Electronic Supplementary Information

Controlled defibrillation of rice straw cellulose and self-assembly of cellulose nanofibrils into highly crystalline fibrous materials

Feng Jiang, Siyuan Han, You-Lo Hsieh*

Fiber and Polymer Science, University of California, Davis, CA95616, USA

Validation of Henry equation and Huckel approximation for zeta potential measurement

The zeta potential (ζ) is determined from the electrophoretic mobility of the particles using Henry equation,

$$M = \frac{\varepsilon\zeta}{\eta} f(\kappa a)$$

where M is the electrophoretic mobility, η and ε are viscosity and dielectric constant of the dispersion media. f(ka) is Henry's function, which equals to 1 (Smoluchowski approximation) when ka>>1 and 2/3 (Huckel approximation) when ka<<1. k is the inverse of Debye length and a is the fibril radius. It has been previously shown that the Henry equation holds for both spherical and particles with higher aspect ratios.[1]

The Debye lengths (k^{-1}) are 9.66 and 966 nm for aqueous solution with ionic strength of 10^{-3} M and 10^{-7} M (pure water), respectively. As no salt was added during the experiments, although the surface carboxyl (maximum 1.3 mM based on the conductometric titration values) would dissociate partially in the solution to increase the ionic strength, the Debye length should fall between 9.66 to 966 nm, much higher than the major fibril radius (less than 10 nm). Therefore, Huckel approximation with f(ka)=2/3 should be used to calculate the zeta potentials of CNFs.

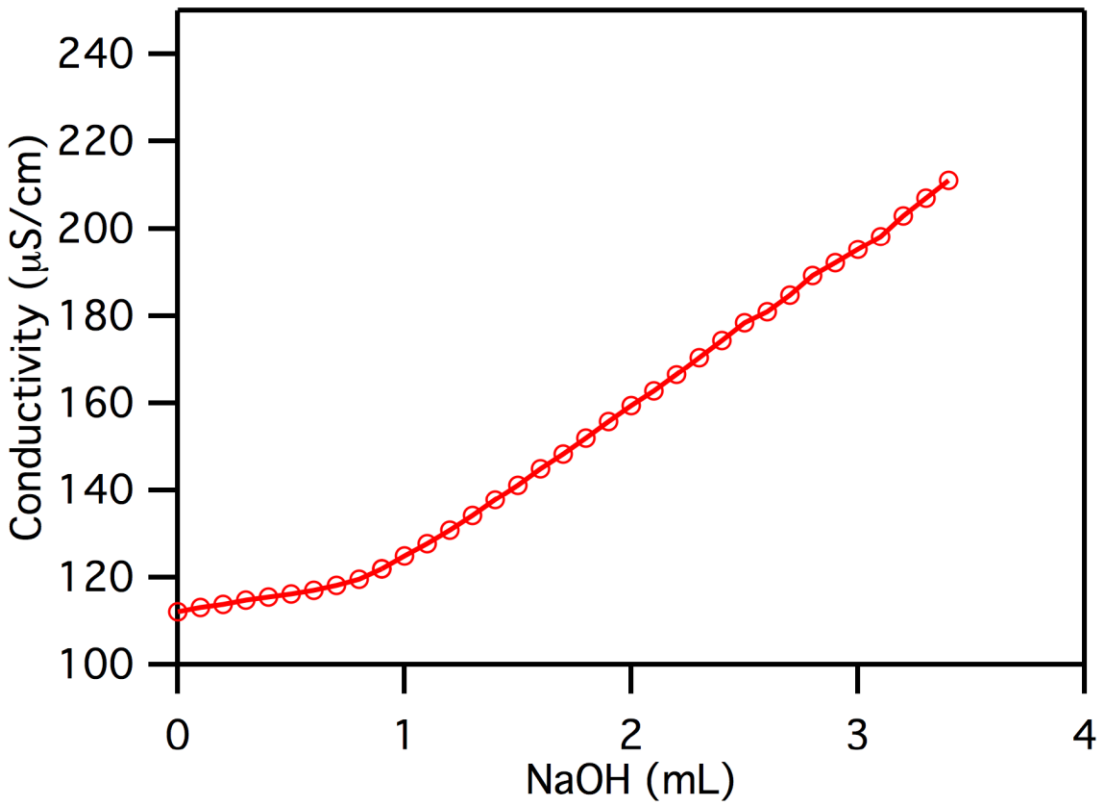
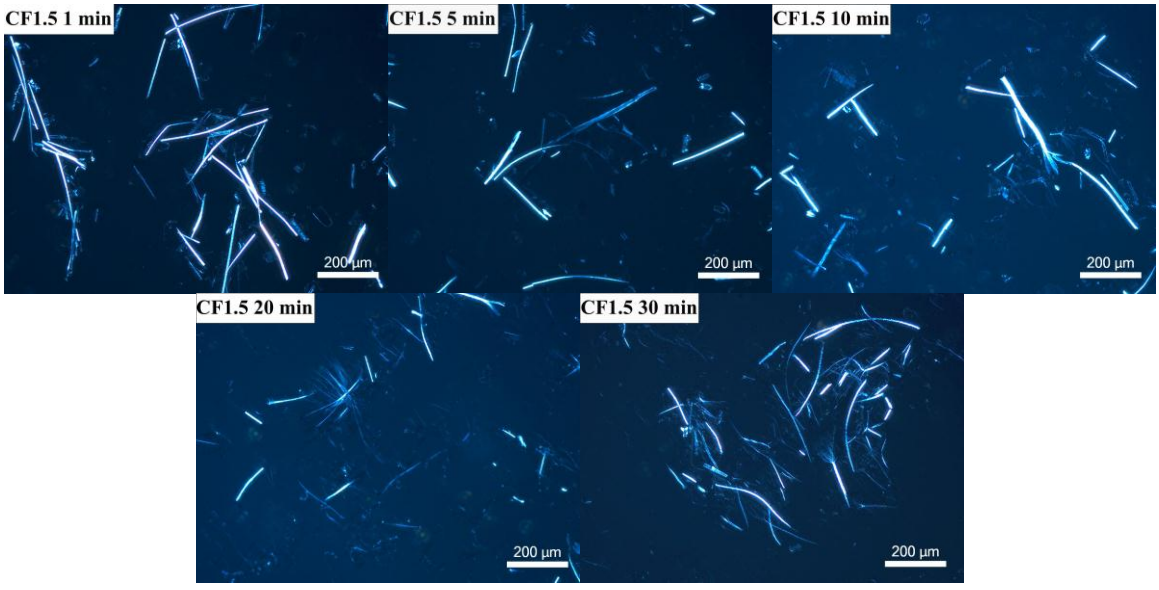


