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Effect of Tobacco Stalk Additive Particle Size on the Bond Strength and Formaldehyde Emission of Urea Formaldehyde Bonded Plywood

Juanito P. Jimenez Jr.[1,2], **Menandro N. Acda**[2], **Ramon A. Razal**[2], **Willie P. Abasolo**[2], **Hidelisa P. Hernandez**[3], and **Arnold R. Elepaño**[4]

[1]Department of Science and Technology
Forest Products Research and Development Institute
College, Laguna 4031 Philippines

[2]Department of Forest Products and Paper Science
[3]Institute of Chemistry; [4]Institute of Agricultural Engineering
University of the Philippines Los Baños
College, Laguna 4031 Philippines

The study investigated the use of native type (Batek) tobacco (*Nicotiana tabacum* L.) stalk particles from two different mesh sizes – passing 100 mesh but retained at 200 mesh coded as N-100m (74–149 µm) and passing 200 mesh coded as N-200m (< 74 µm) – as additive with both extender and filler property, as well as formaldehyde scavenging property in urea-formaldehyde (UF) resins adhesive formulation used to bond veneers [*Falcataria moluccana* (Miq.) Barneby & J.W. Grimes] into plywood. The effect of tobacco stalk particle (TSP) size on adhesion performance, tensile shear strength (TSS), and wood failure (wF), as well as formaldehyde emission (FE) of three-ply plywood, was investigated and compared with traditional additives such as wheat flour (WF), coconut shell flour (CS), and rice hull flour (RH). The glue-mix containing UF resins with TSP at 8% by mass was blended very well. There is no difference in the TSS and wF of plywood bonded with N-200m TSP as additive compared to commercial UF formulations; however, a significant decrease of TSS and wF was observed with N-100m. The addition of N-200m significantly reduced FE by 23%, 33%, and 44% compared to those of WF-CS, WF-RH, and N-100m, respectively.

Keywords: extender, filler, formaldehyde emission, *Nicotiana tabacum* L., nicotine, plywood, tobacco

INTRODUCTION

Tobacco (*Nicotiana* spp.) is one of the highly valued crops in the world that provides job to farmers in rural areas and contribute significantly to the national economy of many countries (Rodgman and Perfetti 2013). It is an annual plant with a stalk height that ranges from 0.8–2.5 m and contractually grown for the cigarette manufacturers by the farmers for its high-grade quality leaf (NTA 2019). The harvesting of mature tobacco leaves involves cutting them off from the fibrous stalk. In the Philippines, about 900 kg/ha of tobacco stalks are either left to rot on the ground, burned in the field, or used as household fuel [Roberto R. Bonoan, National Tobacco Administration (NTA), pers. comm. 2018]. Considering the large volume of tobacco stalks, it is worthwhile to process them into additives for plywood adhesive to add value and provide an additional source of income for the farmers.

*Corresponding Author: jpjimenez4@up.edu.ph
juanito.jimenez@fprdi.dost.gov.ph

The stalk of tobacco consists mostly of 30–40% cellulose and hemicelluloses, 5% lignin, and 40% extractives composed of inorganic salts, tannins, pectins, starch, alkaloids, *etc.* (Hepworth and Vincent 1998; Li *et al.* 2008). Several studies to add value to tobacco stalks were done such as in the production of pulp and paper (Agrupis *et al*. 2000; Shakhes *et al* 2011; Gao *et al*. 2012), energy (Martin *et al.* 2002; Putun 2007; Pesevski *et al.* 2010; Mijailovic *et al.* 2014), composites (Castro *et al.* 1998; Acda and Cabangon 2013), nanocarbons (Garwe *et al*. 2018), and in wastewater treatment to remove heavy metals (Li *et al.* 2008).

Alternative use of tobacco stalks as a potential additive in the glue formulation of UF bonded plywood to replace WF-extender and CS-filler was studied by Jimenez and Acda (2018). The main focus of the previous study was on determining the amount or percentage (4, 8, and 12% by mass) of ground tobacco stalks with particle sizes of < 74 µm (passing 200 mesh) from the native type tobacco (Batek) that can replace WF (extender) and CS (filler) in the glue-mix without affecting the bond shear strength. It turned out that 4–8% of tobacco stalks additive could function as both filler and extender since it replaced WF and CS. In general, plywood additives can be classified as either extender or filler. An extender is a proteinaceous-amylaceous substance with some adhesive properties added to reduce the amount of the primary binder (Pizzi and Mittal 2003). Fillers are generally lignocellulosic materials added to the glue-mix to improve working properties, performance, strength, and other qualities of glue-mix (Skeist 1990; Sellers *et al.* 2005). In plywood manufacture, fillers fill holes and irregularities in the cracked, ruptured veneer surface, thus decreasing their porosity (Sellers 1985).

