Adhesive Bonding Performance of Thermally Modified Yellow Poplar

Abasali Masoumi,[a,]* Francisco Xavier Zambrano Balma,[b] and Brian H. Bond [c]

Thermal modification of wood changes its chemical, physical, and structural properties, which may affect adhesive bondline quality and bonding performance. This research compared the effect of thermal modification on the adhesive bonding performance of poplar (*Liriodendron tulipifera*) wood. Samples were prepared from thermally modified and unmodified yellow poplar using one-component polyurethane (PUR) and polyvinyl acetate (PVA), as they are adhesives used in wood products. Microscopic properties of the bondlines were investigated to understand shear performance and durability. Adhesive line thickness, penetration, shear strength, and moisture durability were measured, and failure modes were recorded. Thermal modification negatively affected the wood and adhesive interaction by reducing penetration (31.2% in PUR and 29% in PVA), therefore creating a thicker adhesive line (70% in PUR and 2% in PVA) and consequently causing a significant reduction in the shear strength of both adhesive types (27% in PUR and 36% in PVA) compared with non-modified specimens. The PUR adhesive had higher shear strength than PVA by 2.7% in non-modified and 14% in thermally modified wood.

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*Contact information: a: Ph.D. Candidate, Department of Sustainable Biomaterials, Virginia Polytechnic Institute and State University, Blacksburg, VA, USA. https://orcid.org/0000-0002-9312-455X; b: M.Sc. Student, Department of Sustainable Biomaterials, Virginia Polytechnic Institute and State University, Blacksburg, VA, USA. https://orcid.org/0009-0002-1737-7536; c: Professor and Associate Dean of Extension, Outreach and Engagement, Department of Sustainable Biomaterials, Brooks Forest Products center, Virginia Polytechnic Institute and State University, Blacksburg, VA 24061, USA; https://orcid.org/0000-0001-6712-8250; *Corresponding author: masoumi@vt.edu*

INTRODUCTION

The use of thermally modified wood (TMW) as decking, cladding, *etc*., in the wood products and construction industry is growing rapidly (Espinoza *et al*. 2015). Thermal modification (TM) is performed by heating the wood in lack of oxygen and without using chemicals at temperatures between 180 and 260 °C (Hill 2006; Can 2020). Thermo-vacuum modification, which is a combination of vacuum drying and heat treatment, eliminates cell wall destruction residue during modifying process. TM changes the chemical composition, physical properties, and mechanical properties of wood. It increases its durability, reducing hygroscopicity, and improves the dimensional stability of wood. During TM, extractives (fats and waxes) migrate toward the surface of the wood; hydroxyl groups (OH) are eradicated in hemicellulose and cellulose, and crystallinity of cellulose changes; lignin degrades and lignin plasticization occurs and new bonds are created between lignin and other components (Hill *et al.* 2021; Esteves and Pereira 2009). Changes induced by TM, which include decreasing its wettability and surface free energy, can negatively affect wood adhesive bonding by creating a hydrophobic surface layer and reducing the wettability depending on the types of adhesives used (Adamopoulos *et al.* 2012).

Many factors such as adhesive type, wood species, surface quality, pressure, and curing condition affect bonding quality. Strong adhesion between the adhesive and the wood can be achieved through adequate wetting, adhesive flow, penetration, and curing (Marra 1992; Pizzi and Mittal 2011). Most wood adhesives contain a large amount of water as a solvent (Sernek *et al*. 2008). Polyurethane (PUR) and polyvinyl acetate (PVA) are water-based adhesives and are among the common industrial and structural adhesives used in wood products (Brandner 2013). For water-based adhesives, the distribution of adhesive on wood surfaces and penetration of adhesive into porous wood structures is highly impacted by the wettability of the surface (Stoeckel *et al*. 2013). Follrich *et al*. (2006) reported that the wettability decreases in TMW due to the surface becoming hydrophobic, less polar, and significantly repellent to water. The low hygroscopicity of TMW also affects the curing of water-based adhesives, since the absorption process performs very slowly (Esteves and Pereira 2009). The movement of extractives toward the surface of the wood during TM makes its surface more hydrophobic (Kral *et al*. 2014). TM also reduces penetration of the adhesive into the porous wood structure (Sernek *et al*. 2008).

