

# European Pallets Fabricated with Composite Wood Blocks from Tropical Species Reinforced with Nanocrystalline Cellulose: Effects on the Properties of Blocks and Static Flexure of the Pallet

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The objective of this study was to characterize the performance of composite wood blocks (CWB) by testing internal bonding, nail extraction resistance, and water absorption. The CWB were glued with two wood adhesives, polyvinyl acetate (PVAc), and urea formaldehyde (UF), modified with 1% nanocrystalline cellulose (NCC). Three tropical species were employed: *Vochysia ferruginea*, *Cordia alliodora*, and *Gmelina arborea*. In addition, the original European pallet in the static flexure test was evaluated. The results showed that the internal bonding relative to solid wood blocks (SWB) increased with both adhesives. Meanwhile, the CWB of *V. ferruginea* with UF and *C. alliodora* with PVAc showed the greatest resistance to nail extraction, while in *G. arborea*, the NCC increased the resistance to nail extraction. The CWB with modified adhesives absorbed more moisture, particularly with PVAc, compared with the SWB. In static flexure tests of the pallets fabricated with CWB, the load at the limit of proportionality and the maximum load increased, while deflections were lower, compared with SWB. The results showed the potential of utilizing NCC in CWB fabricated with tropical species.

*Keywords:* Nanocrystalline cellulose (NCC); Adhesives; Wood; Composite blocks; Pallets

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## INTRODUCTION

Cellulose is the most abundant organic compound on the planet (Thakur *et al.* 2013). It is a naturally formed polymer in all plants, together with other important components such as lignin, hemicelluloses, and waxes (Li *et al.* 2009). Cellulose can be found all over a plant, although it is mostly concentrated in the stem (Lavoine *et al.* 2012) to support the plant (Morán *et al.* 2008).

As a renewable material, and because of its physical and chemical characteristics, cellulose has been of great interest for manufacturing over the last decade (Poletto *et al.* 2014). With dimensions of 20 nm in diameter and 100 nm to 400 nm in length, nanocrystalline cellulose (NCC) can be obtained from cellulose (Morán *et al.* 2008). Alternatively, microfibrillated cellulose (MFC) can be prepared by mechanical treatment, often supplemented by chemical or enzymatic treatments. Both MFC and NCC have been studied and used as reinforcements and filling in various polymers (Poletto *et al.* 2014;

Khanjanzadeh *et al.* 2018; 2019) due to the strength properties conferred by NCCs crystalline nature (Atta-Obeng *et al.* 2013) and their biodegradability (Liew *et al.* 2015).

Wood is a renewable material with good structural resistance that competes with other non-renewable materials (Tsoumis 1968). However, wood has the inconvenience that over 40% of the raw material becomes waste during the industrialization process (Espinoza-Durán and Moya 2013; Jadin *et al.* 2017), which can become a problem unless adequate management measures are taken, particularly in underdeveloped countries like Costa Rica (Gaitán-Álvarez *et al.* 2017). Some industries in more industrialized countries employ wood wastes as a raw material to develop other products, such as wood composites or to use for energy production (Thakur *et al.* 2014).

In addition to the problem of wood waste in Costa Rica, stubble from pineapple plantations is another source of waste. The annual pineapple (*Ananas comosus*) world crop, approximately 24.8 million tons produced (Darnaudery *et al.* 2018), produces large amounts of residues in the forms of unused leaves, stems, crowns, and fruit peels (Rattanapoltee and Kaewkannetra 2014). Nearly 300 tons of stubble per pineapple-sown hectare are produced in Costa Rica (Moya *et al.* 2016), where these wastes, as in other countries, have not yet received adequate management (Moya and Camacho 2014). Because this is a slow-decaying material, toxic herbicides, such as N,N'-dimethyl-4,4'-bipyridinium dichloride (paraquat), are commonly used; paraquat is a soil contaminating herbicide with accumulative toxicity (Moya *et al.* 2016).

One possible application for the pineapple wastes is the extraction of natural fibers for manufacturing ropes and textiles, among other products (Moya and Camacho 2014), or for pulp production (Moya *et al.* 2016). However, recent research presents the possibility of extracting NCC from the pineapple fibers and use of the NCC for reinforcing wood composites (Pirayesh *et al.* 2013; Balakrishnan *et al.* 2018; Rigg-Aguilar *et al.* 2019).

Wood and pineapple residues can be converted into lignocellulosic composites, generally achieving better properties than solid wood. These composites are the result of particle bonding (Rowell 2012), which increases their mechanical properties (Moya *et al.* 2015a, 2015b; Chaabouni and Boufi 2017). The final densities of the wood composites range between 0.6 g·cm<sup>-3</sup> and 0.7 g·cm<sup>-3</sup> (Rangel *et al.* 2017). The particles are moistened with fillings or adhesives that increase their internal bond resistance and are bonded by thermal pressure (Salari *et al.* 2013).

Several studies have shown that the addition of cellulose, either as NCC or MFC, to urea formaldehyde and polyvinyl acetate (two of the most important adhesives) improves their performances in lignocellulosic composites (*e.g.*, wood composites) (Thakur and Singha 2010; Aydemir 2014; Kwon *et al.* 2015; Ayrilmis *et al.* 2016; Mahrtdt *et al.* 2016; Chaabouni and Boufi 2017; Khanjanzadeh *et al.* 2018, 2012).

