Supplementary Information for

Structure property relationships of cellulose nanofibril hydro- and aerogels and their building blocks

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Supplementary figures

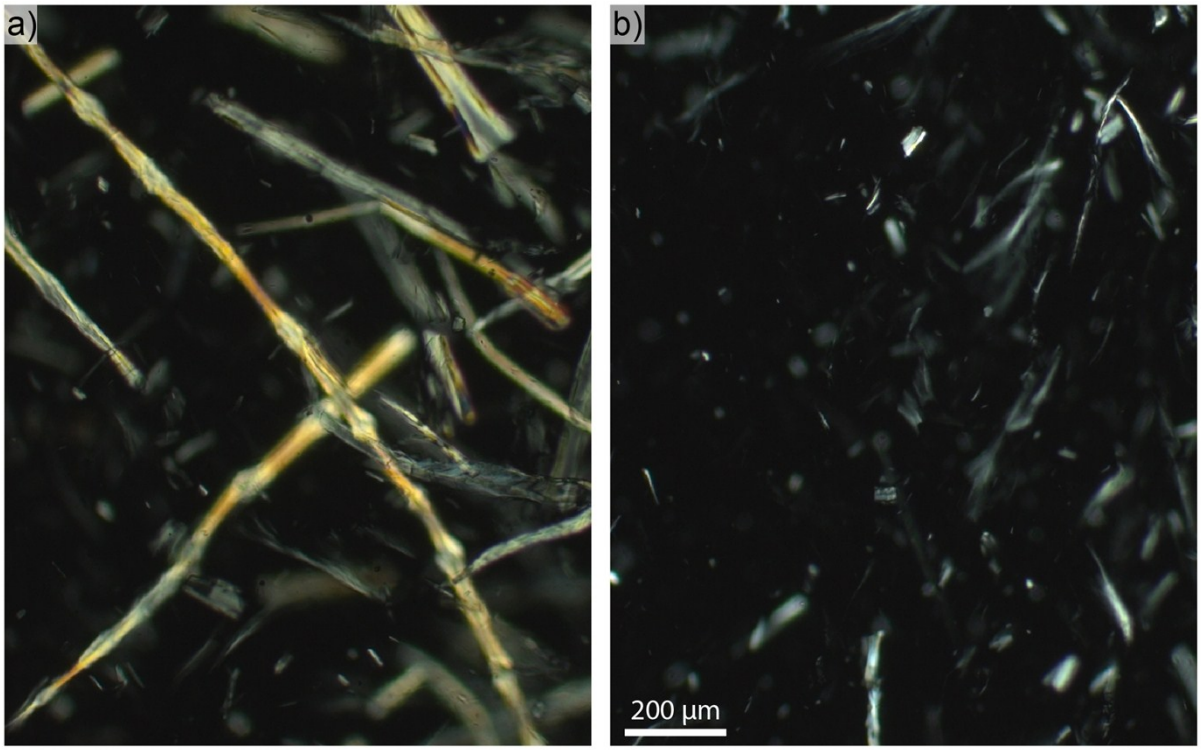

Figure S1. Polarized light micrographs of CNF dispersion (0.5 wt%) from unbleached pulp after 11 min sonication. a) 20 min centrifugation at 4000 rcf and b) after additional 90 min centrifugation at 12000 rcf.

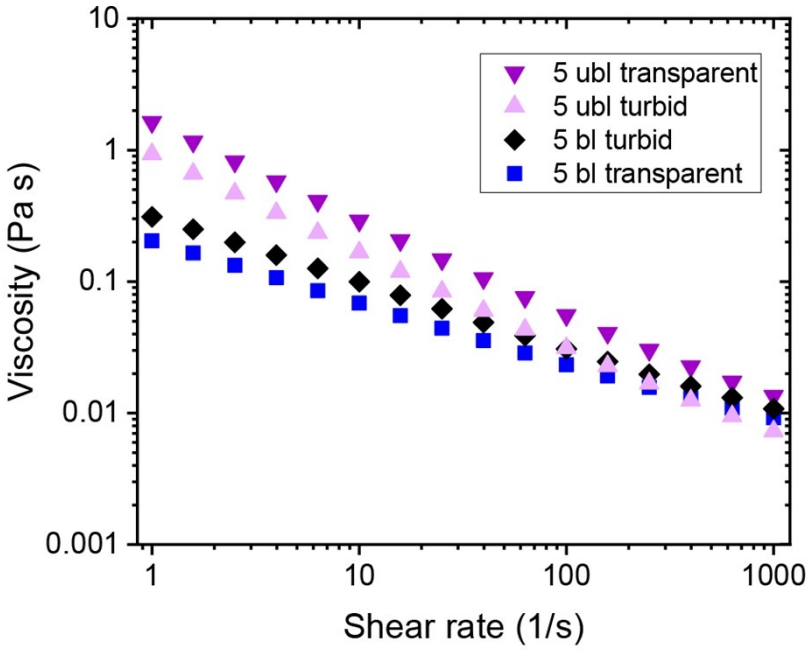

Figure S2. Rheological behavior of transparent and turbid CNF dispersions (0.40 wt%) using a double gap geometry.