In the present paper, the focus is on using the 8% tobacco additive and determining the effect of particle size on the bond strength as well as FE since this had not been done previously. It is a gap-filling research to better understand the effect of the tobacco particles additive in the UF-glue mix.

As production of < 74 µm particle size of tobacco stalks would entail greater energy cost in milling, the NTA suggested to study the effect of particle size on the bond strength as well as FE of the UF bonded plywood. Formaldehyde is classified as carcinogenic to humans based on a meeting in Lyon, France on 02–09 Jun 2004 by the International Agency for Research on Cancer – World Health Organization (IARC-WHO 2006). Such recognition prompted studies on using various plant parts rich in protein and polyphenolic compounds like tannin and pectins to lower the FE of plywood or other wood-based panels bonded by UF adhesive. Examples of these researches are using mimosa bark extracts (Nemli *et al.* 2004), spent tea leaves (Shi *et al.* 2006; Mari *et al*. 2018), stone pine (*Pinus pinea* L.) cones (Buyuksari *et al.* 2010), walnut/almond shells (Pirayesh *et al.* 2013), and giant ipil-ipil [*Leucaena leucocephala* (Lam.) de Wit] seed flour (Chan 1996).

The desire to lower the FE of plywood or other wood-based composite panels bonded by UF adhesives is in compliance with the recent requirement set by the Bureau of Philippine Standards – Department of Trade and Industry (BPS-DTI). The FE limits set by BPS-DTI is parallel to the national requirements of foreign countries such as Japan, Australia, New Zealand, USA, and those in Europe. Table 1 shows the Philippine National Standard on FE limits for plywood.

Table 1. Grade of formaldehyde emissions (FEs).

Grade label	Average value (mg/L)	Maximum values (mg/L)
FE ★★★★	0.3	0.4
FE ★★★	0.5	0.7
FE ★★	1.5	2.1
FE ★	5.0	7.0

Source: PNS 2103:2017

The present paper reports on the use of ground tobacco stalk as both filler and extender in the glue-mix with formaldehyde scavenging property.

MATERIALS AND METHODS

Preparation of Tobacco Stalks

Stalks of native type (*Nicotiana tabacum* L.) Batek (23–34 mm diameter, 1.1–1.5 m in length) were provided by the NTA through their branch office in Rosales, Pangasinan, Philippines. The air-dried tobacco stalks with pith and bark were mechanically chipped into 25–40 mm length, hammermilled to pass through a 4-mm metal screen and further dried using a furnace-type rotary biomass dryer at 70–90 °C to bring the moisture content (MC) to 8% (dry basis). The dried tobacco stalks were hammermilled again to pass through 40 mesh woven stainless wire screen. The flour-like particles were then mechanically sieved through three stages of vibrating screens with mesh # 50, 100, and 200. Particles that pass through 100 mesh (149 µm) screen but retained in 200 mesh (74 µm) (code: N-100m) and those that passed through 200 mesh (< 74 µm) (code: N-200m) were used as additives in this study.

Analysis of Biomaterials

Samples of ground stalks and other additives used in plywood manufacture were obtained from plywood factories. These were analyzed for crude protein following the semi-micro Kjeldahl method (AOAC 978.04), crude fiber (AOAC 978.10), and ash content at 600 ºC (AOAC 930.05). Bulk density of all additives was determined using the ratio of the mass of tapped particles that occupied a 1000-mL glass cylinder. The MC of particles was obtained using a Mettler Toledo moisture analyzer (HC 103) while the pH was measured using a portable pH meter (Mettler Toledo S2). The pH of tobacco stalks, CS, RH, and WF was analyzed using the procedure of Hegde and Nanukuttan (2017). Particle sizes of additives were measured using a Phenom XL desktop scanning electron microscope (Phenom-World BV, The Netherlands) mapping at 10–15 kV.

