Electronic Supplementary Material (ESI) for RSC Sustainability.

Supporting Information

Synthesis and structural design of microspheres comprising cellulose nanofibers and artificial lignin polymer by enzyme-mediated Pickering emulsion templating

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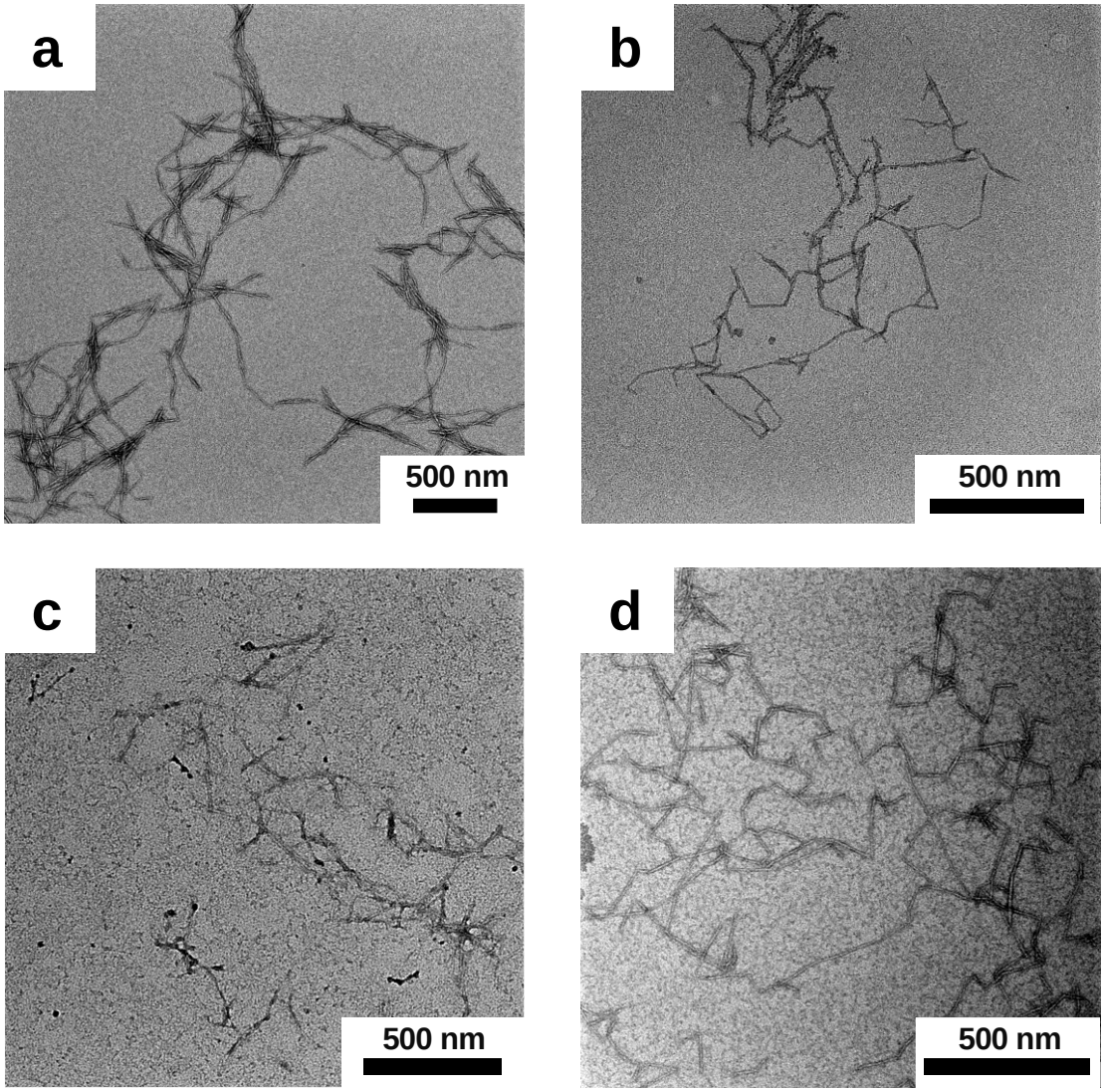

Figure S1 TEM images of a) CNF, b) TOCNF, c) S-CNF, and d) P-CNF. (TEM = transmission electron microscopy; CNF = cellulose nanofiber; TOCNF = 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO)-oxidized CNF; S-CNF = sulfated CNF; P-CNF = phosphorylated CNF).

