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Adhesion and Bonding Characteristics of Preservative-Treated Bamboo (*Gigantochloa scortechinii*) Laminates

¹A. Roziela Hanim, ²A. Zaidon, ²F. Abood and ³U.M.K. Anwar

¹School of International Tropical Forestry, Universiti Malaysia Sabah,
Locked Bag 2073, 88999, Kota Kinabalu, Sabah, Malaysia

²Faculty of Forestry, Universiti Putra Malaysia, 43400, Serdang, Selangor, Malaysia

³Forest Product Division, Forest Research Institute Malaysia, 52109, Kepong, Kuala Lumpur, Malaysia

Abstract: This study were investigate the adhesion and bonding characteristics of bamboo (*Gigantochloa scortechinii*) strips and laminates treated with permethrin-based preservative (Light Organic Solvent-Based (LOSP) and Water-Based (WBP)) formulations, Tributyl Tin Oxide (TBTO) and borax. The bamboo culm were cut into strips and treated with those selected chemicals. The bamboo strips were then glued edge to edge to form a bamboo veneers before fabrication of the three ply perpendicular bamboo laminates. In this research the properties studied include wettability, buffering capacity, shear strength and wood failure. Untreated strips and bamboo strips which were boiled in water (100°C) were also tested for comparison purposes. Those strips treated with LOSP had higher contact angle (3°-9°) which reflects that the surface of the treated strips is less readily wetted. Whereas, borax-treated strips had the highest wetting rate where the value is 1°. In buffering capacity study shows that treated bamboo was more stable towards alkali. This is suggested that a buffering agent (Calcium carbonate) is required in the adhesive formulation to ensure sufficient curing of the resin. Preservative treatments on bamboo strips significantly affect shear strength and wood failure of the laminates. Shear and wood failure of the laminated bamboo were significantly reduced especially in the wet condition where, the range is 0 N mm⁻² (WBP treated) to 0.65 N mm⁻² (boiled-treated) when compared to untreated bamboo laminates (0.79 N mm⁻²). While, in dry condition test, the glue bond strength of were range from 0.64 N mm⁻² (WBP-treated) to 2.04 N mm⁻² (borax-treated). All chemicals and non-chemical treatment generally affects the glue strength of the bamboo laminates especially in wet condition test. In dry condition test there are slightly reductions in glue bond strength but the quality still meets the requirement in the British Standard Part 8: Specification for Bond Performance of Veneer Plywood.

Key words: Permethrin-based preservatives, tributyltin-oxide, borax, shear strength

INTRODUCTION

Bamboo is regarded as eco friendly plants that grows and matures quickly, has a versatile use, unique appearance and potentially as an alternative raw material for wood (Rafidah *et al.*, 2010; Salleh, 1984). Bamboo is a highly renewable resource which may have 40-50 stems in one clump, which adds up to 10 to 20 culms yearly. However, the low durability of bamboo makes it render highly susceptible to fungi, insect and other deterioration agents. According to durability classification (Anonymous, 1982), bamboos fall in class III (non-durable category) with little variation in durability among different species. The starch content in bamboo plays an important role in its durability and service life. Latif *et al.* (1993)

found that the durability of bamboo against deterioration agents is strongly associated with the chemical composition. Bamboo also attractive and tough and thus suitable for conversion to engineered products such as composites and laminated boards (Latif *et al.*, 1993; Razak *et al.*, 1998), ply bamboo (Anwar *et al.*, 2004). *Gigantochloa scortechinii*, known as Buluh Semantan is well known and established in the Malaysian community. It was widely used for traditional uses such as water pipes, poles, flooring and handicrafts. Anwar *et al.* (2005) reported that bamboo such as *G. scortechinii* is suitable for composite materials, laminated boards and plywood. The properties of plywood made from *G. scortechinii* are extremely high in modulus of rupture, bending strength and modulus of elasticity. The ply-bamboo strength ranks

as the highest among all the structural boards and even as good as the solid wood of high-density commercial timber (Chen and Qin, 1985).