Despite advances in the production of wood composites in Costa Rica, the development of composite-based products is limited. Consequently, the use of wastes is scarce (Serrano and Moya 2012). Therefore, an important market for composites can be visualized in this country. For example, 50% of timber consumption in Costa Rica is destined for pallet fabrication (Barrantas and Ugalde 2017), utilizing woody species such as *Gmelina arborea*, *Cordia alliodora*, and *Vochysia ferruginea* (Rigg-Aguilar *et al.* 2019).

As for pallet production, a high percentage are of European type, fabricated with blocks 10 cm × 10 cm × 10 cm obtained from logs with diameters greater than 20 cm (Rigg-Aguilar *et al.* 2019), which can have high commercial value in the fabrication of other products (Serrano and Moya 2012; Jadin *et al.* 2017). This log dimension is large for the diameters produced in fast-growth forest plantations in Costa Rica (Serrano and Moya

2012). Therefore, producers are seeking alternatives for producing blocks from wood composites or other lignocellulosic materials (Zivic *et al.* 2017), as there is experience in other countries manufacturing pallets using blocks produced with wood wastes (Kain *et al.* 2013; Zivic *et al.* 2017).

Consequently, there is interest in the Costa Rican industry to fabricate composite wood blocks (CWB) to produce European-type pallets from timber and pineapple residues *via* extracting the NCC and applying it to the adhesives. Therefore, the objective of this study was to characterize CWB and their use for European-type pallet fabrication. Three tropical forest species (*Cordia alliodora*, *Gmelina arborea*, and *Vochysia ferruginea*) glued with two adhesives, polyvinyl acetate and urea formaldehyde, modified with 1% NCC, were studied. The tests evaluated the internal bond resistance, nail extraction, water absorption, and the static flexure of European pallets fabricated with CWB.

## EXPERIMENTAL

### Materials

The NCC was provided by the Laboratorio Nacional de Nanotecnología (LANOTEC, San José, Costa Rica). It was extracted from pineapple (*Ananas comosus*) peel through a two-stage acid hydrolysis process. The NCC sizes were between 20 nm and 40 nm in diameter, and its concentration was 0.066 g·mL<sup>-1</sup>. The details of the extraction and characterization can be found in Camacho *et al.* (2017) and Rigg-Aguilar *et al.* (2019).

Two adhesive types were employed: polyvinyl acetate (PVAc) and urea formaldehyde (UF). The PVAc (Resistol™ MR 850; Henkel, Düsseldorf, Germany) technical product description indicates that the resin is water-based PVAc, with 54.5% to 55% solid contents and a viscosity between 1.6 Pa.s and 2.0 Pa.s. The UF (Resina CR-560 U-F; Química Centroamericana, Quibor, S.A., Managua, Nicaragua) is also water-based, with four components: resin (51%), water (26%), wheat flour as agglutinant (20%), and ammonium sulfate as catalyzer (3%). The technical description indicates that the pure resin contains 64% to 65% solid contents and has a viscosity of 650 cP to 900 cP. The total solid content of the complete adhesive with the four components is 48%.

The wood used to evaluate the effects of nanocellulose in the two adhesives came from three tropical plantation species in Costa Rica: *Gmelina arborea* Roxb. (melina), *Vochysia ferruginea* Mart. (botarrama), and *Cordia alliodora* Ruiz & Pav. (laurel). These three are commonly used for pallet fabrication (Barrantas and Ugalde 2017) and are commercially prevalent in Costa Rica (Tenorio *et al.* 2016).

The wood utilized came from plantations with ages ranging between 7 y and 9 y. The *C. alliodora* came from CATIE-Turrialba (09° 53' 00" N, 83° 38' 01" W, 602 m a.s.l., 9-years-old), the *G. arborea* came from Bonifacio, Limón (09° 46' 43" N, 82° 54' 59" W, 42 m a.s.l., 8-years-old), and the *V. ferruginea* came from Búfalo, Limón (10° 00' 21" N, 83° 10' 23" W, 25 m a.s.l., 7-years-old). Approximately 7 to 8 trees per species were felled. For the pallets fabricated with solid wood, the *G. arborea* was utilized.

### NCC in wood adhesives

The effects of 1% concentration NCC in the two wood adhesives (PVAc and UF) were evaluated according to the results obtained by Rigg-Aguilar *et al.* (2019). The 1% concentration (w/w) was added according to the percentage of solids in each adhesive (55% for PVAc and 48% for UF).

The mixture of NCC with adhesives was prepared as follows: First, 150 g of PVAc was placed into a receptacle and stirred with an inclined-blade agitator (45°) at 1600 rpm. Then, the NCC gel (approximately 12.5 mL for 1% concentration) was slowly added into the adhesive. Stirring of the NCC-adhesive mixture was continued for 10 min. For the UF, which has 4 components (resin, water, wheat flour, and ammonium sulfate as catalyzer), 76.5 g of the resin was initially mixed with 10.9 mL of NCC and stirred for 5 min at 1600 rpm with an inclined-blade agitator (45°). Then, the rest of the components were added (28.10 to 33.55 mL of water, 30 g flour, and 4.5 mL catalyzer), while stirring was continued for an additional 5 min.

