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Development of preservative-treated cross-laminated timber and lignin-reinforced polyurethane-adhesive for glued laminated timber

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Development of preservative-treated cross-laminated timber and lignin-reinforced polyurethane-adhesive for glued laminated timber

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Interest in the use of mass timber in building and construction is growing worldwide, this is due to the structural integrity and reduced environmental footprint of timber-based structures. Concerns associated with the biological and environmental degradation of mass timber necessitate the development of adequate protection strategies to ensure the durability of these products. Preservative treatment is a proven technique that increases the durability and performance of wood in-service and can also be applied to large-sized timber panels such as cross-laminated timber (CLT). Therefore, this study focused on investigating the feasibility of treating prefabricated 3- and 5-layer CLT panels with Copper-azole type C (CA-C) and micronized copper azole (MCA) preservatives. Further, we studied the effects of panel layup and thickness on the preservative impregnation in CLT. Based on the experimental results, we found adequate preservative penetration and retention in the treated 3- and 5-layer CLT panels, particularly in CA-C treated panels. Also, the lengthwise layup shows better treatment results in both CA-C and MCA-treated panels. In addition to the preservative-treatment of CLT panels, this dissertation covers the development of lignin-reinforced polyurethane adhesive (PUR) for bonding glue-laminated timber (Glulam). Herein, the glulam were fabricated and bonded using lignin-reinforced PUR at different wt% (1, 2, and 3) and tested for shear strength, wood failure and delamination. The lignin-treated PUR samples showed improved adhesion properties via high shear strength and reduced delamination compared to the control specimens. Thus, the lignin-reinforced PUR adhesive shows great potential as a bio-based and environment-friendly wood adhesive for producing glulam used in structural applications.

Keywords: Mass timber, Preservative treatment, Cross-laminated timber, Glue-laminated timber, Polyurethane adhesive, Shear Strength, Delamination.

DEDICATION

I would like to dedicate my dissertation to God who makes all things possible. I would also like to dedicate this dissertation to the Ayanleye family, who has been a continuous source of support and encouragement throughout this journey.

"*With man this is impossible, but with God all things are possible*" (Matthew 19:26)

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INTRODUCTORY CHAPTER

1.1 Introduction

Wood is a valuable raw material for making structural lumber, furniture, mass timber (MT) and other wood-based composites. The attributes of wood such as easy machinability and high-strength properties makes it desirable in structural applications [1]. Also, the light-weight of wood make handling and transportation easier compared to conventional building materials like concrete and steel. However, certain limitations such as dimensional instability and small dimension imposed by wood require the development of high-performance and novel engineered wood products such as MT. MT is a generic term used for a class of wood products made from gluing small timber elements to form a larger panel with better dimensional stability and enhanced strength properties [2]. These primarily include cross-laminated timber (CLT), glue-laminated timber (Glulam) and laminated veneer lumber (LVL). The benefits of utilizing MT in building projects have been reported in the literature [3-5]. Although MT offers the opportunity of using wood-based materials in tall buildings and construction projects, their susceptibility to biodeterioration is often a concern [5]. Correspondingly, the issue with structural bonding of MT such as glulam which is important in the production and safe use of this product is an important research question to be addressed. Therefore, the advancement of research towards developing renewable and durable engineered wood products (MT in this case) is urgent for sustainable construction.

MT is a novel construction material that can be utilized in both residential and non-residential applications. Despite the numerous opportunities that exist for MT in building and construction, the concerns around the quality degradation of the panel, especially when subjected to outdoor environments limit the confidence in the use of these products. While several protection techniques such as preservative treatment (PT) have been implemented for dimensional lumber, their application to MT (such as CLT) is limited. PT is regarded as an effective way of improving the durability and in-service performance of wood and wood-based composites especially for use in structural applications [6-7]. Until recently, the incorporation of PT in CLT manufacturing has received minimal attention. This is partly due to the difficulty of successfully implementing preservative treatment technologies in MT production line. For instance, the treatment of large-sized panels with adequate preservative penetration and retention against biodegradation is a major challenge. This is because of the increased panel thickness and presence of adhesive layers that blocks the flow of preservatives during treatment. Also, cylindrical pressure treating retorts are generally not large enough to efficiently accommodate large panels. Moreover, the lack of treating protocols for engineered wood products such as CLT affects the integration of preservative treatment in industrial environments. Thus, developing an effective preservative treatment procedure for larger dimension panels is crucial to extend the current applications of CLT and expand its market in North America.

Glue-laminated timber (Glulam) is a popular MT often utilized in structural applications. Glulam can be used as beams and columns in roofs, floors and even bridges and has been widely used in Europe, Australia and the US [8-10]. However, the performance of glulam in load-bearing applications is dependent upon its structural bonding [11]. The bonding of wood elements is influenced by the manufacturing parameters (e.g. pressure, glue spread rate, open and close assembly time, temperature, et. al) and the adhesive system. Among the common adhesives used in the manufacturing of engineered wood products such as urea-formaldehyde (UF), phenol-resorcinol formaldehyde (PRF) and polyurethane adhesive (PUR), more attention has been given to PUR adhesives. Compared to the formaldehyde-based adhesives like UF and PRF, the advantages of PUR adhesives (such as high bonding strength and clear bond line) make them a suitable choice in preparing laminated wood products [12-13]. Even though PUR adhesives are mostly preferred in MT manufacture owing to their positive attributes, the drawbacks associated to their usage such as inadequate gap-filling properties and low resistance to delamination require urgent attention [14]. Hence, developing a novel adhesive system with lignin as a reinforcement agent could help solve these issues while reducing the environmental footprint of the adhesive. Moreover, there are several studies on the structural performance of PUR-bonded glulam (determined via shear strength and resistance to delamination) with limited information on those fabricated utilizing lignin-based PUR adhesives. Therefore, the outcome of these investigations will be beneficial to the wood adhesive and MT industry especially when designing high-durable and reliable MT products to be used in structural (and exterior) applications.

1.2 Research Objectives

The use of PT to protect wood from biodegradation has been widely investigated but not yet implemented on MT products such as CLT. The successful treatment of CLT with preservatives is important to expand its applications and increase the market worldwide. CLTs are mostly used in interior or protected conditions where they are less susceptible to biological attack or moisture exposure. However, the use of CLT in exposed or exterior applications will require adequate protection strategies such as preservative treatment to prevent biodeterioration of the panels. Thus, an experimental program was conducted to investigate PT of large-sized CLT panels. The research emphasizes the influence of preservative chemical formulations, panel layup and thickness on the performance of the treated CLT panels. Besides, a simple and effective evaluation method (using color-indicator) was developed to assess the impregnation quality of the panels. In addition, the issue regarding the appropriate sampling technique for evaluating the treatment performance of CLT was addressed.

Alongside the emphasis on the preservative treatment of CLT, this dissertation also investigates the effect of lignin-reinforced PUR adhesive on the bonding quality of mass timber such as Glulam. High-purity lignin was extracted from softwood lignin using fractionation technique. Although this is a preliminary study on the influence of lignin on the adhesion properties of PUR, the experiment was designed to simulate the ideal conditions in industrial environment where wood properties' (such as MC and SG) variation affect the bonding performance of glulam. The lumber used in fabricating the glulam samples were pre-selected to include only those within the same MC and density range.

The general objective of this project was to investigate the feasibility of successfully treating large-sized CLT panels with preservatives. We also examined the influence of lignin-addition on the bonding integrity of glulam bonded using PUR adhesive. This action was taken to contribute to the efforts in replacing fossil-based raw materials utilized in developing wood adhesives with environment-friendly alternatives (such as lignin). Therefore, we explored several specific objectives which include:

1. conduct a comprehensive review on the durability and protection of mass timber products.
2. establish post-treatment procedures for southern yellow pine CLT panels.

3. evaluate the penetration and retention of preservative-treated CLT panels using the color-indicator approach and x-ray fluorescence spectroscopy.

4. evaluate the effect of preservative type, panel layup and thickness on the impregnation quality of CLT panels.

5. develop lignin-reinforced PUR adhesive for gluing wood specimens.

1.3 Dissertation Outline

The dissertation begins with a general introduction to the 3 succinct research projects focusing on the preservative treatment of mass timber such as cross-laminated timber and the development of environment-friendly adhesive system for bonding glue-laminated timber.

Chapter 2 provides a comprehensive literature review on the durability and protection of mass timber structures, where the feasibility of protecting large-sized panels via preservative treatment and wood modification techniques (thermal, chemical modification…) is discussed. Chapter 2 also extends the discussion to include the effect of treatment techniques on the properties (such as wettability, dimensional stability, bending and bonding strength…) of mass timber.

Chapter 3 describes the penetration and retention behavior of preservatives in cross-laminated timber panels treated with copper-based preservatives. Then, the effects of panel layup and thickness on the results are presented and discussed.

In Chapter 4, the effect of incorporating renewable polyol sources (such as lignin) in polyurethane-based adhesive system is introduced. Afterwards, the block shear, wood failure percentage and delamination test results are presented and discussed. The influence of lignin addition (at different wt%) on the bond line shear and delamination results are discussed. Lastly, the FTIR analysis of the control and lignin-treated samples are presented where the interactions between the wood substrate, lignin and PUR adhesive are discussed.

Chapter 5 summarizes the main findings from this dissertation with some recommendations for further research in the development of environment-friendly wood adhesives and highly durable mass timber products.

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CHAPTER II

DURABILITY AND PROTECTION OF MASS TIMBER STRUCTURES: A REVIEW

Ayanleye, S., Udele, K., Nasir, V., Zhang, X., & Militz, H. (2022). Durability and protection of mass timber structures: A review. *Journal of Building Engineering*, 46, 103731. https://doi.org/10.1016/j.jobe.2021.103731. (Republished with permission).

