Supporting Information

Effect of ultrasonication on the size distribution and stability of cellulose nanocrystals in suspension: an asymmetrical flow field-flow fractionation study

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Ultrasonication of cellulose nanocrystal suspension

$$e_{US} = \frac{ultrasound\ power \cdot treatment\ time}{CNC\ mass} \quad \text{(S1)}$$

Table S1: Process parameters during incremental ultrasonication of cellulose nanocrystal (CNC) suspension

Sample name	**CNC-2**	**CNC-4**	**CNC-6**	**CNC-8**	**CNC-10**	**CNC-15**	**CNC-20**	**CNC-40**
Suspension mass /g	243.35	213.32	183.66	153.88	124.05	94.35	64.51	34.70
CNC mass /g	2.43	2.13	1.83	1.54	1.24	0.94	0.64	0.35
Energy input /kJ	4.90	9.13	12.82	15.89	18.36	23.05	26.21	32.81
Treatment time /s	146	274	385	477	551	692	787	985
Targeted e_{US} /kJ g^{-1} CNC	2.00	4.00	6.00	8.00	10.00	15.00	20.00	40.00
Actual e_{US} /kJ g^{-1} CNC	2.01	4.01	6.02	8.02	10.02	14.99	19.91	38.98

Fractionation of CNCs by asymmetrical flow field-flow fractionation

Validity of the set-up

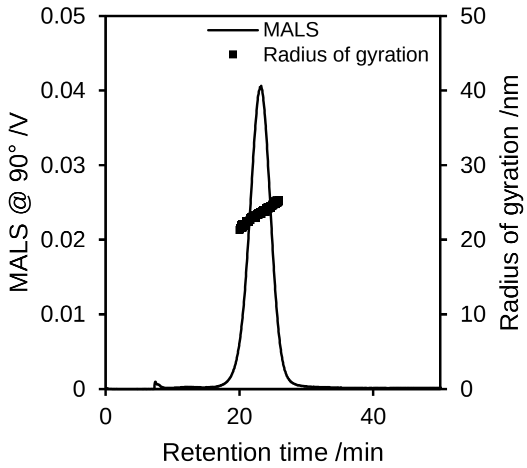

Figure S1: Fractogram of standard polystyrene beads with a nominal diameter of 60 nm (NIST 2021). r_g was evaluated with the NovaMALS software (Postnova Analytics GmbH 2020)

Our set-up enabled an accurate determination of particles with a $r_g \geq 25$ nm that eluted at net retention times, t_R, ≥ 10.6 min (**Figure S1**). Provided that the applied rod model is valid over the full distribution (Mukherjee and Hackley 2018) and CNCs have a L-d ratio of 5–50 (ISO 2017), a r_g of 25 nm corresponds to particles with a rod length of 87 nm and a minimal L-d ratio of 5 limits them to particle diameters below 17 nm. Notably, single elementary cellulose fibrils of plantal sources have diameters in the range of 3 nm (Kubicki et al. 2018) and reported crystallite dimensions of single CNCs from cotton are in the range from 5–10 nm (Dong et al. 1998, Beck-Candanedo et al. 2005). Accordingly, single CNCs with a length of 87 nm and diameters of 3, 5, or 10 nm have respective L-d ratios of 29, 17, or 9. Likewise, minimal evaluable rod lengths of 95 nm (Guan et al. 2012), 101 nm (Mukherjee and Hackley 2018), and 104 nm (Chen et al. 2020) have been reported for the application of AF4-MALS on colloidal CNCs. Shorter rod lengths determined by AF4-MALS have been reported by Ruiz-Palomero et al. (2017) and Espinosa et al. (2017); however, these values are either not unequivocally attributable to CNCs or methodically not inducible, respectively.

