## **Supporting Information**

## High internal phase oil-in-water Pickering emulsions stabilized by chitin nanofibrils: 3D structuring and solid foams

Ya Zhu,<sup>1,†</sup> Siqi Huan,<sup>1,3,†</sup> Long Bai,<sup>1,3,\*</sup> Annika Ketola,<sup>2</sup> Xuetong Shi,<sup>1</sup> Xiao Zhang,<sup>1</sup> Jukka A. Ketoja,<sup>2</sup> Orlando J. Rojas<sup>1,3,\*</sup>

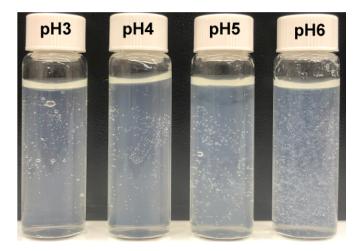
<sup>1</sup> Bio-Based Colloids and Materials, Department of Bioproducts and Biosystems, Aalto University, P.O. Box 16300, FIN-00076 Aalto, Espoo, Finland

<sup>2</sup> VTT Technical Research Centre of Finland Ltd, P.O. Box 1603, FI-40101 Jyväskylä, Finland

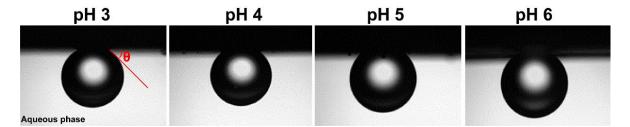
<sup>3</sup> Departments of Chemical & Biological Engineering, Chemistry, and Wood Science, 2360 East Mall, The University of British Columbia, Vancouver, BC V6T 1Z3, Canada

\* E-mails for correspondence: (OR) orlando.rojas@ubc.ca, +1-604-822-3457 and (LB) long.bai@ubc.ca Tel: +1-236-869-0416

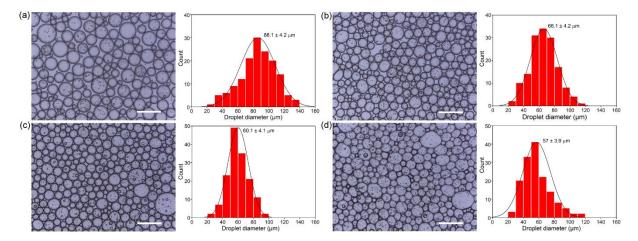
This Supporting Information document contains nine (9) figures in six (6) pages



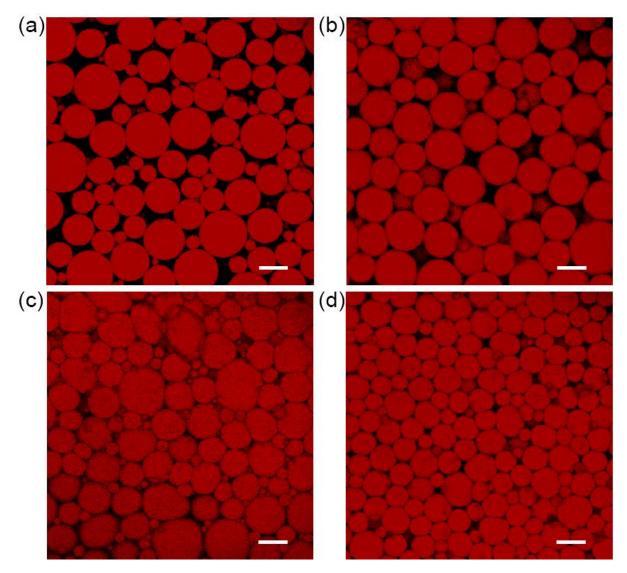
**Figure S1.** Visual appearance of chitin nanofibrils (NCh) suspension at pH value of 3 to 6. Prior to photographing, the samples were sonicated for 3 min in water bath after titrating to designed pH value.



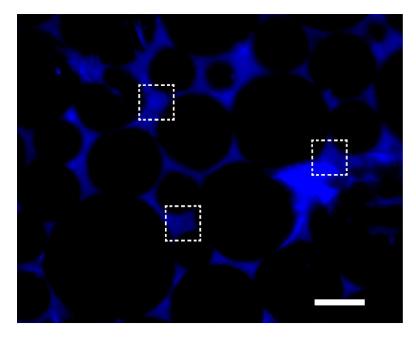
**Figure S2.** Three-phase contact angle (CA) of a sunflower oil droplet on a solid film composed of NCh in Milli-Q water. The NCh films were spin-coated from NCh suspension at pH value of 3 to 6. The red lines in the image indicated the CAs ( $\theta$ ).