The reduction in penetration increases adhesive line thickness, which is a potential area for CO_2 microbubbles to get trapped in crystalized adhesive and play a starting point in fracture in shear stress (Masoumi and Gholamian 2022). Therefore, when curing waterborne adhesives used for bonding TMW, the required open time is much longer than for non-modified wood. There are methods suggested for improving the bonding performance of TMW, such as removing surface layers by planning to mitigate the thermal degradation intensity (Chu *et al*. 2020).

The adhesive penetration into the wood pores affects its bond strength and durability, influencing the performance of wood products (Modzel *et al*. 2011). The penetration of adhesive into the wood makes more contact between the adhesive and cell wall and provides mechanical interlocking between the cells (Mirabile and Zink-Sharp 2017). However, weak penetration is insufficient for chemical bonding or mechanical interlocking. Moreover, penetration affects stress distribution between adherents when subjected to load (Kamke and Lee 2007).

TMW wood has lower shear strength because of its affected bonding quality and changes in the shear strength of the wood structure itself. TMW can be brittle and have decreased affecting the shear strength of adhesive bonds (Kariz and Sernek 2010). The lack of adhesion between wood fibres can cause the failure of the bonded wood even if the adhesive line is intact (Hill 2006). The adhesive bond strength decreases because of poor bonding or due to the reduced strength of heat-treated wood, but it is difficult to determine which factor contributes more (Kariz and Sernek 2010).

Yellow poplar (*Liriodendron tulipifera*) is thermally modified and sold commercially, but a compromise has to be made in its bonding quality and shear strength. The goal of this research was to study the bonding durability of thermally modified yellow poplar wood under thermo-vacuum treatment. Specifically, the objectives were to measure the microscopic quality of bondline, particularly, adhesive penetration, and to compare the shear strength and moisture durability of bonding for two common adhesives of PUR and PVA for TMW/TMW and control/control.

EXPERIMENTAL

Thermally modified and nonmodified yellow poplar lumber was provided by a commercial producer in the Appalachian region. The green lumber was kiln dried and then modified in a thermo-vacuum process at 200 °C by the commercial supplier. The lumber was then conditioned at 12% moisture content (temperature of 23 °C; relative humidity of 50%) for six weeks, machined to remove the hydrophobic surface layer and provide a suitable surface for bonding, and cut to size of 50×8×2.5 cm. Samples were then glued with two different adhesives on their tangential surface: one component PUR and PVA following the manufacturers' instructions (140 g/m^2). The adhesive was applied on the wood surface by roller and pressed under approximately 0.8 (N/mm^2) of pressure using hand clamps and wooden blocks to ensure an even distribution of pressure. Samples were left to dry under pressure for 16 h. Three types of samples were made: a sample with both adherent from non-modified wood as a control (P/P); a hybrid sample with TMW and nonmodified wood (P/T), and samples with TMW as an adherent on both sides (T/T). Fifty boards were prepared for a water soak test. Blocks were cut to have 50×50 cm shear area and with a 1 cm step on it based on ASTM D143. Samples with no visible defects were selected for the shear test. Also, from each type 10 microscopy blocks 10 mm x 10 mm on the surface and 20 mm long were randomly taken.

Microscopy

For microscopic observation, blocks were boiled in water for 5 min to soften the surfaces for microtoming. A GSL-1 sliding microtome, WSL Swiss Federal Institute for Forest, Snow, and Landscape Research, Birmensdorf, Switzerland, used to cut 30 µm thick sections. Three sections of each block were taken and stained for 2 min in a 0.8% aqueous solution of Safranin O stain. Excess stains were washed and blotted from the sections with distilled water. Three sections were mounted on a glass slide using glycerin as the mounting medium. Slides were examined with a Nikon Eclipse LV 100 light microscope equipped with a Nikon DS-Fi1 camera and the Nikon B GFP/D fluorescence filter cube set. NIS-Elements BR software was used for the measurements. Non-stained sections were also prepared to compare with stained sections to select the section with the best contrast to study bondline. Measurements were completed at ten locations on three separate bondline sections on the microscope slides. Adhesive line thickness and maximum penetration were measured with the Nikon software, then average penetration was calculated, and effective penetration calculated based on the authors' proposed method using Eq. 1., (Fig. 1).

$$Peff = A/L \qquad (1)$$

where $Peff$ is the effective penetration (µm), A is the measured area of adhesive that penetrated the wood (µm^2), and L is the length of measured area (µm).