2.1 Abstract

Mass timber (MT), a group of large engineered structural wooden panels such as cross-laminated timber (CLT), glue-laminated timber (Glulam), laminated veneer lumber (LVL), etc., is becoming increasingly popular due to sustainable construction. Despite the numerous benefits of MT-based buildings, such as low-carbon emission, short construction time, and cost-effectiveness, the concerns regarding the durability of MT may limit their market acceptance. In this review, we discuss the advantages and opportunities of applying MT in tall buildings, as well as the durability issues associated with MT application. We examine the traditional wood protection techniques including, preservative treatment, thermal and chemical modification, and discuss the potential of applying these techniques for MT protection. We survey the recent studies on MT durability evaluation, as well as the recent progress in MT structure protection through a moisture control strategy. Finally, we highlight the MT protection strategies through the preservative, thermal, and chemical treatment approaches, review the effects of these treatment methods on the properties of MT such as wettability, glue penetration, bonding strength, etc., and discuss the future of the field.

Keywords: Cross-laminated timber, Mass timber construction, Durability, Wood protection, Preservative treatment, Moisture control, Mass timber building monitoring, Wood modification

Abbreviations

ACC	Acid copper chromate
ACQ	Alkaline copper quaternary
CA	Copper-azole
CCA	Chromated copper arsenate
CCB	Copper chrome boron
CLT	Cross-laminated timber
DLT	Dowel laminated timber
EMC	Equilibrium moisture content
Glulam	Glue-laminated timber
LVL	Laminated veneer lumber
MC	Moisture content
MCA	Micronized copper-azole
MOE	Modulus of elasticity
MOR	Modulus of rupture
MPP	Mass plywood panel
MT	Mass timber
PUR	Polyurethane
SPF	Spruce-pine-fir
TM	Thermal modification

2.2 Introduction

Wood is a widely used structural material due to its desirable characteristics such as high strength to weight ratio, low energy consumption, and reliability in structural applications [1-3]. However, the hygroscopic and anisotropic nature of wood may restrict its use in structural applications, especially in tall buildings. Mass timber (MT) is a group of engineered wood products developed to combat the limitations imposed by small dimensions, dimensional instability, and variability of wood [4-6]. Cross-laminated timber (CLT), glue-laminated timber (Glulam), laminated veneer lumber (LVL), dowel laminated timber (DLT), and mass plywood panel (MPP) are typical MT products (Figure 2.1) [7-8]. The use of these products in multi-story buildings has seen a rapid increase due to the need for sustainable construction coupled with the changes in construction codes [9].

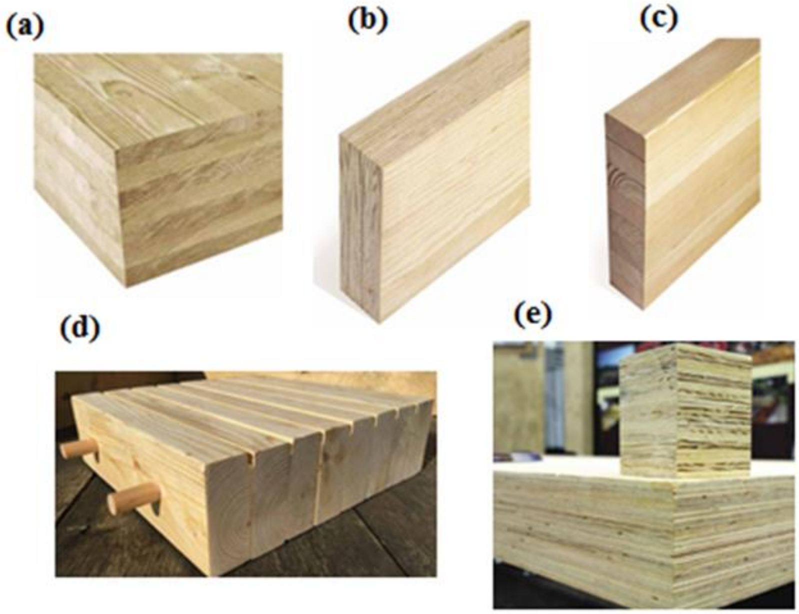


Figure 2.1 Common MT products used in constructions. (a) CLT, (b) LVL, (c) Glulam, (d) DLT, (e) MPP [13]

The benefits of MT products compared to conventional non-renewable building materials (concrete and steel) have been reported [10-12]. Some examples of MT structures include the 18-story tall wood building at the University of British Columbia (Vancouver, Canada) (Figure 2.2) and the 280-foot-tall MT tower in Brumunddal (Norway, Europe), which are constructed with CLT.

Figure 2.2 18-Story Tall wood Building (Hybrid Structure) built with CLT attached to glulam columns by steel connectors, located in Vancouver, Canada [6]

According to the International Mass Timber Report [13], the total annual lumber consumption by the North American MT industry is estimated to be 1.322 billion board feet. Considering the 60 billion board feet of softwood production in North America in 2020, the MT industry consumed 2.2% of North America's softwood lumber production. In fact, the MT manufacturing industry operates at a significantly lower rate than its production capacity [13]. To expand the use of MT in buildings, challenges such as the quality of MT, manufacturing standardization, and durability are critically important to address. Compared to other building materials like concrete and steel, wood's susceptibility to biodegradation, for instance, under weathering and climate conditions or biological attacks (decay fungi and insects such as termites) may limit its market acceptance. Archer and Lebow [14] reported that the annual loss attributed to biodegradation of wooden materials in the United States was approximately $5 billion. Considering the rapid growth of MT buildings in the past decade since the report was published, the current economic loss associated with wood biodegradation can be even higher; hence, MT protection from biodegradation is important [15].

Even though there is an increased interest in MT, limited information on its durability and realistic methods to avert risks undermines confidence in its applications. For instance, recent studies conducted on CLT have shown biodegradation risk upon exposure to outdoor conditions [16-18]. Similar reports on the biodegradation of Glulam can be found [19-20]. Thus, the application of MT products, especially in buildings located in high-risk biodegradation regions, necessitates adequate protection strategies [21-23]. While some articles have reviewed wood biodegradation and protection [2,24-27]; few works have focused on the impact of wood's properties on the durability of large-sized panels such as MT [12,22,28]. Additionally, no comprehensive review is found on MT protection, which is the focus of this paper. Therefore, this review addresses the need to protect MT elements, explores the application of current wood protection protocols to MT, and suggests possible means of implementation. Specifically, this paper starts with a discussion on the recent development of MT in buildings, including opportunities and challenges, while briefly introducing the importance of their protection in construction projects. Then, traditional wood protection protocols are reviewed, the possibility of implementing these strategies in MT protection is discussed. Additionally, recent advances on

MT durability and biodegradation are reviewed, and the MT protection protocols, as well as evaluation methods are also discussed. Finally, the impacts of protection on the properties of MT are reviewed and the knowledge gaps and challenges are outlined. The durability issues addressed in this paper are biological deterioration (decay fungi and insects such as termites, marine borers etc.), environmental degradation (moisture, degradation under weathering) with the exception of fire performance as it can be considered under the scope of safety engineering and not the natural biodegradation caused by environment or biological factors, which is the main focus of this study.

2.3 Need and development of mass timber structures

Timber is a valuable construction material due to its high strength-to-weight ratio, reduced energy consumption, design flexibility, renewability, aesthetic appearance, and reliability in load-bearing applications [3,29-30]. The use of timber in construction could be traced back to early human civilization [31-32] when structures such as the Neolithic long house were erected in 6000 BC. In comparison with conventional structural building materials like steel and concrete, the lightweight of wood makes handling, manufacturing, and transportation easier, thereby contributing to its cost-effectiveness [33-34]. Despite the above-mentioned advantages, the inherent variability of solid wood (due to anisotropy, cell arrangement), dimensional instability, and small dimension limit its utilization and performance in service [6,35]. Also, the competition of other structural materials (i.e., steel and concrete), as well as the public’s perception and government regulations, have limited the use of wood to low-rise, side material or ordinary accessories in buildings [36]. Therefore, the development of engineered wood products such as MT, with larger dimensions, improved strength, and dimensional stability is urgent for sustainable construction.

MT represents a class of framing styles made from pieces of dimensional lumber that are joined and glued together to form large members for wall, floor, and roof construction [10,37]. The representative MT products in the market include CLT, Glulam, and LVL. Compared to traditional wood products, MT offers more benefits such as greater strength and stiffness, increased dimensional stability, and uniformity in structure [6,11]. Moreover, utilizing MT products as alternatives to conventional non-renewable building materials can alleviate the increasing global environmental issues such as global warming and energy depletion. To illustrate, the energy consumption of timber-based buildings is about 15% less compared to conventional buildings [1]. This is because of the reduced energy required for wood production compared to the manufacturing of conventional building materials (i.e., concrete and steel).

To date, the market of building materials is still dominated by non-sustainable reinforced concretes [38], which greatly limits the development of green buildings. Therefore, there is a need for the rapid increase in the development of sustainable materials (such as MT products) for the green building movement [29,39].

2.3.1 Opportunities and challenges of applying MT in tall wood buildings

The interest of engineers, architects, and building professionals in sustainable construction coupled with the projected rise in urban population, as well as the changes in building codes that allow the use of wood in tall buildings, has created enormous opportunities for wide application of MT in building applications [18,40-43]. Figure 2.3 illustrates the benefits of MT in building construction. Constructions using MT are reportedly time-efficient and cost-effective compared to traditional building materials (concrete, steel, or stick framing) due to the reduction in in-situ construction time and on-site labor cost [11,44-45]. MT structures save an average of 20 - 25% in time and 4.2 % in capital cost, compared to conventional reinforced concrete constructions [10-11]. The increasing use of sustainable materials (wood in this case) in constructions has spurred the interest of architects in the use of MT products in urban development, modular housing, and public-use space design [16,46]. For instance, several buildings have been constructed using MT in North America, such as the Brock Common at the University of British Columbia (Figure 2.2, Vancouver, Canada), Promega Corporation reception area (Madison, WI, United States), Candlewood Suites (Huntsville, AL, United States), and 8-story Carbon-12 (Portland, OR, United States). A comprehensive list of the tall wood buildings in Europe, Australia, and North America can also be found elsewhere [12,39].

