

Penetration and Adhesion Strength of Phenol-Tannin-Formaldehyde Resin Adhesives for Bonding Three Tropical Woods

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Abstract

Phenol-tannin-formaldehyde (PTF) resins were prepared by adding bark tannin extracts from mangium (*Acacia mangium*) to phenol-formaldehyde (PF) resin adhesive. The resultant resin was used for bonding wood from three fast-growing tropical species, specifically, mangium, jabon (*Anthocephalus cadamba*), and mindi (*Melia azedarach*). The impact of the tannin extract content on the penetration and adhesion strength of PTF resin adhesives was studied for two different tannin contents (10% and 20%). The resin penetration into wood tissues of the three species was quantitatively measured at the bond line, using a light microscope. Confocal laser scanning microscopy was also used for detecting the resin penetration into wood tissues. The adhesion strength of the PTF resins was determined by measuring shear strength of three-ply parallel plywood constructed from the three tropical woods. As the amount of tannin increased, the depth of resin penetration increased for mangium and jabon wood but decreased for mindi wood. By contrast, the bond-line thickness decreased with the increasing tannin addition for all three wood species. The adhesion strength of plywood was improved as the amount of tannin increased. All plywood bonded with PTF resins using 20 percent tannin met the requirements of the EN 314-2 standard. These results suggest that the addition of tannin extract into PF resin improves the adhesion performance and can partially replace phenol in PTF resins.

The use of adhesives for bonding wood has played a vital role in the production of forest products such as glued laminated timber (glulam), laminated veneer lumber, oriented strand board, and other engineered wood composites. The flow of adhesive over the surfaces and its penetration into the structure of wood are very important because the extent of resin penetration and the bond-line thickness greatly influence the performance of the resultant adhesion. Additional factors, such as wood species, wood density, grain direction, surface wettability, and surface-free energy, also affect adhesive penetration characteristics (Gavrilović-Grmuša et al. 2012). Microscopic techniques are widely used to assess adhesive penetration into larger cell voids, such as the lumen, cell wall cracks, or microcapillaries. Several types of microscope and associated techniques have been used for this purpose, including light microscopy (LM) (Gindl et al. 2005; Singh et al. 2008, 2010; Gavrilović-Grmuša et al. 2012) and confocal laser scanning microscopy (CLSM) (Singh et al. 2008, Gavrilović-Grmuša et al. 2012, Nuryawan et al. 2014). Santoso et al. (2016) reported that merbau (*Instia* spp.) wood extracts contained flavonoid oligomers with up to six repeating units

and that the extracts could be used as a part of the adhesive for glulam manufacturing, especially with medium-density woods. Testing of the physical and mechanical properties of three-ply composite flooring bonded with merbau tannin adhesive showed that the flooring had high performance as a building product and that the shear strength was equal to phenol-resorcinol formaldehyde resin adhesives with low formaldehyde emission (Santoso et al. 2014). Mahogany (*Swietenia* spp.) tannin adhesive had similar adhesion in glulam manufacturing (Lestari et al. 2015). Mangium (*Acacia mangium*), which has a high concentration of

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polyphenolic compounds (34.04%) (Hendrik et al. 2016), could be appropriate for use as a fortifier in phenol-formaldehyde (PF) resin adhesives. Neither Lestari et al. (2015) nor Hendrik et al. (2016) used PF adhesive; only tannin adhesive was used as an adhesive by mixing 10 mL of formaldehyde per 100 mL of tannin extract, with a mixture time of 15 minutes. However, attempts to quantitatively measure the penetration of PF resin adhesives containing different amounts of mangium tannin extract are very limited.

The use of tannin extract for PF resins requires formulating proper viscosity and resin penetration for a strong bond line to be formed (Park and Kim 2008, Abdullah and Park 2009). In general, high-viscosity resins have poor penetration into wood, but low-viscosity resin can cause a starved bond line to form because of excessive resin penetration. In the current study, the extent of penetration of phenol-tannin-formaldehyde (PTF) resin adhesives with different amounts of tannin added was quantitatively evaluated using LM in combination with image analysis processing. CLSM was also used to confirm the presence of resin penetration in the bond line and assess the depth of resin penetration. The adhesion performance of these PTF resins was also investigated by measuring shear strength of three-ply plywood.

Materials and Methods

Materials

Tannin extract was obtained from mangium bark, following a previously reported extraction procedure (Hendrik et al. 2016). Tannin extract was obtained from chipped mangium stem bark through hot water extraction in the range temperature of 70°C to 80°C.