Figure S1. Conductometric titration curve of TEMPO oxidized cellulose at 5 mmol NaClO/g of cellulose without adding HCl.



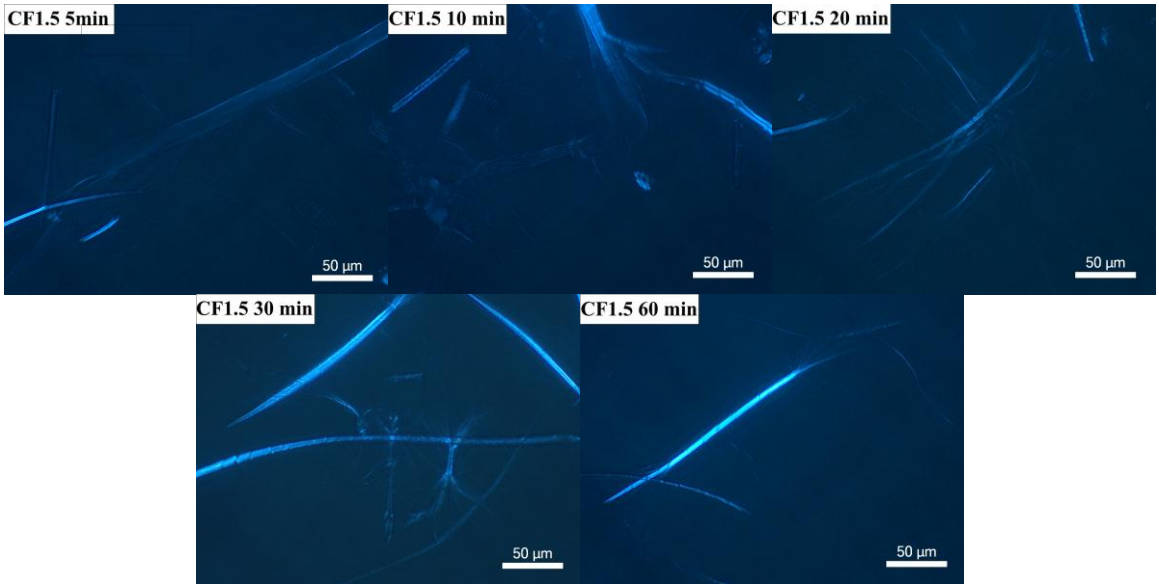


Figure S2. Optical microscopic images under cross polarizer of the precipitates from CF1.5 after various blending times