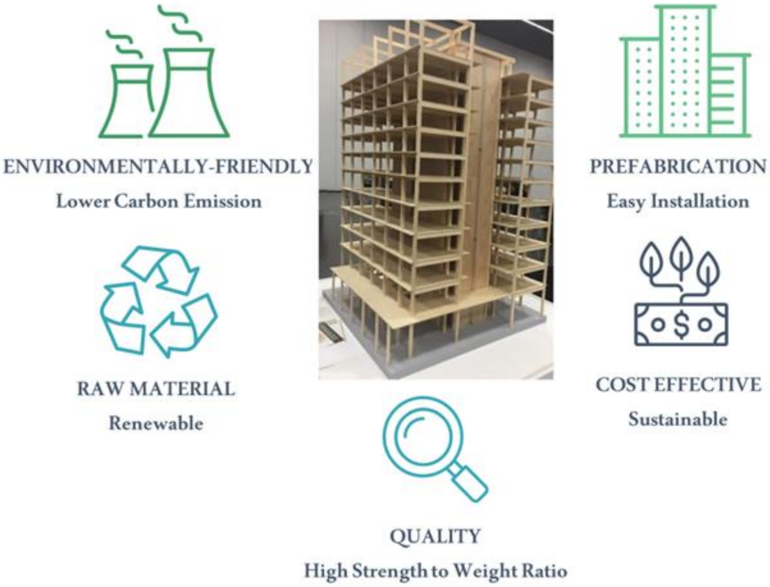


Figure 2.3 Benefits of using MT in buildings and constructions [the image shows a CLT design high-rise building structure in Portland, OR, U.S. [50].

Despite the opportunities of building with MT, several challenges have been documented by researchers worldwide [12,28,34). First, the service life of MT products is greatly impaired when exposed to high moisture conditions because of the fungi decay [37]. Previous studies also revealed the susceptibility of CLT to biodeterioration when subjected to termites. For instance, Singh and Page [47] reported a lower visual rating (6.0 - 7.0) for untreated CLT after fungi decay compared to a boron-treated group with a rating of 10.0. This is similar to other works with a visual rating of 7.5 for untreated CLT samples [48]. The aforementioned studies concluded that there was a moderate to severe decay (rating of 6.0 - 7.5) in the untreated groups and no evidence of decay (rating of 10.0) in the treated CLT. Therefore, adequate design and protection strategies of MT products are important in the acceptance and market growth of MT buildings [46]. Figure 2.4 summarizes some of the main challenges associated with the durability of MT in buildings and constructions.

Figure 2.4 Durability challenges of MT in buildings and constructions.

2.3.2 Need for protection of MT structures

Mass timber construction began in Europe in the 1990s with rapid acceptance in other parts of the globe, especially in North America. In Europe, an important factor has ensured that biological durability has not been a critical structural performance issue. Many of the MT structures are built in northern temperate regions where the risk of degradation is mainly limited to fungi rather than insects, such as termites. Further, building practices in Europe emphasize the exclusion of moisture during construction [28]. However, the recent market expansion and construction of MT structures in areas with high degradation potential coupled with emerging evidence of decay possibility in MT buildings in Northern Europe have made durability a critical issue [12,22,28,49].

As noted in the previous section, adequate protection strategies are needed to enhance MT structures' performance in service, especially in areas with high moisture exposure and degradation potential [23,51]. The sensitivity of MT connections to deterioration due to moisture ingress and cyclic loading has also been reported by the same authors. However, there is limited information on the durability of MT structures, and there are no established methods for minimizing the risks [9]. All structures, including those built with MT, are susceptible to decay, termite, and other insects' activities, particularly for structures in high-risk zones such as Brazil, Australia, and some parts of the U.S. [12,22]. Currently, the MT elements used in constructions are not treated to prevent biodeterioration. Although the current design standards incorporate the use of preservative-treated products for glulam and other wood composites [52], the treatment options for some MT products, particularly CLT, remain unsolved [9]. Therefore, understanding the conditions favorable to attacks of MT elements and identifying possible solutions will be essential for averting risks associated with the use of these products [9].

CLT is a prefabricated engineered wood panel that consists of crosswise stacked boards glued together using adhesive [53-54]. The advantages of CLT, such as homogenized physical and mechanical properties, large-size, and renewability, make it a preferred choice for tall buildings [39,55]. However, CLT is mostly fabricated from less-durable softwood lumber, which is susceptible to the infestation of fungi, insects, and termites, especially when used in warm and humid regions such as the Southern United States [16,56-57]. Several studies on CLT have focused on seismic and fire performance, structural properties, elastic and strength properties [58-60] with little attention on biodeterioration of CLT [12,17]. Some authors have investigated the effect of moisture exposure and consequently fungi and termite attack on CLT's properties. For instance, Stokes et al. [16] have revealed that CLT exposed to subterranean termites was attacked with tunneling along glue lines. Likewise, Wang [61] observed an increase in moisture content (MC) of spruce-pine-fir (SPF) CLT subjected to artificial and natural weathering for 18 days and 60 days, respectively. The author reported an average MC of 24%, which could be favorable for decay fungi and termite attacks. Therefore, the moisture exclusion of exposed CLT elements is highly recommended both in design and practice [62]. To date, the moisture exclusion and monitoring of CLT during construction and usage remain challenging. Thus, the development of preservative-treated CLT with enhanced durability and resistance to high humidity environments is important [63].

2.4 Wood Protection

2.4.1 Natural durability of wood

The high hydroxyl (-OH) content of wood main constituents, i.e., cellulose and hemicellulose, endow its hygroscopic and hydrophilic nature. When exposed to changing moisture/humidity conditions in service, wood and wood-based products are susceptible to deformation and, at higher MC, to biodeterioration in response to moisture absorption and desorption, resulting in quality degradation [64]. The degradation of wood and wood-based products results in undesirable financial implications. These include material and labor costs and in extreme cases, cost arising from litigation as a result of a failed structure (that is, approximately $5 billion annually for the U.S. only). The natural durability of wood is closely related to its extractive type and content, which varies with wood species. The wood extractives, mainly aromatic-based compounds including tannin, flavonoids, quinones, and stilbenes [27,65], act as a natural pesticide that protects living trees against insects' and/or microorganisms' attacks [66-67]. Generally, the heartwood containing higher extractive content increases wood's resistance against biological degradation [68-69]. The natural durability of heartwood from different wood species has been investigated by researchers and scientists worldwide [65,70]. For instance, Kirker et al. [65] investigated the natural durability of several North American softwoods like southern yellow pine, western red cedar, paulownia wood, and black locust against decay fungi (brown- and white-rot). The authors attributed the decay resistivity to the presence of extractives in these species. Likewise, some studies reported the natural decay resistance of hardwood (Tectona grandis- teak) from Togo, India, and Mexico [71-72]. Despite some wood species exhibiting high natural durability, the less availability of naturally durable wood species coupled with leachability concerns of wood extractives have limited the application of unprotected wood in structural applications [73-74]. On the other hand, the durability and service life of wood products could be improved through several treatment methods [75-76]. Wood treatment is a term used to describe any method employed in improving wood's properties including, dimensional stability, resistance to biodeterioration, and weathering [3]. The typical wood treatment techniques include preservative treatment (PT), thermal modification (TM), and chemical modification (CM) [3,24,27,68,77].

2.4.2 Preservative treatment

PT is a method that involves impregnating wood with chemicals that act as a biocide against deteriorating agents for enhancing wood's resistance to decay fungi and termite attack and, in turn, extending its service life [78]. Several methods, including brushing, spraying, dip-treatment, and pressure treatment, have been tested in applying preservatives to wood products [12]. The main goal of these methods is to ensure uniform distribution and adequate retention of preservatives, the factors that are responsible for the increased durability of the products [79-80]. The first generation of wood preservatives such as creosote, pentachlorophenol, and chromated copper arsenate (CCA) were inexpensive and effective against decay fungi and termites and mostly preferred by the wood protection industry for many years [68]. Similarly, some wood preservatives containing copper and chromium compounds have been found to increase weathering resistance in wood products [81-83]. However, the research efforts in the past decades have been geared towards developing more environmentally benign wood preservatives [84]. This results from the tolerance of some fungi to copper-based preservatives and the ban of the first-generation wood preservatives due to environmental concerns [85-86]. Some of the newer formulations include alkaline copper quaternary (ACQ), copper-azole (CA), and micronized copper-azole (MCA) preservatives which have been marketed for residential applications [84,87]. Previous studies revealed the effectiveness of these formulations in protecting wood from biological degradation (fungi, termites, etc.) and environmental degradation (weathering) [88-89]. Tables 2.1 and 2.2 detail some of the common wood preservatives used in North America with their composition and standards for evaluating the treated wood products.