Methods

Characteristics of tropical woods.—The density of wood from three fast-growing tropical wood species was determined by measuring the GARDNER by oven dry and volume of wood samples at room temperature. The pH and buffering capacity were determined by the method described by Johns et al. (1985). Aqueous wood extract solutions were prepared by refluxing 25 grams of ground wood at 100°C for 20 minutes. After extraction, aqueous solutions were filtered and stored in sealed Erlenmeyer flasks; measurements were performed within the next 24 hours after extraction using a digital pH meter (HANNA model HI 9025 C; Hanna Instruments, Ronchi di Villafranca, Italy). For pH and acid buffering capacity measurements, 60 mL of aqueous solution was analyzed at 20°C. Afterward, the aqueous solution was titrated to pH 3 with HCl 0.025 N. Then, a new beaker with another 60 mL of the original aqueous solution was prepared for a second reading of pH and then titrated to pH 7 with 0.025 N NaOH for alkaline buffering capacity measurement.

Synthesis of PTF resin adhesives.—For PF resin preparation, liquid phenol (90%) and paraformaldehyde were used as supplied. PF resins were synthesized in a 500-mL reaction kettle according to the following process. The kettle was charged with phenol (90%) and paraformaldehyde (45.6%) at a formaldehyde/phenol (F/P) molar ratio of 2.0:1.0 under continuous stirring. After heating the components to 40°C in the kettle, sodium hydroxide (50%) was slowly added over 10 minutes to achieve a NaOH/phenol

ratio of 0.3. When the temperature reached 90°C, the reaction was maintained for 2 to 3 minutes before the tannin extract was added. The kettle was cooled to 65°C and then kept for 65 minutes. The temperature was held at 65°C until the viscosity reached 50 to 65 cps. The viscosity was measured with Gardner-Holdt viscosity at 25°C. Liquid PTF resin adhesives with three different tannin additions (0%, 10%, and 20%) were prepared.

Characterization of PTF resins.—The viscosity of PTF resin adhesives was measured using a cone-plate viscometer (DV-II+; Brookfield Engineering, Middleboro, MA, USA) with a No. 2 spindle at 60 rpm at 25°C. Solid content was measured by referring to ASTM D4426-01 (American Society for Testing and Materials 2013) by drying 1 g of the PTF resin in an oven at 103°C ± 3°C until a constant weight was reached. The gel time of PTF resins was measured at 120°C in triplicate for each resin using a gel time meter (Davis Inotek Instrument, Charlotte, NC, USA). The gelation time was measured using viscosity–time data obtained at a constant temperature.

Method of pyrolysis gas chromatography/mass spectrometry analysis.—Pyrolysis gas chromatography/mass spectrometry (GC-MS) (GC6890MS5973; Agilent, Santa Clara, CA) was used to determine specific compounds in PTF resins. The analysis was carried out on a fused silica capillary column (HP-5MS column, 60 m by 0.25 mm with a film thickness of 0.25 µm; Agilent). About 1 µg of resins was inserted without any further preparation into the bore of the pyrolysis solids injector and then placed with the plunger on quartz wool in the quartz tube of the furnace pyrolyzer Pyrojector II (S.G.E., Melbourne, Australia) with a constant temperature of 700°C and a total run time of 20 minutes. The pressure of the helium carrier gas at the inlet to the furnace was 95 kPa. The pyrolyzer was connected to a 7890A gas chromatograph with a series 5975C quadrupole mass spectrometer operated in electron impact ionization mode.

Plywood preparation.—Air-dried veneers from three kinds of fast-growing species, including mangium, jabon (*Anthocephalus cadamba*), and mindi (*Melia azedarach*) were supplied from a local plywood mill in Indonesia. Three-ply, parallel veneer plywood (10 cm by 10 cm by 4.5 mm) was prepared by spreading the PTF resin adhesives over one face of a veneer at 180 g/m² and placing the other veneer over it with the grain running parallel to the first veneer and the same way for the other glue line. The veneers were then cold-pressed with an applied pressure of 8 kgf/cm² and kept at room temperature for 20 minutes to allow the adhesive to penetrate into the wood tissues. Cold-press glue was used for avoiding bubbles during the press, and most wood used was porous so that the cold press was suitable. Finally, the cold-pressed veneers were placed in a hot press at 120°C for 4 minutes under a pressure of 8 kgf/cm².

Shear strength of plywood.—Shear strength of plywood was measured using a universal testing machine (Hounds Co., Redhill, UK) with a specimen size of 2.5 by 8 cm and a crosshead speed of 2 mm/min according to the procedure of the standard KS F 3101 (Korean Standards Association 2006).

Measurement of resin penetration and bond-line thickness.—In the sample preparation, three parallel veneers were bonded to measure resin penetration and bond-line thickness. The bond-line thickness is the adhesive region

between three parallel laminated veneers or the sum distance of two glue lines. Paired plywood was cross-sectioned dry along the bond line on a sliding microtome (Yamato Kohki, Asaka, Japan). Distortion-free 70- μm -thick sections were selected for LM observation. Thin sections were stained with 0.05 percent aqueous toluidine blue O, which permits clear differentiation of wood from the resin (Donaldson and Lomax 1989, Xing et al. 2005). The thin sections were quickly washed with distilled water and then placed under vacuum for 1 hour to remove air bubbles trapped within wood tissues.

