ADHESION AND BONDING PROPERTIES OF LOW MOLECULAR WEIGHT PHENOL FORMALDEHYDE-TREATED PLYBAMBOO

UMK Anwar[1, 2, *], MT Paridah[2], H Hamdan[1], A Zaidon[2], A Roziela Hanim[3] & AS Nordahlia[1]

[1]*Forest Research Institute Malaysia, 52109 Kepong, Selangor Darul Ehsan, Malaysia*
[2]*Institute of Tropical Forestry and Forest Products, Universiti Putra Malaysia, 43400 Serdang, Selangor Darul Ehsan, Malaysia*
[3]*School of International Tropical Forestry, Universiti Malaysia Sabah, Locked Bag 2073, 88999 Kota Kinabalu, Sabah, Malaysia*

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ANWAR UMK, PARIDAH MT, HAMDAN H, ZAIDON A, ROZIELA HANIM A & NORDAHLIA AS. 2012. Adhesion and bonding properties of low molecular weight phenol formaldehyde-treated plybamboo. This study investigated the adhesion of bamboo (*Gigantochloa scortechinii*) strips after impregnation with phenolic resin and the effect of curing time on bonding properties of low molecular weight phenol formaldehyde (LMwPF)-treated plybamboo. The optimum pressing time to produce LMwPF-treated plybamboo was also determined. Properties studied included wettablity, buffering capacity, shear strength and wood failure. The study showed that phenolic-treated strips had higher contact angle and, thus, were more difficult to be penetrated by liquid compared with untreated strips. Buffering capacity showed that bamboo strip was stable towards acid. Shear bond strength of the plybamboo met the requirement of BS EN 314-1. The study concluded that the optimum pressing times were 22 and 33 min for three- and five-ply plybamboo respectively to produce good glue joints.

Keywords: *Gigantochloa scortechinii*, impregnation modification, optimum pressing time

ANWAR UMK, PARIDAH MT, HAMDAN H, ZAIDON A, ROZIELA HANIM A & NORDAHLIA AS. 2012. Kesan perekatan dan ciri-ciri lekatan buluh lapis yang dirawat dengan perekat fenol formaldehid. Kajian ini bertujuan untuk mengkaji perekatan bilahan buluh semantan (*Gigantochloa scortechinii*) serta kesan masa pematangan terhadap ciri-ciri lekatan buluh lapis yang telah dirawat menggunakan perekat fenol formaldehid yang rendah berat molekulnya (LMwPF). Masa pematangan yang optimum bagi menghasilkan buluh lapis dirawat juga ditentukan. Ciri-ciri lain yang dikaji termasuk kadar sebaran, keupayaan penimbalan, kekuatan ricih dan kegagalan kayu. Kajian menunjukkan bahawa sudut sentuhan bagi bilahan yang dirawat menggunakan LMwPF adalah lebih tinggi berbanding dengan bilahan yang tidak dirawat. Justeru, ia lebih sukar untuk ditembusi cecair. Kajian tentang keupayaan penimbalan menunjukkan bahawa bilahan buluh stabil ke arah asid. Kekuatan ikatan ricih buluh lapis telah memenuhi piawaian BS EN 314-1. Kajian menunjukkan bahawa masa yang optimum untuk tempoh pematangan bagi buluh tiga dan lima lapis ialah masing-masing 22 min dan 33 min bagi menghasilkan lekatan perekatan yang baik.

INTRODUCTION

Bamboo and other lignocellulosic materials are unstable when subjected to high moisture and humidity. Impregnation of wood with phenolic resin is one of several promising methods to overcome the weakness of wood-based panel product, in particular its dimensional instability. The use of low molecular weight phenol formaldehyde (LMwPF) resin in plywood, particleboard and oriented strand board (OSB) has improved not only the physical but also the mechanical properties of the boards (Paridah et al. 2006).