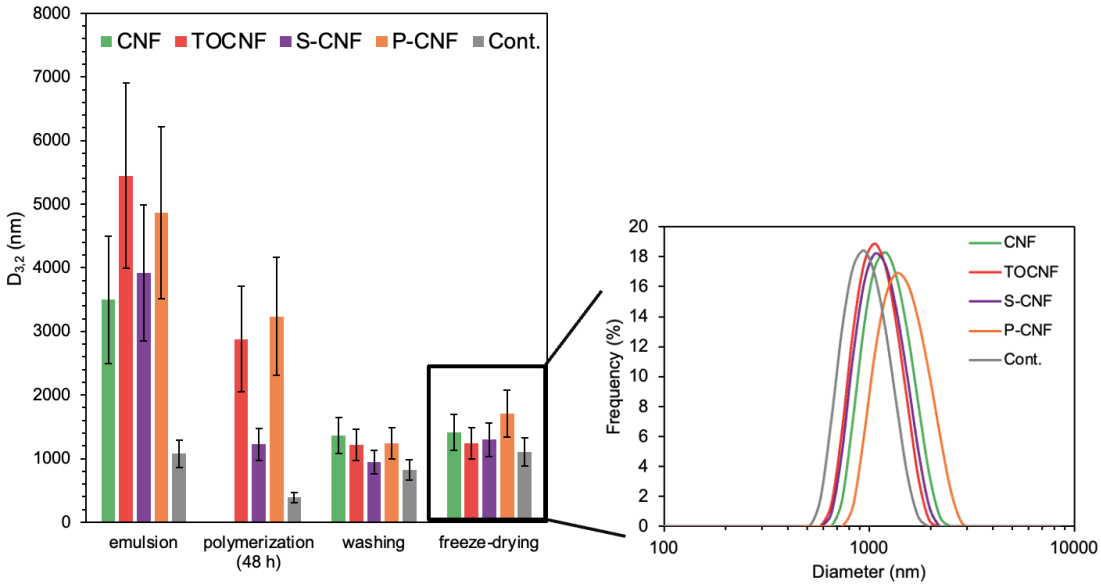

Figure S2 Sauter mean diameters of emulsion and microparticles of ALP/CNFs at each step. (ALP = artificial lignin polymer; CNF = cellulose nanofiber; TOCNF = 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO)-oxidized CNF; S-CNF = sulfated CNF; P-CNF = phosphorylated CNF; Cont.: ALP without CNFs).

