Supplementary information (SI) for

Ion Specific Effects on the Rheology of Cellulose Nanofibrils in the Presence of Salt

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Figure 1S. (A) Dynamic rheology in amplitude dependence at $\omega = 10$ rad/s and of neat CNF suspensions at concentrations indicated in the color bar (0.1 to 7 wt.%) (B) Frequency dependent dynamic rheology moduli measured at a strain amplitude, $\gamma = 5\%$ for neat CNF suspensions of different concentrations (0.1 to 4.5 wt.% as indicated by the color bar). The open and closed symbols are storage/elastic moduli (G') and loss/viscous moduli (G''), respectively. The color bars indicate the CNF concentrations. The 7wt.% CNF sample was omitted here due to the onset at the nonlinear viscoelastic regime (NLVR) beyond $\gamma = 5\%$, so the data was not included to ensure the elastic modulus in the LVR.



Figure 2S. Dynamic rheology of the chloride salts of monovalent cations in the CNF suspensions at 0.55wt.% (left) amplitude strain sweeps, (middle) frequency sweeps, and (right) Cole-Cole plots at (A) 50 mM, (B) 25 mM, and (C) 10 mM of the added salts.



Figure 3S. Dependence of (A) salt concentrations with varying NaCl concentrations and (B) ion enthalpy of hydration on zeta potential of the 0.01wt.% CNF suspensions. The top x-axis labels indicate the cations from the added monovalent chloride salts at 5 mM.



Figure 4S. Dependence of ion hydration of enthalpy on the CNF radius of gyration at 0.01wt.% CNF concentration. The error bars indicate 95% confidence intervals from the measurements. The top x-axis labels indicate the cations from the added monovalent chloride salts at 5 mM.

Table 1S. Salt concentration $({}^{c_s})$ or ionic strength (I), experimental intrinsic viscosity $([\eta])$ values from viscometric measurements, fitted persistence length $({}^{l_p})$ numbers from a combination of Eq. 4 and Eq. 5, and Kuhn length (b) calculated as $b = 2 \cdot l_{p_1}$ for the CNFs in dilute regime.

$c_{s}(mM)$	$[\eta]$	$l_p(nm)$	b (nm)
0.0	416	194	388
0.5	377	176	352
1.0	297	164	328
3.0	241	140	280
5.0	155	130	260

Table 2S. The radii of gyration $({}^{R}g)$ from the static light scattering (SLS) measurements of the dilute CNF suspensions without any additional salt and in the presence of counter ions at 5 mM. The enthalpy of hydration $({}^{\Delta H}{}^{\theta}{}_{hyd})$ are tabulated and obtained from W. E. Dasent's *Inorganic Energetics*.² The radius of gyration $({}^{R}g)$ was linearly fitted and obtained from the Guinier regime along with the 95% confidence intervals (CI) to provide all possible values of the fitted parameter.

Cations	$\Delta H_{hyd}^{\ heta}$	R_g (nm)	
	(kJ/mol)	Mean	95% CI
None	-	88	73-100
Li	-520	80	68-90
Na	-406	85	73-95
K	-320	89	77-99
Rb	-296	81	72-89
Cs	-264	75	60-88

References

(1) Rubinstein, M.; Colby, R. H. Polymer physics; Oxford University Press, 2003.

(2) Dasent, W. E. Inorganic energetics : an introduction; Cambridge University Press, 1970.