Like most lignocellulosic materials, bamboos have a low resistant to biological degradation agents (Zaidon *et al.*, 2000). Thus, there is a need to treat bamboo in order to enhance the service life of the product. The best way is through chemical treatments. Chemical preservation ensures a longer service life for bamboo products and maintains its quality (Satish and Dobriyal, 1992; Anonymous, 2006). The choice of preservatives would determine the success of the bamboo treatment. Chemicals selected should possess insecticidal and fungicidal properties, commercially available, cheap and environmentally friendly and will not affect other properties of the treated bamboo.

Preservatives like Copper Chromated Arsenate (CCA) and boron compounds are becoming less preference due to their toxicity hazards and are not environmentally friendly. New and more environmental friendly preservative formulations are sought to preserve bamboo. Pyrethroids are potential group of preservatives that can be served as an alternative to boron and CCA. It is a synthetic form of pyrethrin that possesses insecticidal properties. They are one of the least poisonous insecticides to mammals (Ray, 1991; Tomlin, 1994). Some of the pyrethroids that have formulated into preservatives include permethrin, cypermethrin and deltamethrin and they are available in the market under different trade names. In earlier study, it has been found that a small concentration of permethrin-based preservatives and Tributyltin Oxide (TBTO) successfully increased the resistance of bamboo towards white rot (*Pycnoporus sanguineus*) and termite (*Coptotermes curvignathus*) attacks. Since the potential usage of bamboo is in lamination form, the adhesion and bonding characteristics of bamboo laminated should then be investigated.

This study was undertaken to investigate the effect of permethrin-based and TBTO preservative treatments on gluing properties of bamboo (*Gigantochloa scortechinii*) strips bonded with Phenol Formaldehyde (PF) resin. Adhesion properties of untreated, water boiled and borax-treated strips were also evaluated.

MATERIALS AND METHODS

Materials: This study was conducted at wood working lab at the Faculty of Forestry, Universiti Putra Malaysia. This research was started on March 2004 until August 2008. Three year-old Buluh Semantan (*Gigantochloa scortechinii*) was used in this study. They were extracted from bamboo plantation research plots managed by FRIM

Table 1: Chemical composition in preservatives used in this study

| Trade name | Composition | Concentration (%) |
|--|----------------------------|-------------------|
| Light Organic Solvent Preservatives (LOSP) | Tributyltin naphthenate | 3.5 |
| | Permethrin | 0.2 |
| | Dichlofuanid | 0.1 |
| Water Based Preservatives (WBP) | Organic solvent | 96.2 |
| | (Total active ingredients) | (3.8) |
| | Disodium octaborate | 10.0 |
| | Benzalkonium chloride, | 2.0 |
| Borax Tributyltin Oxide (TBTO) | Permethrin | 0.2 |
| | Water | 87.8 |
| | (Total active ingredients) | (12.2) |
| | Sodium borate | 5.0 |
| | Tributyltin oxide | 1.0 |

Standard concentration recommended by the manufacturers

(Forest Research Institute of Malaysia) at Compartment 24, Chebar Besar Forest Reserve, Nami, Kedah. The natural stand of bamboo in this area has undergone silviculture treatment to promote the growth of bamboo clumps and individual culms since 1988.

The chemicals used in the study were borax (sodium borate, 5% w/v), pyrethroid based preservatives; LOSP (a.i: tributyltin naphthenate, 3.5%, permethrin, 0.2%) and WBP (a.i: disodium octaborate, 10%; benzalkonium chloride, 2%; permethrin, 0.2%), tributyltin-oxide (TBTO, 1% w/v). The chemicals were selected based on the ability to provide both insecticidal and fungicidal properties, cheap, possess low mammalian toxicity, soluble in water and have an ability to retain the clear and light colored finish of the treated material. LOSP and WBP were natural based preservatives while TBTO and borax was selected based on the previous studies which has been used to treat wood. Boiling treatment of bamboo in water was also carried out for comparison purposes. Table 1 shows the chemical composition and concentration of active ingredients of pyrethroid based preservatives which is LOSP and WBP, TBTO and also borax, the active ingredients and concentration is a recommended by manufacturer.