Table 2.1 Common wood preservatives and their chemical composition in North America

Preservative	**Constituent Elements**	**Composition/Molecular Formula**	**Ref**
Creosote	polycyclic aromatic hydrocarbons (PAHs), phenols, and creosols	N/A	[90]
Pentachlorophenol	Carbon, Hydrogen, Oxygen, Chlorine	C_6HCl_5O	[90-91]
Propiconazole	Carbon, Hydrogen, Oxygen, Chlorine, Nitrogen	$C_{15}H_{17}Cl_2N_3O_2$	[91-92]
Triadimefon	Carbon, Hydrogen, Oxygen, Chlorine, Nitrogen	$C_{14}H_{16}ClN_3O_2$	[91-92]
Acid copper chromate (ACC)	Copper, Hexavalent Chromium	31.8 percent copper oxide, 68.2 percent chromium trioxide	[90-91]
Isothiazolinones	Carbon, Hydrogen, Oxygen, Sulphur, Nitrogen	C_3H_3NOS	[91-92]
ACQ	copper, didecyl dimethyl ammonium carbonate, 2-methyl-4-isothiazolin-3-one, 5-chloro-2- methyl-4-isothiazolin-3-one	**Type A**: copper-ethanolamine (50% CuO equivalent), and DDA chloride (50%) **Type B**: copper-ammonia (66.7% CuO) and DDA chloride (33.3%) **Type C**: copper-ammonia and/or copper-ethanolamne (66.7% CuO) and ADBA chloride (30%) **Type D**: copper-ethanolamine (66.7% CuO) and DDA chloride or carbonate (33.3%)	[90-91]
Borates	Boron, Oxygen	N/A	[90-91]
copper azole	Copper, Boric acid, Tebuconazole	49% copper, 49% boric acid, 2% tebuconazole	[90-91]
Copper naphthenate	Copper, Naphthenic acid	$C_{22}H_{14}CuO_4$	[90-91]

Table 2.1 (Continued)

Preservative	**Constituent Elements**	**Composition/Molecular Formula**	**Ref**
Ammoniacal copper zinc arsenate	Copper, Zinc, Arsenic, Oxygen.	50% copper oxide, 25% zinc oxide, 25% arsenic oxide	[90-91]
CCA	Chromium, Copper, Aresenic, Oxygen	**Type A** (65.5% CrO_3, 18.1% CuO, 16.4% As_2O_5) **Type B** (35.3% CrO_3, 19.6% CuO, 45.1% As_2O_5) **Type C** (47.5% CrO_3, 18.5% CuO, 34.0% As_2O_5)	[90-91]

Table 2.2 Standards in North America for the evaluation of treated wood products

Standard Number	Standard Name
A6-20	Standard Method for the Determination of Retention of Oil-Type Preservatives from Small Samples
A9-21	Standard Method for Analysis of Treated Wood and Treating Solutions by X-Ray Spectroscopy
A12-19	Wood Densities for Preservative Retention Calculations
A16-16	Standard Method for Determination of Didecyldimethyl Ammonium Compounds in Treated Wood by HPLC
A19-19	Standard Method for Sample Preparation for Determining Penetration of Preservatives in Wood
A40-21	Standard Methods for Determination of Boron Trioxide in Treating Solutions and Treated Wood by Potentiometric Titration with Sodium Hydroxide
A48-15 (Reaffirmed 2021)	Standard Method for Analysis of Propiconazole, Tebuconazole and Imidacloprid in Solutions and Treated Wood Products by High Performance Liquid Chromatography
A51-19	Standard Method for Determining Penetration of Protectant in Acetylated Wood
A65-21	Standard Method to Determine the Amount of Boron in Treated Wood Using Azomethine-H or Carminic Acid
A75-18	Standard Method to Determine the Penetration of Pentachlorophenol in Poles by X-Ray Fluorescence
A77-18	Beilstein Test for Determining Presence or Penetration of Pentachlorophenol (PCP) in Wood
E5-21	Standard Field Test for Evaluation of Wood Preservatives to be Used in Marine Applications (UC5A, UC5B, UC5C); Panel and Block Tests.
E7-21	Standard Field Test for Evaluation of Wood Preservatives to be Used in Ground Contact (UC4A, UC4B, UC4C); Stake Test
E8-21	Standard Field Test for Evaluation of Wood Preservatives to be Used in Ground Contact (UC4A, UC4B, UC4C); Post Test
E9-21	Standard Field Test for Evaluation of Wood Preservatives to be Used Above Ground (UC3A and UC3B); L-Joint Test
E13-21	Standard Method to Determine if Lumber has been Pressure Treated with a Water Repellent
E15-17	Laboratory Method for Evaluating the Efficacy of Diffusible or Volatile Remedial Preservatives Against Pure Basidiomycete Cultures: Inoculated Block Test
E16-16	Standard Field Test for Evaluation of Wood Preservatives to be Used Above Ground (UC3B); Horizontal Lap-Joint Test
E18-18	Standard Field Test for Evaluation of Wood Preservatives to be Used Above Ground (UC3B); Ground Proximity Decay Test

Table 2.2 (continued)

Standard Number	Standard Name
E20-21	Standard Method for Determining the Depletion of Wood Preservatives in Soil Contact
E21-18	Standard Field Test for Evaluation of Wood Preservatives to be Used for Interior Applications (UC1 and UC2); Full-Size Commodity Termite Test
E25-21	Standard Field Test for Evaluation of Wood Preservatives to be Used Above Ground (UC3B); Decking Test
E26-21	Standard Field Test for Evaluation of Wood Preservatives to be Used for Interior Applications (UC1 and UC2); Ground Proximity Termite Test
E27-15 (Reaffirmed 2021)	Standard Field Test for Evaluation of Wood Preservatives to be Used Above Ground (UC3B); Accelerated Horizontal Lap Joint Test
E31-18	Standard Field Test for Evaluation of FieldCut Preservatives to be Used in Ground Contact (UC4): Block Test
E32-18	Standard Field Test for Evaluation of FieldCut Preservatives to be Used Above Ground (UC3B): Modified Post and Rail Test
E33-18	Standard Test Method of Evaluating Wood Preservatives Against Decay in Use Category UC2
E34-21	Standard Field Test for Evaluation of Wood Preservatives to be Used Out of Ground Contact: Horizontal Sandwich Method
E35-21	Standard Field Test for Evaluation of the Depletion of Wood Preservatives from Wood Used Above Ground; (US3A and UC3B)

2.4.3 Wood modification

While wood preservatives have been found effective in protecting wood from biodeterioration, the concerns surrounding its impact on aquatic organisms and human health have restricted its use in some applications [93]. Thus, the need to protect the environment and enhance the durability of wood products led to the exploration of other technologies such as wood modification (thermal and/or chemical modifications) [26,94-98]. These techniques help in reducing wood's affinity for moisture, which is the main factor responsible for wood biodegradation, and block the cell walls by penetrating the nanopores in the cell wall [99]. For instance, several TM methods have been developed, involving the exposure of wood to high temperatures (160-230 ^{o}C) under reduced oxygen conditions, leading to permanent changes in its physical and chemical structure (Figure 2.5), thereby improving the decay resistance of wood, which has found widespread application and commercialized in Europe [24,100-102]. Different thermal treatment technologies have been developed, differing in their pressure regimes (Vacuum, non-pressurized, pressurized) or shield gasses.

The influence of thermal treatment on weathering characteristics of wood has been reported. According to the literature [103-105], thermally modified wood showed resistance to degradation caused by weathering in outdoor conditions. Esteves and Pereira [24] reported that thermal treatment above the 200 °C resulted in a hydrophobic wood surface, which reduced the water absorption and therefore enhanced the weathering resistance of wood. Nuopponen et al. [103] discovered that 225 °C high-pressure steam treatment decomposed wood hemicellulose and altered lignin's structure, thus alleviating the UV-light induced free-radical degradation of lignin. The decreased hemicellulose content in the treated wood reduced its equilibrium moisture content, which further enhanced the wood durability by diminishing the leaching of aromatic compounds.

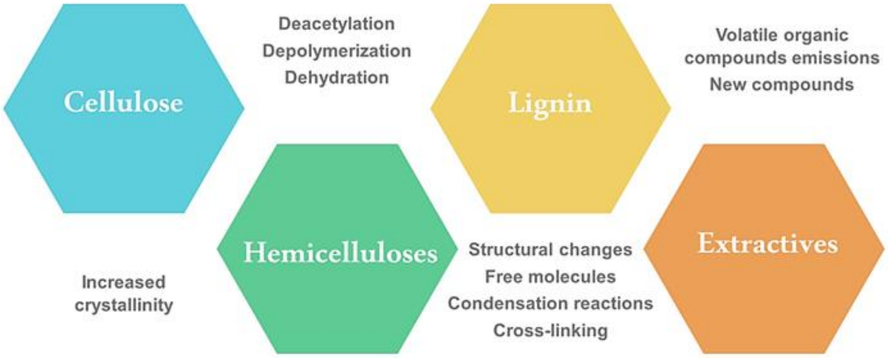


Figure 2.5 Changes in wood’s constituents as a result of TM [24].

However, the reduction of certain strength properties (mainly dynamic properties, impact bending), reduction in the stiffness and modulus of rupture depending on the treatment process [24], and the susceptibility of thermally modified wood to some bio-deteriorating agents such as marine borers and termites limit its application, especially in outdoor and ground-contact applications [106-108]. Alternatively, CM techniques (processes of replacing the hydroxyl group of wood cell wall polymers, responsible for moisture sorption, with other chemical reagents) to improve its decay resistivity and dimensional stability, have been reported in the literature [3]. The effect of wood modification on the wood microstructure is shown in Figure 2.6.

