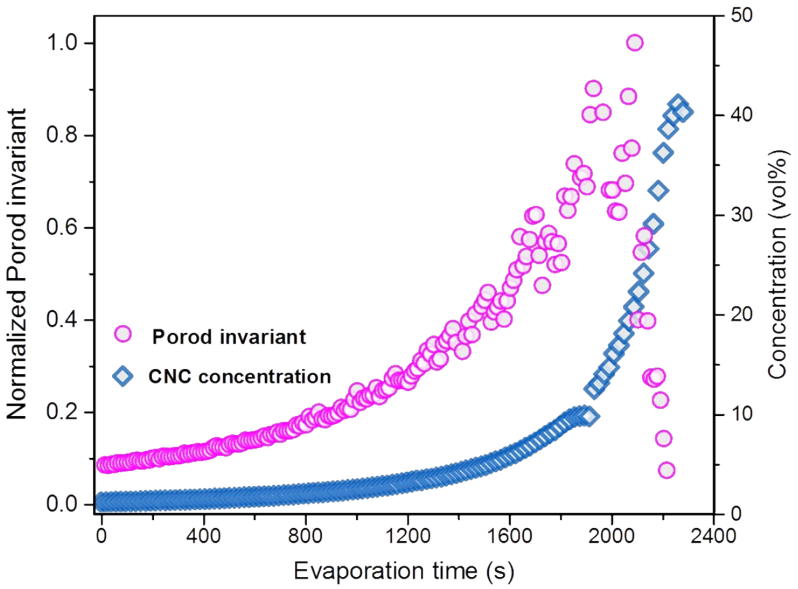

Fig. S8 Change of the normalized Porod invariant and CNC concentration as a function of evaporation time.

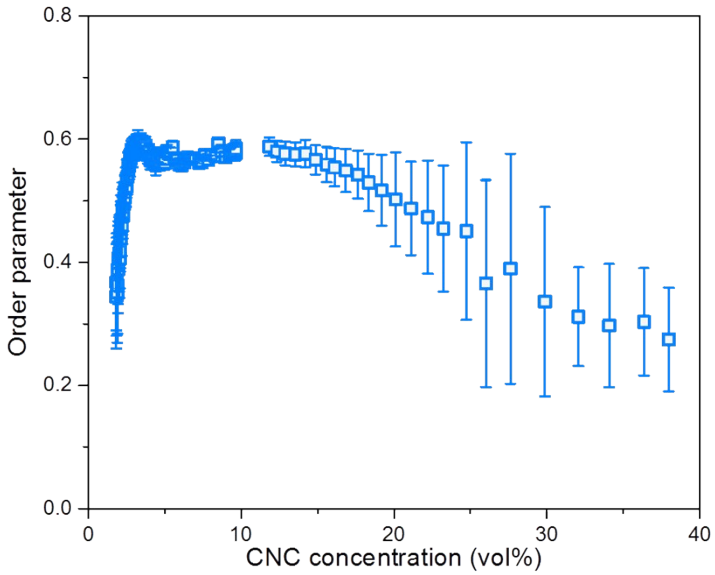

Fig. S9 Change of the order parameter as a function of the CNC concentration.

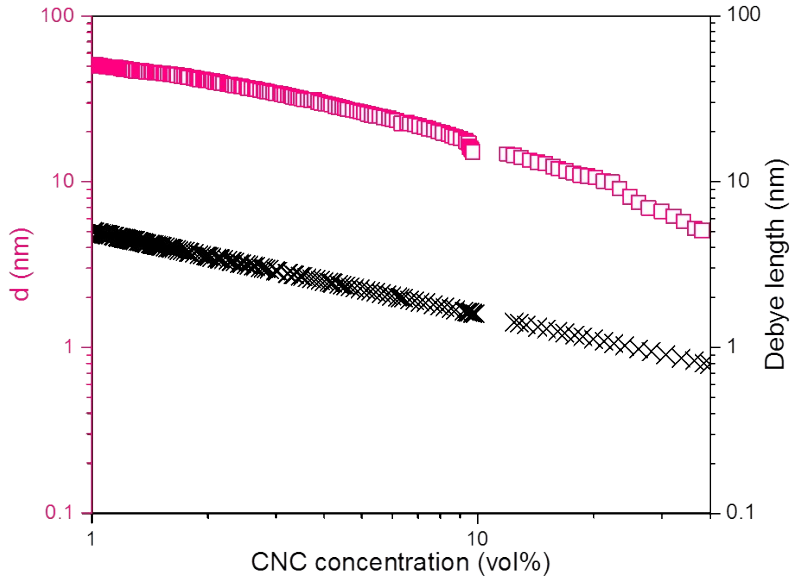

Fig. S10 Change of separation distance (measured by SAXS) and Debye length (calculated) of the negatively charged CNC in the levitating drop as a function of particle concentration.

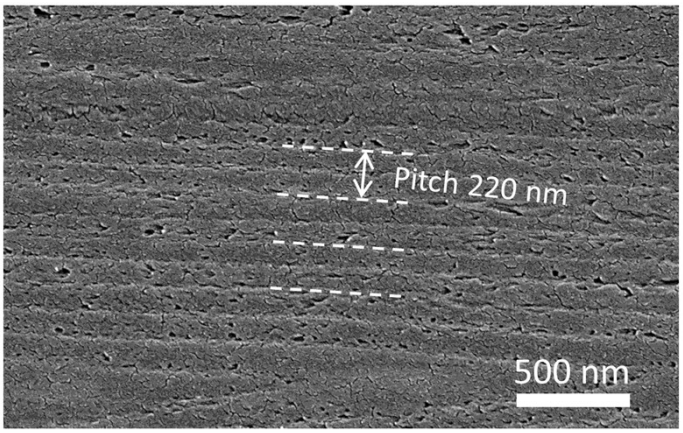

Fig. S11 The SEM image of the cross-sectional surface of a CNC film that has been drop cast onto a substrate using the 1 vol% dispersion.