



## Supplementary Materials

# Dynamic and Static Assembly of Sulfated Cellulose Nanocrystals with Alkali Metal Counter Cations

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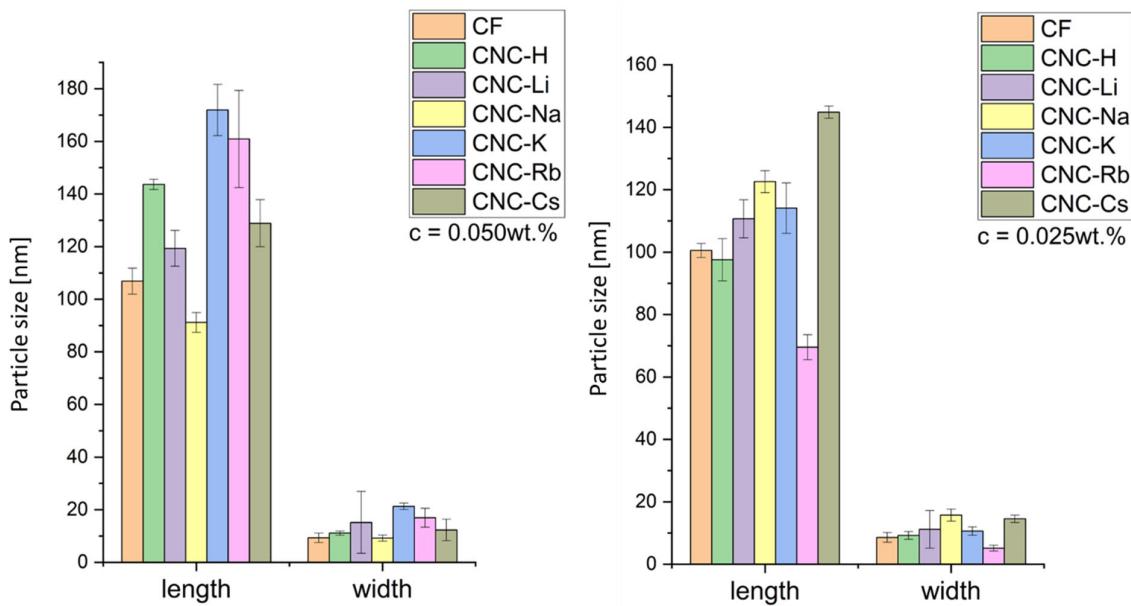
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**Figure S1.** Hydrodynamic diameter of M-CNC water suspensions determined by dynamic light scattering.



**Figure S2.** PLI visualization of K-CNC at  $100\text{ s}^{-1}$  in a zoomed-out version proving the absence of a flow-induced Maltese-cross pattern.



**Figure S3.** PLI visualization of Cs-CNC at  $100\text{ s}^{-1}$  in a zoomed-out version proving the presence of a flow-induced Maltese-cross pattern.

**Table S1.** Atomic composition (at.%) of the CNCs determined using XPS.

	C1s	O1s	Si2p	S2p	Na1s	K2p	Rb3d	Cs3d
H-CNC	53.61		41.54	4.42	0.43			
Na-CNC	57.19		41.50		0.37	0.94	-	
K-CNC	58.65		40.77		0.35		0.23	
Rb-CNC	58.48		40.92		0.37		0.23	
Cs-CNC	57.87		40.49	1.01	0.42			0.20

**Table S2.** Comparison of XPS binding energies of metal sulfates from Wahlqvist et al. [1] with the alkali metal cation modified M-CNC.

	Li1s	Na1s	K2p <sub>3/2</sub>	Rb3d <sub>5/2</sub>	Cs3d <sub>5/2</sub>
M <sub>2</sub> SO <sub>4</sub> [eV]	56.1	1071.6	293.0	109.8	724.3
M-CNC [eV]	n.d.	1072.0	293.1	110.0	724.7

## References

- [1] Wahlqvist, M.; Shchukarev, A. XPS spectra and electronic structure of Group IA sulfates. *J. Electron Spectrosc. Relat. Phenom.* **2007**, *156*, 310–314. <https://doi.org/10.1016/j.elspec.2006.11.032>.