Quantitative measurement bond line with LM.—The thin cross sections were mounted in air-free water on a glass slide, and LM images were digitally captured for 10 different sections, resulting in 10 replications for each experimental unit. For each image, we made 20 measurements for adhesive penetration into wood (interphase) and 10 measurements for bond-line thickness using the LM (UMDOB Olympus Optical Co., Ltd, Tokyo, Japan) with a $\times 109$ objective lens. For image processing and measurement, images in which the adhesive in the bond line was clearly distinguished were selected. For each image, the depth of resin penetration was defined by a linear distance between the interfacial contact point and the wood cells with no resin in the lumen. The depth of resin penetration was then determined by measuring the linear distance at various points along the bond line. The thickness of the bond line was determined by measuring the distance between two interfacial contact points along the bond line. Both the depth of resin penetration and the bond-line thickness were measured using image processing software (IMT solution, Vancouver, BC, Canada). The image processing software was used for image analysis. It was a part of a fluorescence microscope using visible and fluorescence modes to obtain contrast, enabling semiautomated detection of adhesive. The software gave an average value and standard deviation for each image. Figure 1 shows a typical LM image for the measurements of both bond-line thickness and depth of penetration of PTF resins.

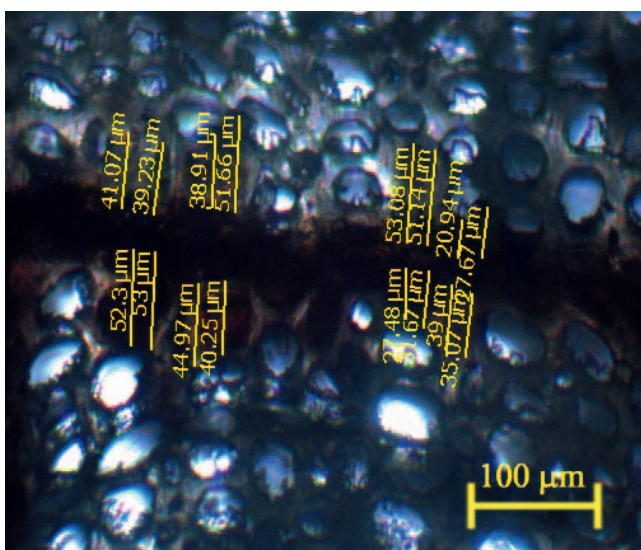


Figure 1.—A representative image from light microscopy for the measurement of bond-line thickness and depth of penetration in the cross section of Jabon wood bonded with neat PF resin.

Qualitative detection of the resin using CLSM.—CLSM (LSM 700, Carl Zeiss, Jena, Germany) at the Scientific Instruments Center of Kyungpook National University was also used to qualitatively confirm the presence of adhesive in the bond line and the resin penetration, enabling a more accurate assessment of the depth of resin penetration. For CLSM, thin cross sections (90 μm thick) cut through the wood–adhesive bond line with the same sliding microtome as for LM were stained with aqueous toluidine blue O and mounted in air-free water on a glass slide prior to examination. CLSM imaging was undertaken in fluorescence mode at two excitation wavelengths of 488 and 555 nm, respectively, and at two emission wavelengths of 420 to 550 and 560 nm, respectively. Figure 2 illustrates a typical image of CLSM for the detection of PTF resins in the bond line. The CLSM image allowed the clear detection of the resin adhesive penetrated into wood tissues.

Results and Discussion

Characteristics of three tropical woods

Table 1 presents the characteristics of the three tropical woods used in this study. Mangium wood had a higher density ($0.61 \pm 0.03 \text{ g/cm}^3$) than mindi wood ($0.54 \pm 0.02 \text{ g/cm}^3$) and jabon wood ($0.46 \pm 0.05 \text{ g/cm}^3$). The wood density of the different wood species was expected to influence the properties of plywoods made with them (Bal and Bektas 2014). Wood density and anatomy control wood porosity, which usually influences adhesive penetration and bond performance. The adhesive penetrates the wood and interlocks several cells, and at higher wood density, the penetration of the adhesive will be lower. However, chemical properties of wood also affect the adhesion, and more polar extracts facilitate adhesive penetration into wood.

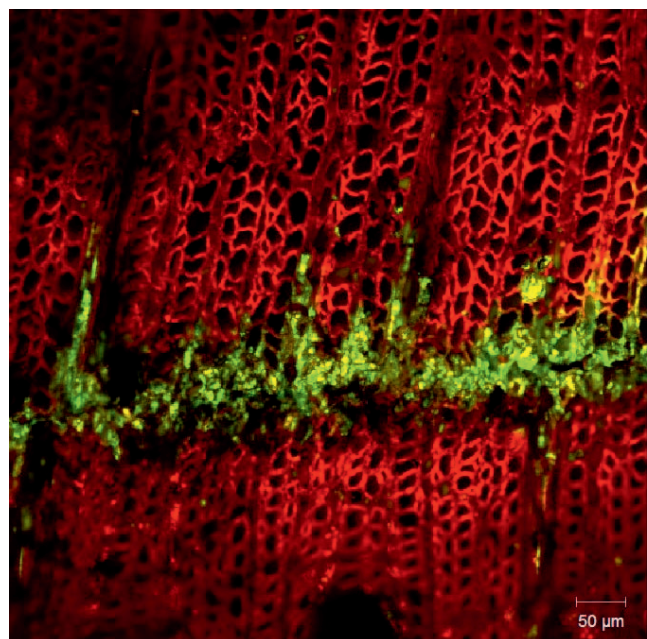


Figure 2.—A representative image from confocal laser scanning microscopy for the detection of neat PF resins in the bond-line from Jabon wood. Red color and green color represent wood and the adhesive, respectively.

