CHARACTERIZATION OF BONDLINES IN CROSS-LAMINATED TIMBER MADE WITH PRESERVATIVE-TREATED LUMBER

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Abstract. The number of mass timber construction projects is rapidly increasing in North America but this technology encounters durability issues where termites are present. One method for minimizing this risk is to incorporate termiticidal treatments into mass timber elements. This study examined the impact of cross-laminated timber (CLT) pre and post layup treatment on bond line integrity. Douglas-fir 2 × 6-in. lumber or CLT panel sections were pressure treated with 1) borates or 2) propiconazole, tebuconazole, imidacloprid, permethrin, and iodopropynyl butylcarbamate (PTIP), or 3) dip treated with a mixture of propiconazole, tebuconazole, and imidacloprid + borate (PTIB). CLT panels were manufactured using melamine formaldehyde or polyurethane resins. The impact of preservative treatment on bondline integrity was tested by delamination and block shear tests. Adhesive penetration was also measured using fluorescence microscopy and surface wettability was measured using a contact angle analyzer. Planing-treated lumber before use in CLT panel assembly reduced actives by 57-94% compared with unplaned lumber containing the same treatment. Panels made with borate-treated lumber were more easily delaminated than panels composed of PTIP-treated wood. Microscopic evaluation of CLT bondlines showed greater resin penetration in panels made with PTIP-treated wood; however, penetration was highly variable across specimens. Borate-containing treatments increased surface wettability which may have contributed to reduced treated

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panel performance. The results help define the challenges associated with incorporating biocidal treatments into panels and identify some mechanisms by which they reduce performance.

Keywords: CLT, preservative, bondline, melamine formaldehyde, polyurethane.

INTRODUCTION

Cross-laminated timber (CLT) is an engineered timber product composed of an uneven number of layers (usually three, five, or seven layers) of lumber glued together in alternating layers offset 90° from one another (Brandner et al 2016; APA 2018). The current form of CLT was first developed in Austria in the 1990s and has gained global use as a sustainable building material (Brandner et al 2016). CLT was first used in building construction in the United States in the early 2000s (França et al 2018). Because it is made from wood, CLT sequesters CO_2 from the atmosphere during its service life. The technology also boasts several other advantages including reduced construction time, easier construction clean-up, strength-to-weight performance comparable to concrete or steel, good seismic and fire performance, and reduced energy consumption (Mallo et al 2014; Crawford and Cadorel 2017; Shahan et al 2021; Ayanleye et al 2022).

CLT is a material whose biological components are degradable if they are wetted above the fiber saturation point (~28% MC) at a wide range of temperatures between 5 and 40°C. It is estimated that the annual loss of wooden materials from biodegradation in the United States is around $5 billion and CLT-based structures will factor into these totals as they age (Ayanleye et al 2022). CLT is largely used for structural wall and floor/ceiling assemblies in buildings in interior protected applications that equate to American Wood Protection Association (AWPA) Use Categories 1 and 2. These exposures face limited risks of wetting and fungal decay provided the vapor barriers and cladding remain functional and intact, but interior framing materials such as CLT are at risk for termite attack. North America is home to several large population centers in tropical and subtropical areas where high annual rainfall and temperatures lead to greater decay risk than those found in Northern Europe (Forest Products Laboratory 2021). Additionally, the southern continental United States and Hawaii harbor a number of subterranean termite species, the most economically important wood-destroying insects (Goodell and Nielson 2023). Hawaii is the only U.S. state to require that all structural timber elements be preservative treated to a specific preservative retention and this requirement will apply to mass timber structures (Hawaii amended 2018 IBC section 2303.1.9).

Wood products can be protected from fungal or termite attacks with chemical treatments applied to wood topically or by pressure treatment (Oliveira et al 2018). The AWPA specifies chemical retention levels for UC1 and UC2 that are sufficient for protection against fungal decay and wood-destroying insects as well as a special level for protection against Formosan termites which requires higher chemical loadings for borate-based treatments (AWPA 2021). However, incorporating fungicidal or termiticidal treatments in CLT can be problematic due to the potential for chemical interactions with wood adhesives and the size of panels relative to commercial treating vessels. Preservatives can be incorporated into CLT panels by using pressure-treated wood to manufacture panels (prelayup treatment) or by applying biocide to panels after a layup by pressure or nonpressure processes (postlayup) (França et al 2018; Wang et al 2018). Lamination using dowels avoids the use of adhesives and should enable the use of pressure-treated lamellae in dowel-laminated timber construction without compromising structural performance. However, dowel-laminated timber is not as widely manufactured in North America as CLT. As a result, termite treatment solutions will have to be incorporated into CLT manufacturing or building management for these materials to penetrate the market more broadly.

Although chemical treatments can prevent fungal and insect attacks, they can also adversely affect CLT panel performance (Tascioglu et al 2003). Postlayup treatments can be applied to whole panels with no need to resurface the treated Panels. However, pressure treatment is limited to panels smaller than the treatment vessel diameter (usually under 2-3 m in diameter). These processes are also limited to organic solvent-based treatments because of the risk of swelling and subsequent deformation. Prelayup treatments can be used to produce panels of any dimension, but the requirement to plane the lumber shortly before resin application removes much of the preservative reducing durability and creating chemically treated wastes. Preservatives can also interfere with resin curing, resulting in reduced bondline performance (Cai et al 2022).

