Performance-Enhanced Cementitious Materials by Cellulose Nanocrystal Additions

Abstract. *In this work, we study the effect of cellulose nanocrystals (CNCs) when they are added into cement paste. Our work encompasses a combined mechanical, microstructural, and chemical investigation of various aspects of CNC/cement mixtures. Results show that CNC additions can increase the flexural strength of cements. Current efforts are focused on understanding the mechanism of increased flexural strength in CNC/cement.*

Keywords. Flexural strength, cement, cellulose nanocrystals.

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Figure 1. *Photograph of a 5-cm-diameter CNC-cement sample produced for the study, from which small test specimens were cut for analytical and mechanical testing.*

Introduction. A quest is underway to develop a new generation of high-performance, multifunctional, renewable, and sustainable infrastructure materials such as cement and concrete to make a radical break with traditional engineering paradigms. On the one hand, nanomaterials offer remarkable opportunities to tailor mechanical, chemical, and electrical properties. On the other hand, intense research in the use of nanomaterials has been criticized due to perceived environmental, cost, health, and safety issues. Currently, there is a growing push for "greener" products, which includes materials made from renewable and sustainable resources. In addition, there is a goal to minimize the carbon footprint of infrastructure materials, which drives interest in biodegradable, non-petroleum-based, low-environmental-impact materials. By increasing the performance of infrastructure materials, it may be possible to reduce greatly the volume of these materials that are needed, thereby reducing the demand for raw materials. The use of higher-performance materials is one way to "do more with less".

Reinforcing brittle cement matrices has always been a challenge. Many reinforcing materials degrade or are difficult to add to the mixture in sufficient volume without causing difficulties in mixing. Furthermore, the costs of the reinforcing materials must always be considered. Cellulose fibers (wood or plant fibers) have been used in cementitious systems to control cracking in fresh systems, to control sound absorption, or to reduce weight and increase the utility of the finished composite (e.g., Hardie plank); however, they generally do not show improvements in toughness [1]. In contrast, cellulose nanomaterials, cellulose nanocrystals (CNC) or cellulose nanofibrils (CNF), offer new possibilities for endowing cementitious composites with improved mechanical performance. Their small size can allow a smaller interfiber spacing and more interactions between the cellulose and the cement system, and as a result, the CNC and CNF have a greater potential to combat microcracking and increase the strength of the system. CNCs are a promising family of nanoscale reinforcing materials for cements in that they have several unique characteristics, such as high aspect ratio, elastic modulus, and strength, low density, and reactive surfaces that enable easy functionalization and are readily water-dispersible without the use of surfactants or modification. In addition, CNCs are extracted from sources that are themselves sustainable, biodegradable, and carbon-neutral, and the extraction processes have low environmental, health, and safety risks. To the best of our knowledge, this is the first study to investigate

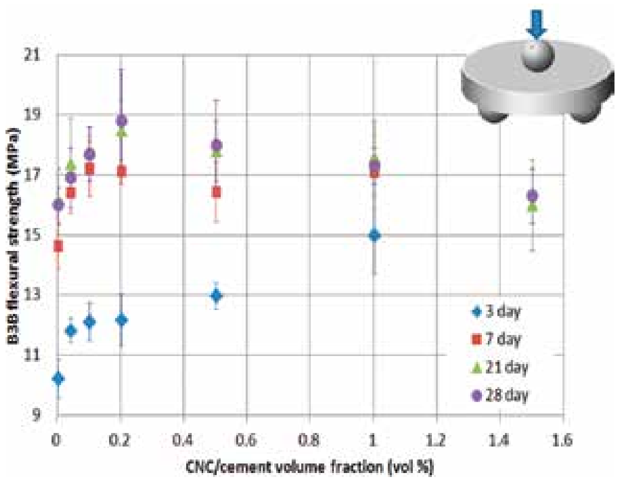

Figure 2. *B3B flexural strengths of CNC-reinforced cement pastes at four different ages*

CNCs in cements. For instance, previous studies have investigated carbon nanotube-reinforced cement composites because of their high aspect ratio and associated capability to bridge nanocracks and increase the strength of cement [2].

We use an integrated approach that combines material preparation, experiments, modeling, and state-of-the-art microscopy to unravel the physical mechanisms that underpin CNC nanomechanics and to relate these to the macroscopic properties of CNC-reinforced cementitious materials.

Methodology. CNC-reinforced cementitious samples were produced by mixing CNC solution (5 wt% CNC aqueous suspension provided by the USFS-Forest Products Laboratory) and Type V cement, maintaining a water/ cement ratio of 0.35 to obtain cement pastes with different CNC concentrations (0 to 1.5 vol% CNCs). Type V cement was chosen because of its compositional purity (i.e., low aluminates and ferrite phases). The cement pastes are prepared by a vacuum mixer at 400 rpm to reduce air entrainment and water evaporation. The mixtures are then cast in plastic cylinders for further curing, producing cylindrical samples as shown in Fig. 1. Sample testing can be grouped into three general categories: (1) the curing process, (2) the microstructure, and (3) mechanical properties. The curing process was characterized using calorimetry and thermogravimetric analysis (TGA) to evaluate the evolution of the degree of hydration (DOH) and to assess the effect of CNC on hydration rate. The microstructure was analyzed by optical and scanning electron microscopy (SEM). The flexural strength at different ages was measured using the ball-on-three-ball method (B3B). The flexural strength was calculated using the closed-form equations given in [3]. The mechanical properties were measured at different ages (7, 14, and 28 days), and at different CNC contents.

Results. Cellulose nanocrystals (CNCs) could be uniformly mixed within cement pastes. Preliminary results show that CNC addition to cement pastes can alter the DOH and can increase the flexural strength. Figure 2 shows the B3B flexural strengths of cement pastes with different volume fractions of CNCs at four different ages: 3, 7, 21, and 28 days. At three days, the strength increases with CNC content. For the other three ages, the strength seems to have an optimum at approximately 0.2 vol.% CNC. For the 28-day aged sample, there was a ~20% increase in flexural strength compared to the neat cement pastes.

Conclusions. Cellulose nanocrystals (CNCs) can be uniformly mixed within cement pastes and were found to alter the degree of hydration and flexural strength of cement pastes. Further details of this investigation can soon be found in [4].

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