Table 1.—Characteristics of the three tropical wood species used in this study.

Wood species	Density (g/cm ³)	pH	Acid buffering capacity (mL Eq/g)	Alkaline buffering capacity (mL Eq/g)
Mangium	0.61 ± 0.02	6.90 ± 0.14	8.11 ± 0.13	7.85 ± 0.07
Mindi	0.54 ± 0.02	5.41 ± 0.08	5.10 ± 0.14	7.50 ± 0.71
Jabon	0.46 ± 0.05	5.84 ± 0.11	5.55 ± 0.49	8.55 ± 0.21

The pH, acid buffering capacity, and alkaline buffering capacity of the three types of woods are also shown in Table 1. The pH of mangium (6.90 ± 0.14) was higher than the pH of jabon and mindi wood, which had similar values. Both jabon (5.84 ± 0.11) and mindi (5.41 ± 0.08) were acidic, with a pH of less than 7. The acid buffering capacity of mangium (8.11 ± 0.13) was higher than those of jabon (5.55 ± 0.49) and mindi (5.10 ± 0.14), making mangium more compatible with PF and PTF resin adhesives because the curing of these resins is accelerated under alkaline pH. By contrast, both mindi and jabon wood were expected to have lower adhesion strength because their low pH will retard the curing of these resins. The alkaline buffering capacity of jabon (8.55 ± 0.21) was higher than those of mangium (7.85 ± 0.07) and mindi (7.50 ± 0.71).

Characterization of PTF resins

Properties of PTF resin adhesives prepared.—Table 2 shows the properties of the PF and PTF resin adhesives synthesized in this study. As expected, the solid content of PTF resins increased as the tannin extract addition increased. In general, the solid content influences the moisture content plywood because water in the resin provides moisture to the veneers. The viscosity of the PTF resins decreased with an increase in the tannin extract addition, and it was lower than that of neat PF resins. This outcome is also due to the water in the tannin extract. Interestingly, the addition of tannin extract into neat PF resin resulted in a short gel time, even though the solid content and viscosity of PTF resins decreased. The shorter gel time of PTF resins reflects higher reactivity than that in the neat PF resin, which results in faster curing and a reduction in the resin's shelf life. Tannin adhesives are known to polymerize faster in a high-pH medium because both the tannin flavonoid units take part in the curing reaction, accelerating curing (Pizzi 1983). In fact, tannin compounds extracted from bark have been widely used as an accelerator for PF resins in the production of particle-board and plywood (Trosa and Pizzi 1997, 2001) because the tannin can reduce both gel time and pressing time.

The addition of mangium tannin extract decreased pH, gel time, and viscosity of PTF resins and increased the solid content of PTF resins. Similar results have been reported for PF resin adhesives, containing 10, 20, and 30 percent derived tannin from *Quercus castaneifolia* (Jahanshahi et al. 2012).

Pyrolysis GC-MS analysis of PTF resin adhesives prepared.—Pyrolysis (Py) GC-MS was employed to understand the chemical species of PTF resins, and a representative Py GC-MS result of neat PF resin is presented in Figure 3. Chromatographs of PTF resins with 10 and 20 percent of tannin extract are presented in Figures 4 and 5,

Table 2.—Properties of the adhesive.

Tannin extract addition (%wt)	pH	Solid content (%wt)	Gel time (s)	Viscosity (mPa/s)
0	10.53 ± 0.01	36.5 ± 0.6	454 ± 20	171.7 ± 0.0
10	10.39 ± 0.01	37.8 ± 0.4	383 ± 51	146.7 ± 0.0
20	10.03 ± 0.01	38.1 ± 0.1	231 ± 26	106.7 ± 0.0

respectively. Table 3 shows the Py GC-MS results of PTF resins containing 0, 10, and 20 percent tannin extract. All Py GC-MS analysis yielded similar results, indicating that phenolic compounds were the dominant components of the adhesive. As the tannin extract increased from 0 to 20 percent in the PTF resin, the phenolic compounds obtained by the Py GC-MS increased from 75.07 percent (w/w) to 77.97 and 75.99 percent. The presence of *o*-cresol in PTF resin was assumed to decrease the pH of the resins because of the addition of –OH functional groups.