A number of factors influence adhesive penetration into wood including wood species, surface characteristics, porosity, surface tension, and pH. Resin characteristics that can affect performance include molecular weight, viscosity, pH value, and curing additives. Excessive adhesive penetration can result in a starved bondline while insufficient penetration produces a thick bondline, with both events leading to an adhesive failure (Kamke and Lee 2007; Ciglian and Reinprecht 2022). The addition of preservatives can further affect adhesive bonding development (Kamke and Lee 2007; Faria et al 2020; Lim et al 2020; Ayanleye et al 2022; Ciglian and Reinprecht 2022; Alade et al 2023).

While preservative treatment may be necessary for the performance of CLT in areas with a substantial risk of termite attack, it will be important to confirm that the treatments do not adversely affect glue line bond properties. The objective of this study was to evaluate the effects of pre and postlayup treatments on bondline integrity of Douglas-fir CLT panels.

MATERIALS AND METHODS

Materials

Untreated CLT panels were made with coastal Douglas-fir 2 × 6-in. nominal lumber obtained in western Oregon. All treated lumber was also made from coastal Douglas-fir. Borate-treated lumber used to make panels was obtained from a commercial facility in western Oregon and was treated with a solution of sodium octaborate tetrahydrate. PTIB-treated lumber was obtained from a commercial facility and was dip-treated in a solution of disodium octaborate tetrahydrate augmented with propiconazole, tebuconazole, and imidacloprid. PTIP-treated wood was pressure treated in a commercial facility using a proprietary solution containing propiconazole, tebuconazole, imidacloprid, permethrin, iodopropynyl butyl carbamate (IPBC), and minor amounts of borates. All lumber used for CLT panel construction in this study was commercially available stock material and contained random proportions of heartwood and sapwood.

Melamine formaldehyde (MF) adhesive was a commercially available preparation that required a hardener to be mixed before application. Polyurethane (PUR) adhesive was a commercially available single-component adhesive that required a primer to be applied to the wood before adhesive application.

CLT Panel Fabrication

CLT panels were made using 51 × 152 mm nominal (2 × 6-in.) Douglas-fir lumber that was either untreated, dip-treated, or pressure-treated with one of three preservative systems and one of two resin systems. All lumber used for panel manufacture was planed 1.6 mm (1/16th in.) before layup using a Leadermac LMC-460PL 4-side planer/molder (Blaine, WA). Three ply 1.52 × 2.44 m (5 × 8 ft) panels were made for each treatment in addition to three untreated panels at the Tall Wood Design Institute at Oregon State University. Panels were glued with either MF or PUR resin in combination with treated or untreated wood as shown in Table 1. The resin was applied using a custom APQUIP Co. resin applicator equipped for one and two-component resin systems (Monterey, CA). Lumber was passed through a resin curtain calibrated to deliver a specified application rate. MF was applied via a two-part applicator with a 1.5:1 resin:catalyst ratio at a rate of 367 g/m^2. Assembly time was 75 min at 21°C, and press time was 5 h at 0.83 MPa (120 psi). The one-part polyurethane was

Table 1. Preservative treatments and resin types used to produce nine three-ply, 5 × 8 ft Douglas-fir CLT panels examined in this study.

Lumber preservative treatment	Preservative components	Adhesive
Postlayup treatments (PTIP)	Propiconazole, tebuconazole, imidacloprid, permethrin, iodopropynyl butyl carbamate	Melamine formaldehyde
Postlayup treatments (borates)	Disodium octaborate tetrahydrate	Melamine formaldehyde
Postlayup treatments (PTIB)	Borates + propiconazole, tebuconazole, imidacloprid	Melamine formaldehyde
Control	Untreated control	Melamine formaldehyde
Organic pressure treatment (PTIP)	Propiconazole, tebuconazole, imidacloprid, permethrin, iodopropynyl butyl carbamate	Melamine formaldehyde
Borate pressure treatment (borates)	Disodium octaborate tetrahydrate	Melamine formaldehyde
Borate/organic diptreatment (PTIB)	Borates + propiconazole, tebuconazole, imidacloprid	Melamine formaldehyde
Organic pressure treatment (PTIP)	Propiconazole, tebuconazole, imidacloprid, permethrin, iodopropynyl butyl carbamate	Polyurethane
Borate pressure treatment (borates)	Disodium octaborate tetrahydrate	Polyurethane
Borate/organic diptreatment (PTIB)	Borates + propiconazole, tebuconazole, imidacloprid	Polyurethane

applied at a rate of 137 g/m^2 to boards that had previously been wetted with a 5% primer solution at 20 g/m^2. Adhesive application rates followed manufacturer recommendations, and these were assumed to be comparable with current industry standards at the time. Lumber was arranged into crosslams randomly with no attempt to normalize laminae for heartwood and sapwood content across panels. Lumber was arranged on a 2.44 × 3.05 m (8 × 10 ft) Minda laboratory CLT press (Minden, Germany). Assembly time was 60 min at 20°C and press time was 2.5 h at 0.83 MPa (120 psi) with pressure applied on the top and sides.

Nine CLT panels were produced for this study. Three panels were made using untreated Douglas-fir lumber and were subsequently cut into nine 457 × 762 mm (18 × 30-in.) subpanels each. Six test subpanels from each were treated with one of three preservative treatments described in the section "Preservative Treatment". Two of the postlayup treated panels per treatment were retained at OSU for testing. The remaining subpanels were treated and sent to Hawaii for separate termite testing.

The remaining 6 panels were produced using pressure or dip-treated lumber that was planed before assembly. Two panels were made per treatment type, one using MF resin and another using PUR resin as described above. The preservative treatments used are described in Table 1.