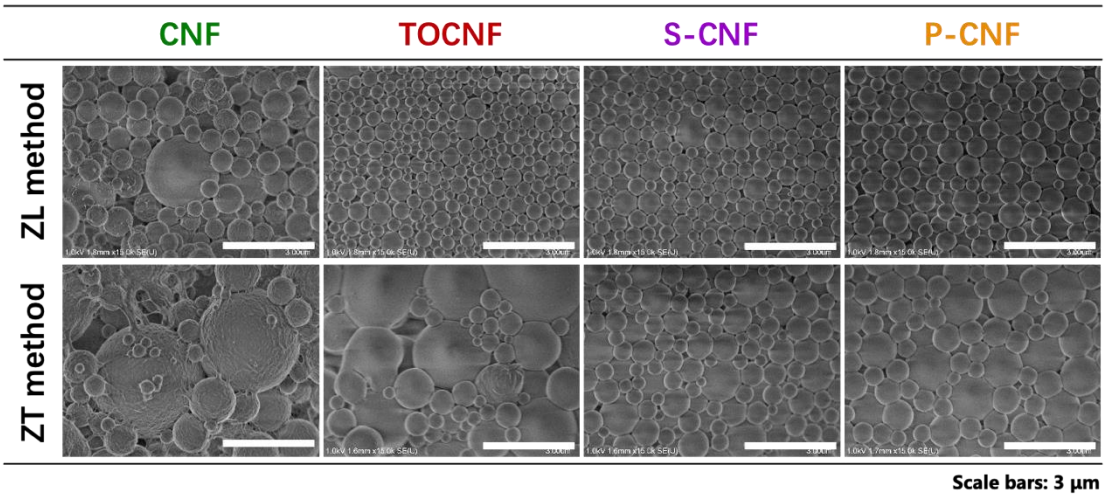

Figure S3 SEM images of ALP/CNF microparticles synthesized by the "bulk" and "stepwise" polymerization, which refer to Zulaufverfahren (ZL) and Zutropfverfahren (ZT) methods, respectively. (SEM = scanning electron microscopy; ALP = artificial lignin polymer; CNF = cellulose nanofiber; TOCNF = 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO)-oxidized CNF; S-CNF = sulfated CNF; P-CNF = phosphorylated CNF).

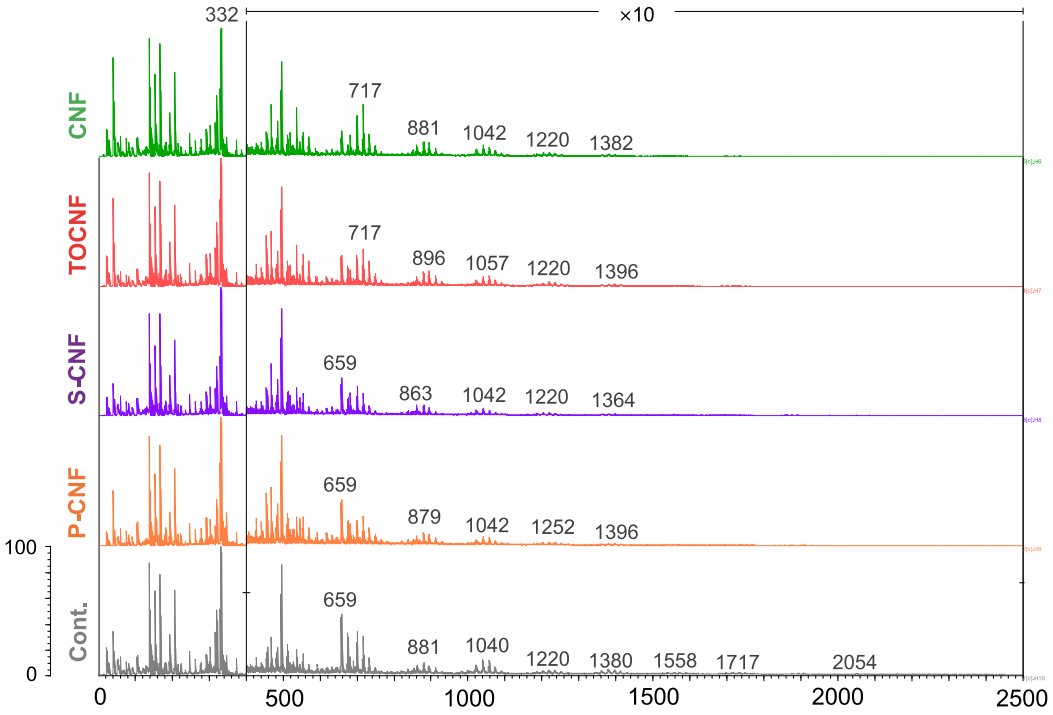

Figure S4 MALDI TOF-MS spectra of ALP/CNFs. Matrix: 2,5-dihydroxybenzoic acid (DHB). (MALDI TOF-MS = matrix-assisted laser desorption/ionization–time-of-flight mass spectrometry; ALP = artificial lignin polymer; CNF = cellulose nanofiber; TOCNF = 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO)-oxidized CNF; S-CNF = sulfated CNF; P-CNF = phosphorylated CNF; Cont.: ALP without CNFs).