Preparation of samples: Bamboo culms were split using splitter knife and were dried to a moisture content of about 10% in a kiln (32.5°C and 60% relative humidity) for two weeks. The epidermis at the outer and inner part of the splits were then removed by using a single face-planning machine and finally dressed into strips of dimensions 150 mm (length)×20 mm (width)×4 mm (thickness). The strips were then treated with the chemicals using vacuum-pressure impregnation method. A full vacuum (56 cmHg) was applied for 30 min and at the end of the evaluation period, an external pressure of 25 kPa was applied and the samples were left immersed for 30 min. The weights of the treated sample were measured before and after treatment for determination of chemical retention.

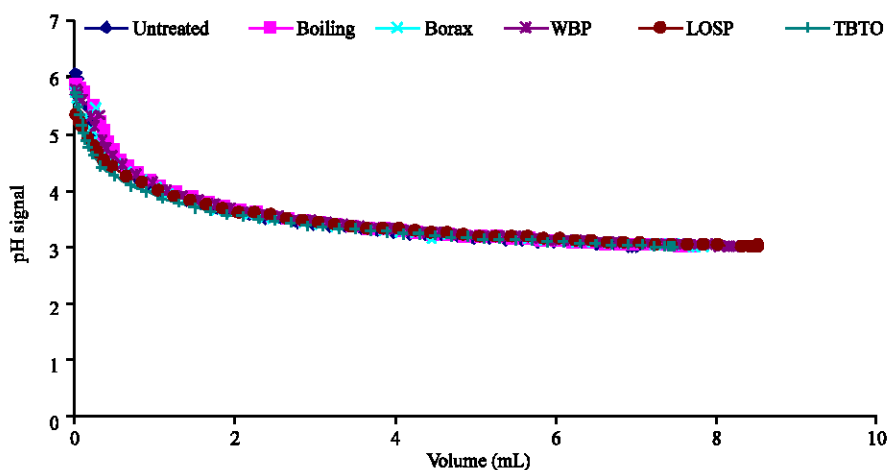


Fig. 1: Variation of pH with respect to addition of acid

Wettability properties of treated bamboo strips: Wetting behavior of bamboo is to observe the movement of liquid to penetrate into the wood, when the good wetting occurs, the contact angle become very small (Minford, 1991). Investigation of wetting properties of the material may help to determine its bonding characteristic with adhesive. The contact angle was measured on the inner and outer surfaces of the bamboo strips. Bamboo strips with dimension of 25×25×4 mm were used for the evaluation of contact angle. Distilled water was used by dropping it onto the surface using injection tube. The AB Lorentzen and Watter (L-W) surface wettability tester was used to measure the contact angle. Prior to determination of wettability, untreated and treated samples were first conditioned at 35°C and 65% RH for about 2 days. The image of the drops was captured by a microscope tube after dropping the droplet on the solid surface. The diameter of contact angle and height of the droplet measured from the glass plate with transparent measuring scale and tangent were calculated. Time of penetration was also recorded.

Determination of buffering capacity of treated bamboo: Investigating the acidity or pH and buffer capacity of bamboo is an important criterion of its suitability for various applications. Knowledge of the pH and buffering capacity of the raw material is important consideration to better understand the effects of raw materials on the curing rate of resin used for panel manufacturing (Cheng *et al.*, 2004). According to Paridah *et al.* (2001), the buffering capacity of wood helps to determine the amount of buffering agent required in the adhesive to prevent changes in pH at the glue line. Bamboo particle in green condition was ground into powder form. Then, 10 g of bamboo powder was refluxed in 100 mL distilled

water for 1 h (solution = 10% w/v). The mixture was filtered with filter paper and washed with 100 mL distilled water. From the 5% distillates, 1 mL was diluted with 100 mL distilled water to produce solution of 0.05%. A final concentration (0.025%) was made by diluting 25 mL distillates with 25 mL distilled water. About 50 mL was then used for the determination of initial pH. The solution was then titrated with 0.01 N HCL and 0.01 N NaOH until the pH reaches pH 3.0 and pH 11. The procedure was repeated to other different chemical treatment. The amount of HCL and NaOH consumed in the titration and pH level was recorded. A graph (Fig. 1, 2), pH versus volume (mL) was plotted to observe the change in pH.