Preservative Treatment

Lumber for the prelayup treatments and test panels used for postlayup preservative treatments were treated on a commercial scale treating plants using pressure or nonpressure processes.

Borate pressure treatment was performed in a commercial cylinder using disodium octaborate tetrahydrate (DOT) to the AWPA U1 Formosan termite retention level for borates of 6.7 kg/m^3 (0.42 pcf) DOT. Lumber and test panels were treated using a full cell process with a 30 min vacuum at -11.9 kPa (26 in Hg) followed by filling the cylinder with a 10% w/w DOT solution. The pressure was applied at 598 kPa (88 psi) within 15 s and then raised to an average of 931 kPa for about 14 min to the target retention. The pressure was reduced to 102 kPa (15 psi) over 60 s and the cylinder was drained before pulling a final vacuum of -11.9 kPa (26 in Hg). Net absorption by gauge was 1693 L (448 gal). The full cross section of borate pressure-treated lumber was penetrated with borates as assessed by the treater.

PTIB dip treatment was carried out by soaking lumber or test panels in a treating solution containing 1.8% PTI and 11% DOT. The treatment targeted a DOT retention of 1.9 kg/m^3 (0.12 pcf). Lumber and test panels were immersed in the treating solution at 43.3°C (110°F) for 15 min PTI retention calculated by solution uptake was 0.37 kg/m^3 (0.021 pcf). The full cross section of PTIB dip-treated lumber was penetrated with borates as assessed by the treater.

PTIP treatment was done in a commercial facility producing lumber used for interior applications in Hawaii. Retentions targeted were suitable for Formosan termite exposure in Hawaii and chemical penetration was assessed at the treating facility.

The active ingredients in lumber or test panels treated with these systems were quantified in the outer 0.4-in. (10 mm) assay zone using the appropriate AWPA standard. Materials were assayed for borates while propiconazole and tebuconazole were assessed for PTIB and PTIP. The assay zone of lumber from each treatment was ground to 20 mesh in a Wiley Mill before analysis. Borates were quantified using the Azomethine method, AWPA standard A65 (AWPA 2021). Propiconazole and tebuconazole were quantified in the organics-containing treatments by high-performance liquid chromatography using a modified version of AWPA standard A48 (AWPA 2021). Permethrin, imidacloprid, and IPBC were not quantified in the organic treatment and the azole fraction was used as a measure of total chemical retention (Table 2). The Formosan termite standard listed includes a sum of propiconazole, tebuconazole, and imidacloprid; however, imidacloprid was not assayed in lumber used in this study.

Bondline Strength Characterization Using Delamination and Block Shear

Three specimens were collected from each 18 × 30-in. subpanel for a total of 12 blocks per treatment according to guidelines outlined in the ANSI/APA PRG-320 standard (APA 2018). One exception to this was for the postlayup treated panels where triplicate samples were only sourced from three of the four replicates per treatment. This was due to unbonded regions after manufacturing in some of the test panels that

Table 2. Retentions of borates, azoles, and imidacloprid in the 0.4-in. (10 mm) AWPA assay zone in pre and postlayup treatments alongside Formosan termite standard retentions listed by AWPA. Prelayup treatments were planed before panel fabrication and show a % percent preservative loss in this assay zone compared with unplaned treated lumber from the same facility.

Treatment	Planed/unplaned	Sample type	Resin system	Boron retention levels (kg/m^3)	% Loss vs unplaned lumber	Tebuconazole (kg/m^3)	% Loss vs unplaned lumber	Propicon azole (kg/m^3)	% Loss vs unplaned lumber	Total azole (kg/m^3)
AWPA formosan termite standard				4.5		0.08		0.08		0.21
Untreated	Planed	Untreated Panel	MF	0.31[a]		0.00		0.01		0.01[a]
Borate	Unplaned	Lumber	N/A	31.90		N/A		N/A		N/A
PTIB	Unplaned	Lumber	N/A	1.98		0.10		0.20		0.30
PTIP	Unplaned	Lumber	N/A	3.47		0.03		0.16		0.19
Borate	Unplaned	Postlayup treated	MF	27.68[c]		N/A	N/A	N/A		N/A
PTIB	Unplaned	Postlayup treated	MF	1.27[b]		0.13	−23.53	0.23		0.36[b]
PTIP	Unplaned	Postlayup treated	MF	2.58[b]		0.07	−97.06	0.31		0.38[b]
Borate	Planed	Prelayup treated	MF	13.42[c]	57.93	N/A	N/A	N/A	N/A	N/A
PTIB	Planed	Prelayup treated	MF	0.12[a]	93.94	0.00	100.00	0.01	96.98	0.01[a]
PTIP	Planed	Prelayup treated	MF	1.21[b]	65.13	0.02	29.41	0.17	−10.97	0.20[a]
Borate	Planed	Prelayup treated	PUR	10.46[c]	67.21	N/A	N/A	N/A	N/A	N/A
PTIB	Planed	Prelayup treated	PUR	0.22[a]	88.89	0.03	70.59	0.05	73.87	0.08[b]
PTIP	Planed	Prelayup treated	PUR	2.32[b]	33.14	0.06	−64.71	0.19	−23.87	0.25[c]

