Electronic Supplementary Material (ESI) for Soft Matter.
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Supplementary Information

Fabrication of highly stiff yet elastic microcapsules incorporating cellulose nanofibrils

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Materials

Polyacrylic acid 250k, butyl acrylate, and 2¬(methacryloyloxy)ethyl]trimethylammonium chloride were obtained from Sigma Aldrich. The cellulose nanofiber (CNF) were obtained in a sodium carboxylate functionalized form at 0.96 wt%. in water from USDA Forest Products Laboratory (Madison, Wisconsin). The fibrils have a diameter of ≈ 5 nm and an aspect ratio of ≈ 100, with a sodium carboxylate density of ≈ 0.65 mmol g^{-1}

Pressure Driven Capsule Transit experiments

The recoverability of CNF/MADQUAT-co-BTA microcapsules was characterized based on pressure driven flow of microcapsules through a tapered capillary. The experimental setup, consists of a capillary tapered to 162 um, tubing, mineral oil bath, and ultra-low pressure regulator (equilibar LPR2). The capillary is connected to the pressure regulator using soft tubing from the non-tapered side and inserted into a mineral oil bath. The capsules are loaded into the capillary using a micropipette and flow towards the tapered part of the capillary by applying a pressure in the range of 0 to 2000 Pa. The pressure was increased quasi-statically from 0-2000 Pa using 7 steps with 30 seconds equilibration time between each step.

Zeta potential measurements

The surface charge of the cellulose nanofiber was characterized from zeta potential measurements, which were obtained by electrophoretic light scattering using a Brookhaven NanoBrook Omni (Brookhaven Instruments Corporation), operating at a wavelength λ=658 nm. Five measurements per sample were collected for 0.16 wt% aqueous solution of CNF. Zeta potential values were calculated using the Smoluchowski equation and determined to be -72.38

A small aliquot of aqueous CNF solution was deposited on a carbon-coated copper grid (200 mesh, Ted Pella). The excess solution was wicked off with filter paper and the resulting sample was then air dried. TEM images were obtained using an FEI Tecnai Osiris TEM with an accelerating voltage of 200 kV.

Figures

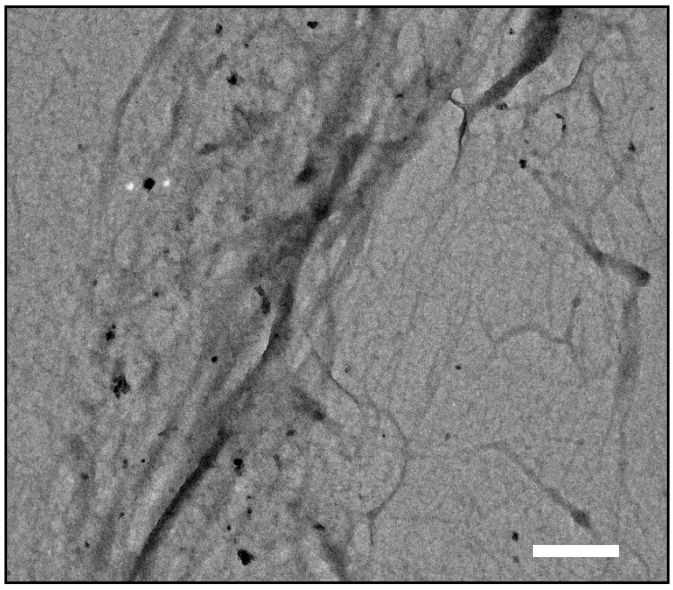

Figure S1. TEM micrograph showing collections of CNF. Scale bar is 500 nm.

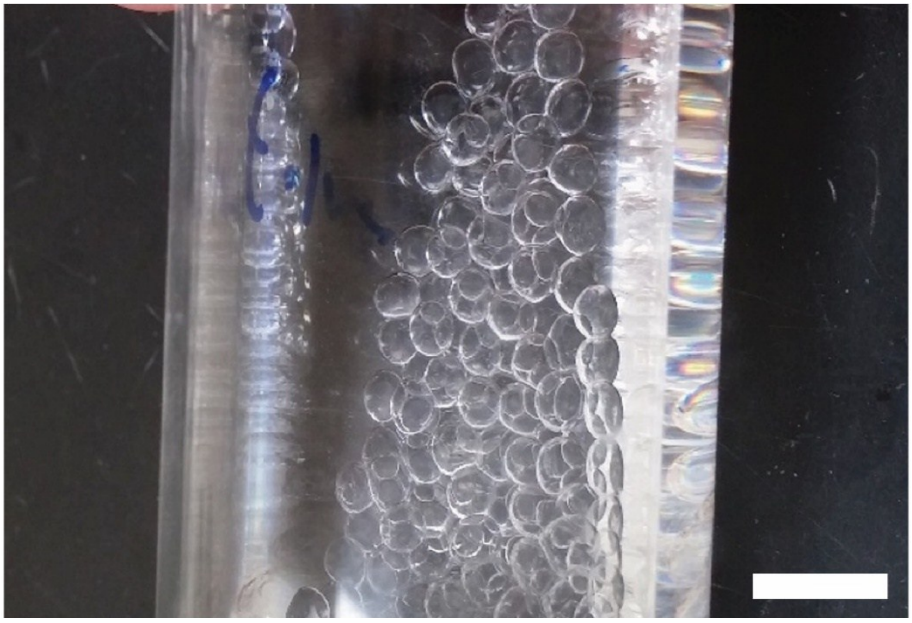

Figure S2. (a) Stable millimeter size CNF composite microcapsules suspended in toluene. The capsules are prepared by dripping an aqueous solution of 0.16 wt% CNF into a toluene solution containing 0.3 wt% MADQUAT-co-BTA. Scale bar is 10 mm.

