Supporting Information for

Hydrophobized Lignin Nanoparticle-Stabilized Pickering Foams: Building Blocks for Sustainable Lightweight Porous Materials

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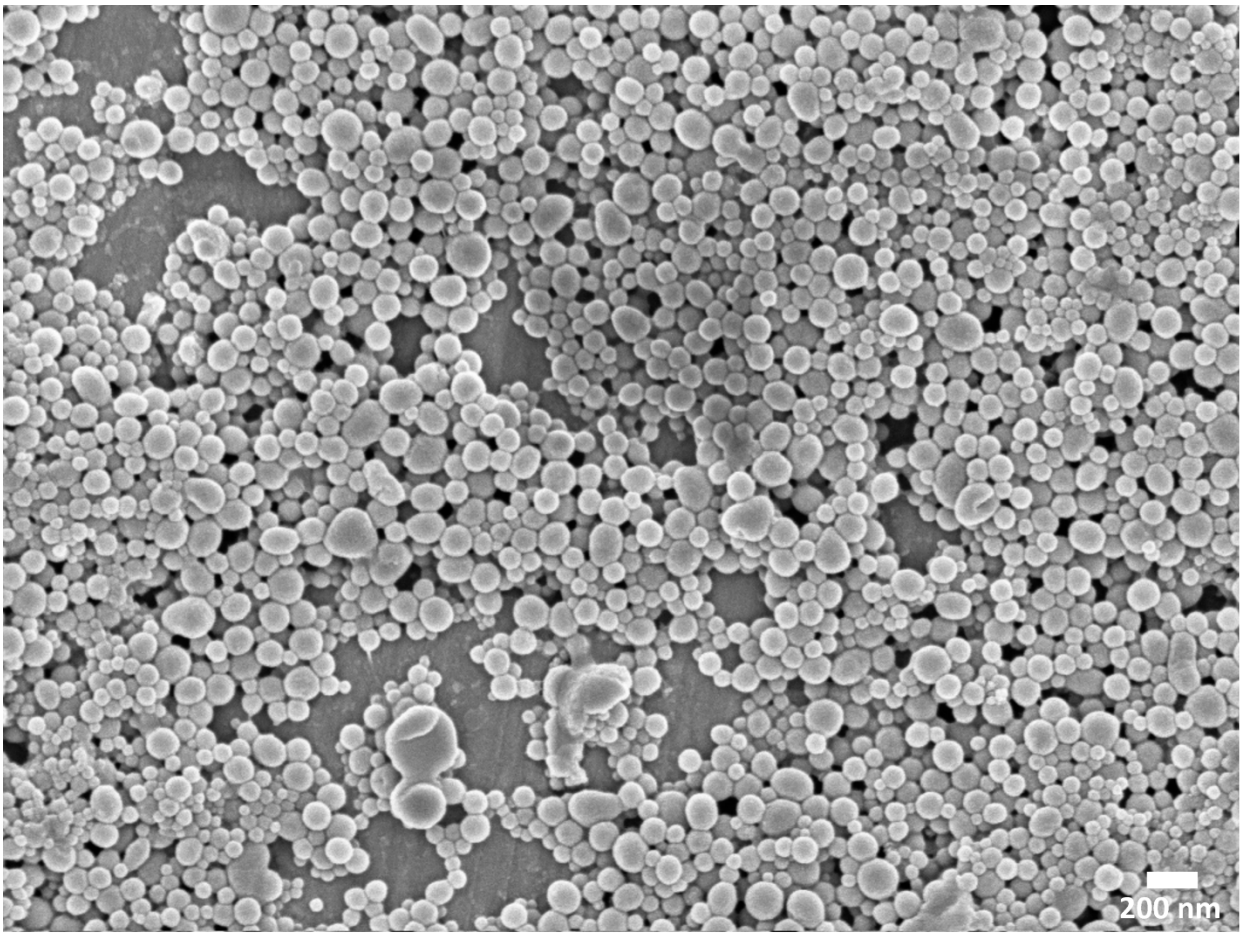

Figure S1. The morphology of LNPs captured by SEM microscopy.

Table S1. Foam formulations at the CTAB to LNP coating ratio from 0 to 15 mg/g. The initial concentrations of LNP aqueous dispersion and CTAB aqueous solution were 5 wt% and 1 wt%, respectively.

