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Research Paper

Reinforcement of natural fiber yarns by cellulose nanomaterials: A multi-scale study

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ABSTRACT

Cellulose nanofibrils (CNF) and cellulose nanocrystals (CNC) were used to improve the mechanical properties of tapes and yarns produced from natural fibers as a new application of cellulose nanomaterials in textile-based composite products. Hemp and flax slivers were used as natural fibers for the production of yarns and tapes. CNC, unground CNF and two different ground CNF suspensions were used as the reinforcement agents. Fiber strands from each natural fiber were soaked in the cellulose nanomaterial suspension and then were processed into tapes or yarns. Individual hemp and flax fibers where also soaked in the same suspensions and dried. SEM microscopy of the surfaces and cross-sections of the yarns and tapes showed that cellulose nanomaterial suspensions affected the morphology of the natural fiber yarns and tapes by filling gaps and adhering the fibers together. Results of studies on tensile properties of single fibers showed improvement in initial modulus and strength of flax fibers after soaking in nanocellulose suspensions especially when CNF was used. Such consistent results were not however seen for hemp single fibers. Yarns and tapes produced by soaking fiber strands in different nanocellulose suspensions had considerably higher tensile properties in terms of strength and initial modulus than controls. Furthermore, it was shown that mixing cellulose nanomaterial suspensions with natural fibers improved the dewatering/drying process, a necessary step should cellulose nanomaterials be used in composite applications.

1. Introduction

Cellulose nanomaterials are a relatively new group of lignocellulose resource-based materials. These materials, which are directly derived from wood or other lignocellulosic feedstocks are a promising class of cellulosic products and are mainly classified into two major groups of cellulose nanofibrils (CNF) and cellulose nanocrystals (CNC) (Turbak et al., 1983). Nanomaterials are used in different industries such as electronics, packaging, medical, and personal care product industries. The key leading point of these materials is their vast range of applications (LLP, 2016) as well as the possibility to impart interesting functionalities and properties at low weight or volume loadings.

CNFs are generally extracted from delignified wood pulp or other lignocellulosic resources as raw material by means of high shearing mechanical attrition in the form of low consistency aqueous nanofibril suspensions. There are several methods to produce CNF including high pressure homogenization, microfluidization, TEMPO oxidation and refining (Dufresne, 2013; Revol et al., 1994; Turbak et al., 1983).CNF is normally produced as a suspension of 97% water and 3% cellulose nanofibrils. This material has great mechanical properties because of its high strength and large aspect ratio (Dufresne, 2012; Dufresne, 2013; Moon et al., 2011). The CNF aspect ratio is highly dependent on the grinding time; the more CNF is ground the higher the aspect ratio of the nanofibrils in the suspension will be and the higher the aspect ratio of the CNF, the higher the surface area (Ghasemi et al., 2017; Moon et al., 2011; Nechyporchuk et al., 2016). These very strong nanofibrils have the potential to be made into strong continuous filament structures. For this purpose, two key issues should be addressed. First, a method should be proposed for removing more than 97% water from the highly hydrophilic CNF and second, the “spaghetti–like” nanofibrils should be aligned unidirectionally and agglomeration should be prevented (Peng et al., 2012; Voronova et al., 2012).

Cellulose nanocrystals on the other hand, are mainly produced by acid (often sulfuric acid) hydrolysis of wood pulp and are highly crystalline suspensions with smaller particle sizes and narrower particle size distributions. High crystallinity, high surface area and stiffness, very low thermal expansion, and many other exceptional properties give CNCs the potential to be used in a variety of applications (Moon et al.,

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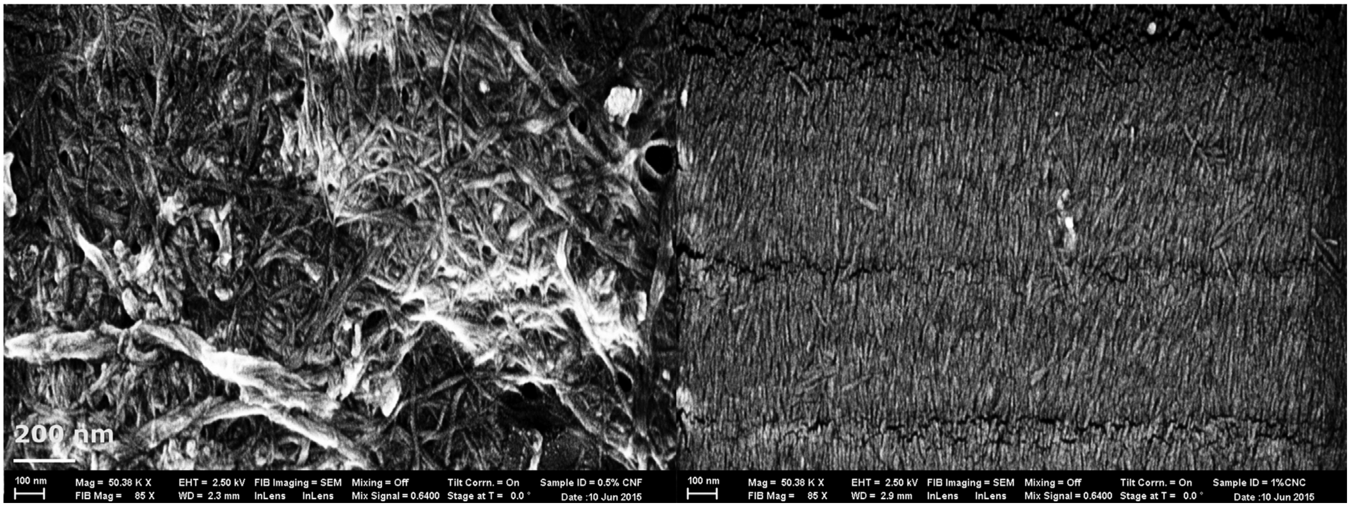

Fig. 1. Surface morphology of a CNF (left) and CNC (right) films showing differences in morphologies.