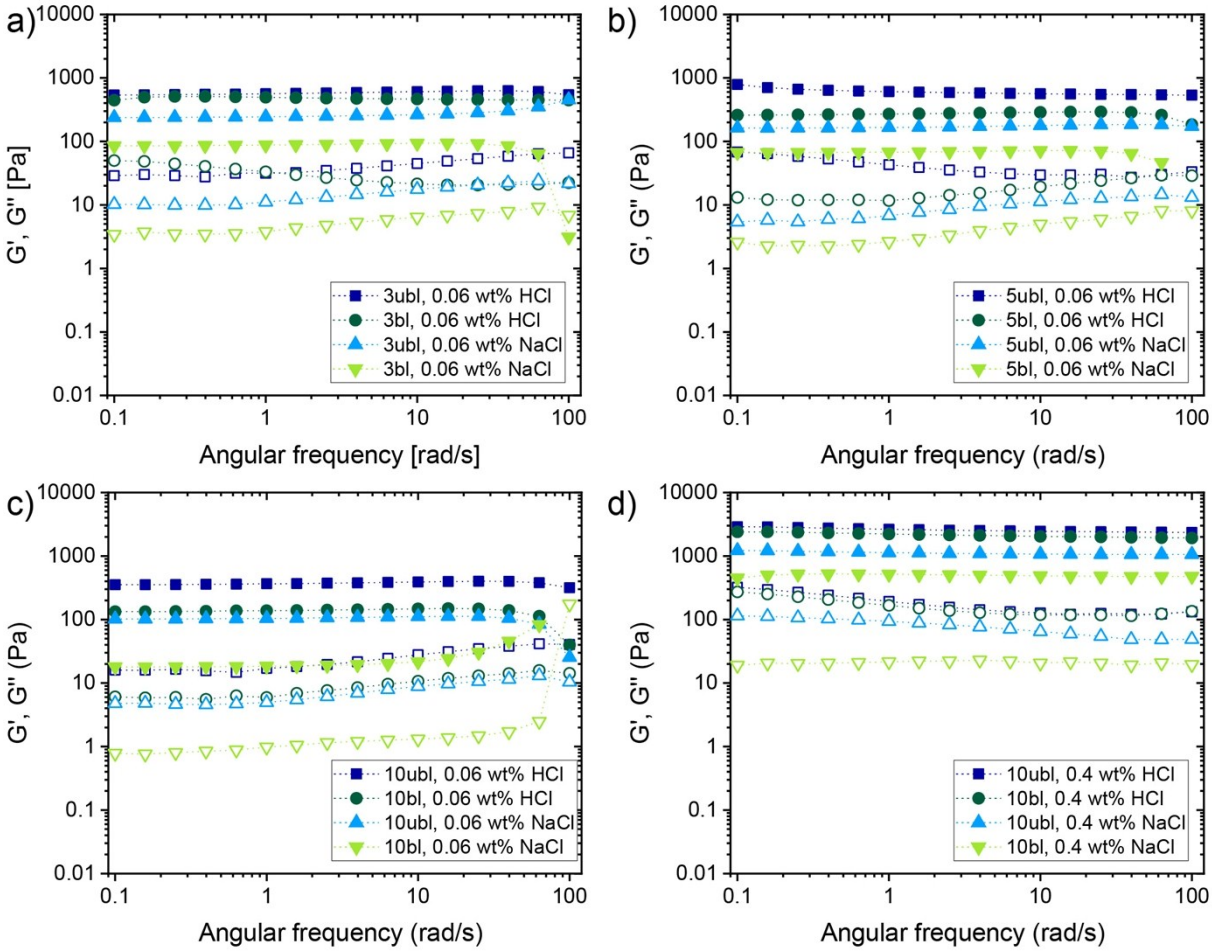

Figure S3. Elastic and viscous shear moduli across the measured angular frequency ω range of a) 3 mmol, b) 5 mmol and c) 10 mmol at 0.06 wt% CNF, and d) 10 mmol at 0.4 wt%.

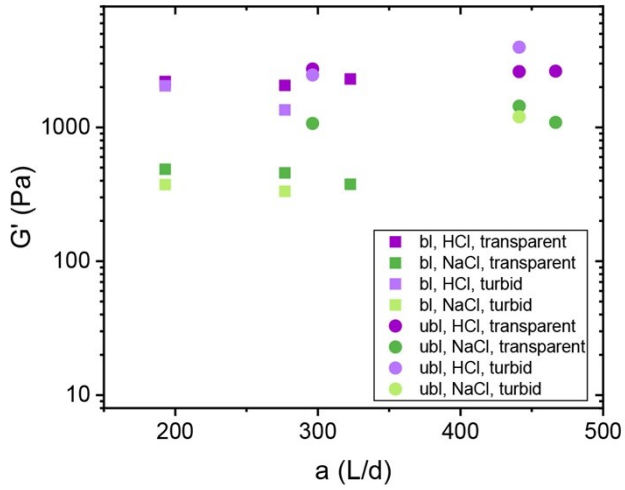

Figure S4. Comparison of elastic shear modulus G' depending on aspect ratio of transparent and turbid CNF dispersions.