prevented the selection of sound test specimens for delamination and block shear tests. Two 3 × 6 in. (76 × 152 mm) by panel thickness samples were taken from near opposing corners at least 1.5 in. (38 mm) from the panel edge and one sample was taken from the center of the panel. Each sample was cut into two paired 3 × 3-in. (76 × 76 mm) samples. One of the samples was reserved for a block shear test and the other for a delamination test. Delamination test specimens were weighed and inspected for preexisting bond issues and marked accordingly before being submerged in water and subjected to a -70 ± 20 kPa (21 ± 6 in Hg) vacuum stage for 30 min, followed by a 520 ± 20 kPa pressure stage for 2 h. The saturated specimens were oven-dried at 70.0 ± 0.1°C for about 14 h until MC was reduced to about 10-15% w/w before soaking. After drying, each specimen was visually inspected for bondline separation and measured using a ±0.01 mm caliper. Delamination was calculated according to Eq 1:

$$D\ \% = \frac{X - L}{L} \times 100\% \qquad (1)$$

where D % is the percentage of delamination of the sample, X is the measured (mm) delamination of the bondline, and L is the total bondline length (mm) before being placed in the pressure vessel. In addition, samples were graded as pass/fail according to the PRG-320 criteria where all specimens must contain less than 5% delamination, otherwise, the whole panel from which samples are taken fails to meet the requirements (APA 2018). A total of 12 blocks were taken from two prelayup panels and nine from postlayup panels within each resin-treatment combination.

Block shear tests were performed according to the ASTM D905 method referred to in PRG-320. Tests were performed on 3 × 3-in. (76 × 76 mm) specimens sampled from the same location as paired specimens tested for delamination. The specimens were then cut into a stair step shape according to ASTM D905. An Instron universal testing machine (Norwood, MA), equipped with a 10kN capacity load cell with an accuracy of ±0.4 N was used to load each bond surface to failure. The ultimate load was recorded. After testing, wood failure (WF) was assessed on both sides of the fractured bond surfaces by illuminating the broken surface with ultra-violent light and photographing the surface. Image J was used to determine the percentage of the surface that contained MF or PUR resin, indicative of adhesive failure, using the plugin Trainable Weka Segmentation (version 3.8.5).

Microscopic Examination of the Bondline

Microscopic analysis of bondline integrity for each treatment listed in Table 1 was done on 25 × 25 × 25 mm blocks taken from near the panel edges (3 on each side) and the panel center (three total). Each sample contained two bondlines and the sections were microtomed to produce 90-μm thick sections across both bondlines in the sample for a total of 18 thin sections per treatment. Each bondline section was dyed with safranin-o red and placed on a glass slide with coverslip (Bastani et al 2016). Samples were then examined under a fluorescent microscope to observe adhesive penetration into the wood using a 40× objective with an excitation wavelength of 450-490 nm. Average cell depth was determined by counting the number of cells from the bondline containing adhesive and dividing by the number of cell rows visible in the photo. Penetration was only measured in the CLT lamina visible in cross section under the microscope.

Surface Wettability

Wettability was measured on treated boards after planing using a Biolin Scientific contact angle analyzer to measure resin droplet contact angle with the wood surfaces by the sensile drop method on all of the treated materials except for PTIP due to loss of samples before the completion of the experiment. The planed boards had ~1.5 mm planed off to represent the process used to manufacture CLT with the treated wood. The boards were cut into 1-in. × 5 ½-in. samples conditioned at 65% RH and 20°C for over a week before testing. The contact angle was examined with MF, PUR resins, and water using a

19-gauge syringe to place drops on the tangential early wood surface of each specimen. Three replicate drops were placed on opposite sides of each specimen for a total of six measurements per piece. The surface tension of the resins was measured by the pendant drop method at 22°C and 65% RH using Young-Dupré Eq 2. Surface energy was found for each treated and nontreated surface.

$$SE = ST_a(1 + \text{Cos}(\text{Ø})) \qquad (2)$$

where SE is the surface free energy of the wood surface, ST_a is the surface tension of the adhesive droplet, and the cos (Ø) is the contact angle of the liquid drop formed on the wood surface.

Statistics

The data were subjected to an analysis of variance (ANOVA) among the control and resin/preservative treatment combinations at $\alpha = 0.05$, using the Excel statistical package. All average data are shown with ±1 SD.

RESULTS AND DISCUSSION

Effect of Planing on Chemical Retention of Wood Treatments

Planing is necessary for all lumber used in the manufacture of CLT to ensure boards fit together flush and to create an activated surface to facilitate bond development. This is a problem for the use of pressure-treated or dip-treated Douglas-fir lumber in CLT because most of the preservative chemical is loaded near the surface of the wood in the outer 10 mm (0.4-in.) assay zone for 50 mm (2-in.) thick lumber. Chemical retention data for the different treatments used in this study are shown in Table 2 along with the calculated losses due to planing relative to nonplaned control lumber of the same treatment. Major losses were seen in the prelayup treatment due to the planing step (Table 1). Planing removed 33-94% of the borates in the AWPA assay zone for the prelayup treatments while borate losses where the principle active preservatives losses ranged from 57 to 94%. The greatest losses were seen in the borate-PTI dip treatment which were 89-94%. This is because nonpressure treatments result in lower penetration leaving the vast majority of preservative in dip-treated lumber in the very outer layers where it was planed off. It is important to note that despite losing 58-67% of the original borates in the AWPA assay zone due to planing, prelayup panels pressure treated with borates still contained more than double the AWPA retention level for Formosan termites. This suggests that despite planing, the borate-treated lumber used in this study would still effectively resist Formosan termite attack.

