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Electronic Supporting Information

Weakening fibril-fibril interactions via on-demand regulation of hemicellulose phase towards facile disassembly of lignocellulose heterostructure into approaching native-state elementary fibrils

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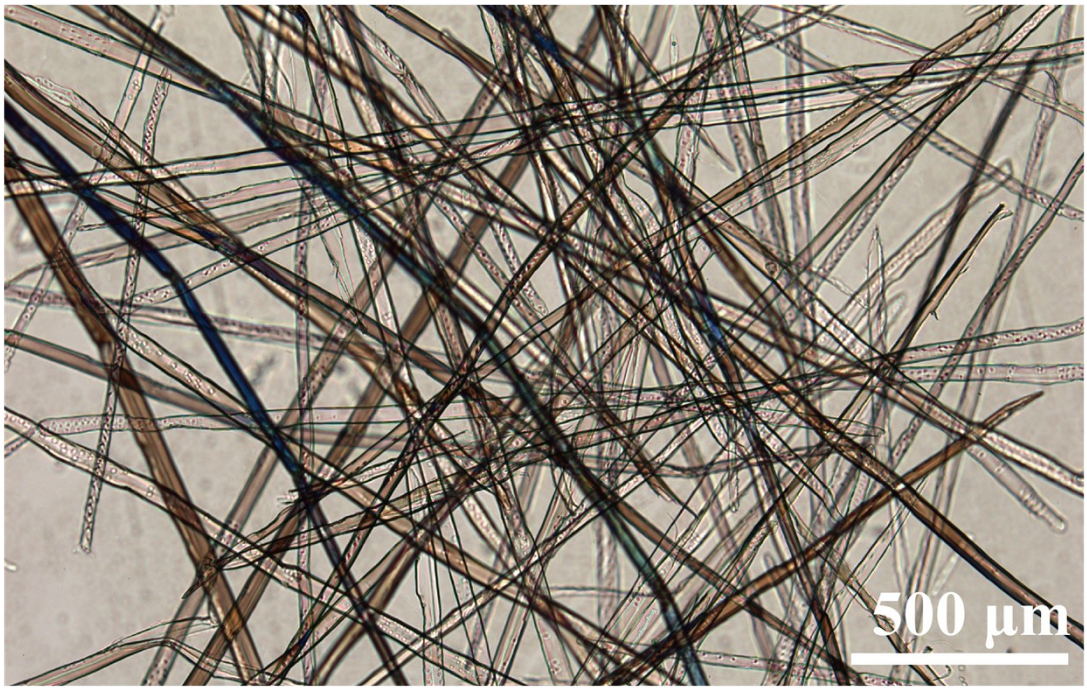

Fig. S1 Optical micrograph of spruce holocellulose fibers.

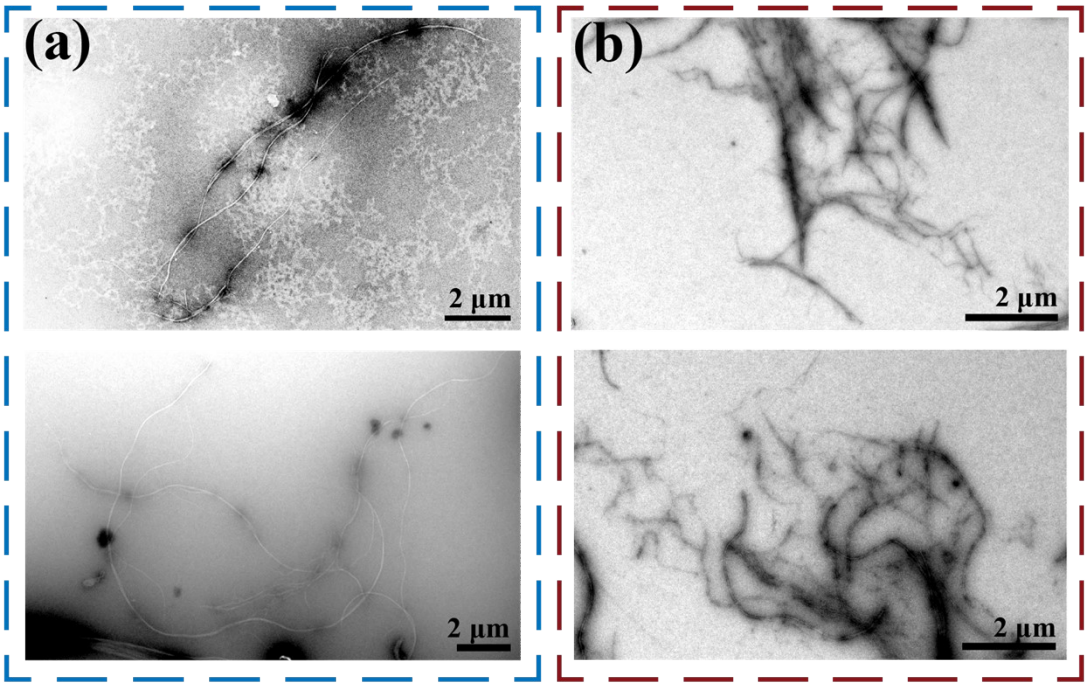

Fig. S2 TEM images of (a) HCNF-24%IL-W, and (b) HCNF-50%IL-W.