CTAB to LNP ratio (mg/g)	**0.0**	**2.5**	**5.0**	**7.5**	**10.0**	**12.0**	**15.0**
LNPs aqueous dispersion (ml)	5.0	5.0	5.0	5.0	5.0	5.0	5.0
CTAB aqueous solution (ml)	0.0	0.1	0.1	0.2	0.3	0.3	0.4
Total solid content (wt%)	5.0	5.0	4.9	4.9	4.8	4.8	4.7

Table S2. Foam formulations with varied CNF fractions. The initial concentrations of LNP aqueous dispersion, CTAB aqueous solution, and CNF aqueous dispersion were 6.5 wt%, 1 wt%, and 2.1 wt%, respectively.

CNF fractions relative to the total solid mass (wt%)	**30**	**40**	**50**
LNPs aqueou dispersion (ml)	8.9	7.6	6.3
CTAB aqueous solution (ml)	0.3	0.3	0.2
CNF aqueous dispersion (g)	11.7	15.7	19.5
Water (ml)	14.1	17.3	21.5
Total solid content (wt %)	2.4	2.0	1.7

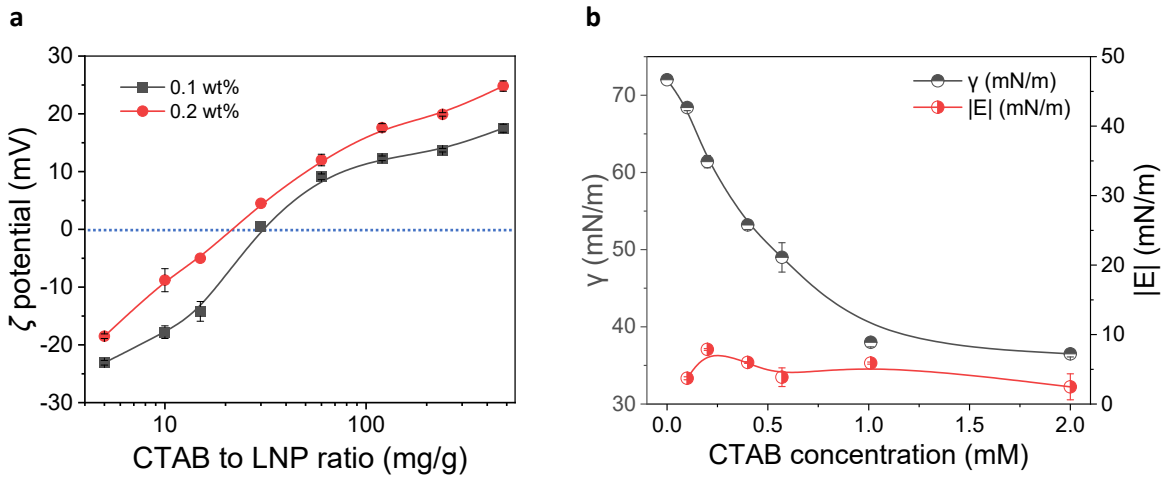

Figure S2. (a) ζ potential of CTAB-LNPs at varied CTAB to LNP coating ratios, measured at the particle concentrations of 0.1 and 0.2 wt%. It is worth mentioning that the ζ potentials of the CTAB-LNPs measured at concentrations above 0.2 wt% were not reliable anymore. The dashed line denotes the ζ potential at 0, and the intersections with the black and red lines are the isoelectric points (IEP) for 0.1 wt% and 0.2 wt%, respectively. Particle concentration at 0.2 wt% shows an IEP at a lower CTAB to LNP coating ratio than for 0.1 wt%, due to less available dissociated carboxyl groups per particle as explained in the main article. (b) Surface tension (γ) and complex viscoelastic modulus ($|E|$) of CTAB aqueous solution plotted against its concentration. The critical micelle concentration is about 1 mM, which is consistent with the literature.[1]