Nearly all of the azoles in the AWPA assay zone were lost from the PTI dip-treated wood due to planing. This indicates low levels of preservative penetration in the dip treatment which is to be expected. Azole levels in the PTIP treatment were higher in planed panels than the nonplaned controls, except tebuconazole in the MF panel. These observations suggest that retentions were still sufficient to maintain Formosan termite protection.

Preservative retentions tend to be higher near the surface and planing removes this material. If the 10 mm assay zone specified in the AWPA Standards is used to measure retentions, then the assay zone on the planed boards is likely to have a lower retention in what becomes the new assay zone. These data indicate that the planing of treated wood for CLT manufacture results in chemical waste and may also result in reduced preservative retentions in the resulting panels. However, retention levels for most of the prelayup pressure treatments after planing were still above those specified for protection against Formosan termites. This is especially notable with the azole retention levels for the PTIP treatment after planing.

It must be noted that retentions on unplaned material were not measured in the same boards used in the CLT layup although they were prepared at the same time. Thus, there may be some variations between the boards. This explains why azole retentions were higher for planed material for some of the prelayup-treated PTIP panels.

Delamination

Some panels had areas with considerable delamination that were avoided when obtaining test samples. Heavily delaminated panels included a subpanel from one of the replicates treated with PTIP after layup. The extensive delamination in this panel limited the number of samples that could be tested for bondline integrity. Borate postlayup treatment also absorbed extensive moisture and had a high amount of preexisting delamination. The delamination could reflect poor handling and storage or manufacturing error creating too much open assembly time before pressing (Long and Morrell 2011; Ayanleye et al 2022; Lukowsky and Nguyen 2023). Minimal planing was done on the panels to try and limit preservative loss. This also may have limited effective bonding in some of the panels. Although it would be difficult to delineate, it is possible that the treatment of the assembled panels induced stresses along the gluelines that contributed to subsequent delamination. Current APA standards preclude pressure treatment of laminated timbers with water-based systems due to these concerns, although there is evidence that the process does not negatively affect flexural properties (Long and Morrell 2011). Test samples for the delamination test were selected to be free of visible delamination and only two borate-treated samples had any preexisting delamination before they were tested.

Delamination results for all of the different treatments are shown in Table 3. Postlayup treatments had lower levels of delamination than prelayup treatments. Organic and borate postlayup treatments performed similarly to the untreated controls (1.7%) while delamination in PTIB was at 4.5%. Previous studies have also shown that postlayup preservative CLT treatment had less effect on bondline integrity since it eliminated the risk of preservative interference with resin curing (Tascioglu et al 2003; Kuka et al 2022, Taylor et al 2022).

The most extensive delamination was observed in prelayup treatments (Table 3). The greatest delamination was observed in borate and PTIB-based treatments, especially with PUR panels. The panels composed of borate-treated lumber and MF resin were the only other panel to have some prior delamination. PTIP-based treatments were associated with the least delamination for either resin (slightly over 2%) (Faria et al 2020).

Table 3. Effect of pre and postlayup preservative treatment of CLT panels on delamination and block shear. Standard deviations are shown in parentheses.

Test	Control	Postlayup			Prelayup					
Delamination	Control	PTIP	Borate	PTIB	MF-PTIP	MF-Borate	MF-PTIB	PUR-PTIP	PUR-Borate	PUR-PTIB
Total # of specimens	12	9	9	9	12	12	12	12	12	12
Predelamination check	X	X		X	X		X	X	X	X
Avg. sample % delamination	1.7% (2.3)	0.8% (.8)	1.1% (1.5)	4.5% (2.8)	2.5% (2.5)	5.1% (3.3)	5.3% (9.2)	2.2% (2.1)	7.7% (6.2)	14.2% (10.1)
% Specimens failed[a]	(10/12) 17% (F)	(8/9) 11% (F)	(8/9) 11% (F)	(7/9) 33% (F)	(9/12) 25% (F)	(8/12) 33% (F)	(9/12) 25% (F)	(11/12) 8% (F)	(7/12) 42% (F)	(5/12) 58% (F)
Block shear										
Total # of specimens	12	12	12	12	12	12	12	12	12	12
Avg. WF of all specimens[b]	94.2% (5.1)[a]	98.1% (3.6)[a]	98.0% (3.2)[a]	96.3% (3.2)[a]	97.9% (3.9)[a]	98.2% (2.7)[a]	96.5% (5.3)[a]	93.0% (9.2)[a]	84.2% (23.4)[a]	75.2% (21.6)[b]
Above 60% WF specimens[a]	100%	100%	100%	100%	100%	100%	100%	100%	(11/12) 8% (F)	(9/12) 25% (F)

[a]F = failure as defined in the PRG-320 standard where if 5% of samples fail the panel does not pass.
**Different lowercase letters indicate statistically significant differences (ANOVA, $\alpha = 0.05$).