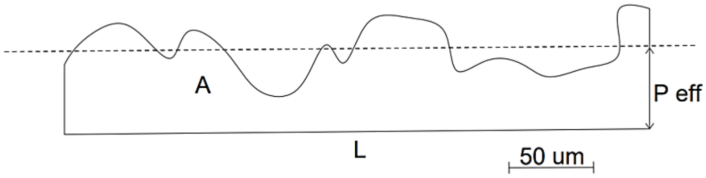

Fig. 1. Schematic of effective penetration in yellow poplar wood with PUR adhesive

Evaluation of Delamination

The HPVA three-cycle dry soak test was used to measure the effect of the wettability of the thermally modified samples with the adhesives. Sixty samples were tested with half being bonded with PUR and half with PVA. The 127 mm to 50.8 mm specimens from each test panel were submerged in water at 24 ± 3 °C for 4 h and then dried at a temperature of 49 to 52 °C for 19 h with sufficient air circulation. The cycle was repeated until all the samples failed, or until the three cycles had been completed, whichever occurred first. A specimen was considered as failing when a gage of 0.08 mm could be introduced between the adhesive gap between the two pieces.

Shear Strength Test

The procedures outlined in the ASTM D143 block shear test were used for measuring the shear strength. The samples tested had on average a shearing area of 50 mm× 50 mm. A machine testing speed of 0.6mm/min was used to apply load until the samples ultimately failed at the bond line or at the wood structure. A transparent laminated sheet with a 50 mm × 50 mm dimension was then used to analyze how much wood and adhesive failure was present.

Statistical Analysis

Statistical analysis was done using JMP Pro 16. A two-way ANOVA was used to identify if there was a significant interaction in microscopic properties and shear between the thermally modified wood and the nonmodified wood, as well as any differences due to the adhesives used. The null hypothesis was that there was not going to be a significant interaction between the thermally treated with non-treated wood and the adhesives. The alternative hypothesis was that at least one of the interactions was going to be significant.

RESULTS AND DISCUSSION

Anatomy Observation

Regardless of having a dark color in TMW, stained samples provided better images in light microscopy than non-stained samples. The contrast in fluorescent light was not sufficient, and it was obscuring some parts of the image and provided less quality than normal light. Therefore, the best images are obtained from stained slides with light microscopy (Fig. 2).

The results of the measured microscopic properties of bondline are presented in Table 1. The TMW samples made with PUR show higher adhesive line thickness than nonmodified wood (Fig. 3), comparing 2.83 µm to 0.85 µm. However, in samples made with TMW and PVA, there was no significant difference at the 0.05 level in adhesive line thickness than non-modified wood. PVA adhesive made a thicker adhesive line compared to PUR, which is in agreement with the research of Mamonova *et al*. (2022). Higher adhesive line thickness is due to less penetration of adhesive into the wood, which causes the accumulation of adhesive between two interfaces. This occurs when the wettability of wood is low, its moisture content is unsuitable for the adhesive type, the adhesive formula or dilution is not suitable with wood anatomy; or the press speed is lower than the increase in temperature in which the adhesive gets cured and gets crystalized before flowing into wood (Masoumi *et al*. 2023).