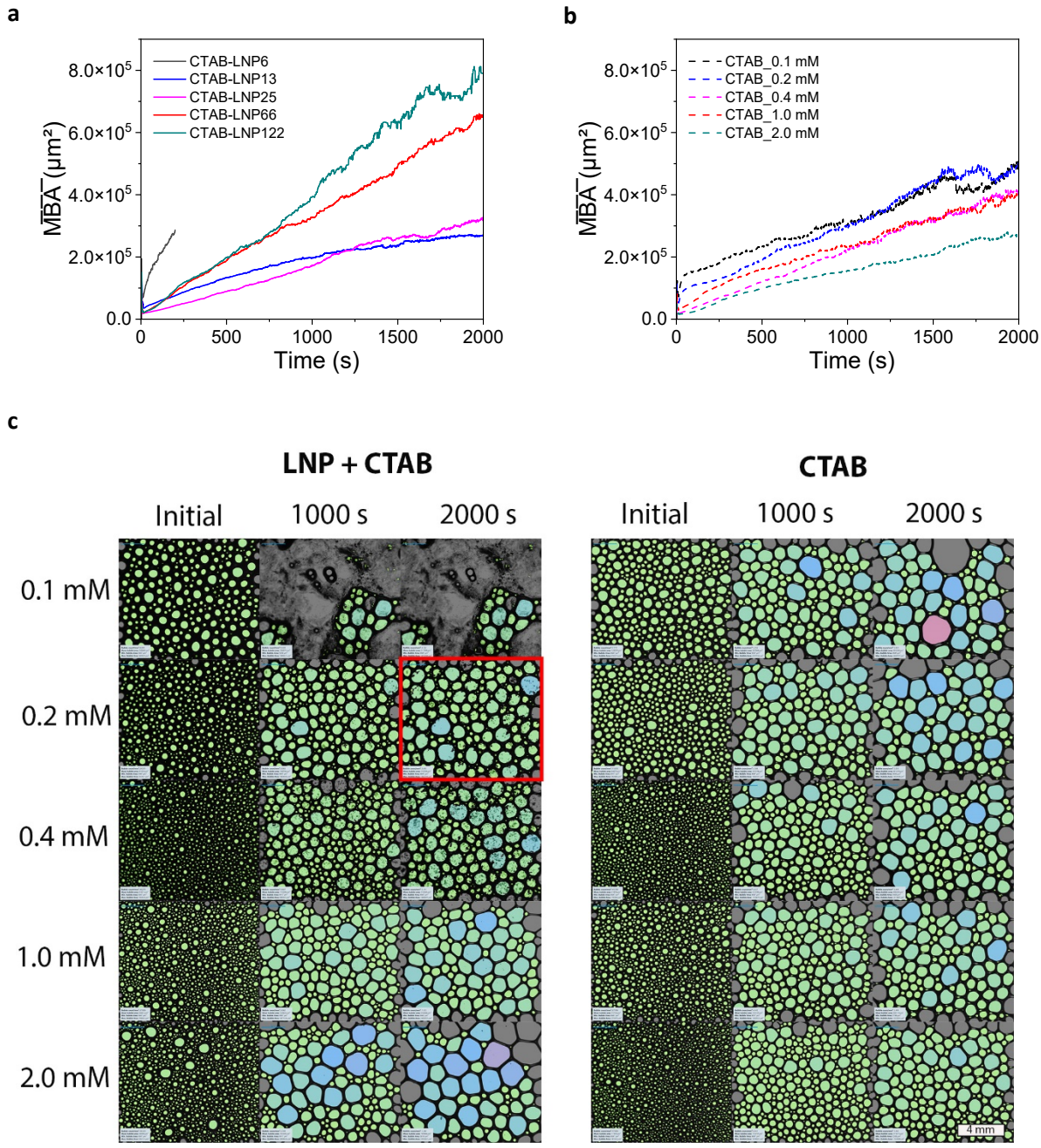

Figure S3. (a-b) Mean bubble size area (MBA) as a function of time of the wet foams stabilized by CTAB-LNPs and the pure CTAB. The LNP concentration was fixed at 0.6 wt% and the CTAB concentration varied from 0.1 to 2 mM. The corresponding CTAB to LNP mass ratios varied from 6 to 122 mg/g. (c) The corresponding bubble profiles of the foams right after foaming (initial), 1000 s, and 2000 s after foaming (scale bar: 4 mm). The red box marks the sample with the smallest

MBA at 2000 s after foaming. Note: the foam was generated by air pumping with Krüss DFA100 and the bubble size profiles were monitored for 2000 s after foaming. A 40 mm-diameter prism column (CY4572) and a FL4551 filter paper (pore size of 12-25 µm, diameter of 32 mm) were used for foaming. The initial liquid volume was fixed at 50 ml, the air was pumping through the filter paper at a flow rate of 0.3 L/min and automatically stopped when the total height (liquid height plus foam height) reached 100 mm (~ 120 mL volume). The air pumping time (foaming time) was found to be between 13 and 14 s for all the samples.