Wood Veneers

Rotary cut *Falcataria moluccana* (Miq.) Barneby & J.W. Grimes veneers were obtained from Orgon Wood Industries, Inc., a veneer and plywood manufacturing plant in Butuan City, Agusan del Sur, Philippines. Straight grain, defect-free veneers (core, 3.0 mm x 300 mm x 300 mm and outer ply, 1.0 mm x 300 mm x 300 mm) with MC of 8% (dry basis) were used for this study.

Adhesive Formulation and Plywood Production

The two TSP N-100m and N-200m were incorporated in the adhesive formulation of a commercial UF resin (Ures® R21-035, 45% resin solids, viscosity 110–300 mPas, specific gravity 1.20, pH 7.6 and 0.5% free formaldehyde) from RI Chemical Corporation, Pasig City, Philippines. The resin reaction was catalyzed with 1.2 % ammonium chloride (NH_4Cl). Formulation with WF (extender) and CS or RH (filler) were used as controls as these are the usual additives used in commercial adhesive formulation in plywood manufacture in the Philippines. Glue mix pH and viscosity after preparation were determined using a portable pH meter (Mettler Toledo S2) and Krebs Viscometer (TQC VK 2000 DV1300), respectively. Viscosity was continuously measured up to 90 min with a reading interval of 5 min using a 220 mL glue-mix placed in a 400-mL beaker.

Three-ply plywood panels measuring 30 cm x 30 cm were fabricated in the laboratory containing 8% TSP by mass of glue-mix (Table 2). This glue-mix formulation was based on the previous study of Jimenez and Acda (2018). A formulation used for commercial plywood production was also prepared as control with the reduced percentage by mass of additives to match the composition of additives in the glue-mix with TSP. All adhesive formulations used in this study were maintained at 35.86% resin content. The glue was applied using a hand roller spreader on both sides of core veneers at a spread rate of 200 g m^{-2}. The veneers were weighed before and after adhesive spreading to determine the exact amount of adhesive used. The panels were consolidated using a hydraulic hot press (120-ton capacity, Kitagawa Seiki Co., Ltd.) maintained at 115 ºC under specific pressure of 10 kg/cm^2 for 2.6 min. The total assembly time was 15 min. After pressing, the panels were conditioned for 2 wk at 23 ºC and 65% relative humidity then trimmed prior to various property testing. Ten (10) replicate panels were made for each adhesive formulation.

Table 2. Glue-mix formulations containing various types of additives and different tobacco particle sizes used for plywood manufacture.

Components	% composition by mass			
	WF-CS	WF-RH	N-100m[a]	N-200m[b]
UF resin (45% solids)	79.7	79.7	79.7	79.7
Hardener (NH_4Cl)	1.2	1.2	1.2	1.2
Wheat flour (WF)	6.4	6.4	0.0	0.0
Coco shell flour (CS)	1.6	0.0	0.0	0.0
Rice hull flour (RH)	0.0	1.6	0.0	0.0
Tobacco stalk particles (TSP)	0.0	0.0	8.0	8.0
Water	11.2	11.2	11.2	11.2
Mix total	100.0	100.0	100.0	100.0
Glue mix resin content	35.86	35.86	35.86	35.86

[a]N-100m (particle size is 74–149 μm)
[b]N-200m (particle size is < 74 μm)

Plywood Testing

Bond shear strength. Ten (10) shear test specimens were cut from the middle of five randomly selected replicate panels for a total of 50 shear specimens per adhesive formulation. The standard shear test specimens (25 mm x 85 mm) specified in PNS-ISO 12466-1 (2016b) were used for dry (DST) and wet (WST) shear tests evaluation. Half of the 50 was used for the dry shear test (odd-numbered specimens) while the other half was used for the wet shear test (even-numbered specimens). Cut DST specimens were conditioned for 48 h after preparation in a room at 23 ºC and 65% relative humidity prior to shear testing. Specimens for WST were soaked in water maintained at 20 °C for 24 h. After soaking, the specimens were paper tissue-dried and tested while wet. The tension shear strength of each specimen was measured using a 5 kN Universal testing machine (AGS-X, Shimadzu, Japan) at a crosshead speed of 5 mm min^{-1}. The load was continuously recorded using data acquisition software

(Shimadzu Trapezium X version 1.4.2 software) running on a desktop computer. The percentage of cohesive wFs on broken shear specimens for DST and WST was estimated using PNS-ISO 12466-1 (2016b). Data was fitted in a completely randomized design with subsamples and evaluated by single-factor analysis of variance using SAS® Analytics Pro software. Means of shear strength were separated by Tukey's Honest Significance Difference test ($\alpha = 0.05$).