Depth of penetration and bond-line thickness of PTF resin adhesive.—The depth of penetration and bond-line thickness of neat PF and PTF resin adhesives are shown in Table 4. Plywood made of mangium wood had a greater depth of penetration (102.51 μm) than the plywood made of jabon and mindi (95.14 and 61.18 μm, respectively). The penetration of adhesive in mangium and jabon wood was greater when the tannin extract addition was higher, but that of mindi wood had the opposite trend. This result was likely influenced by the acidic pH (5.41) of mindi wood (Table 1), which made the alkaline PTF resin adhesives less suitable for their bonding. In general, greater penetration results in thinner bond lines, while less penetration results in thicker bond lines (Nuryawan et al. 2014). In fact, mindi wood had lower penetration (45.63 μm) and a thicker bond-line thickness (24.80 μm) than mangium and jabon wood when the PTF resin contained 20 percent tannin extract. Resin penetration and bond-line thickness are also thought to be affected by the molecular weight of PTF resins, but we did not measure the molecular weight of PTF resin in this study. For example, it is reported that a mixture of low- and high-molecular-weight species in an adhesive provide a strong adhesion bond (Ellis 1993). In other words, a low-molecular-weight species penetrates into wood tissues, contributing to the interfacial adhesion, while a high-molecular-weight species contributes to the cohesive adhesion (Nuryawan et al. 2014). Wood density and anatomy control wood porosity, which usually influences adhesive penetration and bond performance. The adhesive penetrates the wood and interlocks several cells, and at higher wood density, the penetration of the adhesive will be lower. However, chemical properties of wood also affect the adhesion, and more polar extracts facilitate adhesive penetration into wood.

According to the analysis of variance shown in Table 5, the depth of penetration and bond-line thickness were affected by wood species and by the addition of tannin extract in PTF resins. In particular, all parameters were highly significantly different from each other. The interactions between wood species and tannin extract addition were also highly significant, except for shear strength. These results indicate that tropical wood species and tannin extract addition could be optimized for manufacturing plywood.

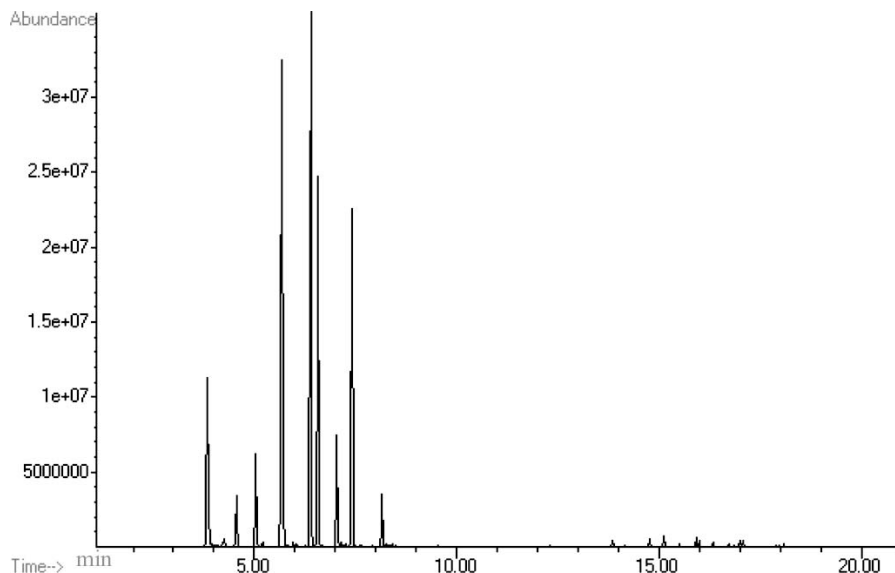


Figure 3.—Chromatogram of neat phenol-formaldehyde resins.

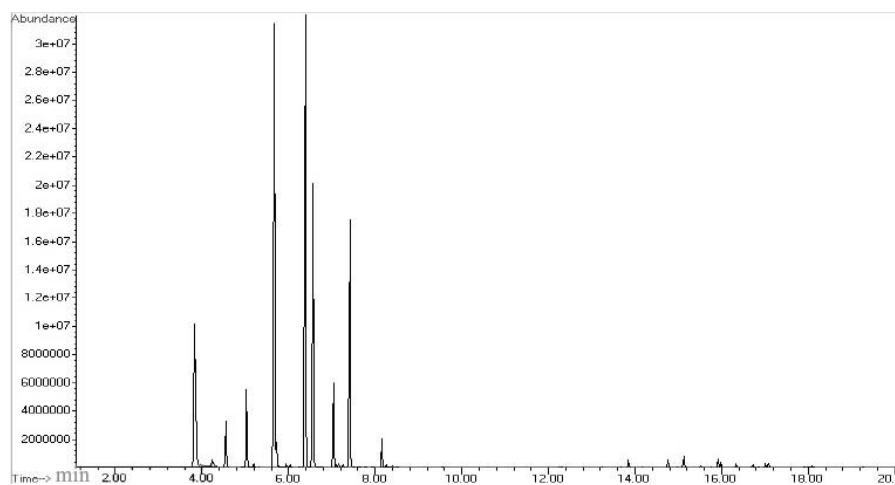


Figure 4.—Chromatogram of neat phenol-formaldehyde resins with tannin extract 10 percent.