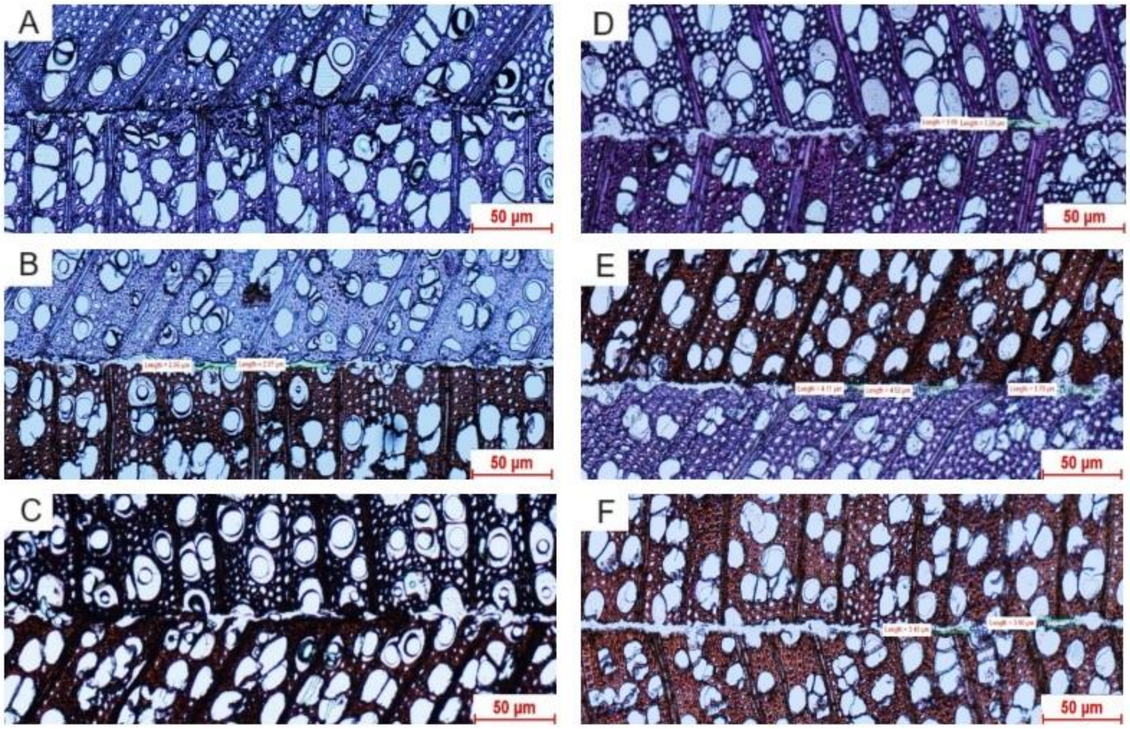

Fig. 2. Microscopic images of bondline samples made with PUR and PVA in thermally modified and nonmodified yellow poplar. A: Both sides non-modified YP with PUR; B: modified and non-modified YP with PUR; C: Both sides modified YP with PUR; D: Both sides non-modified YP with PVA; E: modified and non-modified YP with PVA; F: Both sides modified YP with PUR

Table 1. Microscopic Properties of Bondline

Adhesive Type	**PUR**				**PVA**			
Wood Type	**PP**	**(to P)**	**(to T)**	**TT**	**PP**	**(to P)**	**(to T)**	**TT**
Effective Penetration (μm)	90.8	51.6	38.9	84.2	49.3	55	42.8	31.3
Maximum Penetration (μm)	259.3	240.7	96.9	149	93.6	110.3	92.2	66.3
Average Penetration (μm)	120.2	92.4	33.1	70.2	53.1	42.4	40.3	31.7
Adhesive Line Thickness (μm)	0.85	2.94		2.83	4.2	4.58		4.18

In this table, PP represents a sample with both sides of non-modified poplar; PT is a hybrid sample of non-modified poplar; and TMW and TT is the sample made of both sides of TMW.

The adhesive penetration in TMW was less than in the control samples for both PUR and PVA for all three configurations, P/P; P/T; and T/T. The phenomenon of having lower permeability in TMW, which can be attributed mostly to the creation of a hydrophobic layer by extractives and low wettability in TMW (Chu *et al*. 2020; Hill *et al.* 2021), was well highlighted in hybrid samples, in which there was greater effective penetration than non-modified wood by 25% in PUR and 22% in PVA. Also, in the hybrid sample with PUR, TMW had a maximum penetration of 96.9 μm, which was significantly less than 240.7 μm in non-modified wood. The same difference was observed in the hybrid sample made with PVA, where effective and maximum penetration of 42.8 μm and 92.2

µm were different from 55 µm and 110.3 µm to nonmodified wood. PUR had better permeability than PVA in terms of both maximum, effective penetration and less adhesive line thickness and this result corresponds well with that of Mamanova *et al*. (2022). However, PVA in hybrid samples had greater penetration (9.1%) than hybrid with PUR, as shown in Fig. 4.

