

Supporting Information

Outstanding Synergies in Mechanical Properties of Bioinspired Cellulose Nanofibril Nanocomposites using Self-Crosslinking Polyurethanes

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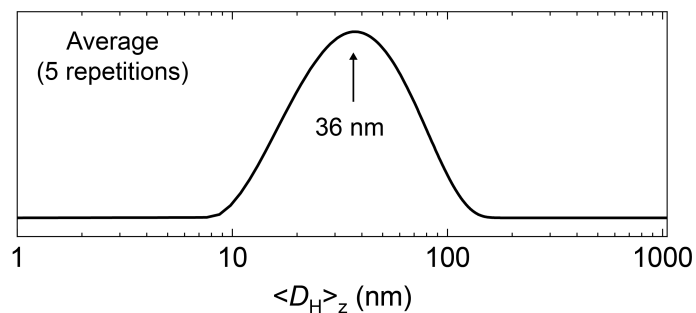


Figure S1. Average size of PU particles measured by dynamic light scattering (DLS)

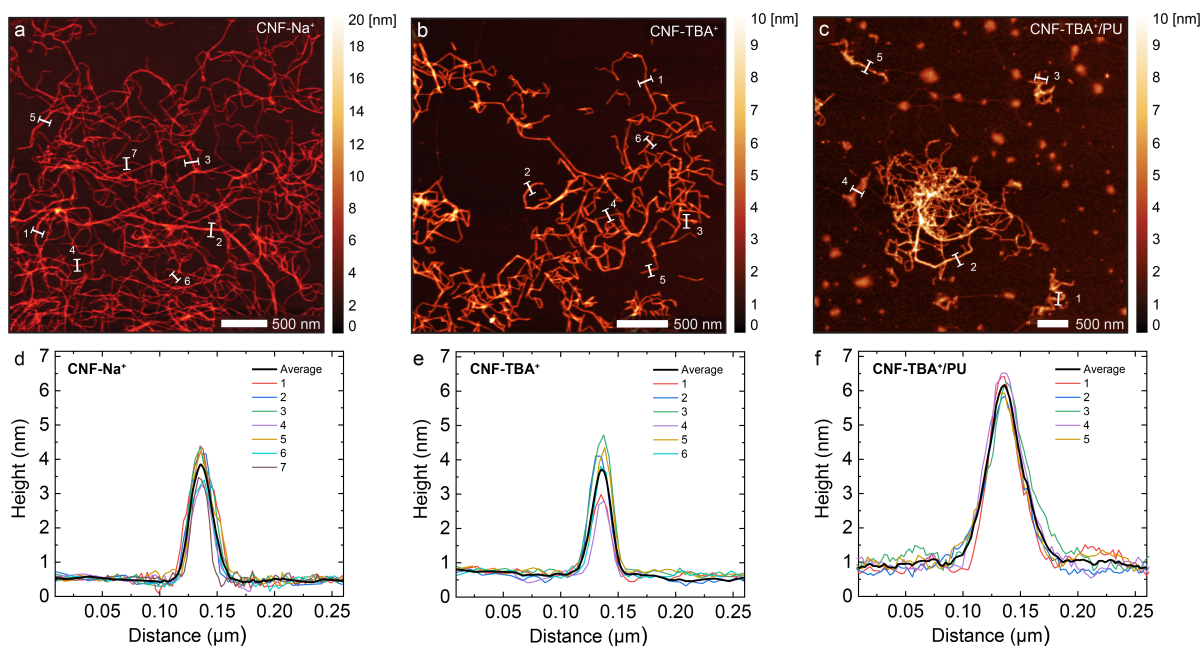


Figure S2. Diameter characterization by AFM (a-c) height images and (d-f) height profiles. The height profiles were obtained by selecting at least 5 random fibrils in the height images for (a,d) CNF-Na⁺, (b,e) CNF-TBA⁺ and (c,f) CNF-TBA⁺/PU. The latter shows the coating effect of the PU over the CNF-TBA⁺.

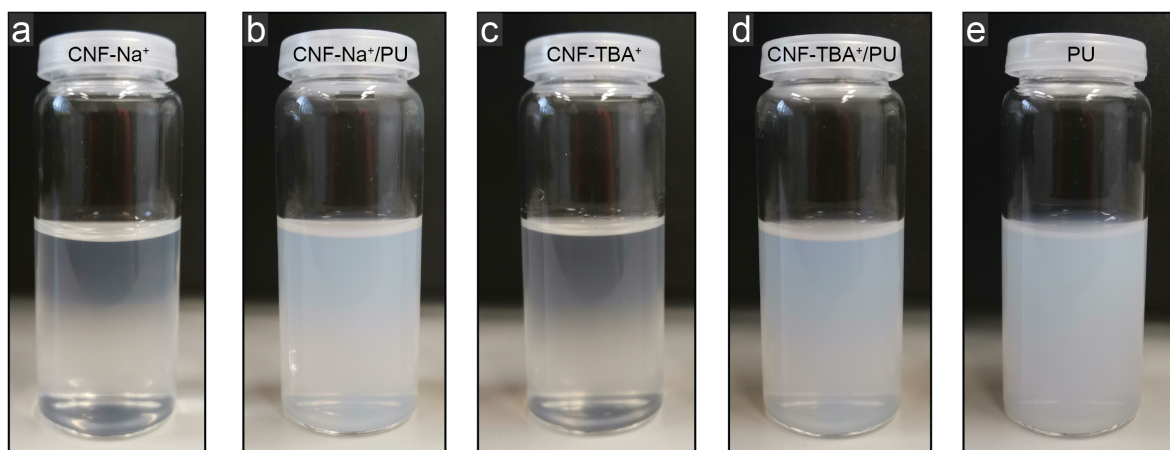


Figure S3. Photographs of dispersions (0.25 wt%) without aggregation and phase separation for (a) CNF-Na⁺, (b) CNF-Na⁺₅₀/PU₅₀, (c) CNF-TBA⁺, (d) CNF-TBA⁺₅₀/PU₅₀ and (e) PU.