The quality of wood composites such as plywood, laminated wood and particleboard as well as the performance of glued joints depend upon good glue bond formation. Good bonding requires good interlinks between substrates and wood, namely, interface–adhesive formation which also involve boundary layers between them (Paridah et al. 2001). An adhesion implies a formation of interface by penetration of adhesive into the wood by mechanical interlocking, thermodynamic adsorption of the glue on the wood and/or chemical (covalent) bonding.

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*E-mail: mkanwar@frim.gov.my

Besides, wettability of wood surface influences bond quality at the interface and boundary layers between wood and resin. Wettability can be measured by contact angle between the solid–liquid interface and liquid–air surface (Anderson 1986). This method has been widely practised and gives quite accurate indication of the wettability of adhesive.

Another property that has great influence on glueline formation is buffering capacity. Buffer capacity measures the resistance of wood in changing pH level either in acid or alkaline liquid (Mansur 2000, Paridah et al. 2001). A wood with high buffering capacity would normally require the addition of greater amount of catalyst to bring the pH to the level (acidic or alkali) required for optimum resin curing (Maloney 1993).

The quality of wood and non-wood lignocelullose-based products is significantly influenced by bonding properties. Mechanical strength, including shear strength, is influenced by bonding quality, which is a function of raw material properties such as wood density or porosity and type the adhesive and its chemical constituents. For example species such as balau (*Shorea* spp.), kasai (*Pometia* spp.) and chengal (*Neobalanocarpus heimii*) have high density and do not have good penetration of adhesive, resulting in poor glueline. Thus, two important factors that need to be taken into consideration when making composite products are wood failure and bonding quality. In certain cases, depending on wood density, although good bonding has been achieved wood failure can still occur due to raw material characteristics (Paridah et al. 2001, Zaidon et al. 2004).

This study evaluated the adhesion characteristics of semantan bamboo (*Gigantochloa scortechinii*) by measuring (1) wettability through its contact angle and (2) buffering capacity. The effects of different pressing times on bonding quality of bonded products were then assessed through plywood shear test. Shear strength and bamboo failure percentage were also examined.

MATERIALS AND METHODS

Four-year-old *G. scortechinii* were felled from the Forest Research Institute Malaysia, Kepong, Selangor. The bamboo culms were cut into splits 20 mm wide using sizing and splitting machine. The splits were planed to 4–5 mm thick to produce bamboo strips and dried to 10% moisture content. In this study, LMwPF resin was supplied by a local adhesive company. The LMwPF resin, with average molecular weight of 600, was used to ensure that the resin could easily penetrate into bamboo strips during impregnation.

Bamboo strips were impregnated with LMwPF resin in a vacuum pressure impregnation processing tank and evacuated at 750 mm Hg for 1 hour. Samples were then dried in an oven at 60 °C for 9 hours and the average weight per cent gain of LMwPF-treated strips was 14%. The treated strips were then used for evaluation of surface wettability, pH, buffering capacity and fabrication of LMwPF-treated plybamboo.

Evaluation of surface wettability

Five samples, each from untreated and LMw™PF-treated strips were used to determine the contact angle of droplets for wettability study. Bamboo strips (20 mm × 40 mm × 4 mm) were conditioned at 20 ± 2 °C and 65 ± 3% relative humidity for a week prior to testing. Using an injection tube, 0.02 mg distilled water was dropped onto first, the inner side followed by the outer surface of untreated and LMwPF-treated strips (Figure 1). Using a surface contact angle tester, the angle made between the droplet and the bamboo surface was measured after 2 s. Then measurements were continuously taken every 1 min for a period of 30 min.