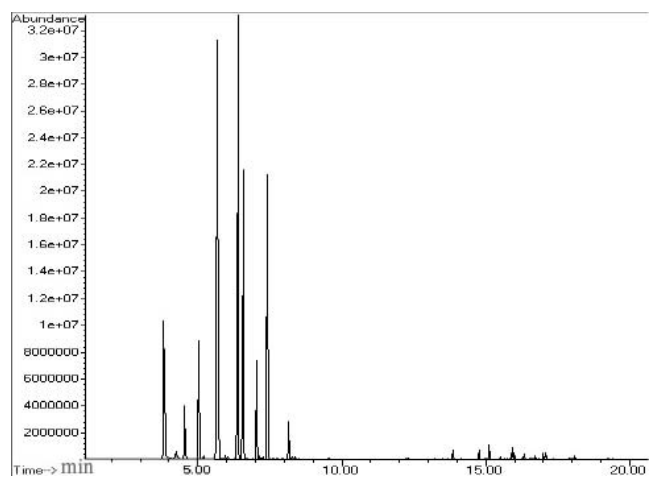


Figure 5.—Chromatogram of neat phenol-formaldehyde resins with tannin extract 20 percent.

Table 3.—Pyrolysis gas chromatography/mass spectrometry absorption bands of phenol-tannin-formaldehyde resins.

Chemical species	Tannin extract addition (%wt)		
	0	10	20
Phenol	25.65	28.67	25.84
<i>o</i> -Cresol	—	22.5	21.92
<i>p</i> -Cresol	12.35	—	12.58
Phenol, 3-methyl-	21.26	12.46	—
Phenol, 2,4-dimethyl-	11.09	10.19	11.04
Phenol, 2,6-dimethyl-	3.20	2.98	3.3
Phenol, 2,3,6-trimethyl-	1.40	1.17	1.31
Phenol, 2-ethyl-	0.12	—	—
Total phenolic compounds	75.07	77.97	75.99

Table 4.—Properties of the panel.

Wood species	Tannin extract addition (%wt)	Depth of penetration (µm)	Bond-line thickness (µm)	Plywood moisture content (%)	Shear strength (MPa)
Mangium	0	92.5 ± 0.8	30.6 ± 1.4	5.55 ± 0.22	1.16 ± 0.04
	10	101.6 ± 11.6	17.0 ± 1.5	5.24 ± 0.04	1.50 ± 0.43
	20	113.3 ± 6.7	13.8 ± 3.1	5.35 ± 0.21	1.32 ± 0.37
Mindi	0	87.2 ± 0.8	98.7 ± 1.2	6.30 ± 0.20	0.57 ± 0.36
	10	50.6 ± 3.9	15.5 ± 3.2	5.53 ± 0.20	0.96 ± 0.18
	20	45.6 ± 1.6	24.8 ± 0.7	6.45 ± 0.06	1.17 ± 0.33
Jabon	0	65.2 ± 1.8	31.7 ± 2.4	5.50 ± 0.25	0.60 ± 0.22
	10	79.0 ± 0.7	20.7 ± 2.5	5.46 ± 0.07	0.97 ± 0.31
	20	141.1 ± 8.0	10.1 ± 1.4	5.57 ± 0.01	1.05 ± 0.18
EN 314-2 (1993) standard				Max. 12	Min. 1

The additional analysis by Tukey’s test (Table 6) showed that wood species differed significantly for the depth of penetration of PTF resin adhesives into the wood tissues. Each wood species was significantly different from the others for the PTF penetration into wood because of several factors, such as porosity, pressing, concentration of tannin extract, and so forth. Furthermore, the effect of depth of PTF penetration into wood can be connected with bond-line thickness in that a greater depth of the penetration will produce smaller bond-line thickness. For jabon and mindi wood, there were no significant differences among bond-line thickness analyzed.

Table 7 shows the results of the analysis of bond-line thickness and depth of penetration of PTF into wood. For concentrations of 0 and 10 percent tannin extract added into PTF, there was an insignificant difference for the depth of penetration, whereas there was a significant difference with the tannin extract concentration of 20 percent. However, there was a significant difference of bond-line thickness for all tannin extracts added.

Table 5.—Analysis of variance of depth of penetration, glue-line thickness, and moisture content and shear strength of the panel.^a

Parameter	Wood species (A)	Adhesive (B)	Interactions (AB)
Depth of penetration (µm)	(0.00)*	(0.00)*	(0.00)*
Bond-line thickness (µm)	(0.00)*	(0.00)*	(0.00)*
Moisture content (%)	(0.00)*	(0.00)*	(0.00)*
Shear strength (N/mm ²)	(0.00)*	(0.01)*	NS (0.70)

^a * = highly significant ($P \leq 0.01$); NS = not significant at a P value of 0.05; values in parentheses indicate confidence level.

