# Facts and Myths about Cellulose Nanofibrils (CNF): What to Expect and What to Not

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## ABSTRACT

This paper provides some facts and myths around cellulose nanofibrils (CNF), its properties and applications and tries to present the "truth" about what to expect from CNF and what to not. Discussions on mechanical properties, processability, water interactions and adhesive properties will be presented. Recent development in the binder application of cellulose nanomaterials, laminate systems and applications in textiles and yarns will be discussed. Future outlook into the potential areas of focus for commercialization will be presented.

***Keywords***: cellulose nanofibrils, applications, properties, binder

## 1 INTRODUCTION

Cellulose nanofibrils (CNF) are perhaps the only type of cellulose nanomaterials that have the potential for both large volume production and large volume applications in near future. Much research has been conducted on the production, property assessment and applications of these interesting materials in many laboratories and the promise for an outstanding, multi-functional renewable material has been widely advertised. While a considerable amount of effort has been made to modify CNF to make it work with systems that it inherently does not interact with much, an alternative approach towards taking advantage of the natural strengths of CNF for applications it is comfortable with seems to be finding traction in the scientific community. Concurrently, research towards using CNF as the bulk material and not as an additive is finding more and more attention for biomedical applications. In parallel, CNF is often confused with other types of cellulose nanomaterials resulting in expectations that cannot be met in practice. Therefore, the ultimate objective of this paper is to provide potential areas of predicted "success" in the use of CNF.

For the purpose of this paper, CNF is defined as the product of mechanical fibrillation of bleached wood pulp by a grinder/refiner with minimum chemical pretreatments. A pilot-scale available example of such material is the CNF that is produced and distributed by the Process Development Center (PDC) of the University of Maine. The PDC has the capacity to produce 1 ton of CNF per day on dry weight basis and has already distributed materials to over 200 entities across the globe.

Figure 1 shows the physical form and microscopic structure of CNF as produced at the Process Development Center, University of Maine. This material is produced by ultra-refining bleached softwood pulp in a specially designed refiner until a pre-determined fine content (95%) is achieved. While the lengths of particles are in the range of micrometers, the diameters are in nano-scale giving the product a very high aspect ratio. No chemical pre-treatment is performed prior to refining.

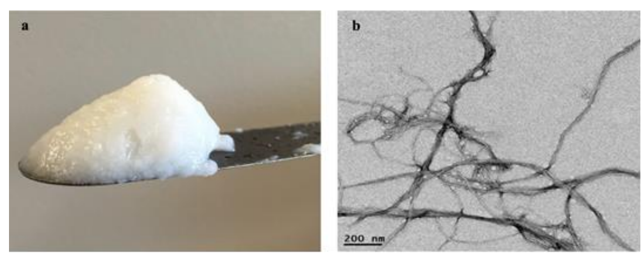


Figure 1: Cellulose nanofibrils: (a) physical appearance of a 3 wt.% solids content slurry; and (b) TEM micrograph.

## 2 CURRENT APPLICATIONS

The current major applications for CNF are reinforcement for polymeric systems, use as additive to paper coating formulations of use as the sole coating formulation, binder applications in composite panels, and barrier films for food and other material packaging. Each application takes advantage of some intrinsic material properties of CNF including that is of significance for the final product's performance in service.

In general, for any new material for which applications are sought, there are two options: either to force the material into applications for which they are not prepared or to take advantage of the strengths of the material and to try to find or develop applications where the natural strengths of the material can be used to their full extent. While efforts to modify CNF to make it compatible with "unfriendly" systems are always needed and encouraged, a closer path to commercialization seems to be the second approach.

## 2.1 Mechanical property considerations

CNF is often times confused with CNC (cellulose nanocrystals) when it comes to mechanical properties. The published mechanical properties of CNC are considerably higher than those of CNF mainly because in the former, all amorphous parts of the cellulose chains are washed away trough acid hydrolysis. The CNF on the other hand contains both amorphous and crystalline components and therefore is not as stiff and strong as CNC when it comes to the mechanical properties of individual fibers [1].

Mechanical properties of cellulose nanomaterials are more often measured by testing free standing films of the material. Here the data obtained from testing are more or less indicative of the bonding between individual fibers rather than their individual properties. CNF makes excellent films that can be easily tested in tension and therefore most mechanical properties available are obtained in tension. With more possibilities in near future to use CNF as the bulk material or the matrix in composites, it will become important to obtain mechanical property data in other modes of deformation including bending, compression and shear. Finally, as CNF materials are hygroscopic and viscoelastic, data are needed to be obtained to quantify the effect of moisture content and temperature as well as time-scale of deformation on mechanical properties.

## 2.2 Reinforcement in polymer composites

Most conventional polymers are hydrophobic and not much compatible with CNF chemically. For CNF to be used in such systems it also needs to be dried. Drying CNF into a powder that retains its nanoscale dimensions or regains them once mixed with a polymer has been a challenge for over a decade. Although great progress in drying procedures has been made [2], the scale-up issue is still there limiting the applications to small scale levels. In addition there will be a considerable reduction in the aspect ratio of CNF particles upon drying leading to inefficiency when used as reinforcement in polymeric systems.