Determination of pH and buffering capacity

Bamboo strips were ground to pass through a 53-mesh sieve. About 1 g of bamboo powder

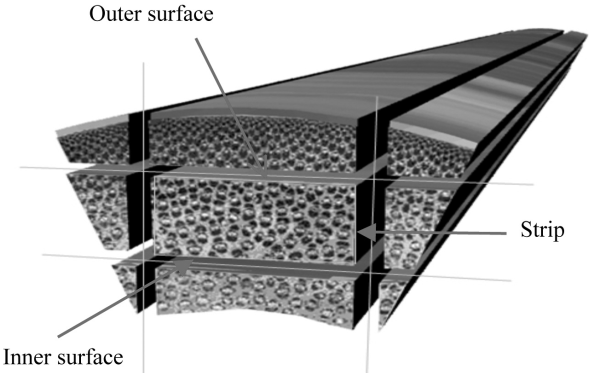

Figure 1 Schematic illustration for contact angle samples of the *Gigantochloa scortechinii* strip

was refluxed in 100 mL water for 1 hour. After refluxing, the mixture was filtered using filter paper and washed thoroughly with 400 mL distilled water. The solutions used during the titration process were 0.01 mol/L HCl and 0.01 mol/L NaOH. Titration was carried out until the HCl and NaOH reached pH 3 and 11 respectively.

Evaluation of bonding properties

A total of 36 panels for LMwPF-treated plybamboo (300 mm × 300 mm × 12 mm or 20 mm) were prepared for the experiment. Six panels of untreated plybamboo were prepared for comparison. For plybamboo fabrication, the treated or untreated bamboo strips were glued together edge-to-edge using phenol resorcinol formaldehyde resin to produce plybamboo veneer. Bamboo veneers were then assembled perpendicular to each other to form three- (12 mm thick) and five-ply (20 mm thick) plybamboo with phenol formaldehyde resin as binder. The plybamboo were hot pressed at 140 °C and pressure of 14 kg m^{-2}. Pressing times of 11, 22, 33 and 44 min were applied for LMwPF-treated plybamboo. For untreated plybamboo, pressing times required to bond three- and five-ply plybamboo were 6.5 and 11 min respectively.

Plybamboo were cut to 100 mm × 25 mm × 25 mm and tested in dry and boiling cyclic conditions according to BS EN 314-1 (BS 2004). Upon completion of the boiling cyclic test, specimens were dried and examined for estimated percentage of wood failure along the glue line. Wood failure of individual specimen was recorded to the nearest 5% and the average shear strength and wood failure were compared with the minimum standard requirement of BS EN 314-1 (BS 2004).

Statistical analysis

Analysis of variance (ANOVA) was employed to evaluate the difference between variables studied. Further analyses of the means for glue bond integrity and dimensional stability were carried out using least significant different method at $p \leq 0.05$.

RESULTS AND DISCUSSION

Wettability of untreated and LMwPF-treated bamboo strips

Surface wettability of *G. scortechinii* strips was examined after impregnation with LMwPF resin. Table 1 shows the ANOVA for initial contact angles. There was significant difference in contact angles between untreated and LMwPF-treated bamboo strip surfaces. Nevertheless, contact angles measured on inner and outer surfaces were not significantly different. Hence, resin filled voids on the surface and inner layer of the cells, resulting in increased surface tension that prevented the treated strip to be wetted easily. This explained why phenolic-treated surfaces had better control of adhesive penetration compared with untreated strips.

Contact angles values at inner and outer surfaces of untreated and LMwPF-treated bamboo strips are shown in Figure 2. After 30 min, contact angles of the inner and outer surfaces decreased from 42.5° and 43.5° to 14.9° and 13.5° respectively. The inner and outer surfaces of treated bamboo behaved similarly as contact angles of water droplets gradually decreased within 1 min. However, contact angles of untreated samples (both inner and outer) could not be measured after 5 s.