Table 6.—Multiple comparisons among wood species for depth of penetration and bond-line thickness.^a

Wood species	Depth of penetration (µm)	Bond-line thickness (µm)
Mangium	102.51 A	20.49 A
Mindi	61.19 C	46.33 B
Jabon	95.13 B	20.83 A

^a Values with the same letters are not significantly different at a P value of 0.05.

Characterization of plywood

Moisture content of plywood.—Table 4 shows that the moisture content of all plywood samples had a different value ($5.66\% \pm 0.42\%$), which met the requirement level (below 12%) of the standard EN 314-2 (European Committee for Standardization 1993). According to the analysis of variance shown in Table 4, the moisture content of plywoods was affected by the wood species and tannin extract addition. The results of Tukey’s test (Table 8) showed that mangium and jabon wood were not significantly different from each other for the moisture content of plywood, but both were significantly different from mindi wood. PTF resins with 0 and 20 percent tannin extract did not have significantly different effects on the moisture content of plywood, but both were significantly different when 10 percent tannin extract was added into PF resin.

Shear strength of plywood.—Shear strength tests for plywood samples were carried out in dry conditions. Table 4 shows that all plywood made with mangium wood and PTF resins at all levels of tannin extract addition met the requirement of the EN 314-2 standard, which is more than 1 MPa. However, plywood made of mindi and jabon wood

Table 7.—Multiple comparisons among tannin extract addition in depth of penetration and bond-line thickness.^a

Tannin extract addition (%wt)	Depth of penetration (µm)	Bond-line thickness (µm)
0	81.68 A	23.68 A
10	77.10 A	17.72 B
20	100.05 B	16.25 B

^a Values with the same letters are not significantly different at a P value of 0.05.

Table 8.—Multiple comparisons among wood species and tannin extract content in moisture content.^a

Wood species	Moisture content (%)	Tannin extract content (%wt)	Moisture content (%)
Mangium	5.38 a	0	5.78 A
Mindi	6.09 B	10	5.41 B
Jabon	5.51 A	20	5.79 a

^a Values with the same letters within a column are not significantly different at a P value of 0.05.

Table 9.—Multiple comparisons among treatments in shear strength analysis of the panel.^a

Wood species	Shear strength (MPa)	Tannin extract content (%wt)	Shear strength (MPa)
Mangium	1.33 A	0	0.78 A
Mindi	0.90 B	10	1.14 B
Jabon	0.87 B	20	1.18 B

^a Values with the same letters within a column are not significantly different at a *P* value of 0.05.

satisfied the requirement level only when the tannin extract content was 20 percent. These results show that mangium wood could be bonded with neat PF resin without tannin extract, while plywood from mindi and jabon wood should be bonded with PTF resins with at least 20 percent tannin extract. This result could be due to the presence of tannin in mangium wood, which accelerates the curing of PF resin in plywood manufacturing (Carvalho et al. 2014). For example, Hendrik et al. (2016) reported that laminated panels bonded with mangium tannin had a shear strength of 3.08 MPa, and they concluded that the addition of tannin could improve the shear strength of the panel.

According to an analysis of variance (Table 5), both wood species and tannin extract content affected the shear strength of plywood. Tukey's test results (Table 9) showed that plywood samples made of jabon and mindi wood were not significantly different from each other, but both were significantly different from mangium wood. Ten percent and 20 percent tannin extract additions did not differ significantly for the shear strength of plywood, but the effects of neat PF resin adhesive were significantly different.

Conclusions

1. This study was conducted to investigate the impact of tannin extract content on the depth of penetration, bond-line thickness, and adhesion performance of PTF resin adhesives for bonding three fast-growing tropical wood species, specifically, mangium, mindi, and jabon. The addition of tannin extract from mangium bark increased phenolic compounds and solid content in PTF resin adhesives, but it decreased pH, gel time, and the viscosity of PTF resin adhesives.
2. As the tannin extract content increased, the depth of penetration increased for mangium and jabon wood, but it decreased for mindi wood. The bond-line thickness of PTF resin adhesives decreased for all three woods, while shear strength of all plywoods increased with an increase in the tannin extract content.
3. All plywoods made of mangium wood satisfied standard requirements. However, plywood from mindi and jabon woods met the requirements only when 20 percent tannin extract was added.

