Mechanical Properties of Cellulose Nanofibril-Wood Flake Laminate

Abstract. *Neat CNF films were laminated onto wood flakes (WF) using phenol formaldehyde (PF) resin. The mechanical properties (elastic modulus (E), ultimate tensile strength (σ_f), and strain to failure (ε_f)) of CNF films, WF, and CNF-WF laminate were measured. The lamination process improved the WF properties in the direction transverse to the wood grain; however, the strengthening phenomenon was not significant in the direction parallel to the wood grain. The improvement in properties as a result of the lamination process was a combination of contributions from the PF resin and the CNF layers.*

Keywords. cellulose nanofibrils, laminate, phenol formaldehyde resin, wood flakes.

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Introduction. Engineered wood-based composites, which are manufactured by binding wood particles, flakes, chips, long flakes, or veneer with adhesives, have been widely used in construction, industrial products, and decorations. The mechanical properties of these composites can be improved by increasing the performance of the starting wood materials. Lamination has been used to fabricate wood composites, e.g., plywood, glued-laminated timber, veneer overlays, and laminated veneer. The use of lamination of wood flakes (WF) or veneers with cellulose nanomaterials (CN) may further improve their properties. In particular, cellulose nanofibrils (CNF) films have high strength (100–250MPa), stiffness (6–20 GPa) and low density (~1–1.5 g/cm^3) [1] and may be a useful reinforcing film for such application. There have been a few studies on the fabrication of CN composites with stacking/lamination processes. Nakagaito *et al*. fabricated high-strength CN composites by laminating PF resin-impregnated cellulose microfibrils [2,3] and wood pulp mats [2]. The fracture and impact toughness of the epoxy laminates were also shown to improve with the addition of cellulose-fiber mats [4]. However, there have been no studies investigating the use of neat CNF films to produce WF- or veneer-laminated composites. As the result, the objective of this study was to produce CNF-WF laminate composites and measure the reinforcing potential of CNF films on WF.

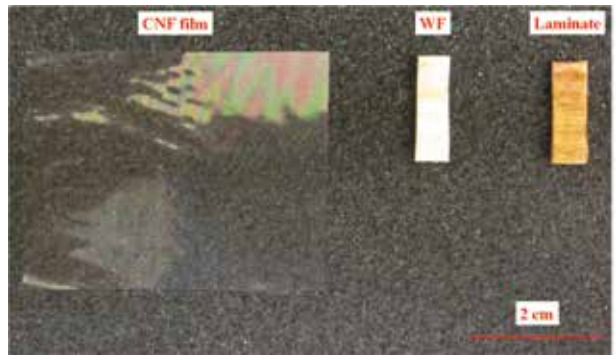

Figure 1. *Photograph of CNF film, wood flakes (WF), and CNF-WF laminate.*

Methodology. TEMPO-CNFs (0.5 wt% CNF aqueous suspension) were provided by the Forest Products Laboratory (FPL). CNF films (~10 µm thick) were produced by solution-casting onto polyethylene terephthalate (PET) substrates. The mechanical properties of dried CNF films were examined with Dynamic Mechanical Analysis (DMA) (Q800, TA Instrument).

CNF-WF laminates were fabricated by laminating CNF films on both sides of the WF (~ 500 µm thick) using PF resin. A thin layer of PF resin was spread on the WF, after which a CNF film was placed on top of the resin. The assembly was then clamped between metal plates and thermally treated (80°C for 24 h). DMA and universal tensile testing machine (MTS Insight, MTS System Corp.) were used to examine the mechanical properties of WF and CNF-WF laminate along the axial direction and the transverse direction with respect to the WF grain orientation.

Results. Neat CNF films were homogeneous and optically transparent (Fig. 1). The average mechanical properties of the CNF films are summarized in Table 1: E = 16 GPa, σ_f = 180, and ε_f = 1.4%. These films were glued onto WF with PF resin to form CNF-WF laminates (Fig. 1), and the stacked films were well bonded on the WF (Fig. 2). The mechanical properties of WF, PF resin-coated WF

Table 1 Mechanical properties summary.

Materials Tested	E (GPa)	σ_f (MPa)	ε_f (%)
CNF films*	16.0 ± 3.0	180 ± 55	1.4 ± 0.5
WF (T)*	0.6 ± 0.3	1.3 ± 0.2	0.7 ± 0.2
WF-PF (T)*	0.9 ± 0.1	3.5 ± 0.5	0.7 ± 0.1
CNF-WF Laminate (T)*	1.9 ± 0.3	5.6 ± 1.0	0.6 ± 0.1
WF (A)**	6.5 ± 1.8	45 ± 7	1.0 ± 0.2
WF-PF (A)**	6.4 ± 1.0	58 ± 13	1.1 ± 0.3
CNF-WF Laminate (A)**	5.8 ± 0.6	67 ± 9	1.3 ± 0.4

E: Young's modulus ; σ_f Ultimate tensile strength ; ε_f Strain of failure
T: specimens tested in transverse direction; A: specimens tested in axial direction
* Tested with DMA; ** Tested with MTS; All tests had 5 replicas.

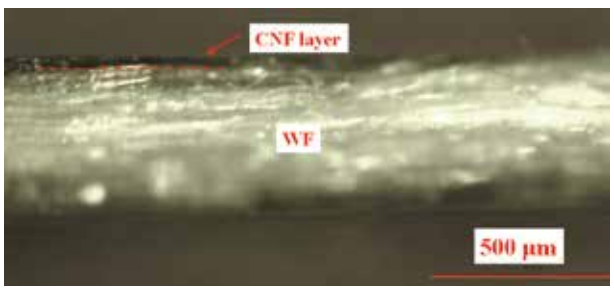

Figure 2. *Optical micrograph of the CNF-WF laminate (T) fracture surface.*

(WF-PF), and CNF-WF laminates were also measured (Table 1). In the transverse direction, WF-PF and CNF-WF laminate had higher E and ε_f in comparison with WF, while the ε_f of all specimens are similar (0.5–0.7%). The CNF-WF laminate shows almost 200% gains in E and a 300% increment in ε_f. The pronounced improvement of WF tensile properties in the transverse direction is due to the weak strength in this direction, where CNF films act as reinforcement layers. In contrast, in the axial direction, the mechanical properties were not enhanced by CNF lamination. This result is considered to result partially from the fact that CNF properties are more similar to those of WF in the axial direction, and therefore the reinforcing potential is reduced. Moreover, the WF-PF results demonstrate that at least part of the WF property modification by the lamination process resulted from the PF resin.

Conclusions. High-strength wrinkle-free cellulose nanofibril (CNF) films were used as reinforcement on wood flakes (WF) to fabricate CNF-WF laminates. The results demonstrated that the E and εf of WF in the transverse direction are improved by 200%–300% after forming the CNFWF laminate. However, in the WF axial direction, the property enhancement was limited because of the better properties of the WF in this direction. The strengthening mechanism is contributed from the combination of CNF addition to the WF and chemical bonding among PF resin, WF, and CNF films.

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Cellulose nanocrystals incorporated within the overcoat varnish of the cover!

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