Fitted regression lines indicated high inverse linear relationship between contact angle and time. The predictive equation for contact angle of LMwPF-treated bamboo strips in this study was highly significant with coefficient determination of 0.988 (Figure 2). Hence, the time required to wet treated bamboo strip surfaces can be predicted by determining the contact angle of water droplet using the following equation:

Table 1 Analysis of variance of initial contact angle

Source of variation	Df	Means square	F value	p > F
Treatment (untreated and LMwPF-treated)	1	2918.8	217.3	0.001***
Surface (inner and outer)	1	0.082	0.01	0.9393
Treatment × surface	1	2.235	0.17	0.6912

*** significant at $p \geq 0.001$

$y = -0.8834 x + 40.521$
where y = contact angle and x = time (min)

Bamboo strips usually generate relatively smaller contact angle within a short time due to their high wettability (Anwar et al. 2005, Seyoum 2005, Hanim et al. 2010). The remarkably low wettability (as indicated by higher contact angle) for LMwPF-treated samples was because the partially cured LMwPF resin still remained on the surface but at the same time filled voids and parenchyma cells (Anwar et al. 2009). LMwPF resin prevented water from penetrating the cells and, thus, wettability of the surface was lower. Similar results were also observed in CCA-treated southern pine (Zhang et al. 1997). The authors reported higher contact angle on CCA-treated samples, which also required longer time to reach 0° (i.e. total penetration) compared with untreated samples. Contact angle of southern yellow pine fibreglass reinforced plastic composites was affected by the amount of preservative used in the treatment (Tascioglu et al. 2004). A positive linear relationship between surface wettability and glue bond strength of various tropical wood glued with urea formaldehyde resin was also reported by Chen (1970). A high degree of wettability in bamboo made bonding difficult (i.e. over penetration of the adhesive caused higher tendency for starved joints). Therefore, the adhesive used for wood cannot be used for bamboo without modifying the formulation (Zhu 1995, Anwar 2005).

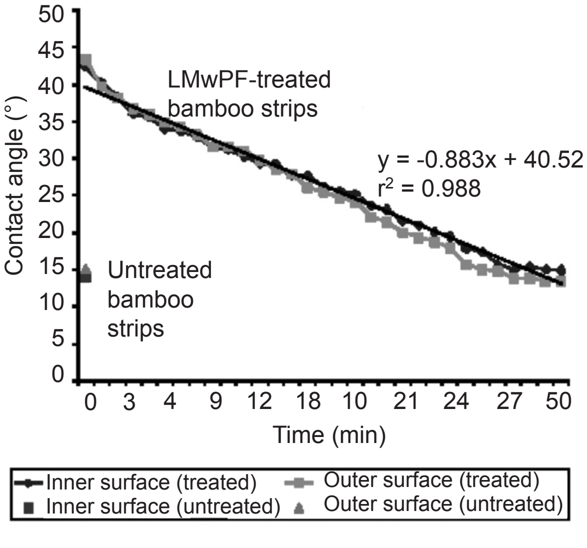

Figure 2 Contact angle vs. time of untreated and LMwPF-treated bamboo strips

Results of this study revealed that when bamboo strips were treated with LMwPF resin, wettability of the treated strips decreased substantially with a trend similar to that of wood. Hence, the adhesive formulation for bonding phenolic-treated bamboo strips may be similar to wood whereby the addition of filler or extender to control adhesive penetration may not be necessary.

pH value and buffering capacity

Initial pH of LMwPF-treated strip in alkali condition was higher than untreated strip but the value was lower in acid condition. The buffering capacity of both untreated and LMwPF-treated strips of *G. scortechinii* was more towards acidic than alkali. It was also suggested that the LMwPF resin did not greatly influence pH values and buffering capacity of bamboo strips. The amounts NaOH required to change the initial pH value from 7.21 (untreated) and 7.84 (LMwPF-treated strips) to 11 were only 1.0 and 1.1 mL respectively (Figure 3). Conversely, the amounts of HCl required to change the pH from 6.42 (untreated) and 5.87 (phenolic-treated strips) to pH 3 were 7.5 and 7.4 mL respectively (Figure 4).

The findings on buffering capacity of bamboo strip are in agreement with Zaidon et al. (2004) and Seyoum (2005). When an alkaline resin (i.e. phenol formaldehyde) is spread onto a bamboo surface, the adhesive at the boundary layers changes its pH to slightly lower than normal, just enough to slow down the curing rate of the phenol formaldehyde resin which normally happens between pH 11 and 12. It was reported that pH had no direct effect on bond strength and wood failure but influenced the adhesive curing process (Sakuno & Moredo 1993). In order for resin binders to cure properly, an optimum level of acidity must be established either on the surface or inside the substrate. The rate of crosslinking in most thermosetting adhesives is pH-dependent (Maloney 1993).