FE values. FE was determined according to the 24-h desiccator method specified by PNS ISO 12460-4 (2016a). This procedure is one of the approved methods for determining the FE of plywood or composite panels. Six specimens (50 mm x 150 mm) per experimental panel were cut for a total of 30 specimens from the five panels. The 30 FE specimens were randomly distributed to the three replicate desiccators per treatment (UF mix). The FE specimens were secured in wire holders set up and allowed to be conditioned for 7 d in a room at 21 °C and 65% relative humidity. After 7 d, each set up was assigned to their respective desiccators with three replicates per UF mix. Desiccators containing no specimens were used as control. A glass dish with 300 mL of distilled water was placed at the bottom of each 11-L glass desiccator. The wire holder with the FE specimens was placed above the glass dish. The set up was kept at 20 °C for 24 h to allow the water to absorb the formaldehyde released from the specimens. The formaldehyde solution contained in the glass dish was treated with acetylacetone-ammonium acetate solution prior to the determination of formaldehyde at 412 nm using a UV spectrophotometer (UV-1700, Shimadzu, Japan).

Table 3. Physico-chemical properties of tobacco stalk, wheat flour, coco shell, and rice hull particles used as an additive for UF bonded plywood.

Parameters	Type of additives			
	Tobacco stalk	Wheat flour	Coco shell	Rice hull
Crude protein, %	11.4	10.1	0.96	5.65
Crude fiber, %	40.5	0.38	55.8	29.4
Ash, %	9.77	0.64	1.51	17.5
Bulk density, kg m^{-3}	260[a] 278[b]	598	470	459
Moisture content, %	12.95[a] 12.98[b]	13.05	9.53	9.33
pH	7.39[a] 7.49[b]	4.95	5.55	5.01

[a] N-100m (particle size is 74–149 µm)
[b] N-200m (particle size is < 74 µm)

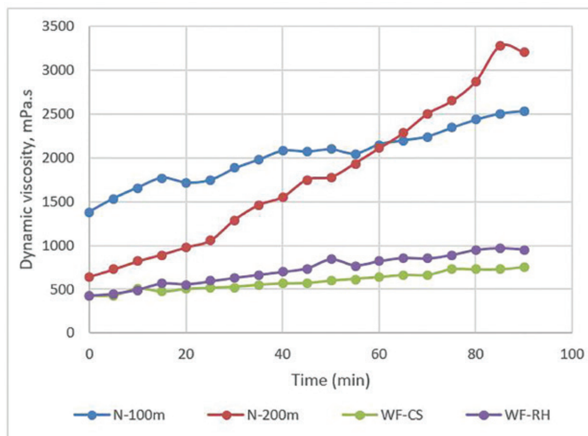

Figure 1. Time-viscosity dependence of the UF adhesive mixtures.

RESULTS AND DISCUSSION

Formulation of Urea Formaldehyde Mix with Various Additives

The glue formulations containing various kind of additives (Table 3) at 8% level by mass blended very well with the UF resin resulting in a light to dark brown glue-mix that is slightly acidic (pH 6–6.5) with satisfactory working viscosity (425–1389 mPas) after mixing. A plot of viscosity over time (Figure 1) was studied to determine the glue-mix gelling time. The dynamic viscosity graph shows that in general, viscosity increases over time due to increased polymerization of the UF adhesive in the presence of the catalyst and the other additives in the glue-mix.

It was observed that the formulations using WF-CS and WF-RH had lower viscosity and still spreadable after the 90-min stand as compared to formulations with only tobacco flour. The acceptable industry standard is only up to 1500 mPas. The glue-mix formulation with TSP N-200m showed that it could last 40 min before the viscosity would rise above the 1500 mPas limit while the TSP N-100m took only 5 min to reach the 1500 mPas. Thus, the glue mix with TSP N-200m should be applied at once, preferably within 30 min.