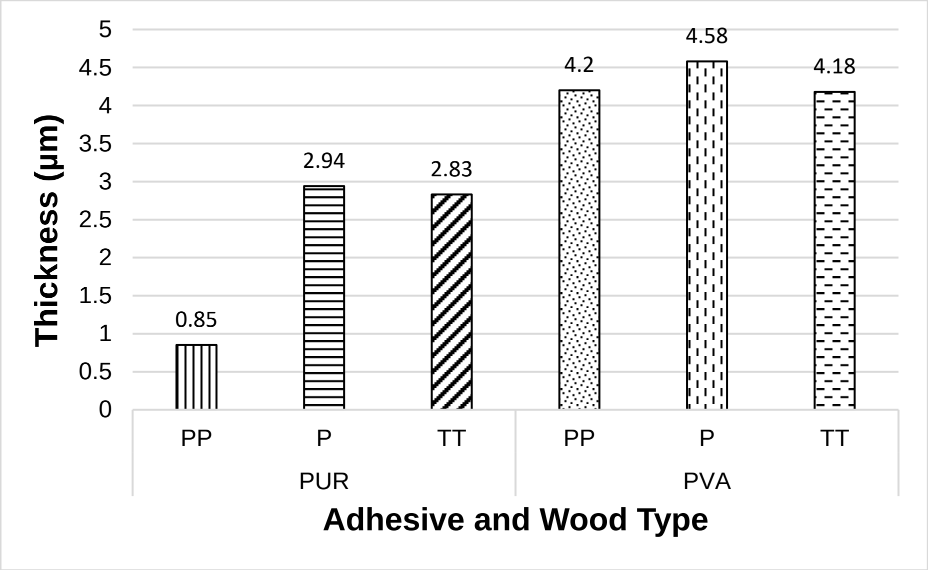

Fig. 3. Adhesive line thickness data. PP represents a sample with both sides of non-modified poplar; PT is a hybrid sample of non-modified poplar and TMW, and TT is the sample made of both sides of TMW.

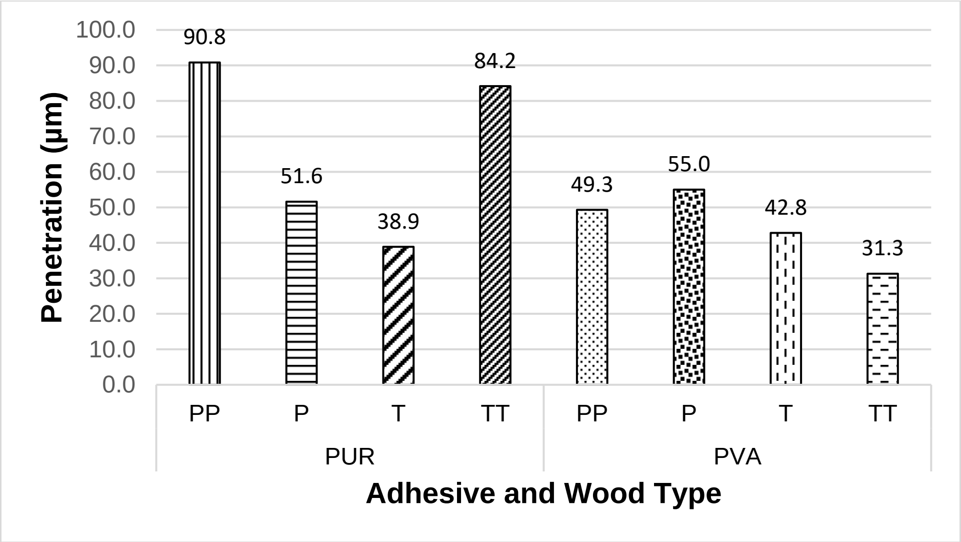

Fig. 4. Effective penetration data. PP represents a sample with both sides of non-modified poplar; PT is a hybrid sample of non-modified poplar and TMW, and TT is the sample made of both sides of TMW.