Fabrication of bamboo laminates: In fabricating bamboo laminates, bamboo strips were glued edge-to-edge using polyvinyl acetate (PVAc) resin to fabricate 150×12× 4 mm bamboo sheets. The bamboo veneer was then glued three-ply perpendicular to the grain using phenol formaldehyde resin to produce a bamboo laminates. Adhesive was formulated by mixing with some other components such as industrial wheat flour, calcium carbonate and water. Adhesive formulation was prepared by mixing the resin with filler, Industrial Wheat Flour (IWF), calcium carbonate (CaCO₃) and water. Table 2 was summarized the mixing formulation of the resin used for fabricating three-ply bamboo laminates. The glue was spread at a rate of 230-270 g m⁻² Single Glue Line (SGL) according to the method specified in British Standard: Part 8: Specification for Bond Performance of Veneer Plywood, (Anonymous, 1986). The glue mixture was applied using a scraper. Three ply bamboo laminates were produced by assembling each bamboo sheet perpendicular to the adjacent ply.

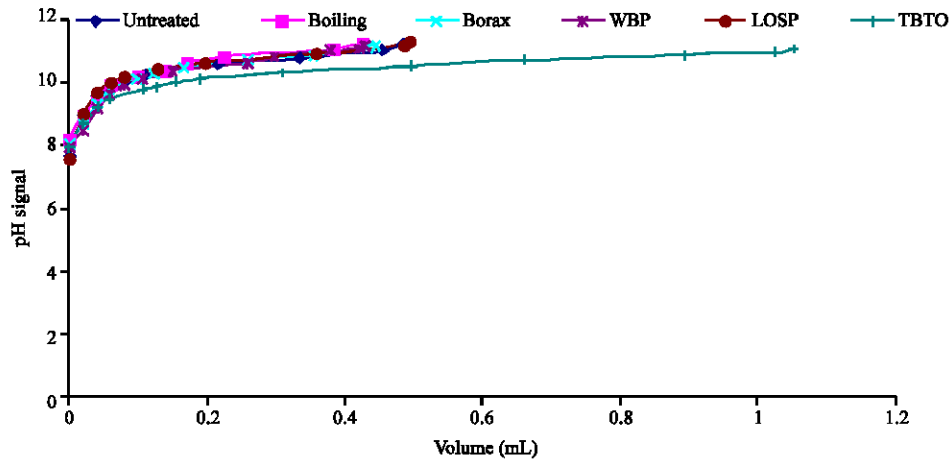


Fig. 2: Variation of pH with respect to addition of alkali

Table 2: Mixing formulation for PF adhesive

| Material | Measurement (g) (weight parts ⁻¹) | Percent |
|--|---|---------|
| Phenol formaldehyde | 100 | 69.10 |
| Filler MF-9 | | |
| Industrial wheat flour | 15 | 10.30 |
| Calcium carbonate (CaCO ₃) | 25 | 17.20 |
| Water | 5 | 3.40 |
| Total | 145 | 100.00 |
| Viscosity | 20 poise | |

Malayan Adhesives and Chemicals Sdn. Bhd (2004)

The bamboo laminates were then pre-pressed at a specific pressure of 10 kg cm⁻² for 5 min. They were then pressed in a hot press at 140°C for 7 min with a specific pressure of 14 kg cm⁻². Finally, the laminates were conditioned in a conditioning room at 25°C and 65% relative humidity (12% EMC) prior to testing. British Standard (BS: Part 8: Specification for Bond Performance of Veneer Plywood) (Anonymous, 1986) was used to investigate the shear strength and glue line between composite elements before and after exposure to cool and boiled cyclic condition.