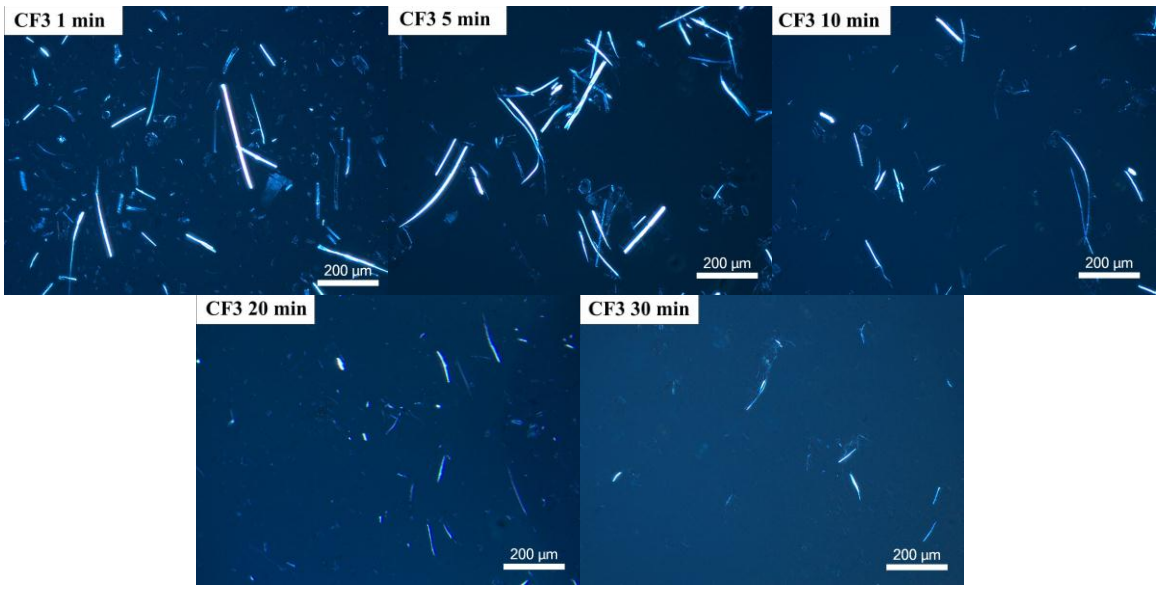


Figure S3. Optical microscopic images under cross polarizer of the precipitates from CF3 after various blending times