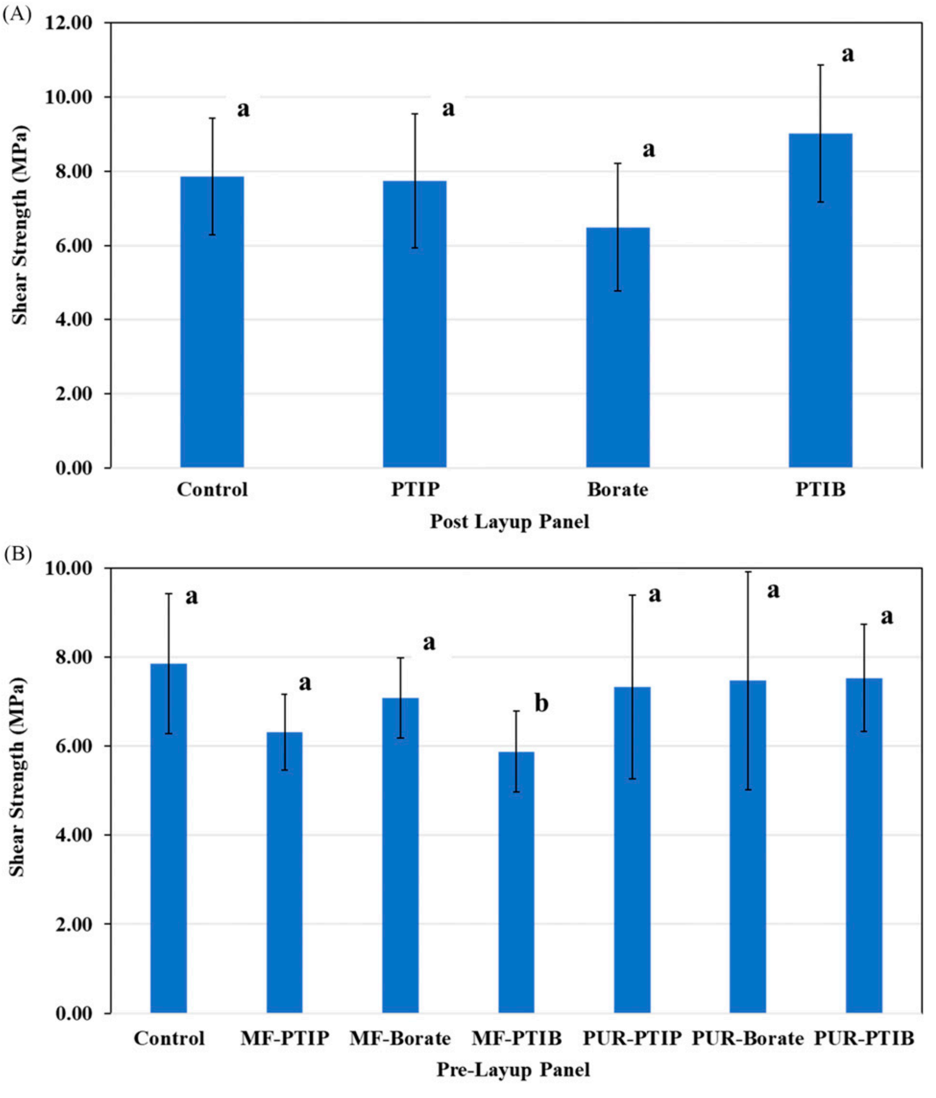

Figure 1. Effects of post layup (A) treatment and pre-layup (B) treatment on maximum shear strength of CLT panels tested. Error bars represent 1 SD while bars with the same letters do not differ significantly from the control at $\alpha = 0.05$.

Mbhamali et al (2022) reported similar negative delamination effects of boron treatments in combination with PUR resins possibly due to the effects of boron on the surface energy of the wood.

Block Shear

Shear strength values were similar among all treatments (Bagheri et al 2022) (Fig 1). The proportion of wood to glueline failure is usually a good indicator of bondline performance (Wang et al 2018). PRG-320 standard requires a minimum of 60% WF in a glueline shear test. WF was above 90% for all treatments except prelayup treatments with borates or PTI. The success of postlayup treatments by this metric could be due to the successful formation of an adhesive bond in the absence of chemical, whereafter chemical interference with the bondline would not be able to interfere with resin curing (Künniger et al 2019;

Kuka et al 2022). Long and Morrell (2011) performed a similar test with nonincised Douglas fir beams and found that post-layup treatment with DOT had the minimum effect on bondline performance and high WF. These findings were consistent with the fact that borates affect resin cure but would be expected to have a negligible effect on cured resin.

PUR was most affected by borate or PTI treatments with WFs of 84.2% and 75.2%, respectively (Table 3). The slightly lower degrees of WF suggest that that adhesive type may play a slight role (Lim et al 2020; Mbhamali et al 2022). Künniger et al (2019) noted that the aromatic backbone of MF resins can form strong crosslinking networks resulting in less ductile failures compared with PUR which may have contributed to less WF.

Increasing boron concentrations in wood composites have been shown to reduce shear strength in PUR- and MF-based composites (Özçifçi 2006; Mbhamali et al 2022). High concentrations of boron have been shown to have negative effects on panel strength (Taylor et al 2022). Organic biocides have been shown to have lower impacts on composite strength than inorganic treatments, although some negative effects have been observed (Antwi-Boasiako and Appiah 2012; Faria et al 2020).