Delamination

The results of the delamination test are shown in Table 2. The resistance to water and moisture increased in bonding when comparing the TMW with the control group of nonmodified wood. This is attributed to the fact that TMW has higher dimensional stability (Hill *et al*. 2021; Birinci *et al*. 2022). The TMW does not shrink or swell as much as the nonmodified wood does, so the samples were less likely to fail in the bondline and its wood structure (Fig. 6). In hybrid samples made of nonmodified and TMW, the fracture was on the nonmodified side. The two adhesives had similar outcomes in terms of maintaining the adhesive bond when soaking and drying the samples in all three types of configurations. Interestingly, the hybrid samples had an intermediate amount of failure between samples made of both sides P/P or T/T.

Block Shear Performance

The average shear strength and percentage of wood failure for different wood and adhesives are presented in Table 2. The shearing stress statistical analysis concluded at 95% confidence that there was a significant difference between the configuration (P value=<0.0001) and the interaction it had with TMW with nonmodified wood and the adhesives used. The impact of TM on the shear was clearly evident when comparing TMW/TMW samples to the control/control samples where the TMW sample resulted in an average of 31.2% lower value for PUR and 29 % for PVA. The wood fracture mostly occurred at the wood interface due to the fact that samples become brittle, reducing their shear strength and hardness due to heat treatment (Can *et al*. 2021; Masoumi and Bond 2023), as demonstrated by the high percentage of wood failure. TMW is reported to have lower shear capacity by Yusoh *et al*. (2022) and Mamonova *et al*. (2022). Also thermally modified poplar wood showed a 20 percent decreased shear strength in research conducted by Chu *et al*. (2020). The shear strength of modified samples had reduction in values from 13.4 to 9.8 MPa in PUR and 13.0 to 8.4 MPa in PVA, respectively as compared to those of the control samples.

Table 2. Average Values for Shear Strength and Face Delamination

Adhesive Type	Sample Type	Shear Strength (MPa)	WFP (%)	Face Delamination (%)
Polyurethane (PUR)	P/P	13.398	92.50	70%
	P/T	11.144	95.00	80%
	T/T	9.789	98.5	100%
Poly Vinyl Acetate (PVA)	P/P	13.025	97.33	60%
	P/T	10.575	94.33	80%
	T/T	8.389	88.00	100%

There was a significant difference (P value=0.0312) when comparing the two adhesives to their wood failure percentage (WFP). The PVA on average decreased the amount of wood failure when thermally modified wood was added to the samples. The average of wood failure found on thermally modified samples was 91.17% in PVA, which was much lower than the average when using PUR, which was 96.8%. This means that the adhesive bond was more robust during the shear test when using PUR than when using the PVA. As presented in Fig. 5, there was a close correlation between adhesive penetration and shear strength in which with the increase in penetration the shear capacity improved. Also based on the results of Table 1 and Table 2, the increase in adhesive line thickness decreased shear strength.

