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Nanoscale Structural Rearrangements in Ultrathin Nanocellulose Films induced by Water

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Cellulose nanofibrils (CNF) as a sustainable biomaterial are excellent building blocks for mechanically exceptional materials and functional coatings. Yet, the water uptake and response to humidity still poses a challenge. We first demonstrate a facile route to prepare large-scale cellulose-based nanostructured thin films with a low surface roughness down to 2.5 nm on (20 × 100) mm^2 substrates. We employ in situ grazing incidence small-angle neutron scattering to study the morphological features within the ultra-smooth CNF thin films under as-prepared conditions as well as their rearrangement under humidification. Increasing CNF surface charge is highly beneficial for the layering mechanism as it directly influences the self-assembly process, which results in a low roughness of the densely packed CNF network. We observe distinct domains of smaller cellulose bundles and larger bundles or agglomerates within the thin film. During in situ humidification and drying of the CNF film, the domains reversibly change from cylindrical to spherical appearance. With decreasing values of surface roughness corresponding to increasing surface charge densities of CNF films, the surface free energy is observed to be tunable. This knowledge can be used to promote the use of polar solvents in applications such as organic solar cells and to further enhance physical properties and materials lifetime.

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