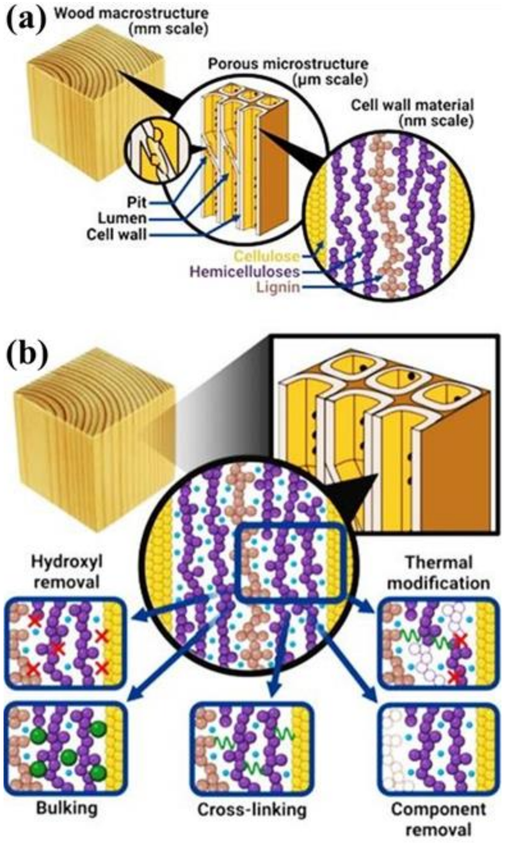


Figure 2.6 (a) Wood macrostructure, porous microstructure, and cell-wall molecular structure. (b) Impacts of wood modification on structures and orientations of main cell wall components, i.e., the cellulose, hemicelluloses, and lignin. CM can cause the removal of hydroxyl groups, the bulking effect to cell walls, the cross-linking among adjacent wood components, and/or specific cell wall component removal. Wood TM results in wood cell wall component removal (mainly hemicelluloses), hydroxyl removal, and cross-linking among cell wall components [109].

Prominent CM systems, including acetylation, furfurylation, and several resin modifications (e.g., DMDHEU, phenol, melamine) were found effective in protecting wood from degradation and were scaled up industrially. For instance, acetylation of wood was found to increase the resistance of wood to biological (i.e., termites and other micro-organisms) and environmental (i.e., weathering) degradation because it greatly decreased the hygroscopicity of wood [110-114] without reducing strength properties. Similarly, furfurylated wood was reported to eliminate wood hydroxyl groups and induced the cross-linking among cell wall components (i.e., cellulose, hemicellulose, and lignin), thus enhancing wood durability [115-116]. A comprehensive report on acetylation and furfurylation can be found in the two most recently published articles [26,117]. Wood modification encourages the use of non-biocide treatment in protecting wood from attacks without harmful effects on the environment [118-119].

While the abovementioned approaches have been proven in improving wood's durability, there are pros and cons of each method. For instance, concerns around the impact of preservative chemicals on aquatic and human health have restricted its use in several applications. Thermally modified wood is effective in protecting wood against biological attack except for termites and marine borers, making it unsuitable for ground contact or exterior applications [108]. Besides, TM may reduce the mechanical properties of wood and cause checking in wood [120-123]. The search for environmentally benign wood protection techniques led to the development of chemical modification; however, it is not cost-effective compared to traditional wood protection strategies such as PT. Table 2.3 summarizes the pros and cons of the different wood protection methods reviewed in this paper.

Table 2.3 Standard methods for wood protection (pros and cons)

Treatment	Methods	Pros	Cons	Ref
PT	Pressure treatment (full cell, modified full cell, empty cell)	• Affordability • Versatility • Durability • High penetration and retention of chemicals • Moisture, fire, and insect resistance • Good strength	• Splinters and checks in treated wood • Color fading • Can not be applied to wood in service • Chemical risks • time required for wood to dry	[90,124]
PT	Non-Pressure treatment (brushing, spraying, pouring and dipping, cold-soaking, steeping, hot and cold bath, diffusion, vacuum process, preservative pads)	• Can be applied on wood in service • Cheaper than pressure treatment • Resistant to insects and fungi • Ease of application • Minimal chemical risk	• Not suitable for timber in ground contact • Several coats of preservatives needed for efficient and maximum protection • less penetration • Time-consuming	• [90,124)
TM		• Chemical-free • Dimensionally stable • Resistance to rot and insects • Better insulating properties • Lighter material • Increased sustainability	• Increased cost • significant strength reduction at higher treatment intensities • does not provide sufficient durability for timber in ground contact • increased embodied carbon due to the pressure and heating process	• [125-128]

Table 2.3 (Continued)

Treatment	Methods	Pros	Cons	Ref
CM	Acetylation, furfurylation	• Increases durability to the highest durability class (class 1) • Shrinkage and swelling are reduced by 70-75% when compared to untreated wood • Resistant to termites and borers • Increase hardness by 15 -30% • No negative impact on strength properties of wood	• Increased cost • Could lead to changes in color depending on the method of modification used	• [117,119,129-130]

2.4.4 Successes with wood treatment

Wood treatability, defined as the ease of liquid penetration into wood's structure, is often affected by the following factors [80,131]:

a- wood physical and chemical properties (porosity, moisture content, proportion of sapwood and heartwood, extractive contents)

b- wood anatomy properties (fibers, vessels, rays, pits, longitudinal tracheids, etc.)

Also, wood quality control processes such as drying could impact treatability via cell wall collapse and pit aspiration [132-133]. The effect of wood's anisotropy (longitudinal, radial, and tangential direction) on the treatability of wood has also been documented. The penetration of liquid, including preservative chemicals, is highest in the longitudinal direction and lowest in the tangential direction [134]. The high conduction of fluids in the longitudinal direction of wood is attributed to the presence of pits joining the fibers together, thereby enhancing the flow of fluids [135]. However, past researchers have found better treatability in the radial direction of certain softwood, which could be due to radial flow caused by the presence of resin canals and wood rays [136].

On the other hand, the treatability of wood-based composites, for instance, CLT and glulam are not only influenced by wood properties but also affected by adhesive layer because it blocks the penetration of preservative chemicals between the adjacent panel layers [137]. Additionally, the lamination increases the panel thickness, thereby the treatability of laminated products [138]. Table 2.4 lists examples of preservative-treated solid wood and wood-based composites (including MT).

Table 2.4 Examples of preservative-treated wood and wood-based composites

Product	**Wood Species**	**Treatment Process**	**Findings**	**Remarks**	**Ref.**
Solid wood	Douglas-fir (*Pseudotsuga menziesii* Franco)	Dip-treatment	Prolonged dipping time and incision influenced the penetration and retention of preservatives.	Incising techniques could be a way to enhance the treatability of wood.	[79]
Solid wood	Southern yellow pine (*Pinus* spp.)	Pressure-treatment	The effect of wood's microstructure on preservative penetration was observed.	Preservatives containing nanoparticles were successfully used in wood treatment.	[139]
Solid wood	Scots Pine (*Pinus Sylvestris* L.) and Norway Spruce (*Picea abies* L.)	Pressure-treatment	Correlations were found between the penetration of Micronized copper azole (MCA) and wood species and the presence of sapwood.	The difficulty of MCA penetration into pine species due to their refractory (difficult to treat) nature was documented.	[131]

Table 2.4 (Continued)

Product	**Wood Species**	**Treatment Process**	**Findings**	**Remarks**	**Ref.**
Solid wood	Scots pine, Turkish fir (*Abies bornmülleriana Mattf)* and Black pine (*Pinus nigra*)	Vacuum-pressure method	Wood samples were treated with copper-azole preservatives. The samples showed resistance to marine borer attack.	Of the species treated, Black pine and fir samples were moderately attacked which could be linked to leaching of the preservative-treated samples	[74]
Modified wood	European aspen (*Populus tremula*) and Birch (*Betula pubescens* Ehrh.)	Dip-treatment	Thermally modified wood was successfully impregnated with preservatives.	Wood's microstructure and permeability influenced the penetration of preservatives.	[75]
CLT	Southern yellow pine	Pressure-treatment	PT influenced the bonding performance of CLT.	The use of pre-treated lumber in CLT fabrication was found effective.	[137]
CLT	Batai (*Paraserianthes falcataria*), sesenduk (*Endospermum malaccensis*), rubberwood (*Hevea brasiliensis*), and kedondong (*Canarium* sp.)	Vacuum pressure method	PT has no significant influence on the bonding performance of CLT.	Treatment of hardwood CLT is possible, but the effect of species, density, and anatomical features should be taken into consideration.	[140]
Glulam	Maritime pine (*Pinus pinaster* Ait)	Pressure-treatment	Preservative retention harms the glulam's bonding performance.	Minimal target retention level of preservatives is vital in producing treated glulam.	[141]
Glulam	Beech (*Fagus* spp.), Hard maple (*Acer* spp.), and Red oak (*Quercus rubra*)	Pressure-treatment	PT decreased the mechanical properties of glulam.	While the mechanical properties decreased, no effect was observed on the adhesive strength of glulam.	[142]

2.5 Development of protection methods for MT structures

2.5.1 Durability evaluation methods for MT structures

Timber remains one of the most used construction materials, and the emergence of MT has contributed to an increased appreciation of the material in the construction industry. Although MT allows the use of timber for tall wood buildings [143], the utilization of MT in the temperate zone with a high risk of biological degradation is often a concern. Even though PT is a potent way of combating this issue, the interference with the bond line, possible chemical toxicity, and potential for increased material costs have impeded its implementation in MT products [140,144]. Currently, the majority of MT products are manufactured without treatment due to a lack of standard procedures and evaluation methods. It is therefore imperative to develop effective methods for MT treatment and evaluation.

While several methods have been established for structural lumber, they are deemed inadequate for MT due to the sizes of the material, the presence of bond lines, and difficulty in creating in-service conditions, which vary considerably between timber and MT applications [12,22,28]. Additionally, because MT elements are typically used in a structural system, it is difficult to evaluate a single element without considering other parts of the structure [12,18]. These issues have been approached differently by several researchers, and although there are no established full-scale tests to evaluate the durability of MT structures, several studies have monitored MT buildings for moisture and decay issues. For instance, Austigard and Mattsson [49] monitored a building for mycelium growth and the amount of time it takes to clean up and repair the building. Similarly, moisture sensors were installed in MT in many studies to monitor moisture changes during and after the construction of the buildings [145-146].