### *CWB manufacturing*

The CWB were 10 cm × 10 cm × 10 cm, and were composed of wood particles from *V. ferruginea*, *C. alliodora*, and *G. arborea*, employing the NCC-modified adhesive. First, chips were obtained from 60 logs (1-m-long and of varying diameter) of the three species. The chips were ground, and the material was sieved to obtain sizes between 3 mm and 10 mm, with moisture contents between 2% and 4%.

Next, 550 g of dry particles were mixed with 100 g of NCC-modified adhesive (18% of total chip weight) for UF and 66 g (12% of total chip weight) for PVAc. The adhesive was slowly added to the particles while stirring over 4 min. The glued particles were placed into a mold and pressed for 5 min with a 60 ton capacity universal testing machine (model: Super L60; Tinius Olsen, Horsham, PA, USA). The mold was then heated to 180 °C, while the pressure was applied for 60 min to 75 min to obtain the CWB. Lastly, the CWB were conditioned to a 12% moisture content for two weeks.

A total of 72 CWB (2 adhesives × 2 concentrations (1% and 0%) × 9 blocks per pallet × 2 samples) were fabricated to manufacture the pallets for each species, with the same number of blocks (2 adhesives × 2 concentrations (1% and 0%) × 9 blocks per pallet × 2 samples) to perform the tests with the composite wood with the modified adhesive.

### *Effect of the NCC in CWB for making European pallets*

In the first stage, flexure in European pallets was tested using CWB with 1% NCC concentration and fabricated with particles from *G. arborea*, *V. ferruginea*, and *C. alliodora*. In the second stage, CWB fabricated with particles of the three species using both adhesives and 1% NCC concentration were evaluated in the following tests: internal bond, resistance to extraction of pallet nails, and percentage of water absorption. Additionally, the CWB fabricated with 1% NCC concentration were compared in the same tests with the blocks fabricated with adhesive without NCC and with solid wood blocks (SWB) of the same species.

## **Methods**

### *Internal bond and perpendicular tension resistance in CWB*

A total of five different CWB fabricated with the two adhesives and modified with 1% NCC were cut according to Fig. 2c, obtaining four samples of 5 cm × 5 cm × 10 cm per block. Thus, 20 samples per treatment were obtained (two adhesives: NCC-modified and NCC-unmodified). The internal bond test was performed in accordance with the ASTM D1037-12 (2018) standard. The bonding strength of the CWB was compared with the grain-perpendicular tension strength of the SWB (Fig. 1d). For this purpose, SWB in green condition for European pallets were obtained from Maderas Bosque Verde S.A. (Cartago, Costa Rica). The SWB were cut according to Fig. 1b, and eight samples of 5 cm

× 5 cm × 6.3 cm were obtained (Fig. 1d). Therefore, 24 samples per treatment were tested, following the ASTM D143-14 (2018) standard.

#### *Resistance to nail extraction in CWB*

Five CWB (10 cm × 10 cm × 10 cm) with both adhesives (PVAc and UF), 1% NCC-modified and unmodified (0% NCC), and five SWB from each species were utilized to measure the resistance to nail extraction. In both cases, the ASTM D1761-12 (2018) standard was followed, inserting pallet nails 5-cm-long (12.5 caliber with a 32° angle spiral) to a depth of 32 mm at each side of the block, with a minimum distance between the nails and the edge of the block of 2.54 cm. The nails were inserted on all sides of the blocks to determine the lateral and transverse resistance. For the composite wood blocks, the transverse directions were defined as the sections where compression was exerted, while the lateral directions were the sections perpendicular to the direction of the compression force at the time of block fabrication. There were 30 tests per treatment.

#### *Water absorption*

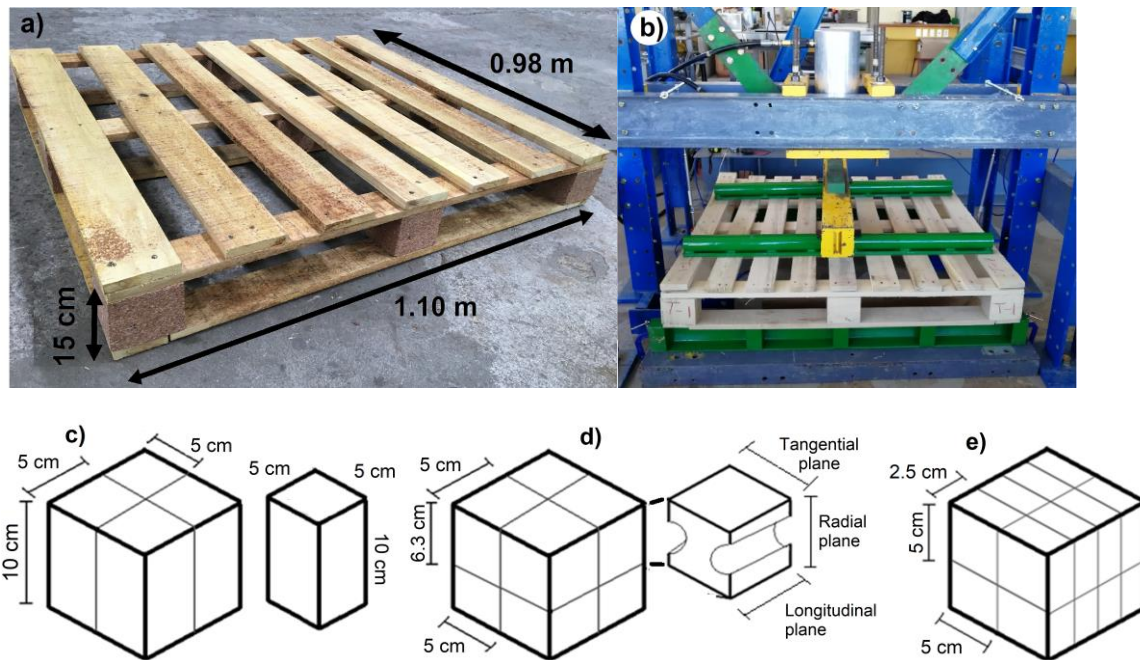
Samples sized 5 cm × 5 cm × 2.54 cm were obtained from the five conditioned CWB per treatment (Fig. 1e). The samples were vertically placed into trays with water covering the bottom up to 2.54 cm for 24 h, in accordance with the ASTM D1037-12 (2018) standard. The weight was recorded before placement into water and after 24 h. Water absorption was calculated for each of the samples according to the ratio of the weight of the water absorbed to the weight of the sample before the absorption test, and expressed as a percentage.