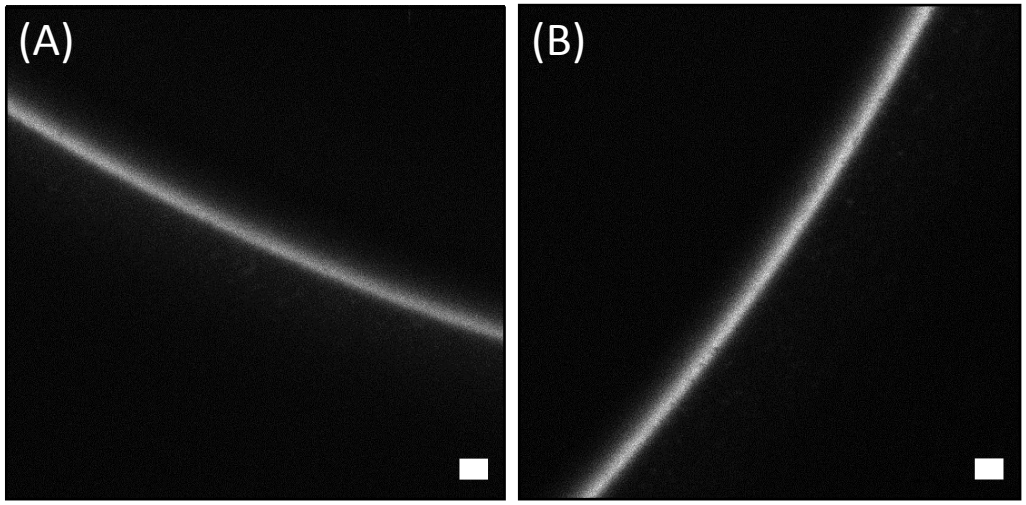

Figure S3. Confocal microscopy images of fluorescently labelled CNF (0.16 wt.%.)/MADQUAT-co-BTA (0.3 wt.%) capsules showing core shell structure prepared with (A) 0.018 wt% PAA with 10.65 µm thick shell. µm. (A) 0.0065 wt% PAA with 11.48 µm thick shell. Scale bar is 18

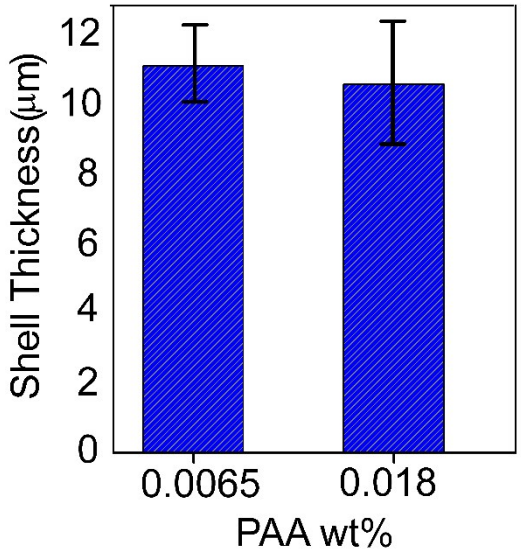

Figure S4. (a) Capsule shell thickness as a function of PAA concentration for 3.1 mm capsules made with. 0.16 wt% CNF and 0.3 wt% MADQUAT-co-BTA. Error bars correspond to standard deviations from a minimum of 9 measurements.

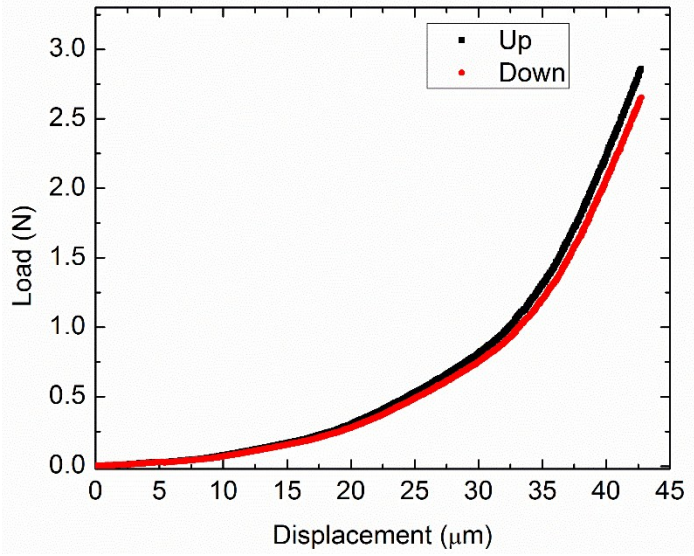

Figure S5. A typical load-deformation curve for CNF/PAA/MADQUAT-co-BTA capsules up to 15% strain showing the reversibility in the loading (reds) and unloading (black) segments.