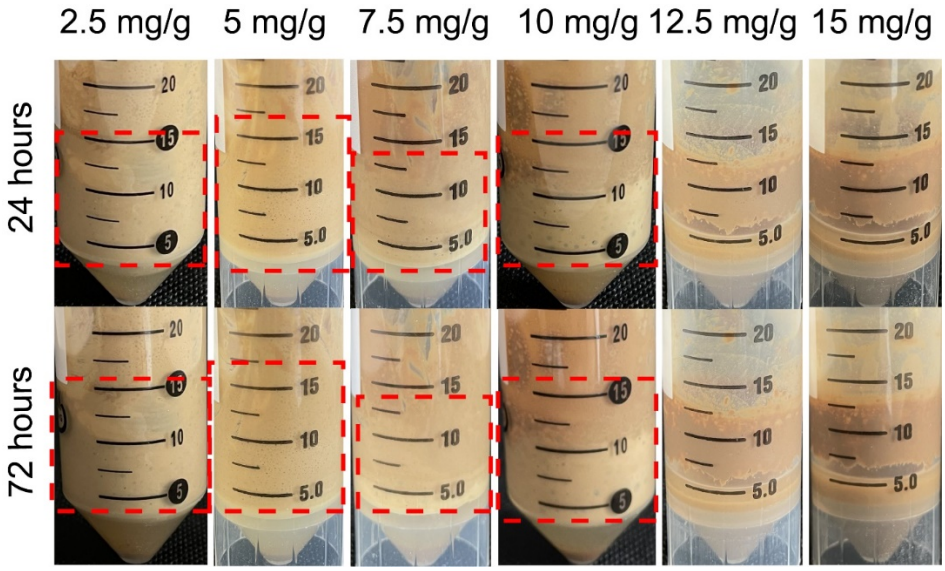

Figure S4. The appearance of the Pickering foams captured 24 and 72 hours after foaming. The foams were stabilized by 5 wt% CTAB-LNPs at the CTAB to LNP mass ratio of 2.5 to 15 mg/g. The dashed red boxes mark foam areas.