## 2.3 Barrier applications

It was found early on that CNF films are excellent oxygen barriers making them a great choice for packaging applications where resistance to oxygen penetration is important. A great number of research projects have been focused on these applications and the results are very promising [3]. The challenge remains where the excellent oxygen barrier properties of CNF are lost at higher relative humidity because of the structural changes in the film. Efforts are being made to improve water vapor transmissivity of CNF films with the hope that it will improve oxygen barrier properties at higher relative humidity levels as well. Barrier applications of CNF seem to be of high potential in near future.

## 2.4 Binder applications

CNF is an excellent binder for wood, pulp and any other lignocellulosic in the production of composite panels. The Laboratory of Renewable Nanomaterials at the University of Maine has been working on binder applications of CNF to produce conventional particleboard and fiberboard panels as well as laminated paper composites. Figure 2 summarizes the concept of using CNF as binder in compressed wood panels. Figure three shows the paper laminates that were produced by using CNF as binder.

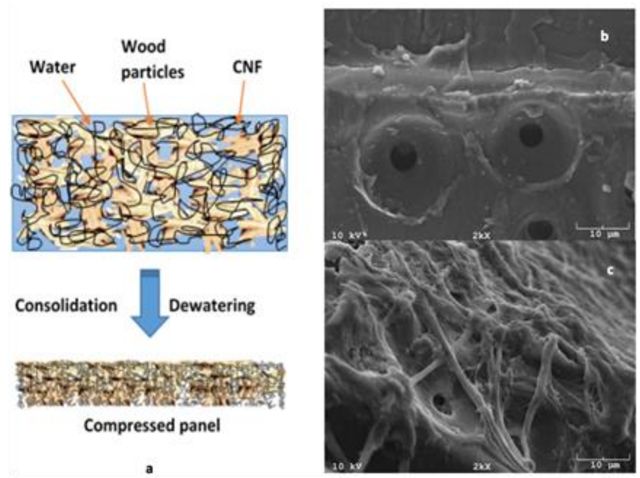


Figure 2: (a) Consolidation and dewatering phenomenon followed by drying led to bond formation at micro/nano scale; SEM image of (b) the surface of a southern pine particle and (c) a southern pine particle mixed with a 3% solids content CNF after air-drying overnight.

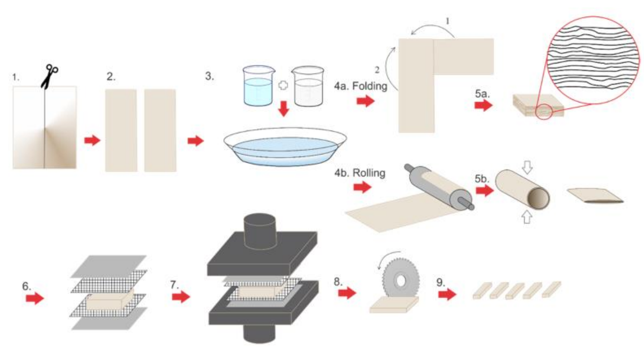


Figure 3: Production of Cellubound (CNF bonded paper laminates) [4]


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## 2.5 Yarns and textile applications

Knowing that cellulose nanomaterials possess high bonding and mechanical properties, their addition to the structure of natural fiber yarns can enhance mechanical properties of such yarns. We have recently examined 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 [5]. Tapes were also made to better understand interactions between nanocellulose and natural fibers and also to evaluate the effect of twist during yarn spinning on the final properties. This novel application of cellulose nanomaterials will provide opportunities for production of textile-based composites with improved mechanical properties.

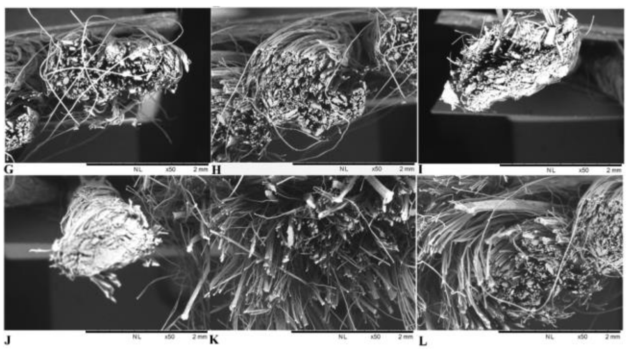

Figure 4: SEM micrographs of surface and cross-section of tapes and yarns: G) CNC flax tape cross-section H) CNC flax yarn cross-section I) CNF flax tape cross-section J) CNF flax yarn cross-section K) Water flax tape cross-section L) Water flax yarn cross-section [5]

Figure 4 shows the effect of adding a small amount of CNF and CNC to yarns and tapes produced from flax fibers. Mechanical properties of tapes and yarns are significantly improved in both cases but the improvements are more significant when CNF is used. This is due to the higher aspect ratio of CNF and the ability to bond natural fiber yarns together in the form of a tight structure with higher density [5].

# 3 CONCLUSIONS AND FUTURE PROSPECTS

CNF materials are versatile and can be used in a wide variety of applications. The key to success in the commercialization of these materials is knowing their properties and limitations and finding applications where the strengths of the material can be well taken advantage of. Attempts to force CNF into applications where it does not naturally belong to might generate interesting research ideas but may also fail to scale up and help the forest products industry that is seeking a way to use large quantities of pulp for which not much demand is expected.

New approaches towards CNF applications say as bulk material for biomedical applications, use as the matrix and not the reinforcement in the formulation of novel composites and applications where CNF slurries can be used in the wet form might pen new horizons for the commercialization of these interesting renewable nanomaterials.

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