A major challenge in the laboratory evaluation of MT durability is the need to use sample sizes whose dimensions are representative of the actual MT products. For this purpose, Cappellazzi et al. [28] suggested the use of an area of at least 0.09 m^2 to get the best result for a 3 ply CLT. Accordingly, a 300 mm × 300 mm three-ply Douglas-fir CLT was tested in the Biodeterioration Lab at the Oregon State University to investigate the impact of wetting and fungal attack [28]. This approach has been adopted by other researchers in the evaluation of durability in MT [18,147]. Similarly, moisture regimes such as leaching, spraying, and full saturation have been used by researchers to imitate real-life conditions to which MT elements might be subjected in-service [147-149]. Furthermore, studies on durability in MT connections have been conducted to adequately describe the effects of moisture and decay on the performance of building systems [18,147]. It was deduced from the aforementioned studies that the evaluation of durability in MT is not a straightforward task, and a proper attempt at it must meet all requirements, including;

a. creating conditions that are not too harsh and are likely to be experienced in real structures
b. using materials with appropriate dimensions
c. creating climatic conditions favorable for biological growth and a complete evaluation of structural systems.

2.5.2 Protection methods for MT structures

MT structures have seen rapid growth in the construction industry over the last decade. The benefits of constructing buildings with MT have been covered earlier in this paper (Section 2.2). Even though MT is a viable ecological and economic alternative to conventional building materials such as steel and concrete, exposure to certain environmental and climatic conditions leaves them prone to degradation militating against their performance in service. Until now, the literature on wood protection mainly focused on solid wood and some wood-based materials with little information on the novel and new building materials such as MT. Here, we provide recommendations regarding the protection of MT, including literature findings on design approaches to combat moisture exposure issues, PT, and other modification techniques that could be applied to improve MT's moisture and weathering resistance, as well as decay resistivity.

2.5.2.1 Moisture control

Moisture remains the principal cause of durability issues in MT, and the elimination or control of moisture can help MT-based buildings remain durable throughout the expected service life [8,150-151]. There are various methods to improve the structural health and enhance the durability of MT's structure, but the avoidance or removal of moisture is the key in all of these approaches. Designing structures so that elements are sheltered from the prevailing wind, the use of roof overhangs to shield elements from moisture, provision of roof drainage systems, and proper architectural detailing in buildings are all valid methods to reduce moisture intrusion in MT structures [152]. Vapor barriers and insulations can also reduce wetting in panels, but they must be used with caution because they slow down drying, which can initiate decay if moisture is trapped within a panel [153]. A more effective way to get rid of moisture would be the use of drained and cross cavities [154]. These systems create pathways for water to leave the MT assembly before they are absorbed by the panels (Figure 2.7). Locations where MT elements are in contact with water sources such as concrete or soil also pose high risks of moisture intrusion. In a study by Zelinka et al. [155], CLT panels on concrete footings were found to have the highest MC during construction. Even though the author stated that these panels later dried up to satisfactory moisture levels, it is important to ensure that MT structures are designed to avoid contact between panels and moisture sources [8].

The proper selection of wood species based on a thorough understanding of the geography of the construction site is also important. To illustrate, Gupta et al. [150] highlighted four insect hazard zones that could be identified in North America; this sort of information is useful in determining if naturally durable or preservative-treated wood would be preferred in MT structures. Although there are chances of moisture intrusion in MT buildings during end-use arising from spills or leaks, prompt cleaning with a slow drying schedule should be employed to bring moisture levels to an acceptable range as well as prevent checks due to differential changes in the individual lamellas. Additionally, moisture sensors should be installed in hidden members to monitor moisture changes throughout the structure's life span. However, many moisture problems in MT buildings are associated with exposure during construction because although the outer portion of the elements might appear dry after wetting, the inner portions may remain at elevated moisture levels. Hence, construction methods, as well as existing climatic conditions in construction sites are important factors to consider. Cappellazzi et al. [28] stated that although expensive and cumbersome, temporary tents would prevent overhead wetting and limit exposure of the materials during construction. Likewise, Schmidt and Riggio [146] suggested constructing MT structures during the dry seasons to prevent wetting, although this could limit design and construction flexibility. The use of water-resistant barriers and sealants either from the factory or on the construction site is also effective for reducing moisture intake by panels [148,154]. It is difficult to completely eliminate moisture intrusion in MT structures either during or after the construction. However, the presence of moisture can be mitigated through reduction of exposure during shipping, proper storage, and protection on-site either with tarps or tents as well as adequate monitoring of panels during end-use either manually or with the help of sensors.

Timely design, construction, and building enclosure should also be encouraged when building MT structures. Prefabrication and modular construction could prove effective in this regard.

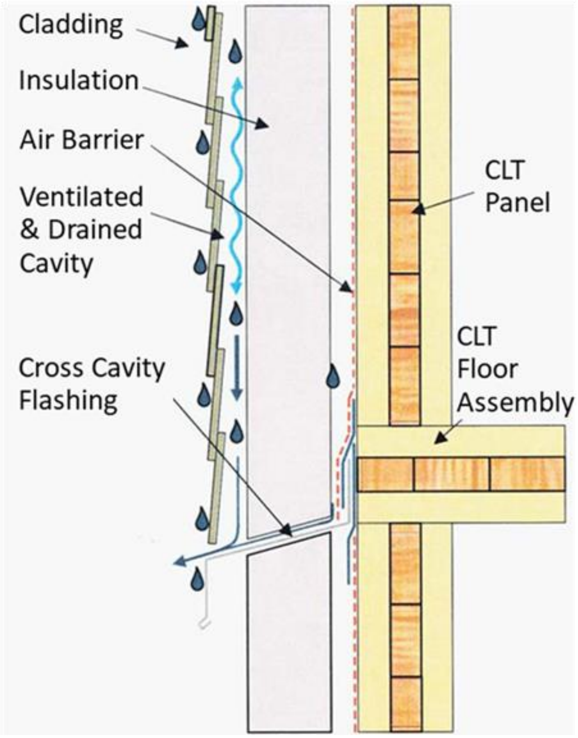


Figure 2.7 Moisture control practices in CLT walls [154].

A detailed description of every path of the system in Figure 2.7 is given below;

Cladding: The first level of protection that helps to deflect moisture away from the structure.

Ventilated and drained cavity: It creates a pathway for the moisture which gets into the cladding to leave the system and reduces moisture transfer from the cladding to the rest of the structural assembly.

Insulation: It improves indoor comfortability and protects the assembly from moisture which might have bypassed the exterior cladding and drained cavity.

Water-resistive barriers: The final level of protection against moisture which might have bypassed other stages of protection.

Cross cavity flashing: This is an internal pathway created for absorbed moisture to leave the assembly.

The following is a summary of practices to address moisture intrusion and control:

- Using naturally durable wood (costs and availability are factors)
- Reduction of exposure during shipping, proper storage, and protection on-site either with tarps, tents, wrapped in plastic, and storing under cover
- Applying water repellent coatings or surface fungicide [28]
- Prefabrication and modular construction
- Developing moisture monitoring programs

As stated above, developing a framework for moisture monitoring in MT structures is critical. This requires employing the following steps: (1) wood moisture meter; (2) identifying the monitoring locations with a high risk of moisture intrusion; (3) data acquisition, storage, cleaning, and processing; (4) developing a monitoring schedule and benchmark data for other applications and cross-reference with other projects [156]. According to Riggio et al. [156], spotting the high-risk locations depends on different factors such as:

- Construction schedule
- Duration of zone exposure
- Exposure to precipitation and wind-driven rain
- Presence of waterproofing elements and weather sealants
- Presence of connectors and cuttings
- Potential for wicking from wood end grain panel ply depth

A method for handling monitoring data of MT buildings is offered by Baas et al. [157]. A unified strategy on moisture monitoring in MT structures should be proposed and practiced by researchers to achieve more standardized planning towards moisture management and control in MT buildings. Cappellazzi et al. [28] stated that a more unified data development approach is needed for monitoring and management of MT durability. Protocols should be developed to identify the potential high-risk locations and methodology for their online monitoring. One of the key challenges could be collecting big data from multiple sources that may not be necessarily synchronized and are typically polluted by noise. Another concern is the processing tools needed to extract the meaningful features that can be correlated with the moisture and other decay indicators. Machine learning or deep learning models may need to be employed depending on the size and complexity of the collected data [158]. Further research can be performed on MT buildings to develop a systematic plan to monitor the durability of these structures and degradation by moisture intrusion and decay.

2.5.2.2 Protection of MT through PT (pre- and post-treatment)

The use of MT in outdoor applications with the risk of biological and environmental degradation requires appropriate protection techniques to ensure product integrity and expand the market for these products [87,159]. Protective methods such as PT in the manufacturing of MT, including, CLT, Glulam, and LVL, have been proposed by several researchers [149,160-161]. For instance, the efficacy of preservative-treated CLT was noted by some authors [149], where no evidence of decay was observed in boron-treated CLT after several weeks of exposure to *Oligoporus placenta* and *Antrodia xantha.* Figure 2.8 shows the comparison between untreated and boron-treated Radiata pine CLT after 12 weeks of exposure. Likewise, some studies revealed the impact of PT on the decay resistivity of poplar LVL compared to their counterpart (untreated

LVL). The results indicated that treating LVL with copper-azole preservatives enhanced its durability against brown and white-rot fungi [161].