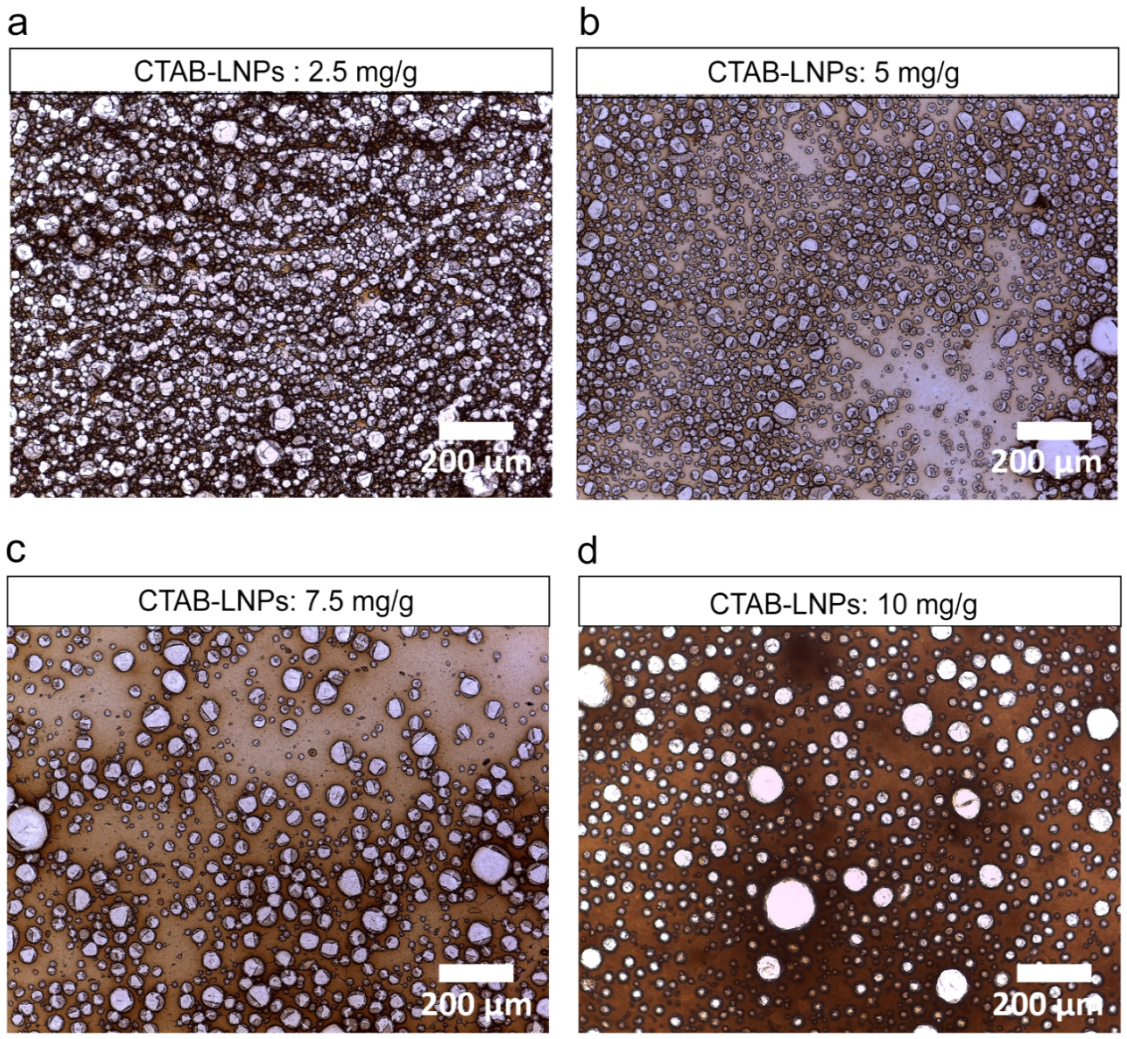

Figure S5. Optical microscopic images of the wet Pickering foams stabilized by 5 wt% CTAB-LNPs (the mass ratio of CTAB to LNP varies from 2.5 to 10 mg/g). The images were captured 3 hours after foaming. The foams were mostly dried while being captured by the optical microscope, indicating the strong stability of these Pickering foams against rupture upon drying.

