Supporting Information

On the Roles of Cellulose Nanocrystals in Fiber Cement: Implications for Rheology, Hydration Kinetics, and Mechanical Properties

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Content (6 pages, 4 figures, 5 tables)

This supporting information (6 pages) presents a set of figures (Figures S1 – S4) and tables (Tables S1 – S5) to show the information discussed in the manuscript.

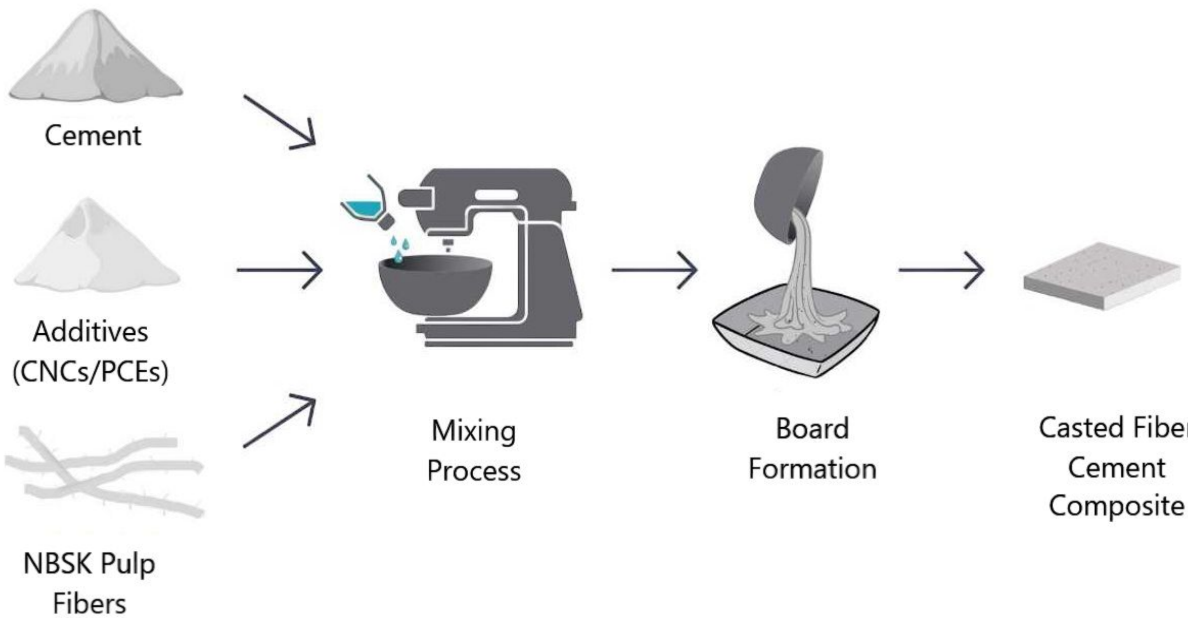

Figure S1. Schematic illustration of the fiber-reinforced cement composites containing additives (CNCs/PCE).

Material Properties

Table S1. The refined fiber properties of NBSK pulp used for this research.

The bleached pulp fibers (NBSK from Canfor, Canada) were soaked in 'RO' water for 8 hours and then passed through a disintegrator for 600 revolutions. The disintegrated pulp was then dewatered and subjected to refining by feeding them into "Pulp and Fiber Research Institute" mill, commonly denoted as PFI mill.[1] After that the fiber properties were measured using a Fiber Quality Analyzers (FQA) from OpTest, Canada (FQA–360)

Refined fiber properties	Dimension (μm)
Fiber length (L)	1386
Fiber width (W)	27.7

Aspect ratio (L/W)	50

Table S2. The physical properties of cellulose nanocrystals (CNCs) produced *via* sulfuric acid hydrolysis and spray dried.