Glue bond integrity of plybamboo pressed at different pressing time

Mean values of glue line shear strength and bamboo failure of untreated and LMwPF-treated plybamboo (three- and five-ply) at different pressing times are tabulated in Table

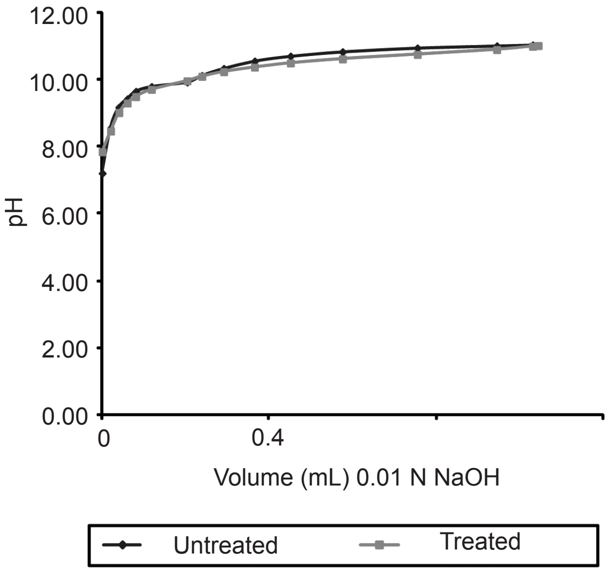

Figure 3 Buffering capacity of untreated and LMwPF-treated bamboo strips in alkaline condition

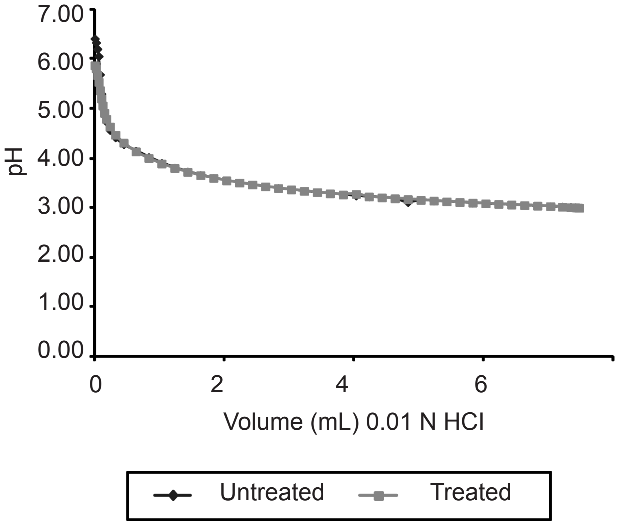

Figure 4 Buffering capacity of untreated and LMwPF-treated bamboo strips in acidic condition

2. Generally, there was significant difference ($p \leq 0.05$) between untreated and LMwPF-treated plybamboo in cyclic boil resistance test, strength retention and bamboo failure. Results also showed that samples met the minimum shear strength requirement of BS EN: 314-1 (BS 2004). Dry shear strength of three-ply LMwPF-treated plybamboo pressed at 140 °C for 33 min pressing time was higher than that of 11 and 22 min as well as untreated plybamboo but the difference was not significant.

Shear bond strength and percentages of bamboo failure after the boards were exposed to cyclic boiling resistance test were markedly reduced (Table 2). The boiling and drying treatment created extensive stresses in bamboo and gluelines which, upon shearing, were easily broken due to the release of internal stresses. When subjected to cyclic boiling resistance test, shear retention of the three-ply LMwPF-treated plybamboo that was pressed for 22 min performed better than those pressed at 11 and 33 min. The high shear strength retention (87%) as well as high bamboo failure (91%) obtained from this plybamboo suggested that its bond integrity was of high quality. Bamboo failure of LMwPF-treated plybamboo is shown in Figure 5.