High viscosity – in the case of N-100m, as compared with N-200m – is probably caused by the large surface of the ground tobacco stalks capable of absorbing and binding the water in the glue-mix. Figure 2 shows the scanning electron micrograph (SEM) and relative sizes of the particles. The SEM images D and E show the sizes of the tobacco particles used in the glue mix of N-100m while SEM image F pertains to the sizes of N-200m.

The glue-mix combination of WF-CS and WF-RH had lower viscosity than the mix with TSP because it had

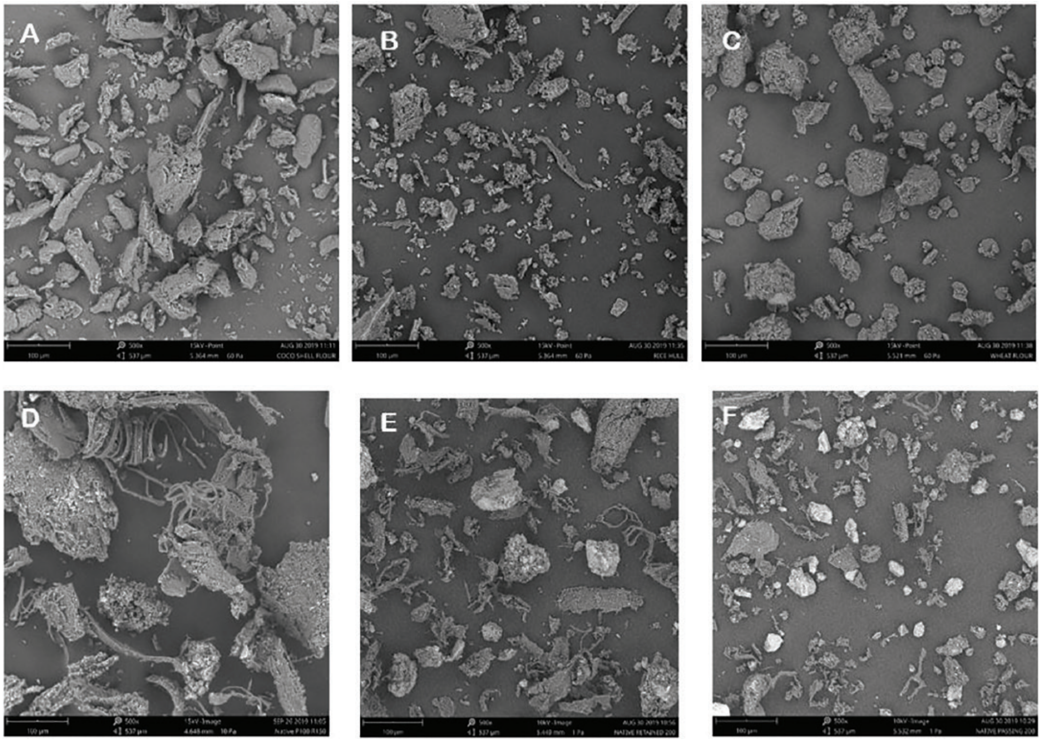

Figure 2. SEM at 500X of different additives used in the glue-mix. A) CS, B) RH, C) WF, D) native tobacco particles (NTP) passing 100 mesh retained 150 mesh, E) NTP passing 150 mesh retained 200 mesh, F) NTP passing 200 mesh

more extender (6.4%) than filler (1.6%). In addition, their bulk density was higher and particle sizes were smaller than the N-100m; hence, a lesser volume of materials was added. Further, WF is slightly soluble in water (Saeid *et al.* 2015), thus resulting in lower viscosity for the combined extender-filler additive. The addition of more water is recommended to lower the viscosity of the glue-mix formulation with TSP additive. This, however, would need further testing to determine the plywood properties.

Bond Shear Strength

Veneers from *F. moluccana* were made into plywood in the laboratory. This species is one of the fast-growing, low-density wood intensively cultivated as an industrial tree plantation species in Mindanao (Alipon *et al*. 2016). It is extensively used to produce veneer and plywood in the Philippines, which accounts for 50–70% of the veneers used in plywood production (Jimenez *et al*. 2015).