Appearance	White
Sulfur content (in dry CNCs)	0.85 wt% Sulfur
Solid content	98 % dry powder
Dimension (nm)	150 –200 (L) 5 – 20 (W)
Aspect ratio (L/W)	10–30

Table S3. Physical properties of the commercial (Sigmal Aldrich, USA) polycarboxylate (PCEs) based superplasticizer (a petrochemical-based additive)

Appearance	White to off-white
M_W (kDa)	520
M_n (kDa)	150

Table S4. Power law fit (shear-thinning regime) parameters based on rheological characterization (viscosity versus shear rate) of the fiber-cement slurry (non-Newtonian fluids) containing additives (CNCs/PCEs).

Additive content (wt%)	Consistency Coefficient '*k*' (CNCs)	Flow index '*n*' (CNCs)	R^2 Fit (CNCs)	Consistency Coefficient '*k*' (PCEs)	Flow index '*n*' (PCEs)	R^2 Fit (PCEs)
0	3.424	0.5608	0.9985	3.424	0.5608	0.9985
0.02	4.838	0.5388	0.9989	4.568	0.553	0.995
0.06	4.995	0.5272	0.9931	4.356	0.5984	0.9972
0.2	6.747	0.519	0.9975	7.606	0.6189	0.9973
0.6	8.195	0.5127	0.9996	8.61	0.6293	0.9978
1	11.29	0.4958	0.9945	7.072	0.6383	0.9998
2	53.45	0.2789	0.9962	8.613	0.6586	0.9998
3	107.4	0.184	0.9189	7.731	0.6819	0.9991
4	113.2	0.1787	0.9554	16.53	0.7187	0.9998

Rheological Characterization (Ordinary Portland Cement Paste)

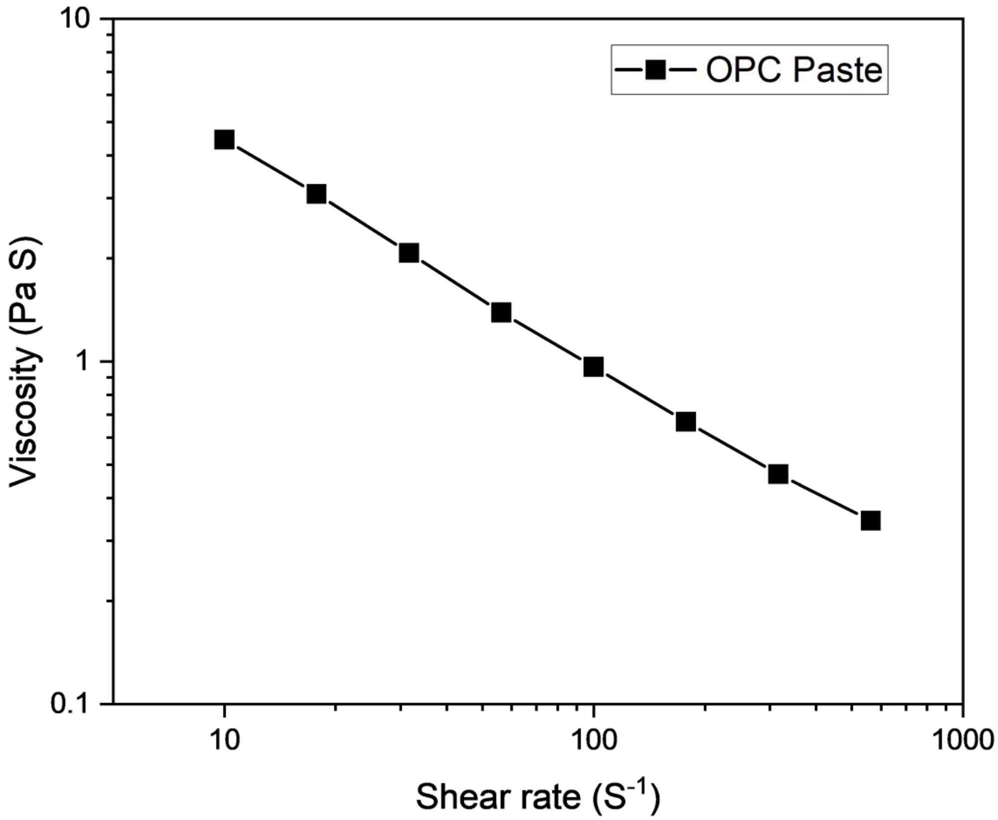

Figure S2. Rheological characterization of Ordinary Portland Cement (OPC) pastes without any reinforcing bleached fiber and additive. Variation of viscosity as a function of shear rate. Note that a water/cement ratio of 0.5 (used in this study) was employed for testing the steady state viscometry of OPC paste.