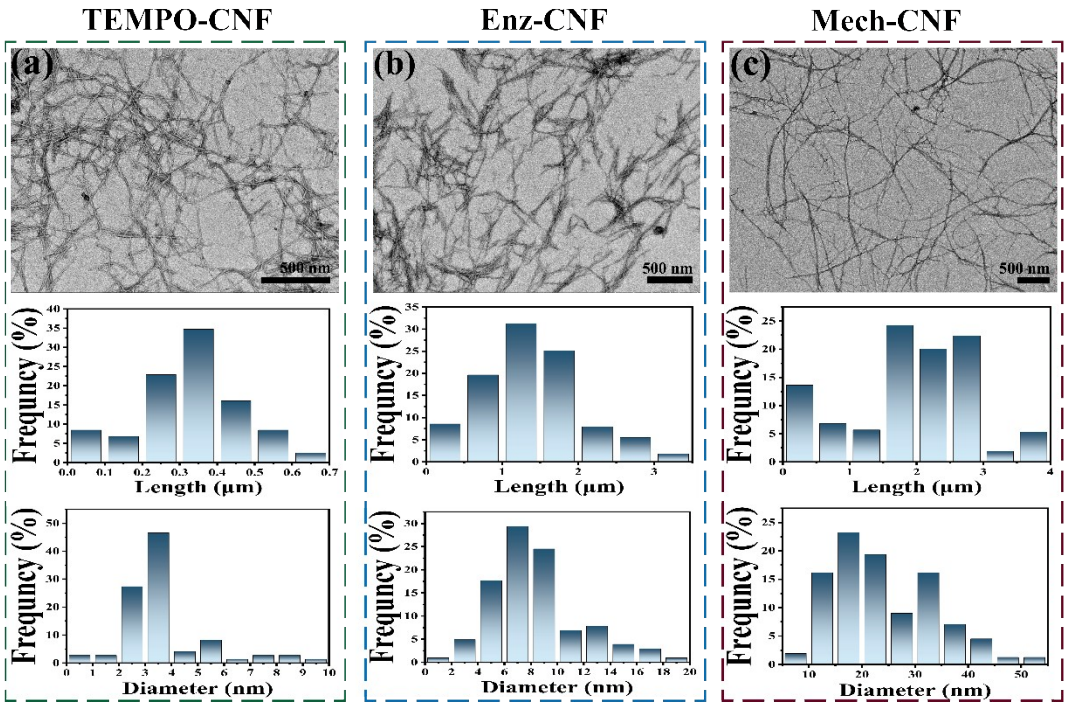

Fig. S3 TEM images and TEM-based length and diameter distributions of (a) TEMPO-CNF, (b) Enz-CNF, and (c) Mech-CNF.

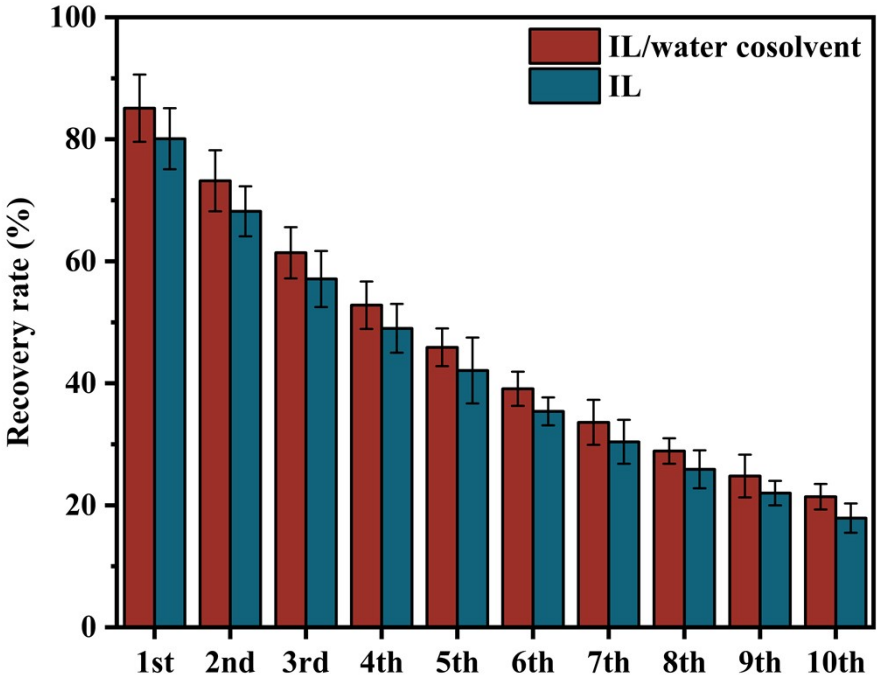

Fig. S4 The recovery rate of IL/water cosolvent and IL at 1st~10th cycles.