#### *Pallet fabrication*

The pallets were fabricated at Maderas Bosque Verde S.A. (Cartago, Costa Rica). The European pallets were 1.10-m-long, 0.98-m-wide, and 0.15-m-high (Fig. 1a). The upper boards were 10.1-cm-wide and 1.8-cm-thick; seven boards were placed. In both cases, *G. arborea* was utilized. The pallets were nailed employing a 6 bar pressure pneumatic pistol (Unicorn Fasteners Co., Ltd., Tianjin, China), 2 nails per board (5-cm-long, 12.5 caliber, and 32° angle spiral). The lower side of the pallet featured three *G. arborea* boards supporting the blocks and three boards perpendicular to the first three, measuring 8.3-cm-wide and 1.8-cm-thick. Each block was nailed with two nails to the lower boards.

#### *Static flexure of the complete pallet*

The pallets fabricated with CWB, as well as with SWB, were tested for their flexural strength according to the ISO 8611-1 (2013) standard, in which the pallet is placed on its sides while a double load is applied on the sections between the blocks (Fig. 1b) at 60 N/min. A crackmeter sensor was placed at each side of the pallet to measure the vertical deflection at the central section of the pallet. During the test, the load and the deflection were recorded in periods of 4 s until the pallet failed. These data were utilized to calculate the load and deflection at the limit of proportionality ( $L_{LP}$  and  $D_{LP}$ ) and the maximum load and deflection ( $L_{max}$  and  $D_{max}$ ) of the pallet based on the calculations of ISO 8611-1 (2013). At the end of the test, the different failure types that occurred to the pallet were recorded.



**Fig. 1.** (a) Dimensions and aspect of the pallet fabricated with composite wood; (b) pallet flexural test; cutting patterns to obtain the samples for tests: (c) internal bond, (d) perpendicular tension in SWB, and (e) water absorption

### Statistical analysis

Assumptions of normal data distribution were evaluated by the Shapiro–Wilk test, with the variance homogeneity evaluated by Levene’s test and the InfoStat software (InfoStat Company, version 2017, Buenos Aires, Argentina). Then, to determine differences among the means of the tests, an analysis of variance (ANOVA) test was applied, where the tests performed were the response variables, and the NCC concentration was the independent variable. Then, this analysis was accompanied by the Tukey test at 99% reliability to determine the differences between both NCC concentrations within the species per adhesive. As the internal bond and water absorption tests did not meet the assumptions, non-parametric tests were used: the Kruskal–Wallis test to determine differences between NCC concentrations and Steel’s test to compare the CWB with the control (SWB).

## RESULTS AND DISCUSSION

### Internal Bond Resistance and Resistance to Nail Extraction of CWB

Table 1 shows the internal bond resistance of the blocks used for pallets. Compared with tension parallel to the grain, the SWB showed statistically greater internal bond resistance values than those of the CWB with the unmodified adhesive and with NCC-modified adhesive in the three species. Modifying the adhesive with NCC did not affect the internal bond resistance of CWB fabricated with PVAc, as no significant differences were observed in the three species between the adhesives with or without NCC.

**Table 1.** Internal Bond / Perpendicular Tension, Water Absorption, Pallet Nail Resistance, Moisture Content, and Density in Blocks Used in European Pallets Fabricated with *V. ferruginea*, *C. alliodora*, and *G. arborea* Particles Glued with PVAc and UF Adhesives With and Without NCC