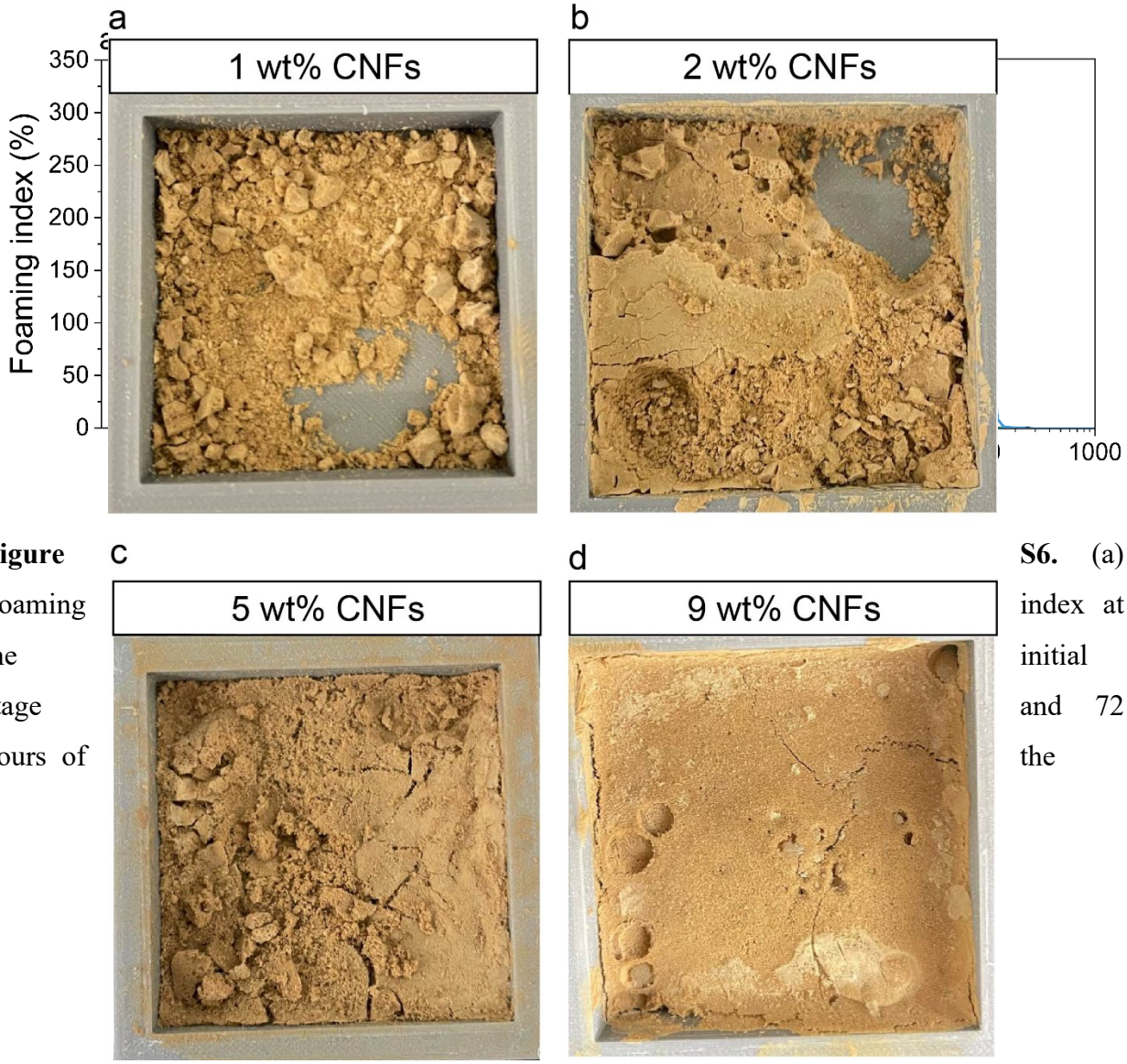

Figure S6. (a) Foaming index at the initial and 72 hours of the stage

Pickering foams stabilized by 5 wt% CTAB-LNPs (5 mg/g) with the addition of CNFs (1 to 9 wt% relative to the total solid mass). (b) Bubble size distributions of the foams measured 3 days after foaming.

Figure S7. Appearance of the dry foams at the CNF fractions of (a) 1 wt%, (b) 2 wt%, (c) 5 wt%, (d) 9 wt%. The foams were freeze-dried from the Pickering foams described in Figure S6.