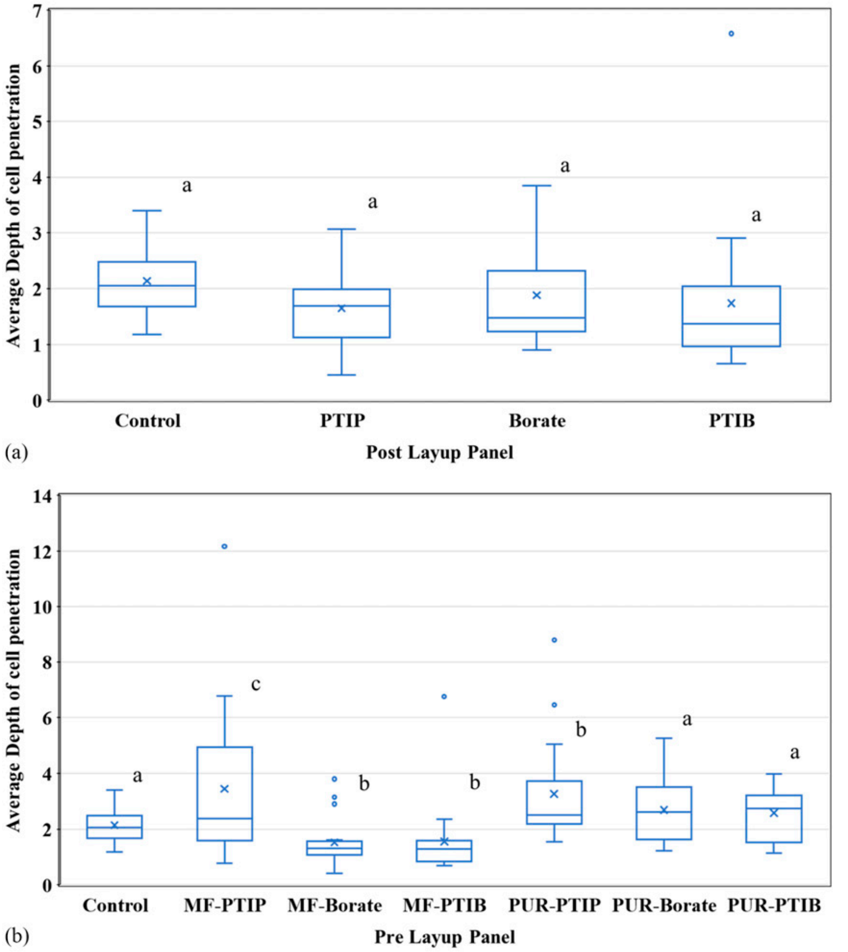

Figure 2. Effect of pre (A) and postlayup (B) preservative treatment of CLT on adhesive penetration represented as the average number of cells containing visible resin from the glueline.

Alipon et al (2018) found comparable results to the current study with deltamethrin + propiconazole and various resins.

Wettability and Adhesive Penetration

Adhesive penetration in CLT panels can be readily visualized by fluorescence microscopy since the resins fluoresce while the wood remains darkened red by safranin staining. Adhesive penetration varied widely between samples even within treatments, which sometimes made it difficult to compare different treatments (Fig 2). Preservative treatments further complicated the analysis.

Adhesive penetration did not differ among the postlayup treatments reflecting the fact that the resin cured before treatment (Fig 2[a]). However, there was evidence of bondline damage with noticeable voids, which could be due to the partial weather cycle during pressure or dip treatment as the water-borne preservative swelled the wood and stressed the bondline (Fig 3[a]).

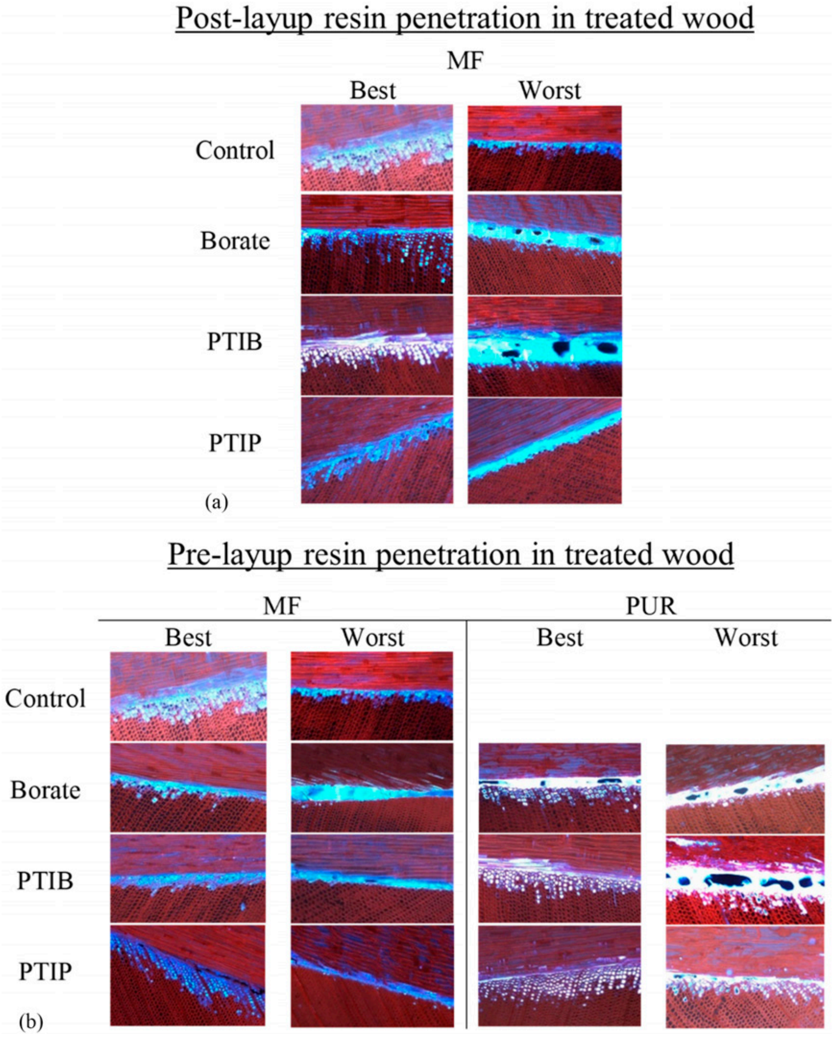

Figure 3. (A) Post layup, melamine formaldehyde (MF) adhesive. (B) examples of safranin-O-stained fluorescence micrographs of CLT bondlines taken from panels made with wood treated with one of three biocides and two adhesives MF or polyurethane (PUR).