Species	Type of Block (Adhesive, % NCC)		Internal Bond Resistance (MPa)	Water Absorption (%)	Pallet Nail Resistance		Moisture Content (%)	Density (g/cm <sup>3</sup> )
					Lateral (N)	Transverse (N)		
<i>V. ferruginea</i>	SWB		2.09 <sup>A</sup> (0.771)	39.0 <sup>A</sup> (5.38)	398.4 <sup>A</sup> (92.0)	241.9 <sup>A</sup> (59.4)	18.38	0.30
	CWB with PVAc adhesive	0.0	0.14 <sup>B</sup> (0.041)	196.5 <sup>B</sup> (27.72)	370.2 <sup>AB</sup> (90.7)	252.5 <sup>A</sup> (47.0)	9.07	0.57
		1.0	0.16 <sup>B</sup> (0.049)	179.3 <sup>B</sup> (14.93)	285.3 <sup>A</sup> (73.9)	224.3 <sup>A</sup> (28.3)	8.28	0.58
	CWB with UF adhesive	0.0	0.07 <sup>A</sup> (0.033)	94.5 <sup>A</sup> (4.96)	532.5 <sup>B</sup> (60.9)	419.2 <sup>B</sup> (104.5)	10.03	0.62
		1.0	0.59 <sup>B</sup> (0.123)	131.2 <sup>B</sup> (11.47)	337.7 <sup>A</sup> (66.6)	176.1 <sup>A</sup> (35.8)	8.18	0.61
<i>C. alliodora</i>	SWB		1.89 <sup>A</sup> (0.622)	48.8 <sup>A</sup> (7.85)	391.3 <sup>A</sup> (119.2)	200.8 <sup>A</sup> (36.9)	17.05	0.31
	CWB with PVAc adhesive	0.0	0.06 <sup>B</sup> (0.028)	251.5 <sup>B</sup> (27.25)	246.9 <sup>B</sup> (71.0)	82.7 <sup>B</sup> (25.6)	9.28	0.55
		1.0	0.06 <sup>B</sup> (0.013)	240.8 <sup>B</sup> (17.99)	393.5 <sup>A</sup> (96.5)	131.1 <sup>B</sup> (21.0)	8.19	0.57
	CWB with UF adhesive	0.0	0.05 <sup>A</sup> (0.030)	120.0 <sup>A</sup> (9.43)	464.1 <sup>A</sup> (114.3)	285.6 <sup>B</sup> (63.7)	10.20	0.60
		1.0	0.22 <sup>B</sup> (0.058)	164.1 <sup>B</sup> (17.96)	308.0 <sup>AB</sup> (57.0)	192.3 <sup>AB</sup> (49.4)	7.47	0.61
<i>G. arborea</i>	SWB		2.00 <sup>A</sup> (0.654)	64.9 <sup>A</sup> (14.07)	437.3 <sup>A</sup> (125.4)	222.5 <sup>A</sup> (72.3)	40.87	0.39
	CWB with PVAc adhesive	0.0	0.01 <sup>B</sup> (0.003)	151.6 <sup>B</sup> (31.88)	107.3 <sup>B</sup> (36.6)	40.2 <sup>B</sup> (12.4)	11.00	0.56
		1.0	0.03 <sup>B</sup> (0.009)	176.7 <sup>B</sup> (27.26)	154.5 <sup>B</sup> (38.2)	65.7 <sup>B</sup> (20.4)	7.88	0.58
	CWB with UF adhesive	0.0	0.10 <sup>B</sup> (0.032)	67.5 <sup>AB</sup> (3.81)	251.0 <sup>B</sup> (48.7)	186.7 <sup>A</sup> (47.4)	10.83	0.62
		1.0	0.45 <sup>A</sup> (0.064)	100.6 <sup>B</sup> (8.70)	223.6 <sup>B</sup> (56.3)	173.5 <sup>A</sup> (43.5)	8.24	0.63

Values in parentheses represent standard deviations; average values identified with different letters are statistically different at  $\alpha = 0.01\%$

In contrast, in the CWB fabricated with UF adhesive and modified with 1% NCC, the internal bond resistance increased significantly in relation to the CWB fabricated with unmodified adhesive (Table 1).

Meanwhile, the analysis between NCC concentrations of the CWB for each species per adhesive showed that, for *C. alliodora* and *G. arborea* with PVAc adhesive, nail extraction resistance increased in both sections with 1% NCC, which was statistically different only for the lateral section in *C. alliodora* (Table 1). In the rest of the cases, neither reinforcement from NCC nor statistical differences were observed in most of the comparisons.

The CWB of *V. ferruginea* and *C. alliodora* glued with 1% NCC presented resistances to nail extraction that were statistically equivalent to SWB of *V. ferruginea* and *C. alliodora*, both laterally and transversely, with the exception CWB with 1% NCC of *C. alliodora*, which had a lower transverse resistance than the SWB (Table 1). However, there was a slight increase in the resistance to nail extraction in the CWB without NCC with both types of adhesives, but this increasing was not statistically significant. There were exceptions, where CWB with PVAc adhesive without NCC had statistically lower lateral and transverse resistances than the SWB for *C. alliodora*, as well as lower lateral and transversal resistance than the SWB for *V. ferruginea*. In *G. arborea*, the SWB showed the greatest values of resistance to nail extraction compared to the CWB bonded with both adhesives, while the CWB with PVAc were statistically different from the control treatment.

Strength in the CWB is affected by varying aspects (Hoadley 2000; Carvalho *et al.* 2003), one of which is the size of the product. In this case, the blocks were 10 cm × 10 cm × 10 cm, which is rather large to be densified by compression. Vital *et al.* (1974) and Leng *et al.* (2017) mention that the strength of the products increases if the material is adequately densified, especially if performed by compression, as were the blocks in this project. Furthermore, densification by compression at temperatures above 100 °C facilitates plasticization of the wood particles (Hunt *et al.* 2017), thus increasing the internal bond strength. However, the CWB cannot achieve the values observed for SWB (Rangel *et al.* 2017).