RESULTS AND DISCUSSION

Wettability and contact angle: Wettability of bamboo indicates the rate and how fast a liquid can wet and spread on it. In relation to contact angle when good wetting occurs, the contact angle becomes very small. On the contrary, if the contact angle is higher, there is lack of wetting. Contact angle of *G. scortechinii* on the outer part was 9° for LOSP-treated strips, 4.03° for TBTO-treated strips, 3° for WBP-treated strips, 2° for untreated strips, 1° for boiled-treated and borax-treated strips respectively. Contact angle for the inner part was 5° for LOSP-treated bamboo strips, 3° for WBP-treated bamboo strips and 3° for TBTO treated bamboo strips, respectively. There was no contact angle recorded for untreated, boiling and borax

Table 3: Contact angle (°) on the outer and inner layer surfaces of *G. scortechinii* strips

| Treatments | Contact angle (°) after 2 sec | |
|---------------------------|-------------------------------|-------------|
| | Outer layer | Inner layer |
| Untreated | 2 (1.97) | 0 |
| Boiling (Water) | 1 (1.88) | 0 |
| Borax (5% w/v) | 1 (0.72) | 0 |
| LOSP (Total a.i 3.8% w/v) | 9 (0.95) | 5 (1.16) |
| WBP (Total a.i 12.2% w/v) | 3 (5.42) | 3 (0.61) |
| TBTO (1 %w/v) | 4 (1.27) | 2 (0.11) |

No. of samples, 10 for each treatment. Values in parentheses are standard deviations

due to fast absorption. The smaller contact angle or complete wetting suggested that the surface was easier to be wetted. Zhu (1995) also stated that higher wettability resulted in poor bonding due to greater tendency for starved joints. Thus, the adhesive used for wood has to be modified. In this study, some modification on glue mixture was made to optimize the bonding process. In all cases, the amount of filler was kept at 27% of the amount of resin. The contact angle of untreated and treated bamboo strips are summarised in Table 3.

The highest contact angle value was found with LOSP-treated strips. This was related to the chemical properties of LOSP, which is an organic solvent-based fungicidal preservative (Anonymous, 2000). Hunt and Garret (1967) also mention that hydrocarbon solvents would restrict the penetration of liquid into the bamboo substrate. Tascioglu *et al.* (2003) reported in their research about composite joist treated with oil-borne preservatives had a low surface wettability due to the presence of hydrocarbon solvent on the surface of the material.

The contact angle between the outer and inner surface of bamboo differs significantly. From the results, regardless of the treatment, the outer layer of material was easier to wet than the inner layer. This was due to the difference in specific gravity between the two surfaces.

Vessel size also contributed to the variation of contact angle value. This finding also supported by Sekhar and Bhartari (1960), Razak *et al.* (1998), Jamaludin *et al.* (1997) and Anwar *et al.* (2005), the contact angle is strongly related to the size of vessel or pores. In comparison to the vessel size for inner and outer part of the culm, the vessels size was very small at the outer surface and become larger when it goes inward (Liese, 1998).

Buffering capacity: The results on buffering capacity shows that the treated and untreated bamboo was more stable towards alkali (Fig. 1), the amount of 0.1N NaOH required to change the pH value range from 7.51 (TBTO)-8.14 (Boiling) to 11 was only 0.5 mL. Conversely, the amount required to change the pH 7 to 3 was using 0.1 N HCL was 8.5 mL (Fig. 2). Bamboo is sensitive to an alkali-based adhesive, such as PF, a buffer is required in the adhesive formulation to ensure sufficient curing of the resin (Sakuno and Moredo, 1996). The results imply that the adhesive formulation used for bamboo laminates for all treatments would behave the same in terms of curing rate as long as the pH value is within the alkaline region. Malonney (1993) also stated that an acidic condition might affect the curing rate or pressing time due to the combination of pH, buffer capacity and the existing or potential of total free volatile acid content of the material it is suggested that there is a needed for a longer pressing time to ensure sufficient curing time.