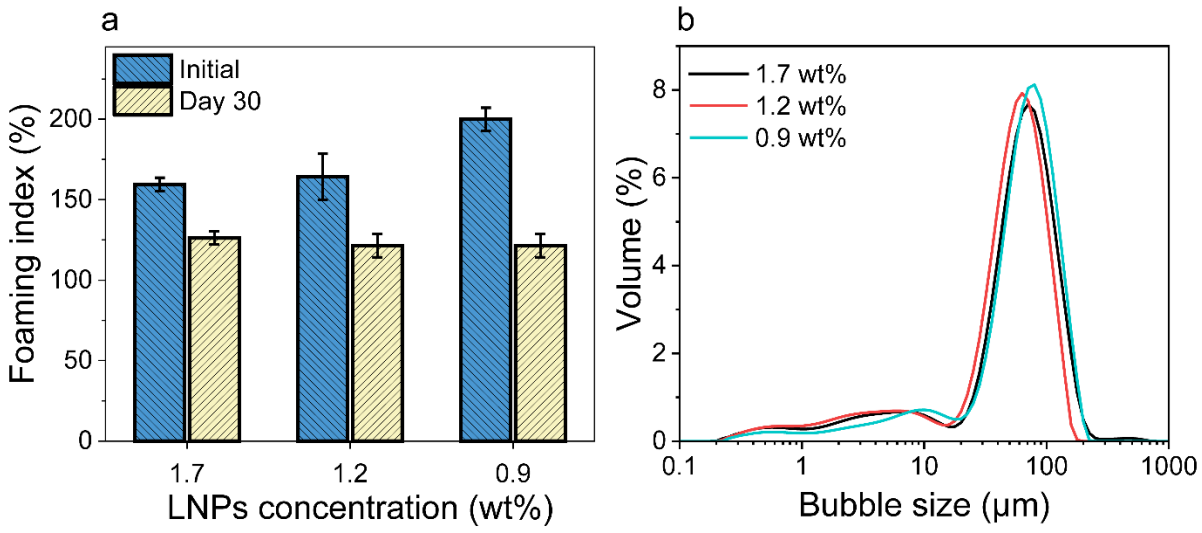

Figure S8. (a) Foaming index at the initial stage and day 30 of the Pickering foams stabilized by CTAB-LNPs (5 mg/g) at the concentrations of 0.9, 1.2, and 1.7 wt%. (b) Bubble size distributions of the foams measured 3 days after foaming.