The increases in the internal bond resistances of the CWB with NCC-modified UF adhesive in the three species (Table 1) is supported by the works of Veigel *et al.* (2011) and Zhang *et al.* (2011), who found increases in the internal bond strength of UF adhesive with added NCC. Kwon *et al.* (2015) mentions that the increase in the strength of NCC-modified adhesives is due to the fact that the cellulose fibers in the glue line merge with the adhesive inside the wood cavities, allowing better penetration; consequently, greater strength is needed to separate two wood pieces. Likewise, the reinforcement effect of the wood-adhesive interface is the result of the formation of a network between the NCC and the polymer chains of the adhesives (Grishkewich *et al.* 2017). Ramires and Dufresne (2012) and de Almeida Mesquita *et al.* (2018) explain that NCC's high contact area, high resistance to traction, and rigidity allow the formation of a crystal network that interacts with the adhesive, increasing its mechanical resistance.

Meanwhile, in thermoplastic adhesives such as PVAc, the addition of NCC can improve the low rigidity of the polymers, as well as adhesion in the glue line (Gindl-Altmutter and Veigel 2014). Different adhesion mechanisms occur within the interface of wood particles and adhesives, as the adhesive can penetrate the wood, allowing interlinking and greater internal adhesion. Moreover, internal strength within the adhesive allows the adhesive layer in the interface to remain bonded, improving the interactions and increasing



the bonding strength (Gindl-Altmutter and Veigel 2014).

Modifying the PVAc adhesive with NCC did not improve the properties of the CWB in relation to CWB without NCC (Table 1). This result can be due to NCC producing a cracking effect inside the adhesive, decreasing the strength of the glue line (Gindl-Altmutter and Veigel 2014), which was reflected in the diminution of the resistance to nail extraction and of the internal bond strength of the blocks. Therefore, PVAc application to CWB had no beneficial effect on the bond resistance.

### Water Absorption

The CWB compared to SWB presented greater water absorption, being statistically equivalent in the CWB with UF without NCC in the three species (Table 1). No statistical differences were observed in water absorption among the CWB with the PVAc adhesive. Contrastingly, the CWB with UF adhesive in *V. ferruginea* and *C. alliodora* presented statistical differences between NCC concentrations, as water absorption was greater with 1% NCC concentration.

The hydrophobic nature and porous structure of the wood favor water absorption, as wood particles have a greater contact area (Salari *et al.* 2013). Moreover, neither of the two adhesives present good performance against water (Yang *et al.* 2006), so greater water absorption in the CWB compared to SWB was expected (Table 1).

However, the addition of NCC to the PVAc adhesive did not improve block performance against water absorption. These results disagreed with Chaabouni and Boufi (2017) and Jani and Izran (2013), who report that the decrease in water absorption in products using PVAc is explained by the interlinking between the OH groups and the adhesive matrix, preventing water intrusion inside the polymer matrix. In contrast, the same result cannot be obtained with the UF adhesive due to the reduced interaction between the NCC-modified adhesive and the wood particles, which deteriorates the adhesion forces and prevents the formation of an adhesive barrier against water absorption, resulting in an increased amount of absorbed water (De Almeida Mesquita *et al.* 2018).

Notably, CWB are affected by moisture absorbed from the environment, which impairs the performance of the composite, as moisture penetration can weaken the internal bonds of the wood particles (Rofii *et al.* 2016). The CWB, once compressed, are extracted from the mold completely dry. During their conditioning, the CWB absorb humidity from the environment, especially in a tropical region such as Costa Rica. Due to water absorption, the CWB expand slightly, weakening the internal bonds of the adhesive or the wood-adhesive interface, or the resistance to nail extraction (Gerhards 1982).

### Evaluation of the Pallets

The flexural test showed that  $L_{LP}$  and  $L_{max}$  increased for the pallets fabricated with CWB of the three species and with the two adhesives (PVAc and UF) modified with NCC at 1% (Table 2). The increases in  $L_{LP}$  varied from 8% to 37%, while the increases in  $L_{max}$  ranged from 42% to 59%, with the lowest increase observed in *C. alliodora* and the greatest increase in blocks fabricated with *V. ferruginea* (Table 2).

For deflection,  $D_{LP}$  was lower for pallets fabricated with CWB of *C. alliodora* and *G. arborea* and the two nanocellulose modified adhesives. The decrease in  $D_{LP}$  ranged from 1.0 mm to 3.1 mm, and the lowest deflection values were observed in the CWB of *C. alliodora*. In contrast, the pallets built with CWB of *V. ferruginea* presented higher  $D_{LP}$  than the pallets built with SWB. Meanwhile, the  $D_{max}$  in pallets fabricated with the three species increased between 14.5 mm and 23.3 mm, while the greatest deflection was

observed in pallets of *C. alliodora* and *G. arborea*, with the lowest deflection in pallets fabricated with *V. ferruginea* (Table 2).