2011). The negative surface charge, which is resulted by the chemical procedure used during production helps with better dispersibility and abundant hydroxyl groups in the structure provide opportunities for chemical modification (Peng et al., 2012). Fig. 1 shows scanning electron microscopy (SEM) micrographs of the surface of a typical CNF and CNC film where morphologic differences are readily evident.

Although, cellulose nanomaterials have the potential to be used in a wide range of products, presently very few of the proposed applications have been scaled-up to commercialization. Cellulose nanomaterials are produced in the form of low consistency aqueous suspensions with an average water content of ≥90%. Thus, the first requirement for many of these applications is to find a feasible and economical way to remove the water while maintaining the nano-scale dimensions of the material. Many of the proposed methods to dry cellulose nanomaterials are either too expensive to be used for large scale production or are unable to dry the material without causing further agglomeration of nanoparticles (George and Sabapathi, 2015; Moon et al., 2011; Xu et al., 2013). Applications that can target using cellulose nanomaterials in their natural aqueous state by skipping the initial drying process are one important step closer to commercialization.

Over the past two decades, growing attention to environmental conservation and use of abundant biodegradable resources has led to the introduction of a class of products made from natural fibers in different applications mainly as short fiber reinforcement in polymeric systems (Bledzki and Gassan, 1999; Eichhorn et al., 2001; Hinchcliffe et al., 2016; Kim et al., 2012; LLP, 2016). Using natural fibers in place of synthetic ones is a great step towards the production of biodegradable products and brings about significant environmental benefits.

Natural fibers have been traditionally used for various applications including those in the form of yarns. One of the main parameters restricting the utilization of yarns produced from natural fibers in most applications is their low mechanical properties compared to that of the stronger yarns such as nylon, Kevlar, carbon or glass (Taha and Ziegmann, 2006; Yu et al., 2006). Reinforcement of yarns by applying different treatments for improving properties specifically the strength of the yarn has been the focus of a number of recent research (Robles et al., 2015; Pickering and Aruan Efendy, 2016; Wambua et al., 2003). For example, a cyclic loading treatment for ramie yarns showed improvements in strength and stiffness of these yarns through increasing crystallinity and imparting orientation (Zhu and Goda, 2008). Some improvement in the mechanical properties of natural fiber yarns after applying winding and heat setting were also reported (Zhu et al., 2010). In addition, alkali treatment has been shown to improve tensile properties of yarns produced from natural fibers (Bledzki and Gassan, 1999).

CNF and CNC are 100% cellulose nanomaterial suspensions that have the potential to be used for production of filaments or yarns as the basic element for producing textile structures and composites. Different works have reported nanocellulose applications in textile industry (Hebeish et al., 2016; Minko et al., 2016). There are also recent publications on the production of continuous filaments from nanocellulose using wet-spinning (Hakansson et al., 2014; Iwamoto et al., 2011; Lundahl et al., 2016) and dry-spinning methods (Ghasemi et al., 2017; Hooshmand et al., 2017; Hooshmand et al., 2015; Hooshmand et al., 2014). Nanocellulose also has the potential to enhance mechanical properties of natural fiber yarns. The high amount of water in the suspensions however is regarded as a limiting factor. Interestingly, because of the abundance of hydroxyl groups on the cellulose nanomaterial surfaces, strong hydrogen bonding is expected to form when they are mixed with other polar materials leading to considerable water release in a phenomenon recently reported by (Tajvidi et al., 2016). In this phenomenon known as contact dewatering, it is speculated that hydroxyl groups on the surface of cellulose nanomaterials will form hydrogen bonding with cellulose in the natural fibers, thereby releasing a significant amount of the water from the cellulose nanomaterial suspension. Since natural fibers such as hemp and flax have similar chemical compositions (hemp 68% cellulose, flax 71% cellulose) (Dufresne, 2013), it is expected that a similar phenomenon would take place when these fibers are in contact with cellulose nanomaterials. This will be a significant advantage to enable using wet CNF or CNC in a final product and maintain the nanoscale dimensions while eliminating the initial drying process needed for many composite applications.

Knowing that cellulose nanomaterials possess high bonding and mechanical properties, their addition to the structure of natural fiber yarns may enhance mechanical properties of such yarns. In this work, the influence of different types of cellulose nanomaterials on tensile properties of natural fiber yarns as well as their effect on dewatering and drying process of cellulose nanomaterial suspensions when in contact with natural fibers is presented. Tapes were produced for better understanding of interactions between nanocellulose and natural fibers as well as comparing the influence of the presence of twist during yarn spinning on the final properties of the structure. This is a new application of cellulose nanomaterials, which once optimized will provide opportunities for production of textile-based composites with improved mechanical properties. This work is the first attempt to study the reinforcing influence of nanocellulose on yarns and tapes produced from natural fibers.