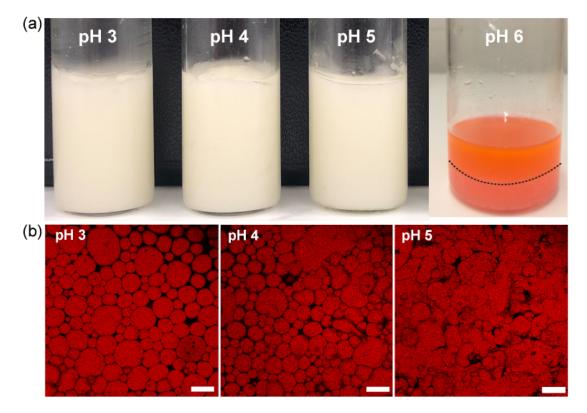
**Figure S3.** Optical images and droplet diameter of HIPPEs stabilized by NCh (pH 3) at oil volume fraction of (a) 66, (b) 74, (c) 80, and (d) 88%. The concentration of NCh in continuous phase is 0.5 wt%. The diameter for each sample was indicated in each histogram. The scale bar is  $200 \,\mu\text{m}$ .



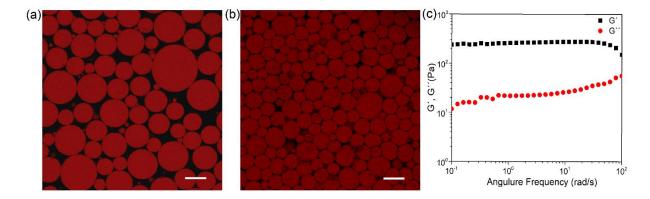
**Figure S4.** Confocal images of HIPPE stabilized by NCh (pH 3) at sunflower oil volume fraction of (a) 66, (b) 74, (c) 80, and (d) 88%. The concentration of NCh in continuous phase is 0.5 wt%. The HIPPEs were stored at room temperature for 24 h. The oil phase was dyed by Nile red during HIPPE preparation. The pH of NCh suspension was 3 for sample preparation. The scale bar is 80 µm.



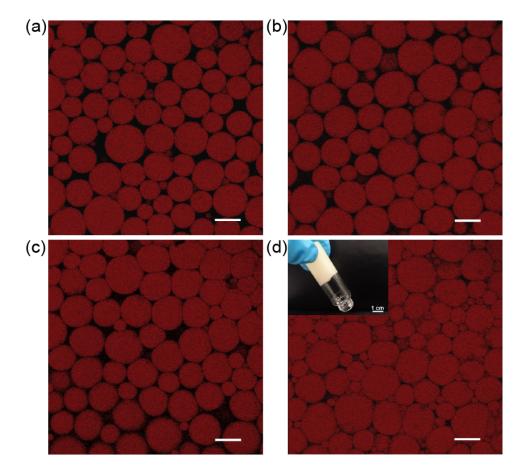
**Figure S5.** Fluorescence micrograph of HIPPE at 88% oil volume fraction (0.064 wt% NCh and pH 3). NCh was stained by Calcofluor white prior to observation. The dashed boxes in the image indicate the contour of the oil droplets. The scale bar is  $50 \mu m$ .



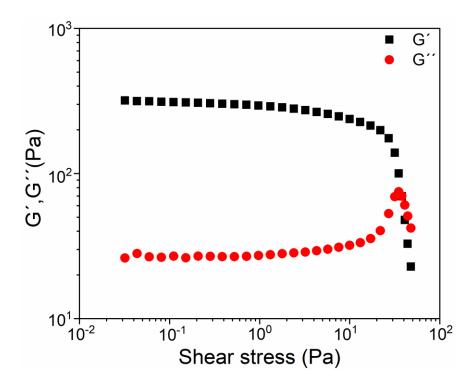
**Figure S6.** (a) Visual appearance and (b) confocal images of HIPPEs stabilized by NCh with pH value of 3 to 6. The oil volume fraction for all samples were 88%. The concentration of NCh in continuous phase is 0.5 wt%. Oil was stained by Nile red for pH 6 sample to better visualize the phase separation, and the black dashed line indicates the separation. The confocal was measured after storing HIPPEs at room temperature for 24 h. The scale bar is 80 µm.



**Figure S7.** Confocal images of HIPPE stabilized by long-fibril NCh (pH 3) at oil volume fraction of (a) 66% and (b) 88%. The concentration of NCh in continuous phase is 0.3 wt%. The HIPPE was stored at room temperature for 24 h. The oil phase was dyed by Nile red during HIPPE preparation. The scale bar is 80 µm. (c) Moduli of HIPPE in (b).



**Figure S8.** Confocal images of HIPPE stabilized by NCh (pH 3) at oil volume fraction of (a) 66%, (b) 74%, (c) 80%, and (d) 88%. The concentration of NCh in continuous phase is 0.5 wt%. The HIPPE was stored at room temperature for 3 months. The insert in (d) shows the inverted vial for HIPPE containing 88% oil after 3 months. The oil phase was dyed by Nile red during HIPPE preparation. The scale bar is 80 µm.



**Figure S9.** Oscillatory rheology of HIPPE at 88% oil fraction volume (0.064 wt% NCh and pH 3). Shear yield stress is indicated by the intersection of storage and loss moduli.