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ADVANCED MATERIALS

Supporting Information

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Flexible Photonic Cellulose Nanocrystal Films

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Electronic Supporting Information

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Supplementary Figures





(b)





Figure S1. (a) ¹H-NMR of Na-CNC (curve I) and CNC-DMAPS suspensions at mass ratio 1:0.41 prepared using H-CNC (pH~3) (curve II) and Na-CNC (pH~7) (curve III). The fourth curve (IV) represents CNC-DMAPS suspension at mass ratio 1:1 using Na-CNC (pH~7), while curve (V) represents the DMAPS solution. (*) peaks are assigned to CNC protons. An increased scale for curve (IV) is given in (b). It is evident from the above that the CNC-DMAPS complex is present in the solutions at varying mass ratios and pH values, which confirms successful adsorption of the zwitterionic surfactant onto the CNC surfaces.



Figure S2. PXRD patterns of evaporation-induced self-assembled (EISA) CNC-DMAPS films shows well defined peaks assigned to the (002) plane at $2\theta \approx 22.7^{\circ}$ and the (101) and (10 \overline{I}) planes at $2\theta \approx 14-17^{\circ}$, as well as a contribution from the (040) plane at $2\theta \approx 34.3^{\circ}$. Shown are patterns for CNC:DMAPS mass ratio 1:0.41 (blue) and 1:1 (red) vs. neat CNC (black) confirming CNC structural integrity within complex. Neat CNC intensity curve is scaled.



Figure S3. Pictures depicting dispersion of DMAPS-CNC complexes in various solvents, with their corresponding polarity index. DMAPS-CNC complexes, like pure CNC, are dispersible in polar protic solvents, like water, and some polar aprotic solvents, like N,N-dimethylformamide (DMF) and pyridine, but not in non-polar solvents, like toluene.



Figure S4. (a) Representative image of the CNCs (b) Length and height distributions of the CNCs. The average length and height are respectively 100 nm and 3.8 nm, for a total number of measured rods equal to 2331 for the length and 2303 for the height.





Figure S5. Bright and dark field micrographs, acquired for the left-, LCP, and for the right circular polarisation, RCP for the Na-CNC films with increasing amount of DMAPS. Scale bars are $100 \,\mu$ m.





Figure S6. SEM images depicting the cross section of (a) pure CNC and DMAPS:CNC films prepared at (b) 1:0.1, (c) 1:0.41 and (d) 1:1 CNC:DMAPS mass ratios. No CNC/DMAPS segregation is visible confirming the excellent homogeneity of the fabricated films, whereas irregularities are due to film breaking-by-hand. Scale bars are $1\mu m$.