Shear strength and wood failure: Glue line shear strength is one of the basic mechanical and durability indicators for laminated composite boards. By using the British Standard (BS: Part 8: Specification for Bond Performance of Veneer Plywood) (Anonymous, 1986), the shear strength and bamboo failure between composite elements were assessed in Dry Condition (DT) and after Cyclic Boil Treatment (CBR). In dry condition test, except for borax

treatment, the results show that the shear and wood failures of the bamboo laminate were significantly reduced. The values of the shear strength were in the range of 0.64 to 2.04 N mm⁻², as compared to untreated bamboo laminates (2.66 N mm⁻²). For wet condition test, the range was from 0.48-0.69 N mm⁻² compared to 0.78 N mm⁻² for untreated bamboo laminates. The finding on the shear strength and bamboo failure at dry and cyclic boil test is summarized in Table 4.

Anwar *et al.* (2005) and Zaidon *et al.* (2000) revealed that the shear strength of untreated laminates was relatively higher compared to treated laminates. In addition, Anwar *et al.* (2005) in their findings found that the shear strength of *G. scortechinii* plywood in dry condition was 3.4 and 1.7 N mm⁻² in wet condition. Similarly, Zaidon *et al.* (2000) found that the shear strength was 2.36 N mm⁻² (parallel) and 2.68 N mm⁻² (cross-ply laminates in wet condition) while in dry condition the shear strength was 1.51 and 1.24 N mm⁻², respectively. The percentage of the shear strength reduction is summarized in Fig. 3.

Treatment with borax were found to reduce the shear strength of the laminates from 2.66 to 2.04 N mm⁻². i.e., 23% reduction in the shear strength properties. While for TBTO-treated bamboo laminates the strength decreased by 32%, water boiled-treated laminates 44%,

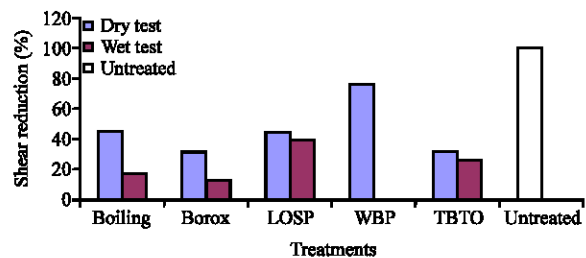


Fig. 3: Shear reduction of treated bamboo laminates calculated against untreated laminates

Table 4: Mean shear strength and bamboo failure percentage of bamboo laminates

| Treatments | DT ¹ | | CBR ² | |
|---------------------------|--------------------------------------|--------------------|--------------------------------------|--------------------|
| | Shear strength (N mm ⁻²) | Bamboo failure (%) | Shear strength (N mm ⁻²) | Bamboo failure (%) |
| Untreated | 2.66±0.40 ^a | 14 ^a | 0.79±0.46 ^a | 2 ^a |
| Boiling (water) | 1.48±0.40 ^b | 16 ^a | 0.65±0.65 ^a | 3 ^{abc} |
| Borax (5% w/v) | 2.04±0.44 ^{ab} | 28 ^a | 0.63±0.62 ^a | 4 ^a |
| LOSP (Total a.i 3.8% w/v) | 1.47±0.96 ^b | 10 ^a | 0.48±0.37 ^{ab} | 3 ^{bc} |
| WBP (Total a.i 12.2% w/v) | 0.64±0.35 ^c | 8 ^a | 0 ^b | 0 ^d |
| TBTO (1% w/v) | 1.80±0.40 ^b | 14 ^a | 0.58±0.53 ^{ab} | 3 ^{abc} |
| British standard | Shear strength | | Wood failure | |
| | 0.35<τ<0.7 | | >75 | |
| | 0.70<τ<1.7 | | >50 | |
| | 1.70<τ<2.5 | | >25 | |
| | 2.50<τ | | >15 | |

No. of samples, 70.±is standard deviation; Means within a column with the same letter are not significantly different at p = 0.05. ¹DT (Dry Condition Test), ²CBR (Cyclic Boiled Resistance)

LOSP-treated by 45% and WBP-treated by 76% when tested in dry condition. The same trend happened when tested in wet condition. The shear strength of the laminates decreased for all treatments. Boiling-treated laminates decreased by 17%, borax-treated laminate by 21%, TBTO-treated laminates by 26%, LOSP-treated 39% and lastly, for WBP-treated laminates it was delaminated after exposure to wet condition test.