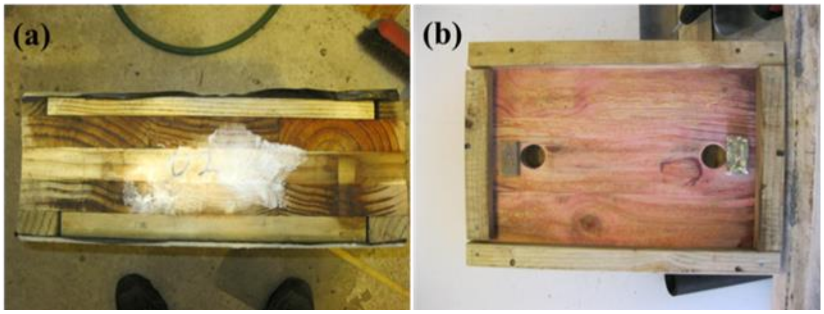


Figure 2.8 (a) Untreated CLT after 12 weeks of exposure and (b) Boron-treated CLT with no trace of decay after 12 weeks of exposure [149].

The incorporation of PT in MT fabrication could be achieved through pressure and non-pressure treatments. These treatment techniques may be used to treat laminates with preservatives before product assembly (pre-treatment) or after assembly (post-treatment) [162]. Pre-treatment involves treating the laminates before gluing and fabrication, while post-treatment introduces the preservative solution to the already glued and large-size laminated materials. In the manufacturing of MT, the former approach is often recommended to ensure deep and uniform penetration of the preservatives into the MT elements [22,152,163]. Such an approach was successfully implemented in producing CLT composed of softwood and hardwood species. Lim et al. [137] investigated the influence of PT and adhesive system on the bonding quality of CLT produced from southern yellow pine lumber. The results showed the feasibility of fabricating CLT from treated lumber without compromising the bonding quality, especially when polyurethane (PUR) adhesive is utilized. In the same way, Adnan et al. [140] manufactured CLTs from four ACQ treated Malaysian hardwood species (batai, sesenduk, rubberwood, and kedondong). They found no effect of PT on the bond performance (block shear and delamination) of the CLTs. While the pre-treatment technique could be used in MT manufacturing as demonstrated by the aforementioned studies, there are precautionary measures to be considered [138,164-165]:

- influence of preservative retention level on adhesive penetration
- impact of wood surface preparation on the preservative-treated laminates
- the blockage of surfaces where adhesive-wood bonding develops by the preservative

As earlier mentioned, the injection of PT into laminated materials (such as CLT, Glulam, and LVL) after fabrication is also worthy of consideration. In the past, Tascioglu et al. [138] established the likelihood of such an approach in laminated products. However, the post-treatment technique has not been applied to MT. Recently, we successfully treated southern yellow pine CLT panels with copper azole preservatives (CA-Type C and MCA) (Figure 2.9). We found that drying the treated panels is critical, and inappropriate handling and drying would generate wood defects (such as checking and splitting) and even delamination of the panels. Ongoing researches focus on the evaluation of preservative penetration and retention, as well as adhesive bonding strength of the post-treated CLTs.

Figure 2.9 Post-treated CLT panels manufactured from southern yellow pine lumber.

Some attempts made in integrating PT into MT manufacturing are summarized in Table 2.5. As demonstrated in this section, there is an opportunity to implement PT as a protection method for MT. Despite the great potential of this approach, more data are needed on the durability of preservative-treated MT elements to determine their resistance to biological and environmental degradation in real-life applications. Thus, the development of appropriate protocols for treating large-size MT elements is important and can lead to the market expansion of MT products worldwide.

Table 2.5 Some attempts made in fabricating MT products using PT

MT Product	Preservative method	Parameter studied	Findings	Reference
CLT	Brushing	Decay resistance	PT improved the decay resistivity of radiata pine CLT.	[149]
CLT	Pressure treatment	Block shear and delamination	Low preservative retention reduced the block shear strength.	[137]
CLT	Pressure treatment	Rolling shear strength	PT reduced rolling shear strength but increased rolling shear modulus.	[63]
CLT	Vacuum pressure treatment	Block shear and delamination	PT did not influence the bonding strength of treated CLT samples.	[140]
Glulam	Pressure treatment	Block shear and delamination	The preservative retention level influenced the bonding performance, by increasing the delamination (See Table 4).	[141]
Glulam	Pressure treatment	Block shear and delamination	Creosote treatment had a negligible effect on bonding quality (shear strength and delamination).	[166]
Glulam	Pressure treatment	Bending strength and stiffness	The bending stiffness decreased with an increase in retention level (See Table 4).	[167]
LVL	Dipping	Bending and compression strength	PT did not affect the bending and compression strength of LVL.	[168]
LVL	Vacuum pressure and dipping	Delamination	There was no evidence of delamination in preservative-treated LVL.	[169]
LVL	Non-pressure soaking	Decay resistance	The decay resistivity of poplar LVL was significantly improved by PT.	[161]

2.5.2.3 Wood modification (use of modified wood to fabricate MT)

As mentioned in Section 3.3, thermal and chemical modifications of wood are environmentally benign alternatives to PT for increased wood's resistance to biodegradation. The commercialization of some thermally and chemically modified wood in Europe further attests to the effectiveness of these methods [105]. While wood modifications have been successfully implemented [105,170-172], information regarding their potential applicability in the fabrication of MT products is lacking.

One wood modification approach that could be used in fabricating MT products (CLT, Glulam, and LVL) is TM. This technique relies on the alteration of the chemical structure of wood's cell wall due to degradation of hemicellulose and reduction in water sorption sites, which would limit the risk of biological attack [102,173]. The performance of MT produced from thermally modified timbers when exposed to outdoor conditions has been documented. For instance, Kržišnik et al. [174] reported an improved decay resistance and reduced moisture absorption for some thermally modified glulam beams. A similar study on LVL made from thermally modified maple and poplar showed a reduction in moisture absorption by 29.3%, which is justifiable due to the degradation of hemicelluloses during TM [175]. Moreover, the potential of incorporating modified wood in manufacturing MT is evident in the recently approved guidelines for use of thermally and chemically modified wood in the North American Market by the American Wood Protection Association [163,176-177].

A major challenge associated with TM is the reduction in the strength properties (such as bending strength and stiffness, even more, impact bending) of the modified products, which could limit their use in structural applications. Aro et al. [178] studied the impact of TM on the mechanical properties of LVL. In comparison with unmodified samples, thermally modified

LVLs (post-treatment) made from poplar, beech, and maple exhibited a 41.7%, 45.5%, and 45.7% bending strength reduction, respectively, as well as 41-53% decrease in the bending stiffness. Similar results were also reported by Kohl et al. [179] and Sahin Kol et al. [180]. A study by Widmann et al. [181] also confirmed the poor bending strength observed in glulam beams fabricated by thermally modified wood. The mechanical strength reduction can be attributed to the degradation of cell wall components (such as celluloses and hemicelluloses) that might have weakened the wood's structure, thereby reducing the rigidity of the material [178]. Cappellazzi et al. [28] suggested that thermally treated lumber may affect the bonding strength of MT, and more research needs to be conducted on this topic. The outcomes from these studies show the need to understand the impact of TM on MT's properties before its implementation

CM has been proved as an effective approach to improve the moisture and weathering performance as well as decay resistivity of wood and wood-based materials [182-183]. The influence of CM such as acetylation on wood's durability has been reported. A general agreement in the literature is that acetylated wood with a weight percentage gain (WPG) of about 20 % is enough to keep wood safe from biological degradation [184]. However, the fabrication of MT using chemically modified lumber has not been explored yet, partly due to the high cost of the CM process. A recent review [28] reported the likelihood of utilizing chemically modified wood products in the external facades of MT elements, indicating the potential of this approach in MT-based buildings. Some examples exist of the use of acetylated wood in load-bearing constructions like bridges (Figure 2.10).

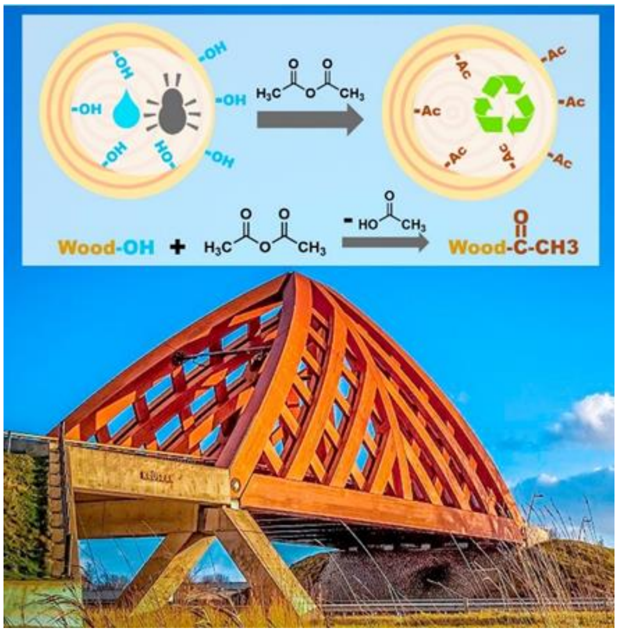


Figure 2.10 Glulam bridge made from acetylated wood. In acetylation, the replacement of the hydrophilic hydroxyl (OH) groups with the hydrophobic acetyl groups (Ac) through acetylation with acetic anhydride makes the wood more durable and dimensional stable [3]

The successful implementation of this system in MT manufacturing line would be beneficial since CM imparts improved properties to wood-based products. Furthermore, the future of CM as an environmentally friendly treatment method makes it even attractive for building professionals. Thus, this approach should be considered in the manufacturing of MT and could offer lasting solutions to the problems encountered when MT is utilized in outdoor applications.