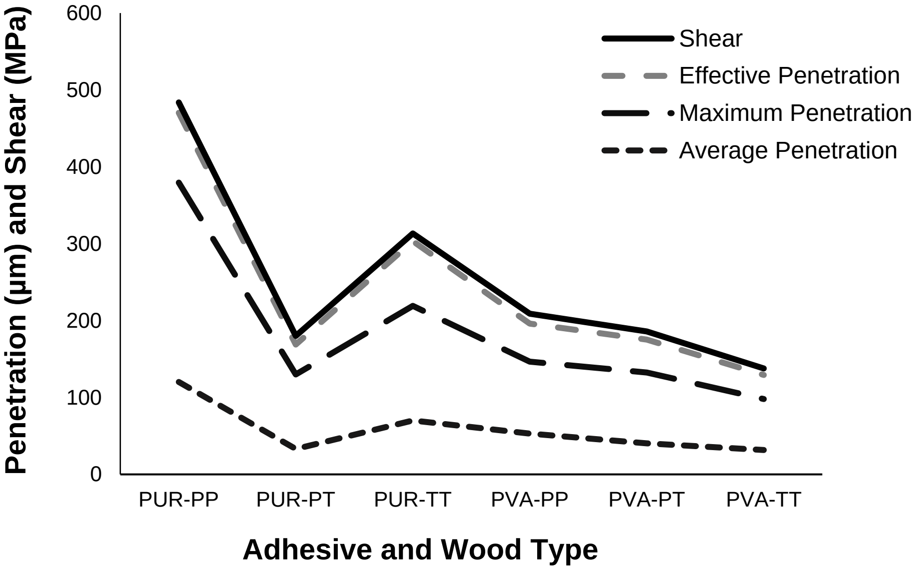

Fig. 5. Bondline property and shear strength

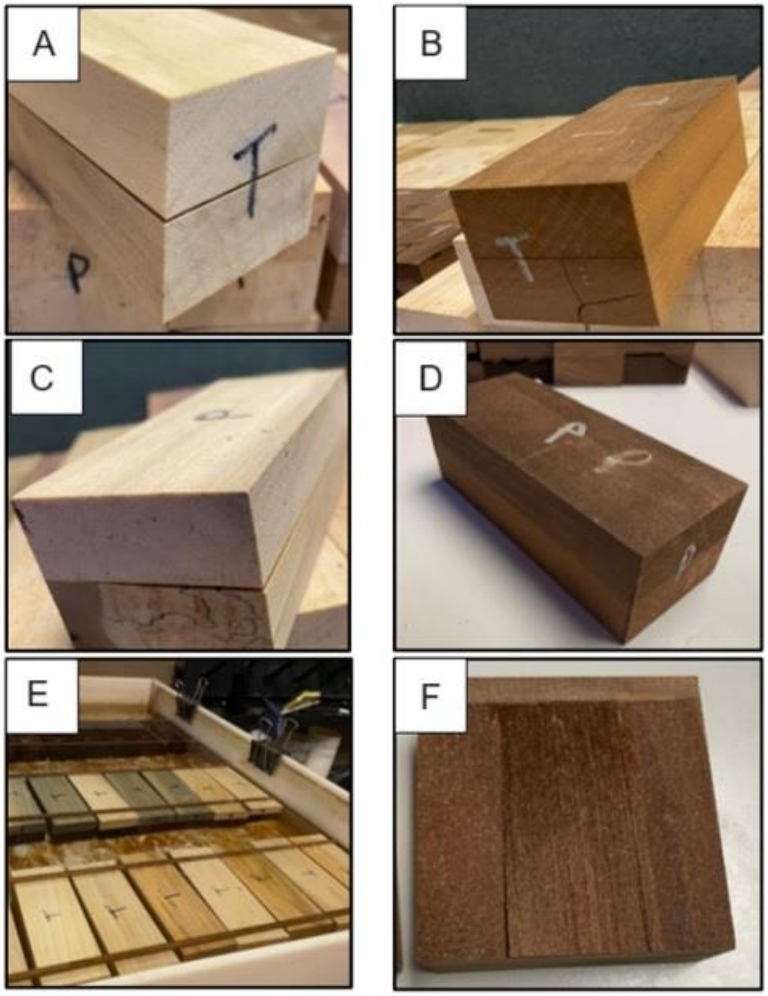

Fig. 6. A: Both sides non-modified YP with PVA; B: Both sides modified YP with PVA; C: Both sides non-modified YP with PUR; D: Both sides modified YP with PUR; E: Dry-soak procedure; F: Wood failure of sample made of both side modified YP in block shear test

TMW is being used in many different applications, and new applications are being developed where the ability to join pieces using common adhesives is desirable. The result of this study showed that both PUR and PVA are suitable for bonding TMW, as adhesive bondline played its role and transferred the shear stress to wood as an adherent in which the failure as WFP was observed. Also, the combined study of microscopic, shear, and delamination analysis revealed that the higher adhesive penetration leads to stronger bonding. Understanding the quality of adhesive bondline, particularly adhesive penetration in the quality control part of the wood products industry, could help to find an appropriate solution to low-quality products.

CONCLUSIONS

1. Microscopic observations of bondlines showed that the adhesive penetration into thermally modified wood including maximum and effective penetration decreased significantly compared to the control tests.

2. The thickness of the adhesive line increased for specimens of thermally modified wood particularly when using polyurethane (PUR).
3. Hybrid samples made of modified and nonmodified wood maintained an intermediate amount of bonding qualities.
4. In yellow poplar, as a case in this study, the shear strength of the specimens of PUR and polyvinylacetate (PVA) adhesives in thermally modified (TM) wood was lower than unmodified wood. However, the bondline in TMW remained intact and suffered the shear stress until failure occurred in wood.
5. Both PUR and PVA were shown to be suitable for bonding TMW, as demonstrated by the high wood failure observed.

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