Borates used as a preservative for wood composite panels bonded with phenol formaldehyde (PF) resin often reduce resin gel time, not allowing the resin to flow and cure sufficiently. This problem is related to the functional methylol groups on resin molecules and their interaction with borate ions (Sean *et al.*, 1999). This interaction is detrimental to bond performance and ultimately reduces physical properties of the panel. Vick (1990), investigated preservatives compatibility with PF resin for thirteen non-acidic waterborne preservatives using retreated aspen veneers. The results indicated that the borate containing preservatives tested caused poor bonds, as did an emulsion of copper naphthenate.

Treatment on bamboo strips altered the shear strength and wood failure properties of the bamboo laminates. In particular, WBP showed a tremendous reduction in the glue bond shear strength and bamboo failure after exposing the bamboo laminates to both dry and cyclic boiling test. The properties of the formulations and treatment that were water-based or hydrocarbon-based may have influenced the compatibility of the adhesive used in bonding the bamboo laminates. However, the wood failure test of the treated material is not significantly different compared to untreated bamboo laminates where the wood failure test ranged between 8% to 28% in the dry condition test and 2-4% in the wet condition test. However, in the dry condition test, the glue bond quality of untreated and treated laminates met the minimum standard requirements of the British Standards: Part 8: Specification for Bond Performance of Veneer Plywood (Anonymous, 1986).

Shear strength and wood failure reflected on the glue bond quality of the laminated bamboo. Whereas one of these values is high and the other is low, it indicates that either the bamboo strips are of low strength quality or the adhesive bond is poor. Anwar *et al.* (2005) stated that the strength of glue line is also related to the glue mixture itself. Even though the bamboo failure was relatively low (ranging 40-60%), this does not imply the ply-bamboo is of inferior quality. This is because the adhesives were found to have penetrated into the bamboo substantially, forming a good adhesion and anchorage between the bamboo layers. Zaidon *et al.* (2000) reported that shear strength is affected by grain orientation. However, it was

observed that dry wood failure percentage was relatively higher in the parallel-ply laminates (75%) than in the cross-ply laminates. Strips arranged perpendicular to each other experienced a relatively higher reduction in shear strength after cyclic soaking in boiled water and after long soaking in boiled water. As reported by Sulaiman *et al.* (2006) shear strength of bamboo laminates was reduced by heat treatment, where oil was used as the heating medium. The reason for the loss in strength may have been due to the presence of oil in the cells. Presence of oil may reduce the wetting of the surface, thus reducing the absorption of adhesive by the surface. This effect may eventually reduce the adhesion to the surface. Formaldehyde adhesives are usually water-borne resins such that the curing process is not only polymerisation, but also the loss of water used as the solvent. Polymerization process evolves too much water in the bond lines and this retards the reaction. Insufficient water prior to polymerisation reduces the mobility of the resin and limits collision needed for polymerization, in addition to limiting heat transfer. Controls of both open and close assembly times are important in controlling the penetration and water content in the bond line (Rowell, 2005).

CONCLUSION

The result on wettability shows that there a need to modify the adhesive formulation so that the adhesive can penetrate adequately into the bamboo substrate. The buffering capacity for all treatments behaves the same in terms of curing rate as long as the pH value is within the alkaline region. Chemicals and non-chemical treatments affected the bonding properties of laminates slightly. Shear strength was adversely affected by the chemicals treatments. In dry condition, except for borax acid-treated laminates, the shear strength of other treated laminates were significantly reduced. Boiling-treated laminates had superior shear strength compared to other treated laminates tested in wet condition. Among the preservatives, WBP affected the bonding properties most. The wood failure of treated laminates was not significantly different from the untreated. As a whole, the bonding property of the treated bamboo laminates surpassed the minimum requirement of British Standard: Part 8: Specification for Bond Performance of Veneer Plywood.

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