Dry (DSS) and wet (WSS) shear strengths, as well as percentage cohesive wF of *F. moluccana* plywood incorporating TSP and other additives in the UF glue-mix, are shown in Figures 3 and 4. There is a statistically significant difference among the DSS of plywood specimens and the kind of UF mix additive in the formulation (F = 2.24, p-value = 0.0378). The UF mix with N-200m additive has a DSS that is comparable with

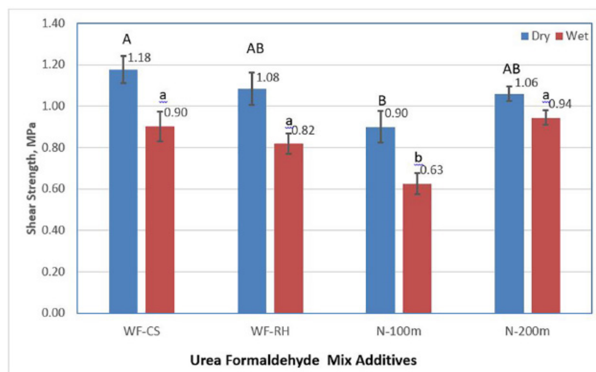

Figure 3. DSS and WSS of UF resin with various mix additives.

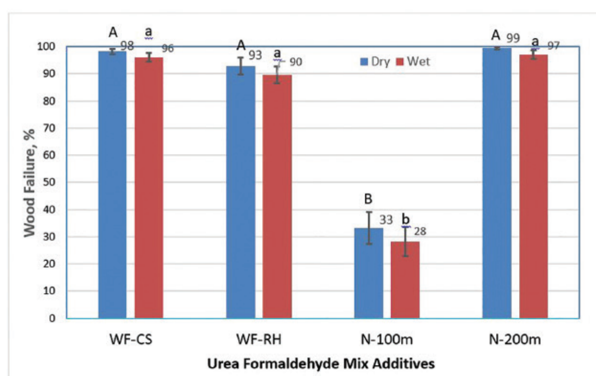

Figure 4. Dry and wet wF of UF resin with various mix additives.

the two control formulations (WF-CS and WF-RH). In contrast, the UF mix with N-100m additive has a DSS that is lower than all the UF mixes. With WSS, the UF mix with N-200m was again comparable with the controls but the N-100m was found to be significantly lower than all the UF mixes (F = 8.26, p-value < 0.0001).

The significant reduction in WSS of N-100m may be attributed to two factors. First is the weaker cohesion of UF adhesive with N-100m TSP as compared with N-200 m TSP. The larger particles probably caused gap or weak points in the UF resin cohesion and polymerization, hence resulting in lower bond shear strength of the plywood. Second is the poor interface adhesion between wood veneer and the UF adhesive with N-100m TSP due to the absence of mechanical interlocking on the veneer substrate, as observed in Figure 5 where wF is very low for N-100m as compared with N-200m.

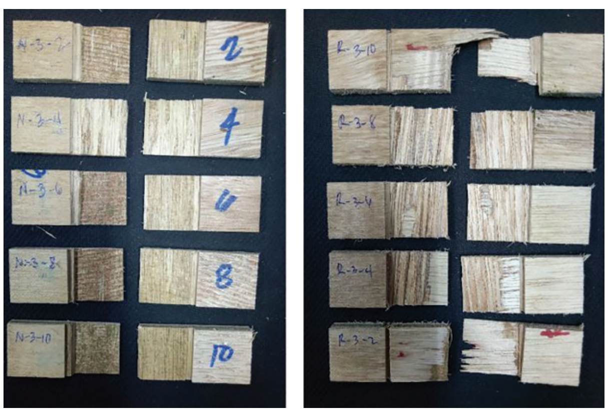

Figure 5. Cohesive wF of the Falcata plywood for N-100m (left) and N-200m (right). wF of N-100m with four test pieces shows poor adhesion caused by bigger TSPs that is even visible on the sheared veneer surface while N-200m shows 100% wF.

This observation is similar to the findings of Chen *et al.* (2018), who concluded that smaller particle size of peanut meal adhesive was beneficial to the adhesive water resistance strength as smaller particle size penetrates easily into the wood surface for the improvement of adhesive strength. Due to the larger particle size of N-100m, the glue spread unevenly in some areas of the veneer (Figure 6), which resulted in a weak bond owing to the thick glue line and agglomeration of the particles. In addition, the difficulty in spreading is due to higher initial viscosity of the glue-mix for N-100m, as previously shown in Figure 1.