However, pressing the three-ply LMwPF-treated plybamboo for 33 min appeared to excessively cure the LMwPF resin, so much so that it became brittle due to high crosslinking in the system. Such brittleness resulted in poor glueline, hence significantly increased the adhesive failure. Bamboo failure was 78% in the dry treatment and cured resin was less affected by the cyclic boil resistance treatment. The bamboo was able to maintain its structure with failure of only 61%. Nonetheless, the brittleness of the cured resin had some effect on bond integrity of the plybamboo, which is shown by the low (68%) shear strength retention. Similar findings were observed by Winandy and Lebow (2001) and Winandy and Krzysik (2007). The authors noted that prolonging pressing time affected strength properties of medium density fibreboard.

For the five-ply LMwPF-treated plybamboo, bonding strength and bamboo failure increased when the press time was increased from 22 to 33 min but decreased after 44 min. Apparently, 33 min of pressing time produced high bond strength for dry and cyclic boil resistance test samples. Average dry shear strength and bamboo failure ranged from 2.5 to 3.2 N mm^{-2} and 50 to 76% respectively.

Bonding mechanism of LMwPF-treated plybamboo can be explained using the hypothetical chain of nine links. LMwPF resin affected overall gluing properties of plybamboo by changing the rate of resin cure at the boundary layers between veneers. This analogy (Figure 6) was based on Marra (1961) who established a hypothetical chain during wood bonding. It was interpolated that when bamboo strips were treated with LMwPF resin and later end-jointed

Table 2 Average values of plybamboo shear strength and bamboo failure of LMwPF plybamboo at different pressing times

Plybamboo	Total pressing time (min)	Shear strength ($N mm^{-2}$)		Strength retention (%)	Bamboo failure (%)	
		Dry	CBR		Dry	CBR
Three-ply						
LMwPF-treated	11	3.17 a (0.6)	2.58 a (0.5)	81 ab	83 (13)	42 (26)
	22	3.52 a (0.6)	3.06 a (0.6)	87 a	91 (10)	40 (18)
	33	4.23 a (0.8)	3.18 a (0.6)	68 b	78 (16)	61 (22)
Untreated	6.5	3.08 a (0.7)	1.52 b (0.5)	49 c	27 (11)	17 (9)
Five-ply						
LMwPF-treated	11			Failed		
	22	2.55 b (0.4)	1.56 b (0.2)	58 ab	76 (21)	53 (23)
	33	3.16 ab (0.3)	2.03 a (0.2)	67 a	80 (18)	65 (17)
	44	2.61ab (0.1)	1.56b (0.1)	60 ab	50 (22)	35 (19)
Control	11	3.26 a (0.3)	1.55 b (0.3)	45 b	28 (9)	16 (11)

Values are averages of 160 specimens; values in parentheses are standard deviations; means followed by the same letters in the same column are not significantly different ($p \leq 0.05$); Dry = specimens tested in dry condition; CBR = cyclic boil resistance

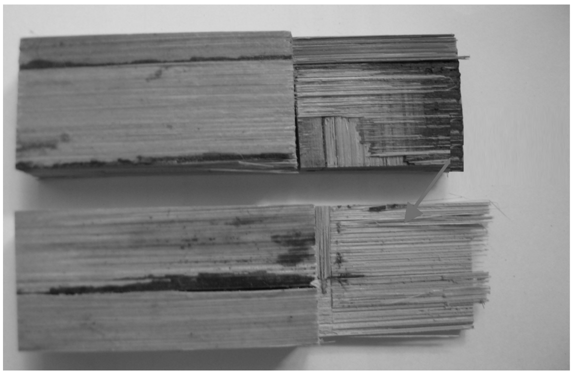

Figure 5 Bamboo failure of LMwPF-treated plybamboo

into bamboo veneers to form plybamboo, an additional link was formed within the nine links. The link occurred either from within (between bamboo strips) or on the surface (between bamboo veneers) of plybamboo. Optimum curing time (during pressing) plays an important role in determining bonding strength of the LMwPF-treated plybamboo. Failure of any one link will cause failure of the hypothetical chain. Similarly, failure at any one of the actual location in the bonded assembly may cause failure to the entire assembly.