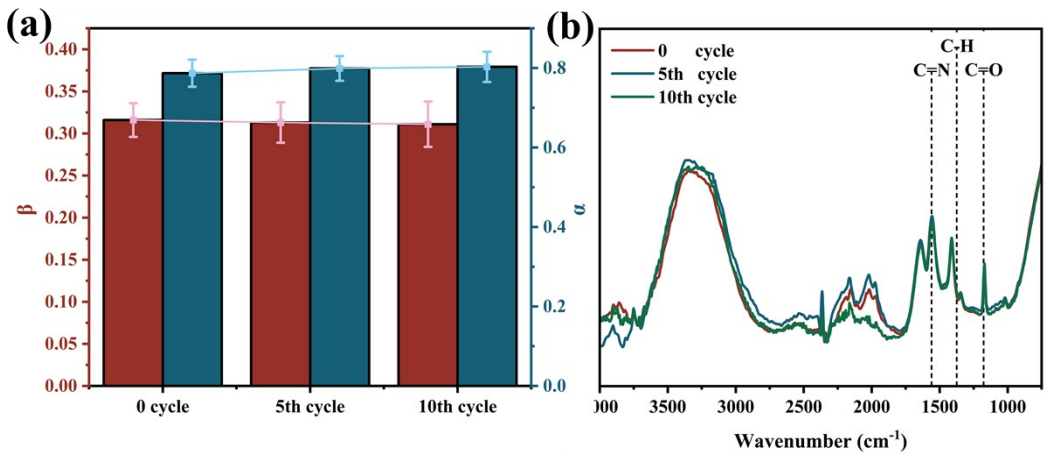

Fig. S5 (a) Kamlet Taft hydrogen bond basicity β and hydrogen bond acidity α values of the recovered IL/water cosolvent; (b) ATR-FTIR spectra of the ILs from the recovered IL/water cosolvent.

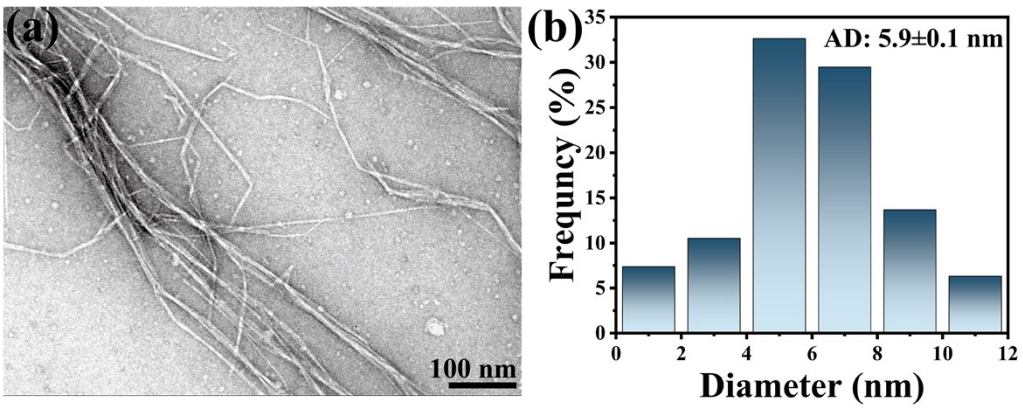

Fig. S6 (a) TEM images and (b) TEM-based diameter distributions of the HCNF produced by using the 10-times recycled IL.