This becomes evident with the evaluation of wF for both the DSS and WSS tests (Figure 4) where only the N-100m UF mix did not pass the glue line requirement of PNS-ISO 12466-2 (2016c) for apparent percent cohesive wF. At least 40% wF should be attained by the panels bonded with N-100m UF mix for it to pass the standard. The

Figure 6. N-100m (left) shows the agglomerated TSP (yellow oval) with glue spreading difficulty causing uneven glue spread. N-200m (right) showing uniform and even spread of the glue-mix with TSP.

dry and wet wF also show that the N-100m UF mix was significantly different from all the UF mixes (dry wF: F =45.93, p-value <0.0001; wet wF: F =45.33, *p-value* <0.0001).

The result of the bond test confirmed the earlier study of Jimenez and Acda (2018) that particles (< 74 µm) from native type (Batek) of tobacco stalk could be used as an additive replacement for WF and CS or RH flour in the UF glue-mix. The intention of the present paper to study the larger particle size (74–149 µm) of tobacco stalk as an additive in the UF glue mix was not technically feasible due to lower bond strength and poor wF owing to a thick glue arising from the agglomeration of the particles. Several studies have shown that thicker glue line could result in poor bond strength (Ramazan 2006; Hajdarević and Šorn 2012; Jimenez and Natividad 2019). According to Marra (1992), there is a strong relationship between glue line thickness and loss of joint strength – as glue line thickens, glued joint decreases in strength.

FE

The plywood specimens with various additives in the UF mixes had significantly different FE (F = 29.07, p-value = 0.0001). Figure 7 shows that the particle sizes of tobacco stalks affect FE. The plywood with N-100m ground tobacco stalk has higher FE probably because the large-size particle absorbed much of the water in the

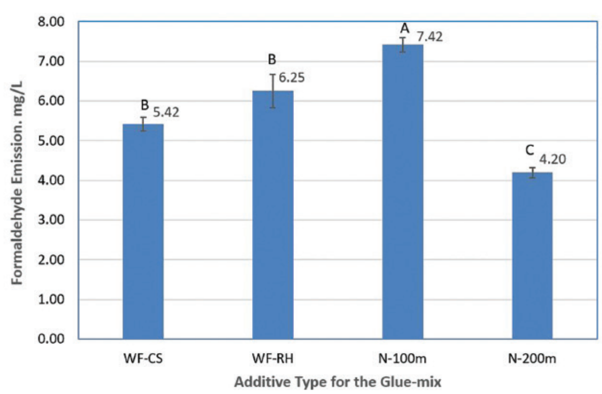

Figure 7. FE of UF bonded plywood with various mix additives.

glue-mix. This action resulted in lesser water in the mix, which might have caused an increase in FE similar to the findings of Réh *et al.* (2019).

The TSP N-200m has the lowest FE (4.20 mg/L) among the various additives and is the only one that passed the minimum requirement for FE of PNS 2103:2017 of having less than 5 mg/L of FE. The FE reduction by the tobacco additive compared with the traditional additive was 23% (WF-CS) and 33% (WF-RH). Relative to the larger particle size of the tobacco additive, the reduction was 44% (N-100m). The lower FE of the N-200m tobacco particles may be attributed to the capacity of smaller particles to block the tiny passages within the cellular structure of the plywood to prevent excess formaldehyde gas from escaping. Further, it could be hypothesized that the smaller tobacco particles (N-200m) are more reactive because they are more numerous than the N-100m and the reduction in its size enables increased exposure of more functional groups, making it possible for the nitrogenous (protein and alkaloids) compounds to adhere and chemically bond with the excess formaldehyde. The crude protein content (11.4%) of tobacco stalks, as per analysis shown in Table 3, was the highest among the other additives (WF = 10.1%; CS = 0.96%; RH = 5.65%). Wang and Pizzi (1997) and Hoffman *et al*. (2015) showed that nitrogenous compounds such as protein can scavenge the excess formaldehyde and integrate it into its chemical structure. Glue-mix additives with high protein content such as spent tea leaves (Mari *et al.* 2018) and giant ipil-ipil [*Leucaena leucocephala* (Lam.) de Wit] seed flour (Chan 1996) had been shown to significantly reduce FE of UF- bonded plywood.

