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The influence of cellulose nanocrystals on the microstructure of cement paste

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5 Abstract

6 This paper reports the influence of raw and sonicated cellulose nanocrystals (CNCs) on the microstructure 7 of cement paste. A novel centrifugation method is designed to measure the concentrations of the adsorbed 8 CNCs (aCNCs) on the cement surface, and the free CNCs (fCNCs) which are mobile in water. It is found 9 that, the majority of the CNCs (>94%) are aCNCs. More importantly, sonication does not significantly 10 reduce the amount of aCNCs (reduction of less than 2%). We surmise that, after sonication, the aCNCs 11 are primarily dispersed over the cement surface, instead of becoming fCNCs via sonication. Isothermal 12 calorimetry and energy-dispersive X-ray spectroscopy (EDX) results support this theory. The water 13 desorption tests show that the total porosities of cement pastes with raw and sonicated CNCs are 14.8% 14 and 14.4%, which showed a reduction from 16% for the plain cement paste. The porosity reduction is a 15 result of an increase in the degree of hydration. The advantage of sonicated CNCs is they are dispersed, 16 avoiding therefore agglomerates that can lead to pores, voids, and air entrapment. The nanoindentation 17 results show that the reduced indentation modulus on the interfacial regions between cement particles and 18 the low density CSH is increased when CNCs are used.

19

20 Keywords

21 cellulose nanocrystal, cement, agglomeration, ultrasonication, flexural strength, nanoindentation

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23 **1. Introduction**

24 Cellulose nanocrystal (CNC) is the crystalline part of the cellulose materials, which can be extracted from 25 trees and a variety of plants [1]. As a reinforcement material, CNC has a few extraordinary advantages, 26 including biodegradability, high abundance, low cost, and excellent mechanical properties [1]. Recently, 27 CNCs have been found to improve the flexural strength of cement composites due to an increase in the 28 degree of hydration (DOH) [2] which is possibly aided by a mechanism referred to as short circuit 29 diffusion (SCD). The basic prerequisite for SCD is the adsorption of CNCs on the surface of cement 30 particles, acting as the pathway to preferentially transport water from pores to unhydrated cement cores. 31 Such capability can be attributed to the high hydrophilicity and hygroscopicity of the CNCs [3]. A 32 competing mechanism has also been identified for CNC concentrations larger than 0.2 vol. % where 33 agglomerates induce stress concentrators limiting the strength of the cement pastes [2]. This issue was

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