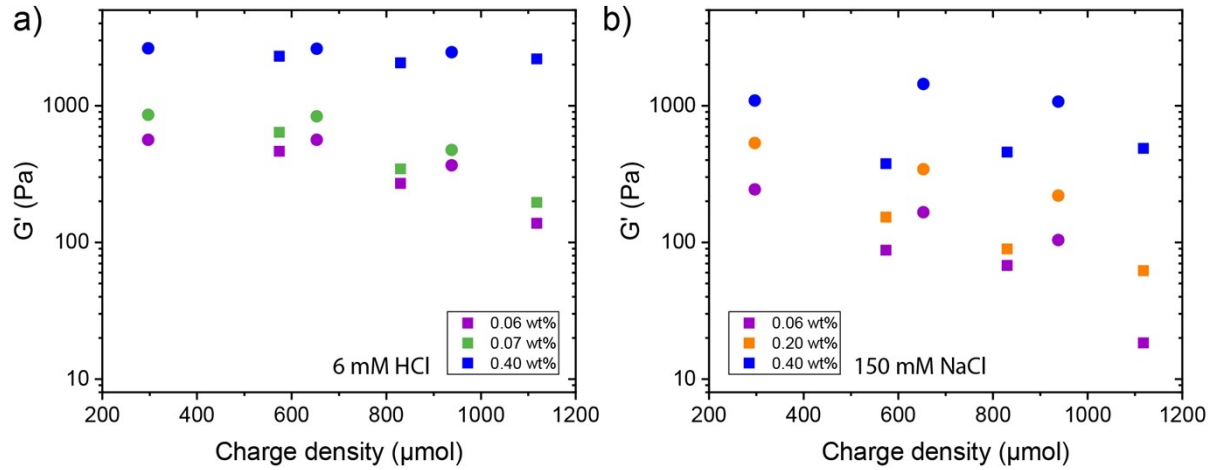

Figure S5. Dependency of the elastic shear modulus (G') on charge density of the gels at ω = 1 rad/s, γ = 0.5% and 20 °C for different concentrations after adding a) 6 mM HCl and b) 150 mM NaCl. The squares represent CNFs from bleached pulps, the circles unbleached, respectively.

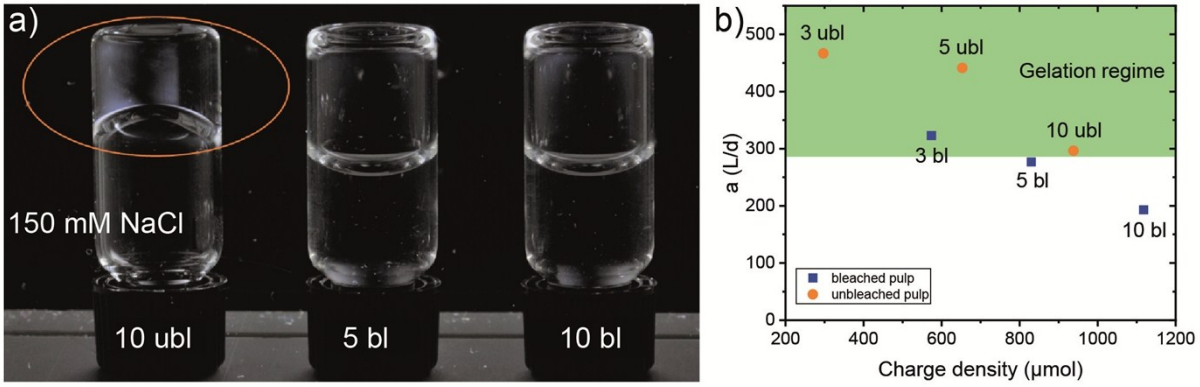

Figure S6. a) Inversion test and b) gelation dependency on aspect ratio as function of charge density of CNFs from bleached and unbleached pulp at 0.06 wt% due to addition of 150 mM NaCl.

Supplementary table

Table S1. Parameters obtained from AFM image analysis for all different CNF samples. Data adapted from ref 1 where the mean, standard deviation (stdD) and standard error (stdE) has been from sample sizes of n > 200.

Sample name	Mean length (nm)	stdD	stdE	Length weighted length (nm)	stdD	stdE	Inverse kink density	Average segment length (nm)	Number of kinks per fibril	Charge density (µmol/g)
3bl	292	283	11	567	395	15	239	132	1.22	570
5bl	272	242	14	486	323	19	257	132	1.06	830
10bl	204	166	8	339	214	10	340	127	0.60	1100
3ubl	454	408	23	819	548	31	253	163	1.79	230
5ubl	459	381	24	774	495	31	303	183	1.51	650
10ubl	268	261	17	520	363	24	411	162	0.65	940

References

1 M. Arcari, E. Zuccarella, R. Axelrod, J. Adamcik, A. Sánchez-Ferrer, R. Mezzenga and G. Nyström, *Biomacromolecules*, 2019, **20**, 1288–1296.