In general, cement paste does exhibit both shear thinning and shear thickening behavior. However, the extent of this behavior is dependent on the cement composition, which includes C_2S (Belite), C_3S (Alite), Calcium sulfates, Gypsum, C_3A and C_4AF, to name a few, and process parameters, which includes, water to cement ratio, mixing time, and curing time.[2]

It is important to note that in the compositional paradigm, Gypsum is an important component in OPC, that is added to delay cement setting. However, in the absence of Gypsum cement would rapidly set, causing excessive cracking and loss of strength to the cured cementitious material. Furthermore, excessive gypsum could cause shear thickening behavior in cement pastes due to the formation of a large amount of plaster set, due to the presence of anhydrite gypsum in the cement and a higher water-cement will free up the spaces between cement particles hence contribute to shear thinning effects.[2]

Regarding the process parameter, for water-to-cement ratio of 0.5 (used in this study) the viscosity vs shear rate plot for cement pastes does indicate a shear thinning. However, the addition of pulp fibers increases the "pot life" of the fiber cement system, which means keeping the viscosities of the fiber cement paste in a usable regime (prevent the rapid increase of viscosities with time). In other words, it increases the handling time of the fiber cement pastes.

In addition to this, the shear thinning effect of the cement pastes is improved with the addition of pulp fibers as fiber flocks tend to break down and align in the direction of shear, resulting in decreased viscosity with the increase in shear rate.[3]

Mechanical Characterization

Table S5. Mechanical characterization of cured cementitious sample based on the 3-point bending tests. Calculated modulus of rupture (MOR) values of the OPC and fiber cement board (thickness: ca. 8 mm). OPC board does not contain any reinforcing pulp fibers and additives while the FC board contain 8 wt% of pulp fiber (reinforcement) and 5 wt% CNCs (additive).

Cured Sample (cement board)	Modulus of Rupture (MOR)	Standard Deviation (S.D)
OPC (without reinforcement and additive)	4.8	0.81
FC_CNCs(*5*)wt% (reinforced and with additive)	5.38	0.998

Microstructural Chareceterization

Field Emission Scanning Electron Microscopy (FE–SEM)