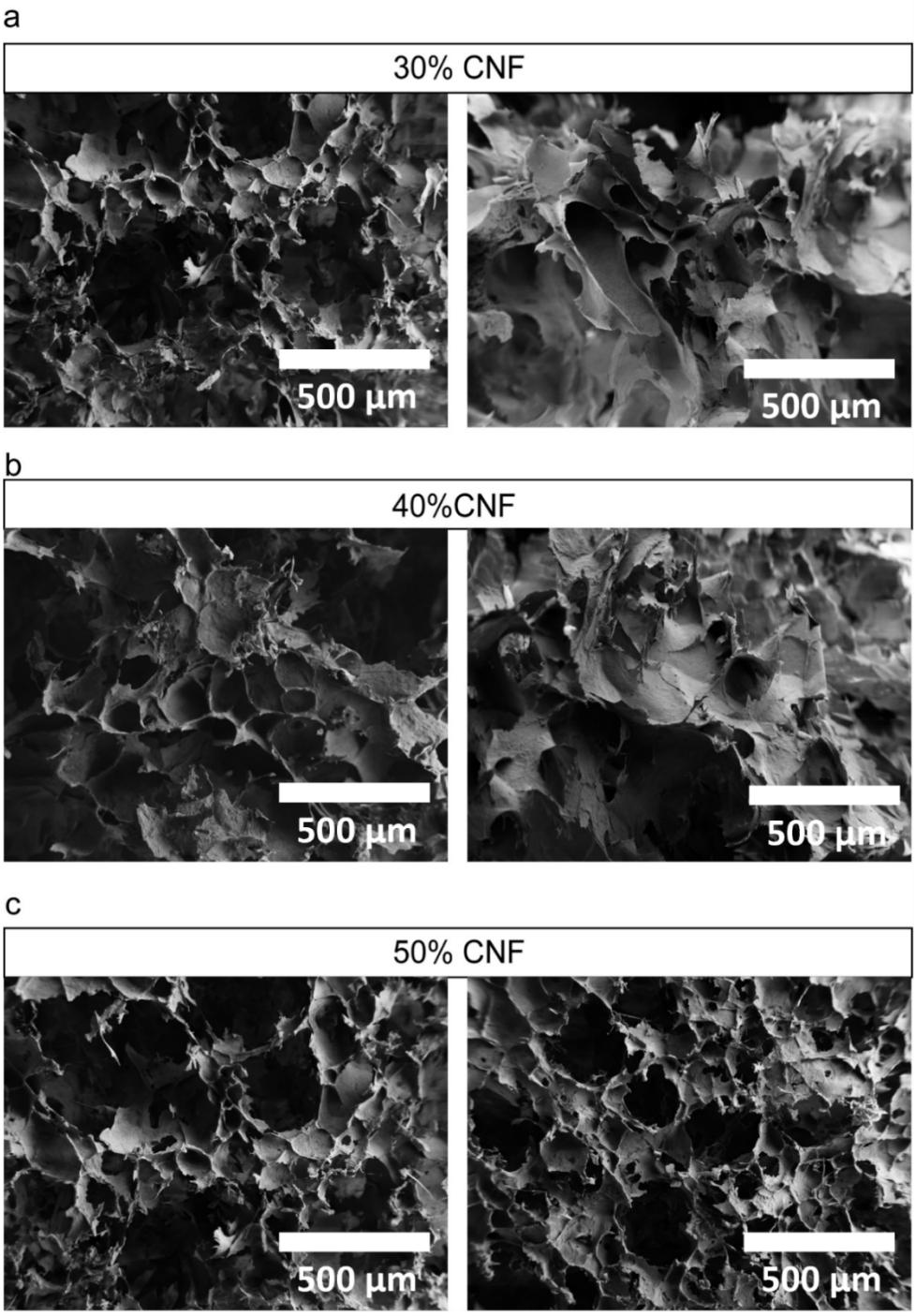

Figure S9. SEM images captured from the cross sections of the dry composite foams comprised of CTAB-LNPs (5 mg/g) and CNFs at the CNF fractions of 30 wt%, 40 wt%, and 50 wt% (relative to the total solid mass), respectively.

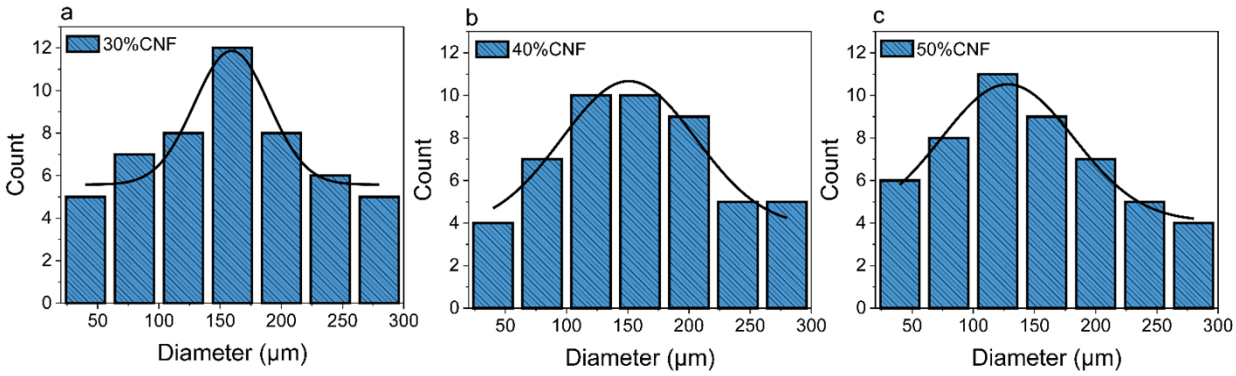

Figure S10. The cell size distributions of the foams calculated based on the SEM images (Figure 4b, f, j and Figure S9) and fitted using a Gaussian function.

Figure S11. The thermal insulation experimental setup. (a) IR thermal camera. (b) Hot plate with a copper plate on it. (c) Thermometer to ensure the constant temperature of the hot plate at 120℃.

Table S3. The average surface temperature of the foams at 1 min and 60 min after being heated on a hot copper plate at 120 °C.

Sample	Time (min)	Average (°C)	Minimum (°C)	Maximum (°C)	Span	Standard deviation
30% CNF	1	48.02	39.22	66.69	27.47	4.10
	60	63.56	34.45	96.89	62.44	6.89
40% CNF	1	47.81	32.40	64.46	32.06	4.04
	60	65.17	41.75	93.62	51.87	7.55
50% CNF	1	45.92	34.41	72.19	37.78	4.62
	60	62.69	40.64	97.12	56.48	4.47
Rigid PU foam	1	41.06	37.86	77.55	39.69	3.92
	60	51.34	28.59	72.96	44.37	3.58

Reference

(1) Li, N.; Liu, S.; Luo, H. A New Method for the Determination of the First and Second Cmc in Ctab Solution by Resonance Rayleigh Scattering Technology. *Anal. Lett.* **2002**, *35* (7), 1229–1238. https://doi.org/10.1081/AL-120005975.