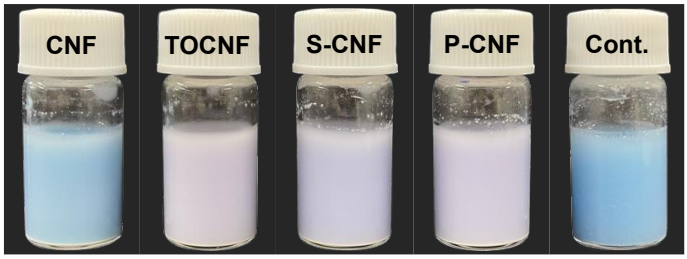

Figure S5 Metachromatic color change of the aqueous dispersions of ALP/CNFs with 0.01 mg mL^{-1} TBO (toluidine blue O). (ALP = artificial lignin polymer; CNF = cellulose nanofiber; TOCNF = 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO)-oxidized CNF; S-CNF = sulfated CNF; P-CNF = phosphorylated CNF; Cont.: ALP without CNFs).

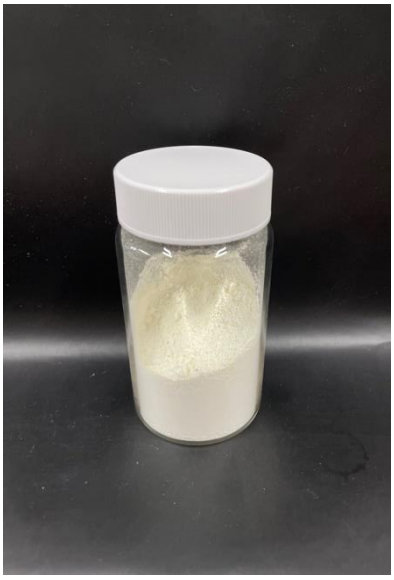

Figure S6 Digital image of ALP/CNF microparticles. (ALP = artificial lignin polymer; CNF = cellulose nanofiber).

Table S1 Description of the sample for SPF determination. (SPF = sun protection factor).

	Raw materials	Constituents	Content %
A	Purified water	Water	50.66
	BG	1,3-BG	5.00
	Glycerin	Glycerin	1.00
	EDTA-2Na	EDTA-2Na	0.04
	Phenoxyethanol	Phenoxyethanol	0.30
B1	BENTONE GEL ISD V	Isododecane, Disteardimonium Hectorite, Propylene carbonate	2.00
	IPM-100	Isopropyl myristate	8.00
B2	Uvinul MC80	Ethylhexyl methoxycinnamate	3.00
B3	KF-6038	Lauryl PEG-9 Polydimethylsiloxyethyl Dimethicone	1.00
	KF-995	Cyclopentasiloxane	5.00
	KSG-16	Dimethicone/Vinyl Dimethicone Crosspolymer, Dimethicone	4.00
C	DIS-11A	Titanium oxide, hydroxylated Al, Silica, Hydrogen Dimenthicone, Cyclopentasiloxane	10.00
	DIF-3ST2	Zinc oxide, Silica, Hydrogen Dimenthicone, Cyclopentasiloxane	5.00
D	**Powder sample**	**Microparticles**	**5.00**
		Total	100

The sample preparation method was as follows: (1) component A was weighed and dissolved; (2) components B1, B2, and B3 were weighed and dispersed; (3) component C was weighed, added to component B mixture, and dispersed; (4) component D was weighed and added to components B + C and stirred; and (5) component A was added to components B + C + D and dispersed (2400 rpm, 30 s).