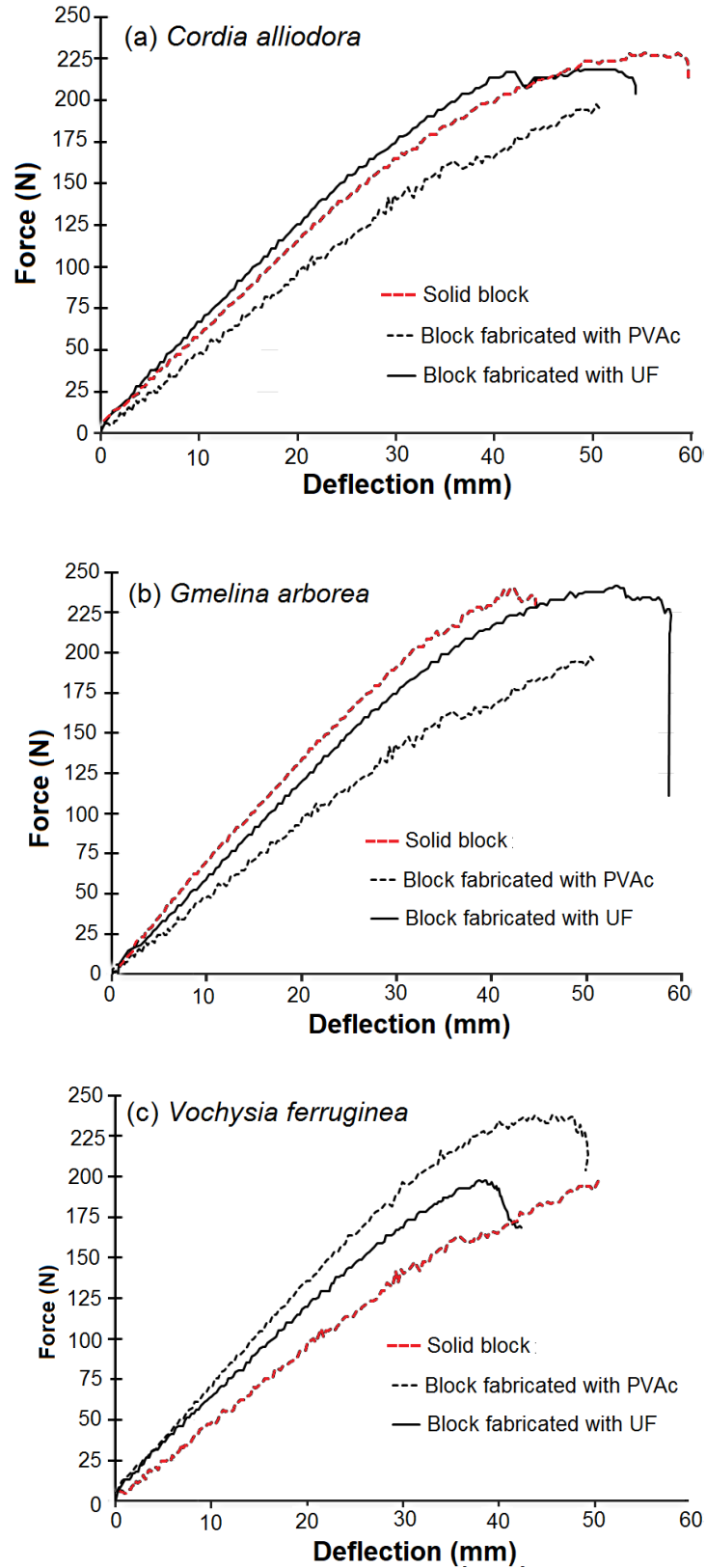
**Table 2.** Failure Types in *V. ferruginea*, *C. alliodora*, and *G. arborea* Bonded with PVAc and UF Adhesives Under Two NCC Concentrations

Parameter	Adhesive	Value for Pallets Built of SWB	<i>Cordia alliodora</i>	<i>Gmelina arborea</i>	<i>Vochysia ferruginea</i>
$L_{LP}$ (kg)	SWB	1465	-	-	-
	PVAc	-	1645 (12%)	1708 (17%)	2007 (37%)
	UF	-	1580 (8%)	1635 (12%)	1702 (16%)
$D_{LP}$ (mm)	SWB	28.5	-	-	-
	PVAc	-	25.9 (-2.6)	27.1 (-1.4)	29.8 (+1.3)
	UF	-	25.4 (-3.1)	27.5 (-1.0)	28.7 (+0.2)
$L_{max}$ (kg)	SWB	1556	-	-	-
	PVAc	-	2377 (53%)	2392 (54%)	2483 (59%)
	UF	-	2202 (42%)	2428 (56%)	2258 (45%)
$D_{max}$ (mm)	SWB	31.24	-	-	-
	PVAc	-	52.6 (+21.4)	48.7 (+17.5)	46.3 (+15.0)
	UF	-	49.4 (+18.2)	54.5 (+23.3)	45.7 (+14.5)

Values in parentheses correspond to the percentage of load increase for  $L_{LP}$  and  $L_{max}$ . For  $D_{LP}$  and  $D_{max}$ , the values in parentheses represent the deflection change of the pallet: less deflection (-) or greater deflection (+).