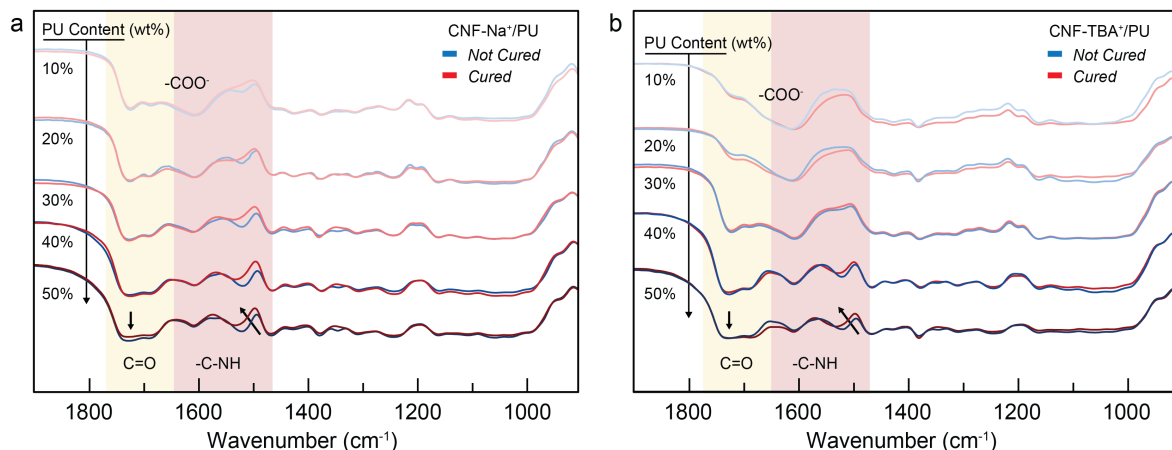


Figure S4. FTIR spectra of not cured (blue) and cured (red) nanocomposites for **(a)** CNF-Na⁺/PU and **(b)** CNF-TBA⁺/PU with different PU content (from 10 to 50 wt%). The black small arrows show the curing effect of the PU in the nanocomposite, explained in the main text. Increasing bands for CNF can be observed for lowering the PU content, see main text for explanations.

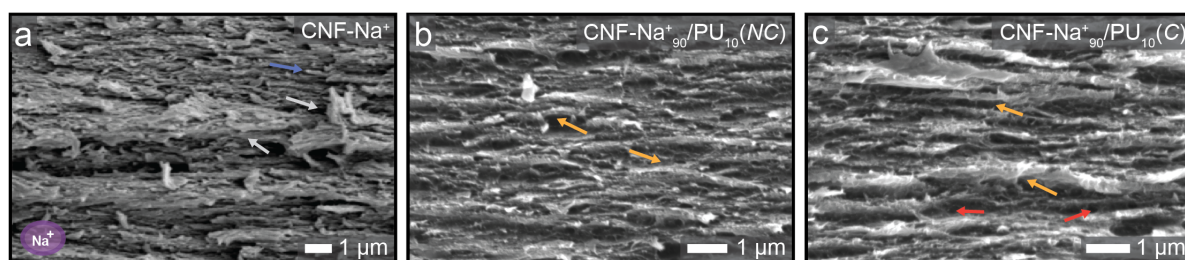


Figure S5. SEM of the cross section of fracture samples to illustrate their deformation mechanisms. The transition of their behavior is shown for **(a)** pure CNF-Na⁺, **(b)** CNF-Na⁺₉₀/PU₁₀ (NC) and **(c)** CNF-Na⁺₉₀/PU₁₀ (C). The grey arrows show bundles and agglomerates of fibrils, the blue arrows show individual fibrils, the orange arrows show pull-out of mesoscale layers and the red arrows show a polymeric phase holding two layers.

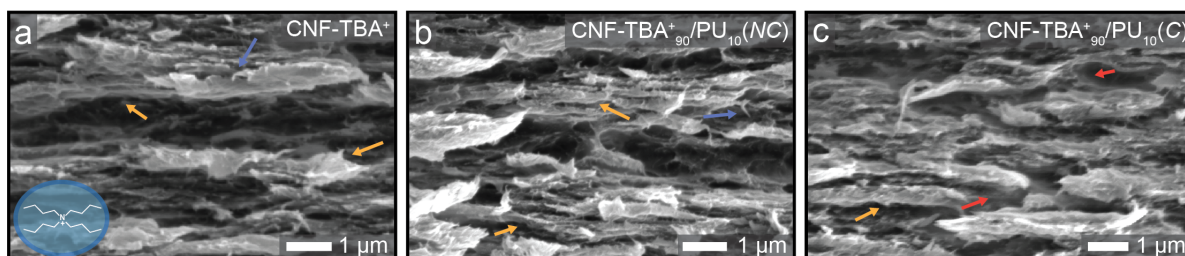


Figure S6. SEM of the cross sections of fracture samples to illustrate their deformation mechanisms. The transition of their behavior is shown for **(a)** pure CNF-TBA⁺, **(b)** CNF-TBA⁺₉₀/PU₁₀ (NC) and **(c)** CNF-TBA⁺₉₀/PU₁₀ (C). The blue arrows show individual fibrils, the orange arrows show pull-out of mesoscale layers and the red arrows show a polymeric phase holding two layers.