Table 4. Contact angle and surface energy for adhesives on treated/nontreated early wood planed boards. Stats compared with control and respective resin treatment combination within the same group.

Sample	CA (θ)	SE (mN/m)
MF-Cont.	82 (14)[a]	69 (12)[a]
MF-PTIP	N/A	N/A
MF-Borate	77 (12)[a]	75 (12)[a]
MF-PTIB	78 (16)[a]	73 (17)[a]
PUR-Cont.	35 (9)[a]	44 (2)[a]
PUR-PTIP	N/A	N/A
PUR-Borate	35 (6)[a]	44 (2)[a]
PUR-PTIB	34 (8)[a]	44 (2)[a]
Water-Cont.	57 (23)[a]	118 (24)[a]
Water-PTIP	N/A	N/A
Water-Borate	7 (2)[b]	152 (1)[b]
Water-PTIB	11 (2)[b]	151 (4)b

Adhesive penetration varied widely in CLT composed of pretreated lamella. Adhesive penetration for MF-Borate and MF-PTI-treated panels trended lower than the controls although there were examples of the opposite trend. This was surprising since contact angle tests using MF adhesive on treated wood used in this study showed that treatments, particularly borates and PTI, were associated with decreased contact angles, indicating greater wettability (Table 4).

Previous studies indicate that boron limits adhesive penetration into the wood surface and increases gelation (Gao et al 2016; Alade et al 2022, 2023). Other studies have shown that lower borate concentrations can have a greater negative effect on bondline quality than higher concentrations by reducing adhesive penetration, creating a thicker bondline, and reducing bond strength (Qin et al 2019; Lim et al 2020).

Borate and PTI treatments were also associated with increased wood surface wettability in CLT constructed with PUR. Microscopic analysis suggested that average PUR penetration was deeper than MF for each treatment type. PUR tends to have lower polarity and a higher viscosity which would limit penetration into the wood cells (Ciglian and Reinprecht 2022). Other preservative systems have been shown to increase wood wettability such as micronized copper azole-treated southern yellow pine using PUR (Cai et al 2022). Composites made with this material had the thinnest bondlines compared with control and the lower retention treatments. Ciglian and Reinprecht (2022) found that treating spruce composites containing PUR with various inorganic treatments reduced surface wettability except with boric acid. Adhesive penetration was also increased over control for all preservatives with boric acid having the highest penetration overall. A similar mechanism could be driving the poorer bondline performance in borate-containing treatments in this study.

Increased resin penetration may occur via several mechanisms. Preservatives can negatively affect resin penetration by physically or chemically blocking resin movement (Lorenz and Frihart 2006; Kamke and Lee 2007; Lim et al 2020). Resin penetration can also be affected by latewood and earlywood differences which can also affect preservative distribution. Earlywood is more permeable and creates a better wettable surface than latewood. Adhesives will follow the path of least resistance during pressing and flow more into earlywood cells than latewood which can cause air pockets to form and create an uneven bondline (Cai et al 2022; Ciglian and Reinprecht 2022). PUR curing is highly dependent on wood moisture and releases carbon dioxide that can create an inner vapor pressure driving more adhesive further into the wood and away from the bondline (Bastani et al 2016). This might explain why PUR panels in our study appeared to have more voids in the bondline than MF (Fig 3[b]). It is also important to note that this study only focused on one commercially relevant wood species used in CLT manufacture, Douglas-fir. The adhesive penetration patterns observed here in combination with treated Douglas-fir may vary with other wood species due to anatomical differences in species that could impact adhesive flow. Chemical distribution can also differ greatly between wood species and these differences may cause changes in how and where adhesives interact with preservative chemicals during the bonding but were not assessed in this study.

The deepest resin penetration was noted with organic treatments for both resin types. In fact, by visual observation, organic treatments had the thinnest bondlines for both the best and worst slide samples (Fig 3[b]). Alipon et al (2018) found that organic treatments such as propiconazole, deltamethrin, and permethrin outperformed inorganic treatments with various adhesives and provided full protection against termite attacks. Cai et al (2022) noted that thin bondlines usually can outperform composites with thicker bondlines as the more highly concentrated treatments accelerated curing reactions and created extensive branching within the wood structure. This could also explain why some authors observed increased shear strength as preservative concentration increased which produced a consequent increase in wettability (Tascioglu et al 2003; Lim et al 2020).

CONCLUSIONS

Prelayup planing of treated wood reduced the active preservative concentrations in MF and PUR CLT panels, average levels were still sufficient to protect pressure-treated materials against Formosan termites on average. This suggests that panels made with pressure-treated laminae (borates and PTIP) would provide good protection against Formosan termites. The presence of preservatives in the wood did impact bondline quality by increasing wettability and starving the bondline. Of the treatments, PTIP, appeared to have less of an effect on the bondline integrity even though the bondline was the thinnest of those in preservative-treated wood. All panels made with treated laminae in this study had significant issues with delamination that would have resulted in them failing quality control according to the PRG-320 standard. This finding indicates there are significant hurdles to manufacturing CLT with currently available preservative-treated wood, although one of the treatments, PTIP had a lower negative effect on panel performance.

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