The N-200m, although having almost similar particle sizes with WF and RH (Figure 8), resulted in lower FE because more filling materials are present in the glue-mix (with only tobacco additive) than the WF-RH and WF-CS formulations. At the same amount or quantity of ground tobacco stalk additive in the glue-mix, the smaller particle size (16–71 µm) of N-200m are relatively more numerous than the bigger particle size (45–172 µm) of N-100m. In other words, when the glue-mix is spread on the surface

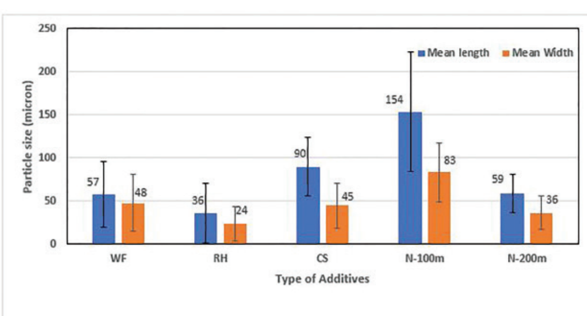

Figure 8. Particle size of different additives.

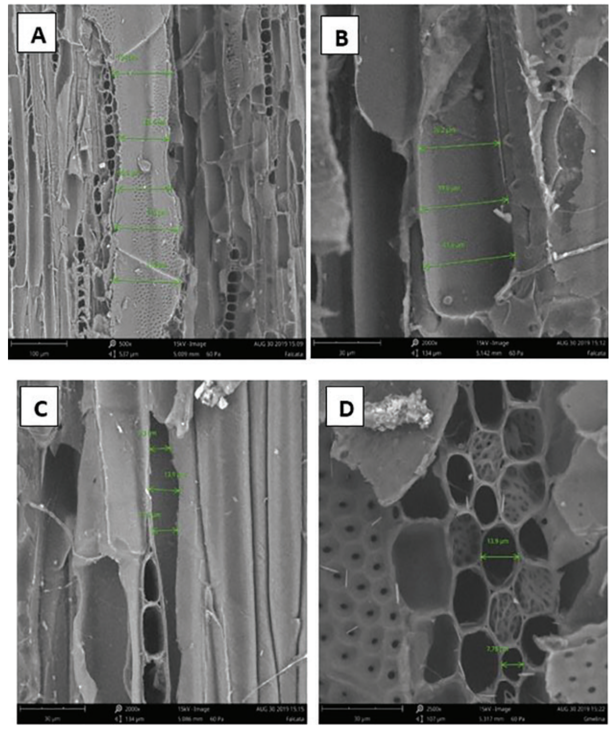

Figure 9. Measurements of some cell cavities in the veneer used in the study. A & B) vessel, C) fiber, D) ray cell.

of the veneer, the smaller particle size N-200m can cover a wider or bigger area than the larger N-100m particle size. In effect, it can perhaps presumably cover and block the excess formaldehyde gas from escaping through the tiny cavities within the veneer cells. Measurements of the cavity diameter (Figure 9) of the vessels, fibers, and ray parenchyma cells range from 36–118 µm, 9–14 µm, and 8–14 µm, respectively. During pressing of the plywood assembly, the TSP of the N-200m sizes can easily fit or be forced to fit into the tiny cavities of the wood cells. The N-100m larger particles can cover the minute cavities of the cells wholly but it will not fit in the minute cavities since it is larger than the cavities of the wood cells. Also, N-100m is lesser in numbers or quantity and cannot cover a bigger surface area of the veneer as compared to the N-200m.

CONCLUSIONS AND RECOMMENDATIONS

The present study shows that TSP additives consisting of small particle sizes (N-200m) produce comparable shear strength and wF with the combination of traditional extenders such as WF, CS, or RH, and effectively passed the requirements of PNS-ISO 12466-2 (2016c). By contrast, the larger particle size of tobacco particles

(N-100m) was not comparable and did not meet the bond quality requirements of the standard. FE also showed that smaller tobacco particles could effectively lower the emission to an acceptable level of the PNS 2103:2017 standard. Although good results were obtained, further research is needed in the UF glue-mix formulations with tobacco particle additives. Viscosity needs to be adjusted to increase the working time of the mix as production in the factory is sometimes delayed. The addition of more water in the formulation with TSP additive is recommended to lower the viscosity of the mix. Also, the addition of tobacco leaf dust may be tried in combination with the TSP. This, however, would need further testing to determine the plywood properties.

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