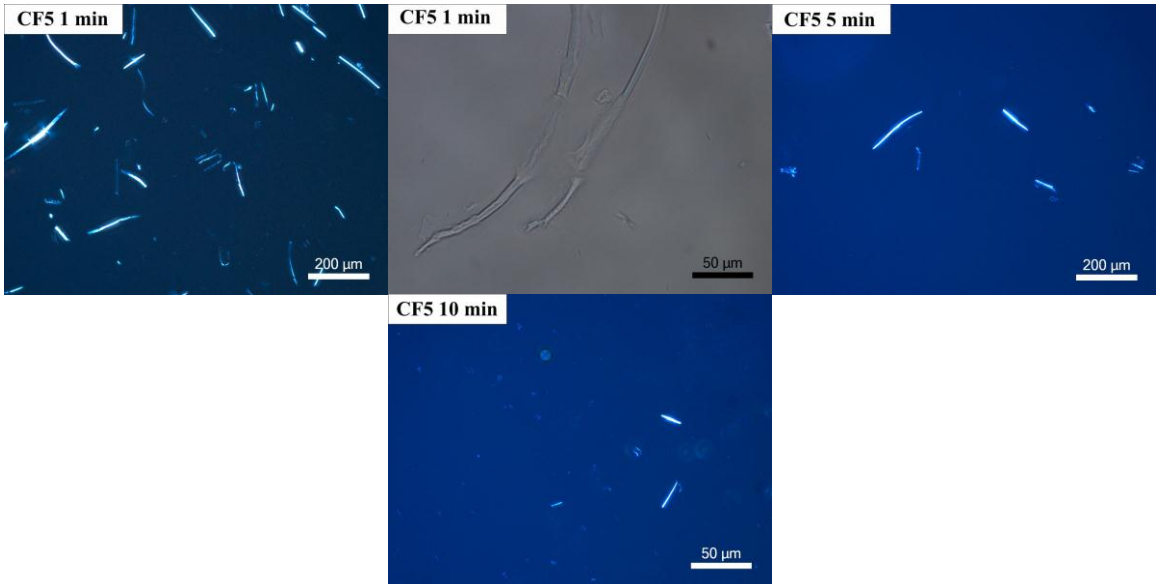


Figure S4. Optical microscopic images under cross polarizer of the precipitates from CF5 after various blending times

Table S1. Crystallite dimensions and calculated surface C6 primary hydroxyls for rice straw cellulose and CNFs according to eq. 3&4 from the deconvolution of XRD spectrum.

Sample		Cellulose	CNF1.5	CNF3	CNF5
Crystallite	$1\bar{1}0$	2.98	2.61	2.46	2.71
dimension	110	3.39	3.38	2.83	2.47
(nm)	Average	3.19	3.00	2.65	2.59
Surface C6 primary hydroxyls (OH/AG)		0.257	0.268	0.291	0.296

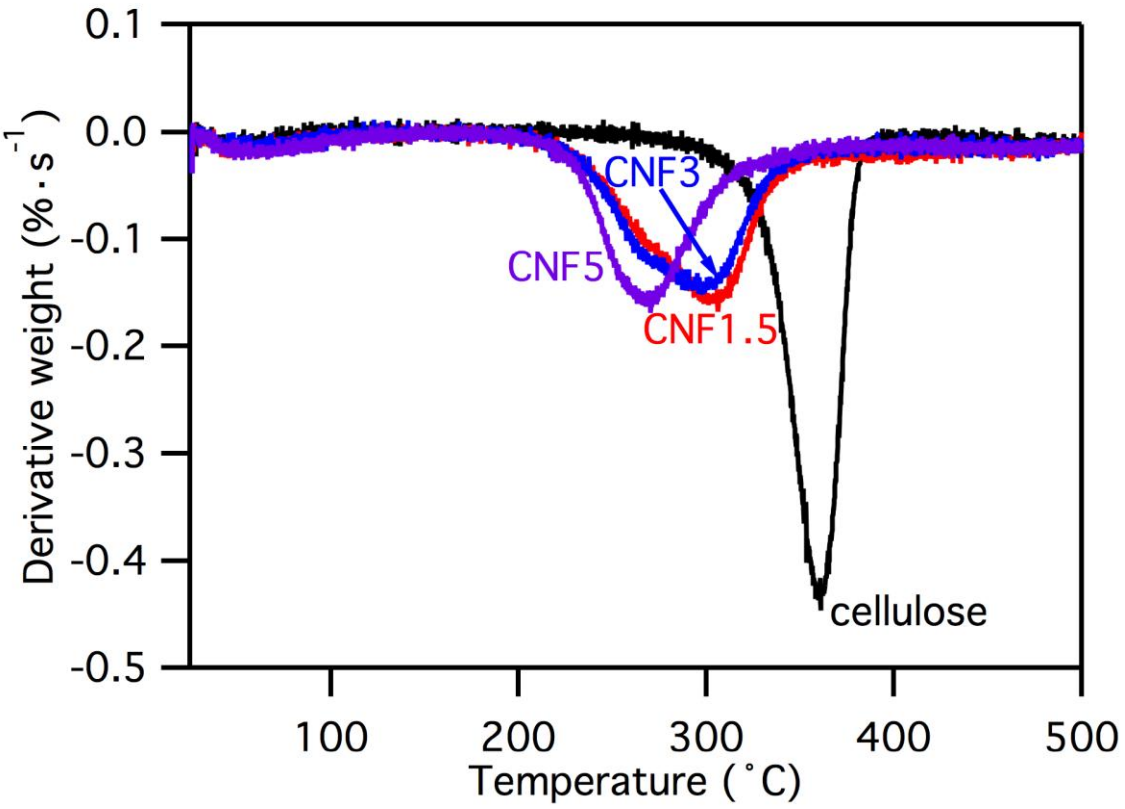


Figure S5. DTG curves of rice straw cellulose (black) and freeze-dried TEMPO oxidized and defibrillated CNF1.5 (red), CNF3 (blue) and CNF5 (purple).