Using this new analogy, we concluded that there were 11 link chains for a typical bamboo-to-bamboo assembly compared with 9 in untreated bamboo bonding. The additional two represented LMwPF resin that was embedded in the bamboo strips. Links 10 and 11 represented strength of pure bamboo that had direct link with 8 and 9 which represented the cohesive strength of LMwPF-treated bamboo (Figure 6). Link 1 was the bulk cohesive strength of adhesive film from phenol formaldehyde and also supported by LMwPF resin (between these two phenolic-treated veneers). Links 6 and 7 acted as boundary layers of the two adjacent LMwPF-treated

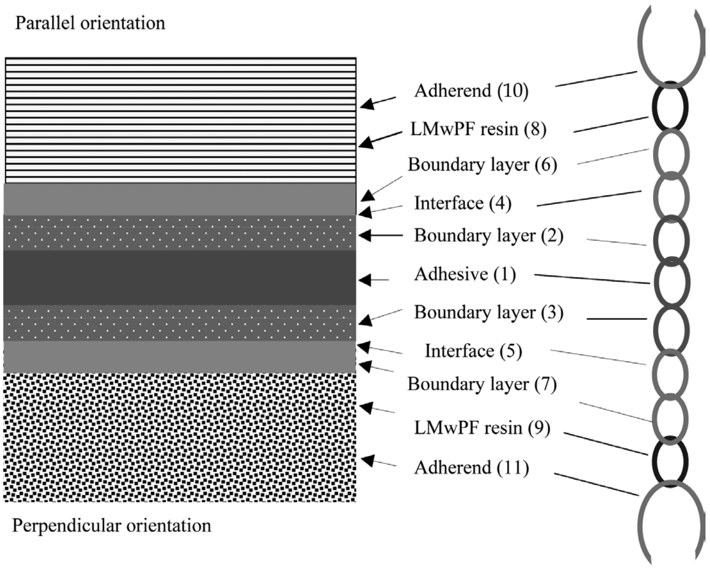

Figure 6 Proposed schematic representation of an adhesive joint showing various links (numbers) in the chain analogy in LMwPF-treated plybamboo

bamboo strips as the former might be altered. This may be due to damage in bamboo fibres caused by machining, drying or the presence of LMwPF resin before bonding of LMwPF-treated plybamboo.

Links 2 and 3 represented boundary layers in the adhesive film, shown by the presence of adherend bond while links 4 and 5 represented adhesive–adherend interface where the actual adhesion occurred. Weak adhesive forces or weak boundary layers in the film caused failure either in the treated bamboo members (links 8, 9, 10 and 11) or in the boundary layers of the treated bamboo (links 6 or 7).

In this study, sheared bamboo failed at the boundary layer (links 6 and 7) and interface (links 5 and 4). High shear strength, followed by high bamboo failures indicated the presence of strong adhesive-bonded joints between phenol formaldehyde and LMwPF resin, between cellulose and phenol formaldehyde, and between LMwPF and cellulose.

CONCLUSIONS

The treatment of bamboo strips with LMwPF resin changed the wettability but not the buffering capacity of the strips. Treated *G. scortechinii* had relatively low wettability compared with untreated strips. Contact angle of water droplet was higher in treated strips than untreated. Results obtained from buffering capacity indicated that samples were more stable towards acidic rather than alkali. Since phenolic-treated bamboo had similar wetting properties with that of wood, the phenol formaldehyde resin formulation used to bond LMwPF-treated bamboo veneers was the same as for wood. Generally, all LMwPF-treated plybamboo had good dry and cyclic boil resistance glue bond shear strength, which met the minimum requirement of BS EN 314-1.

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