Evolution of maximal multi-angle light scattering intensity

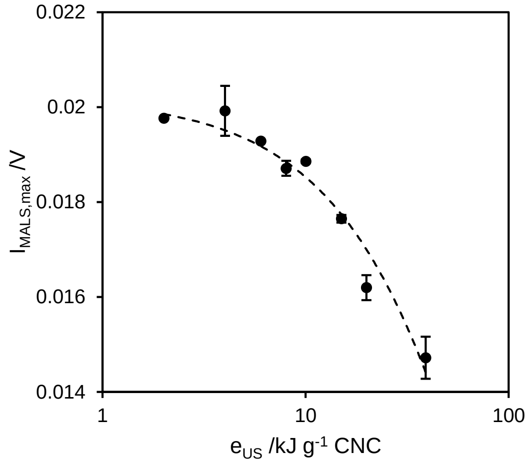

Figure S2: Maximal intensity of the 90° signal decreases exponentially with increasing ultrasound energy density

Effect of CNC treatment with ion-exchange resins

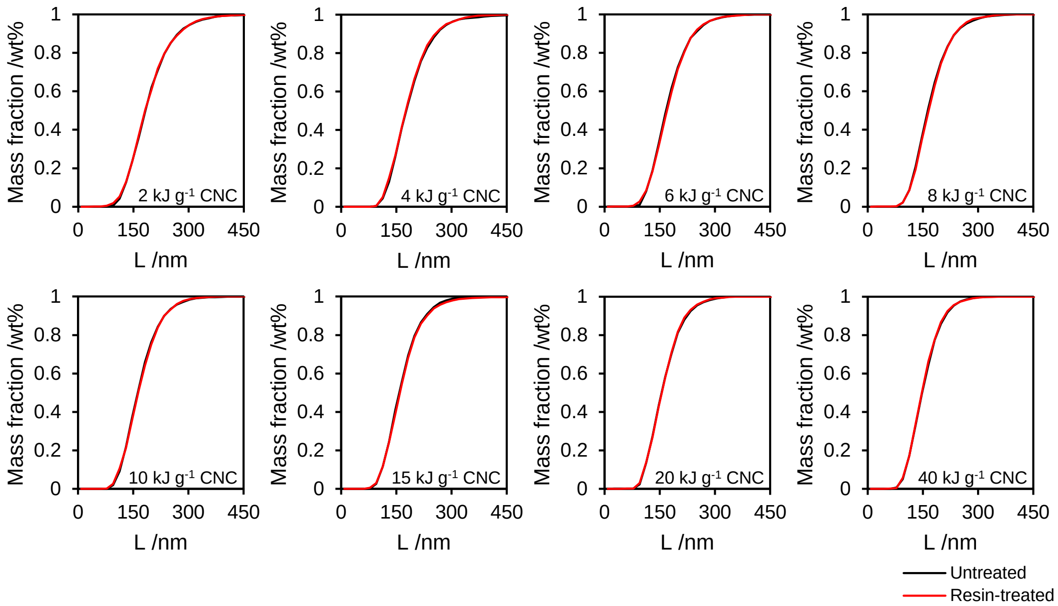

Figure S3: Particle size distributions of colloidal CNCs before (black) and after (red) treatment with ion-exchange resins at each energy input

Fractograms after CNC conditioning for six months

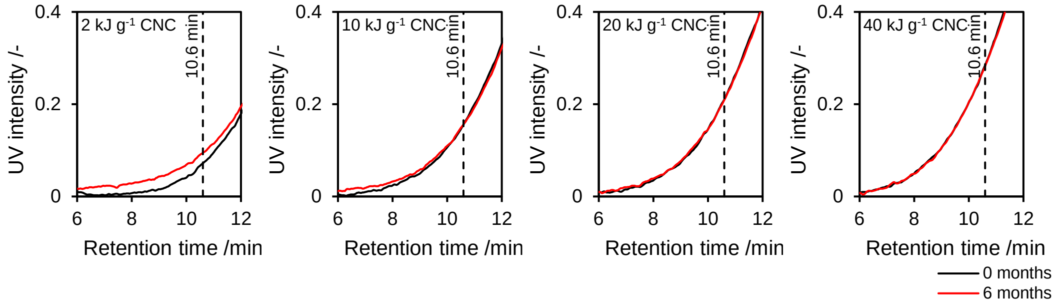

Figure S4: Normalized UV density distributions before (black) and after (red) conditioning of CNC suspensions for six months

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