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Literature Cited

- Abdullah, Z. A. and B. D. Park. 2009. Hydrolytic stability of cured urea-formaldehyde resins modified by additives. *J. Appl. Polym. Sci.* 114:1011–1017.
- ASTM International. 2013. Standard test method for determination of percent nonvolatile content of liquid phenolic resins used for wood laminating. ASTM D4426-01. ASTM International, West Conshohocken, Pennsylvania.
- Bal, B. C. and I. Bektas. 2014. Some mechanical of plywood produced from eucalyptus, beech, and poplar veneer. *Maderas. Cien. tecnol.* 16(1): 99–108. DOI:10.4067/S0718-221X2014005000009
- Carvalho, A. G., A. J. V. Zanon, F. A. Mori, R. F. Mendes, M. G. da Silva, and L. M. Mendes. 2014. Tannin adhesive from *Stryphnodendron adstringens* (Mart.) Coville in plywood panels. *BioResources* 9(2):2659–2670. DOI:10.13140/2.1.3543.8725
- Donaldson, L. A. and T. D. Lomax. 1989. Adhesive/fibre interaction in medium density fibreboard. *Wood Sci. Technol.* 23:371–380.
- Ellis, S. 1993. The performance of waferboard bonded with powdered phenol-formaldehyde resins with selected molecular weight distributions. *Forest Prod. J.* 43(2):66–68.
- European Committee for Standardization. 1993. Plywood-bonding quality: Requirements. Part 2. EN 314-2. European Committee for Standardization, Brussels.
- Gavrilović-Grmuša, I., M. Dunky, J. Miljkovic, and M. Djiporovic-Momcilovic. 2012. Influence of the viscosity of UF resins on the radial and tangential penetration into poplar wood and on the shear strength of adhesive joints. *Holzforschung* 66:849–856.
- Gindl, W., A. Sretenovic, A. Vincenti, and U. Müller. 2005. Direct measurement of strain distribution along a wood bond line. Part 2: Effect of adhesive penetration on strain distribution. *Holzforschung* 59:307–310.
- Hendrik, J., Y. S. Hadi, M. Y. Massijaya, and A. Santoso. 2016. Properties of laminated panels made from fast-growing species glued with mangium tannin adhesive. *BioResources* 11(3):5949–5960. DOI:10.15376/biores.11.3.5949-5960
- Jahanshaei, S., T. Tabarsa, and J. Asghari. 2012. Eco-friendly tannin-phenol formaldehyde resin for producing wood composites. *Pigm. Resin Technol.* 41(5):296–301.
- Johns, W. E., R. M. Rammon, and J. Youngquist. 1985. Chemical effects of mixed hardwood furnish on panel properties. In: Proceedings of the Nineteenth International Particleboard/Composite Materials Symposium, T. M. Maloney (Ed.), March 25–27, 1985; Washington State University, Pullman, Washington. pp. 363–377.
- Korean Standards Association. 2006. Ordinary plywood. KS F 3101. KSA. Seoul.
- Lestari, A. S. R. D., Y. S. Hadi, D. Hermawan, and A. Santoso. 2015. Glulam properties of fast-growing species using mahogany tannin adhesive. *BioResources* 10(4):7419–7433. DOI:10.13576/biores.10.4.7419-7433
- Nuryawan, A., B. D. Park, and A. P. Singh. 2014. Penetration of urea-formaldehyde resins with different formaldehyde/urea mole ratios into softwood tissues. *Wood Sci. Technol.* 48:889–902. DOI:10.1007/s00226-014-0649-9
- Park, B. D. and J. W. Kim. 2008. Dynamic mechanical analysis of urea-formaldehyde resin adhesives with different formaldehyde-to-urea molar ratio. *J. Appl. Polym. Sci.* 108:2045–2051.
- Pizzi, A. 1983. Tannin-based wood adhesive. In: Wood Adhesive: Chemistry and Technology. M. Dekker, New York. pp. 177–246.
- Santoso, A., Y. S. Hadi, and J. Malik. 2014. Characterization of merbau wood extract used as an adhesive in glued laminated lumber. *Forest Prod. J.* 66(5–6):313–318. DOI:10.13073/FPJ-D-15-00080
- Santoso, A., Y. S. Hadi, A. Pizzi, and M. C. Lagel. 2016. Characterization of merbau wood extract used as an adhesive in glued laminated lumber. *Forest Prod. J.* 66(5–6):313–318. DOI:10.13073/FPJ-D-15-00080
- Singh, A. P., B. Dawson, C. Rickard, J. Bond, and A. Singh. 2008. Light, confocal and scanning electron microscopy of wood-adhesive interface. *Microsc. Anal.* 22(3):5–8.
- Singh, A. P., T. Singh, and C. L. Rickard. 2010. Visualising impregnated

- chitosan in *Pinus radiata* early wood cells using light and scanning electron microscopy. *Micron* 41:263–267.
- Trosa, A. and A. Pizzi. 1997. Stability and performance of tannin-accelerated PF resins for plywood. *Holz Roh- Werkst.* 55(5):306.
- Trosa, A. and A. Pizzi. 2001. A no aldehyde emission hardener for tannin-based wood adhesive for exterior panel. *Holz Roh- Werkst.* 59(4):266–271.
- Xing, C., B. Riedl, A. Cloutier, and S. M. Shaler. 2005. Characterization of urea-formaldehyde resin penetration into medium density fiberboard fibers. *Wood Sci. Technol.* 39:374–378.