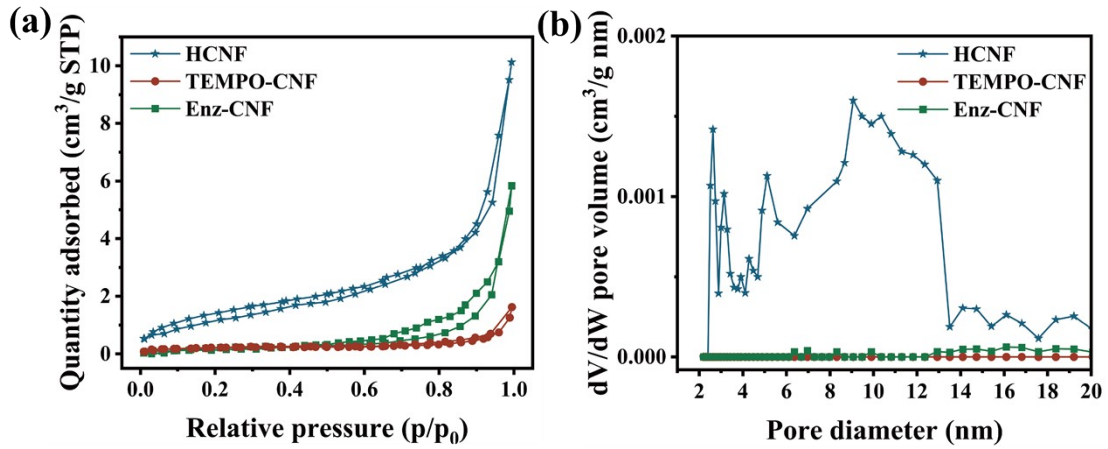

Fig. S7 (a) N_2 adsorption-desorption isotherms and (b) BJH pore size distribution derived from N_2 adsorption for HCNF, TEMPO-CNF and Enz-CNF nanopapers.

Table S1. Kamlet Taft parameter of IL/water cosolvent at different IL concentration.

IL	KT-parameter at 25°C			
(wt%)	β	π^*	α	β-α
0	0.142	1.330	1.121	-0.979
24	0.316	1.318	0.787	-0.471
50	0.492	1.306	0.636	-0.144
100	1.107	1.039	0.474	0.633

Table S2. Morphological characteristics for the high-aspect-ratio CNFs produced by

using different strategies.

Precursor fibers	Protocol	Length (μm)	Diameter (nm)	Aspect ratio	DP	References
Bleached bamboo pulp fibers	DES (ZnAc/ChCl) treatment and subsequent homogenization	14.3	16.2	882	\	Ling et al.[1]
Bleached spruce pulp fibers	Carboxymethylation and subsequent mechanical blending	3.8	3.9	1000	1300	Zhou et al.[2]
Bleached sulfite softwood pulp fibers	Maleic anhydride esterification followed by mechanical blending	~1.3	~3.2	390	\	Zhang et al.[3]
Spruce holocellulose fibers	Mechanical blending	~2.1	~5.0	530	3460	Yang et al.[4]
Softwood holocellulose fibers	High-pressure microfluidization	~3.0	~5.0	600	3800	Galland et al.[5]
Spruce holocellulose fibers	Mechanical blending	~2.0	~3.4	580	3400	Yang et al.[6]

Table S3. Estimated costs of materials and unities per ton of HCNF.

	Categories	Consumption (ton/kW·h)	Price (CNY·ton^{-1})	Cost (CNY)	Sources
Materials	Spruce holocellulose fibers	1	13400	13400	Guangxi Jianing Pulp Co., Ltd.
	1-Ethyl-3-methylimidazolium acetate (99.5%)	8	50000	400000	Guangzhou Ruishi Biotechnology Co., Ltd.
Utilities	Water	75	5.59	420	Xixiangtang District, Nanning, China
	Electricity	3293	0.83	2733	Xixiangtang District, Nanning, China

Table S4. Estimated costs of materials and unities per ton of TEMPO-CNF.

	Categories	Consumption (ton/kW·h)	Price (CNY·ton^{-1})	Cost (CNY)	Sources
Materials	Bleached pine pulp	1	7750	7750	Qingdao Junuo International Trade Co., Ltd.
	TEMPO (98%)	0.016	2516000	40256	Shanghai Aladdin Reagent Co., Ltd.
	NaBr (99%)	0.1	200000	20000	Shanghai Aladdin Reagent Co., Ltd.
	NaClO (5%)	1.12	34000	38080	Shanghai Aladdin Reagent Co., Ltd.
	NaOH (99%)	2	4000	8000	Maoming Xiongda Chemical Co. Ltd.
Utilities	Water	940	5.59	5255	Xixiangtang District, Nanning, China
	Electricity	35168	0.83	29189	Xixiangtang District, Nanning, China

Table S5. Main technical data of the equipment used for producing TEMPO-CNF and HCNF.

Equipment	Model	Technical data			Manufacturers
Electric blender	ZXY-90	Speed (rpm)	Capacity (t)	Power (kW)	Anhui Shuangjie Manufacturing Co., Ltd.
		30000	3.3	6～7	
Supermasscolloider	MKZB20-100J	Speed (rpm)	Capacity (Kg/h)	Power (kW)	Masuko Sangyo Co.,Ltd.
		1500	1200～6000	38～76	
Centrifugal machine	4-5KL	Speed (rpm)	Capacity (Kg)	Power (kW)	KingHwa Medical Scientific Co.,Ltd.
		13500	3	1.2	
Circulating water vacuum pump	SHB-III	Maximum vacuum (MPa)	Air volume per head (L/min)	Power (kW)	Nanning Boyu Instrument Co.,Ltd.
		0.098	10	0.18	

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