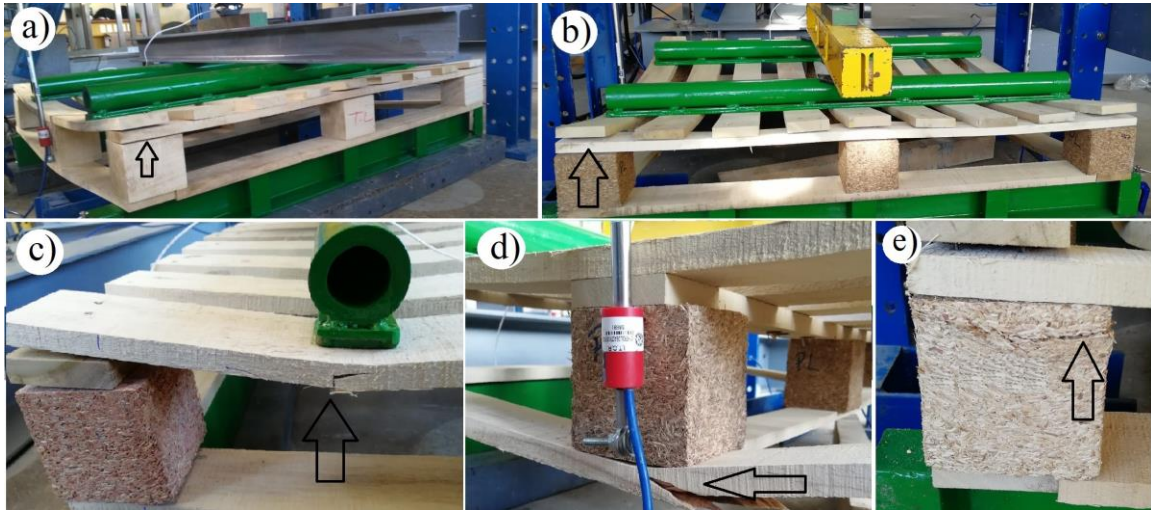
Figure 2 shows the behavior of the load applied to the pallets fabricated with the different types of blocks (SWB or CWB) relative to deflection. In most of cases, the load-deflection curves of the SWB pallets were below the load-deflection curves of the pallets constructed with CWB glued with the two adhesives (PVAc and UF) modified with nanocellulose. This result means that the pallets fabricated with these blocks manufactured with PVAc or UF modified with nanocellulose will present less deflection compared to the pallets built of SWB, for the same load magnitude. Furthermore, these load-deflection curves also showed that there was less deflection when using the UF adhesive in CWB than in CWB glued with PVAc in *G. arborea* (Fig. 2b). However, the CWB with UF showed greater deflection than the CWB with PVAc in the same species (Fig. 2b). For *V. ferruginea* wood (Fig. 2c), the CWB with UF showed greater deflection than the CWB with PVAc and the load-deflection curves showed that there was less deflection when using the UF adhesive in the CWB. Meanwhile, the deflection for the same load in the wooden blocks of *C. alliodora* was similar for the two adhesives tested (Fig. 2a).

The pallets constructed with CWB of the three species presented the same failures shown by SWB pallets. There were four failure types. In type I, upper boards detached at the corners of the pallets, both from the lower boards and from the blocks themselves (Figs. 4a and 4b). In type II, the upper boards were broken (Fig. 3c). In type III, failure was caused by tension in the lower boards of the central block of the pallet (Fig. 3d). In type IV, internal block bonding failed (Fig. 3e).



**Fig. 2.** Load curves vs. deflection in the flexural test for pallets fabricated with CWB of *Cordia alliodora* (a), *Gmelina arborea* (b), and *Vochysia ferruginea* (c) with two different wood adhesives

Most pallets presented failure type I (Fig. 3a). Only one pallet fabricated with composite *V. ferruginea* blocks and another pallet built with composite *C. alliodora* blocks presented failure types II and IV (Figs. 3c and 3d), respectively. In the two pallets fabricated with CWB of *G. arborea* with the NCC-modified PVAc adhesive, the blocks failed due to internal bond failure (Fig. 3e).



**Fig. 3.** Failure types in pallet flexural tests: Type I, upper boards detached (a and b); Type II, upper board broken (c); Type III, failure due to tension (c); Type IV, upper boards were broken (d); and failure in internal block bonding (e)

Although it was observed that CWB has much lower internal bonding resistance, the pallets fabricated with CWB, the load at the limit of proportionality and the maximum load increased, while deflections were lower, compared with SWB. NCC-modified adhesives indicate that the increase in the resistance of is because the cellulose fibers in the glue line merge with the adhesive inside the wood cavities, allowing better penetration; consequently, greater strength is needed to separate two wood pieces (Kwon *et al.* 2015; Grishkewich *et al.* 2017). Then pallets fabricated with CWB jointed with nails showed the improvement of the proprieties of wood composites.

Finally, the pineapple wastes present the possibility of extracting NCC from the pineapple fibers for reinforcing wood composites and joint with wood residues produced in a sawmill can be converted into lignocellulosic composites, generally achieving better properties than solid wood. Then such composites can substitute for solid blocks from sawlogs, and they can be used in high-value products. Composite blocks for pallets are commonly used in pallet fabrication, and its equipment and technology are widely development. Thus, it is possible to fabricate pallets with wood composites having a high beneficial effect of NCC.

## CONCLUSIONS

1. The addition of 1% NCC to PVAc and UF adhesives for fabrication of CWB with *Vochysia ferruginea*, *Cordia alliodora*, and *Gmelina arborea* wood produced an increase in the internal bonding and in the nail extraction test of the blocks. Therefore,

the NCC improved the resistance properties of the wood adhesives. However, although the level of water absorption increased with added NCC, it remained higher than in SWB.

2. The CWB fabricated using NCC-modified PVAc and UF adhesives and wood from the tropical species commonly used for pallet fabrication in Costa Rica showed improved performance in static flexure tests of the European pallets, specifically in maximum load and deflection. This result, together with the tests conducted on CWB, indicates that substituting SWB with CWB and adding 1% NCC to the adhesives is a viable option that improves the structural performance of the European pallet.

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