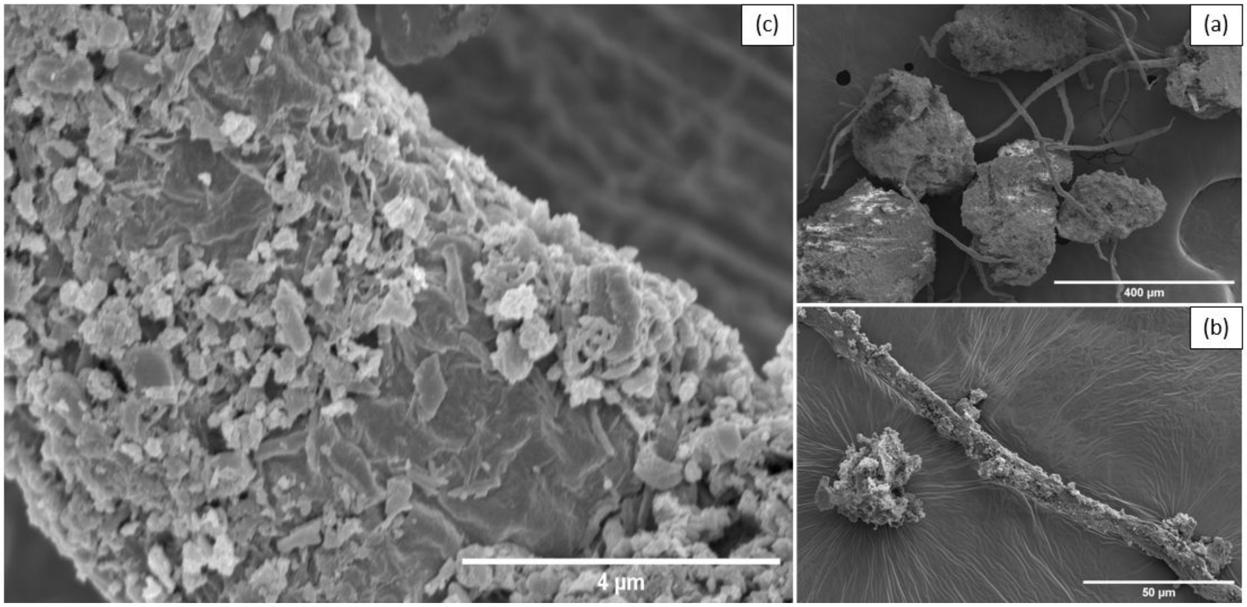

Figure S3. FE–SEM micrographs of fiber–cement interface (from low to high magnification (Figure S3 (a-c)), representing the fiber-cement interfaces containing CNCs.

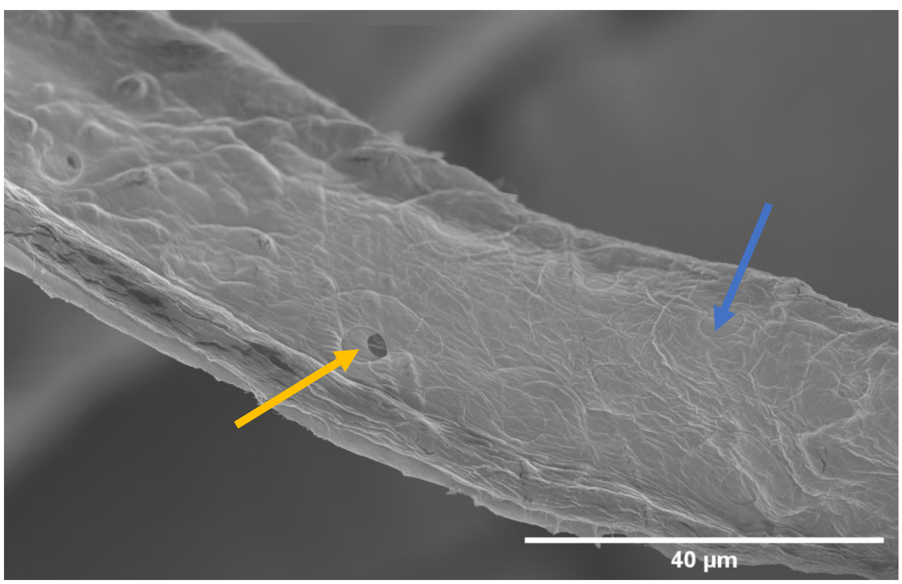

Figure S4. FE–SEM micrograph of fiber–CNCs interface. The image depicts voids (pores) filling effect (indicated by the blue arrow) of CNCs deposited to the bleached pulp fiber. The unfilled void (pore) of the pulp fibers is indicated by the yellow arrow.

References:

(1) Gharehkhani, S.; Sadeghinezhad, E.; Kazi, S. N.; Yarmand, H.; Badarudin, A.; Safaei, M. R.; Zubir, M. N. M. Basic Effects of Pulp Refining on Fiber Properties—A Review. *Carbohydr Polym* **2015**, *115*, 785–803. https://doi.org/10.1016/J.CARBPOL.2014.08.047.

(2) Greenberg, S. A.; Meyer, L. M. Rheology of Fresh Portland Cement Pastes Influence of Calcium Sulfates.

(3) Derakhshandeh, B.; Kerekes, R. J.; Hatzikiriakos, S. G.; Bennington, C. P. J. Rheology of Pulp Fibre Suspensions: A Critical Review. *Chem Eng Sci* **2011**, *66* (15), 3460–3470. https://doi.org/10.1016/J.CES.2011.04.017.