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## High-strength cellulose nanofibers produced via swelling pretreatment based on a choline chloride-imidazole deep eutectic solvent

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Table S1. Fiber width and CWT of cellulose fibers after treatment with CCIMI DESs at various cellulose/solvent mass ratios for 1 h.

Entry	Cellulose/DES mass ratio	Temperature (°C)	Fiber properties	
			Width (µm)	CWT (µm)
1	1:50	60	19.3	6.4
2	1:50	80	19.5	6.3
3	1:50	100	19.5	6.2
4	1:33	60	19.3	6.3
5	1:33	80	19.6	6.5
6	1:33	100	19.7	6.3
7	1:25	60	19.2	6.2
8	1:25	80	19.2	6.3
9	1:25	100	19.6	6.5

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Table S2. The number average molar mass  $(M_n)$ , weight average molar mass  $(M_w)$ , and polydispersity (PD) of cellulose before and after treatment with CCIMI at 100°C for 180 min and CCUrea 100°C for 120 min

Sample	$M_{n}\left(g/mol\right)$	$M_{\rm w}(g\!/mol)$	PD
Original pulp	47 000 ± 3000	$476\;000\pm 29\;000$	10.2 ± 0.3
CCIMI 100C 180 min	51 000 ± 2000	543 000 ± 85 000	10.6 ± 1.7
CCUrea 100C 120 min	52 000 ± 13 000	$728000\pm79000$	14.4 ± 2.2

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Figure S1. DRIFT spectra of DES treated fibers: a) CCIMI 15 min 60 °C, b) CCIMI 180 min 60 °C,



Figure S2. Aerogels produced by freeze-drying from CNFs obtained from DES treated cellulose fibers, from left to right: CCIMI 15 min 60 °C; CCIMI 180 min 100 °C; CCIMI 180 min 100 °C; CCUrea 120 min 100 °C

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Figure S3. The histograms of the diameters of cellulose nanofibers produced from DES pre-treated fibers. a) CCUrea 60 °C 15 min b) CCUrea 60 °C 180 min c) CCUrea 100 °C 15 min d) CCUrea 100 °C 180 min e) CCUrea 100 °C 120 min

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Figure S4. TGA curves of original pulp and CNFs produced from CCIMI and CCUrea treated fiber measured in air.

Recycling times	Mass after evaporation (g)	Water content (%) <sup>a</sup>
1	263	4.9
2	264	5.3
3	260	3.8
4	260	3.8

Table S3. Yield and the water content of the CCIMI after recycling

<sup>a</sup> Calculated from the original mass of the DES



Figure S5. DRIFT spectra of CNFs produced using pristine CCIMI (a) and after 1 (b), 2 (c), and 3 (d) recycling.