2.6 Effect of Treatment on MT Properties

While protection methods enhance the durability of MT structures against biological and environmental degradation, they also impact the physical properties and mechanical performance of MT products. Apart from the durability of MT under biological/environmental deterioration, there are some other key features to be considered when designing these products. For instance, glue penetration, wettability, and bonding strength and some mechanical properties such as the rolling shear, stiffness or flexural strength are of critical importance in design and manufacturing of MT products. Thus, it is important to study the impacts of protection methods on these features. Such an impact will be discussed in this section for different protection methods and MT products.

2.6.1 Effect of PT on MT properties

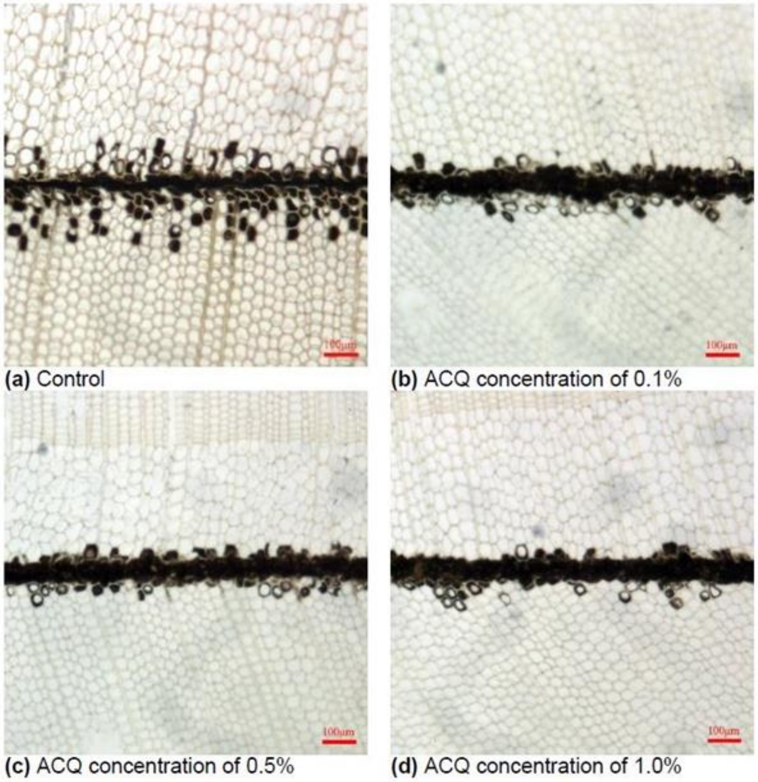
2.6.1.1 CLT

Although PT could help improve CLT's durability and extend its use in structural applications, it may compromise other important properties of CLT such as mechanical and bonding performances, processability, etc. In other words, the complex interactions between wood properties, PT, and adhesive may impose challenges on applications of preservative-treated MT panels.

The impacts of wood properties and PT on gluing properties (i.e., surface roughness, wettability, etc.) of glued wood products were reported. To illustrate, a direct relationship was found between wood density and preservative solution uptake in wood, which could affect the wood's surface roughness. Low-density wood is characterized by more voids, creating a pathway for impregnating solutions into the wood cells [185]. Likewise, wood anisotropy was found to affect the bonding of glued wood products in that preservative impregnation moves faster in the longitudinal direction compared to tangential and radial directions [186]. Therefore, the interaction between wood properties (such as density, anisotropy), surface characteristics (roughness), and bonding need to be well understood if CLT with adequate preservative penetration and bonding strength is desired.

Several researchers evaluated the influence of wood species, PT, surface roughness, type of adhesive, and glue spread rate on the performance of treated CLTs. For instance, Lim et al. [137] investigated the effect of preservative retention levels and wood adhesives on the bonding performance of treated CLTs. An increased rate of delamination was observed for specimens with a higher retention level, especially for panels glued by melamine-formaldehyde and resorcinol-formaldehyde adhesives. In contrast, PUR bonded CLTs provided low delamination rates (<1%) at both low and high retention levels, indicating the suitability of such adhesive in bonding treated CLTs. This is in line with the results of Kuka et al. [186] that found adequate penetration of copper-azole preservatives in PUR-bonded specimens without compromising the bonding strength. Brunetti et al. [187] noted the influence of wood species on the bonding of CLT, where hardwood beech gave poor bonding strength compared to softwood spruce. This was because beech has higher water penetration capability and higher volumetric mass than spruce, which not only induced a higher water absorption rate for beech but also caused a higher deformation upon water sorption.

PT is reported to increase the lumber wettability for adhesives in CLT manufacturing because of the improvement in lumber surface roughness [188]. The high surface roughness usually observed in treated lumber could be because PT raised the wood fibers and deposited chemicals on wood surfaces [189]. Adnan et al. [140] reported increased shear strength for CLT manufactured by ACQ-treated lumber in comparison with untreated ones, which is probably related to the increased wettability of treated lumber. Qin et al. [144] evaluated the influence of preservative penetration on the bonding quality of glued wood products. In this case, lower glue penetration was observed for the ACQ-treated specimens than the untreated specimen (Figure 2.11). As a result, the products manufactured from ACQ-treated specimens exhibited lower shear strength than the untreated ones. Nevertheless, this study also concluded that enhanced adhesive penetration is responsible for better bonding properties. However, it is worth noting that the enhanced wettability can also cause the over-penetration of adhesive that eventually affects the bonding performance of rougher specimens [190]. Therefore, more studies are needed to be conducted to address the relations among PT, wettability, glue penetration, and CLT's bonding performance.



(a) Control
(b) ACQ concentration of 0.1%
(c) ACQ concentration of 0.5%
(d) ACQ concentration of 1.0%

Figure 2.11 Image showing the penetration depth of adhesive in treated and untreated bonded assembly; the treatment reduced the penetration depth in treated samples and the bond line thickness restricts the flow of adhesive into wood cells [144].

The mechanical properties of treated CLT panels are essential for their utilization in load-bearing applications. While some preservatives (such as CCA) undermine wood's strength, several studies reported little to no decrease in the mechanical properties of treated wood samples [191-192]. The strength properties of some CLTs treated with MCA preservatives were assessed by Lim et al. [63] and compared with untreated controls (Figure 2.12). They found no impact of PT on rolling shear modulus but recorded a decrease in rolling shear strength. The authors [63] concluded that even though there was a decrease in the rolling shear strength of the treated CLTs, there was no statistical significance between the rolling shear properties of both treated and untreated CLTs.

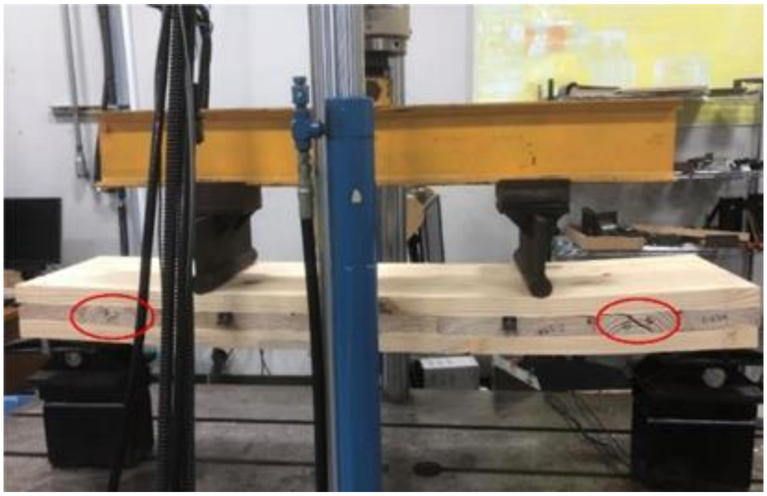


Figure 2.12 Image showing the rolling shear failures of CLT specimen; the red lines indicates the shear zone at core layers of the panels [63]

In summary, evaluating the bonding performance of treated CLT depends on several parameters such as wood anatomy that impacts the roughness, wood treatment, type of adhesive, adhesive spread rate, etc. and can hardly be generalized. Thus, a better understanding of the relationships between these factors and how they affect bonding is important. More studies should be conducted with the focus on optimizing the manufacturing parameters to produce preservative-treated CLT with satisfying mechanical and bonding performance.

2.6.1.2 Glulam

Previous studies reported the blockage of preservative solution due to the wood anisotropy and glue line could affect the impregnation of preservatives in glulam [174]. For instance, low preservative uptake was observed in the tangential direction of some glulams produced from Radiata Pine regardless of the treatment regime compared to other directions (radial and longitudinal) [160]. Also, a recent study by Kuka et al. [186] revealed the role of glue lines in preservative penetration of glued wood products (bonded using PUR adhesive). The reports showed that Cu preservative was restricted in the fabricated glued wood product due to the presence of glue lines. Similar observations were reported for treated glulam bonded using formaldehyde-based adhesives [4,20].

Moreover, Hansel et al. [193] noted the influence of surface preparation on the gluing properties of glulam due to Physico-chemical changes caused by wood machining that reduces its wettability and bonding quality. Further, the effect of surface characteristics on the bonding quality of copper chrome boron (CCB) treated glulam was investigated by some authors [194]. Although low values were reported for the shear strength, they were attributed to the interaction between the adhesive and preservatives as well as the deposition of solutions on treated wood surfaces, which affected the bonding quality. Another factor that affects the bonding of glulam is glue-line thickness, in that low-viscous adhesive flows freely on the wood surface, leading to starved glue joint compared to the high-viscous adhesive that results in adequate bonding [194-195].

Several studies focused on the effect of PT on the mechanical properties of glulam. However, there was a disparity in the results reported by different authors. To illustrate, a reduction in the strength properties (such as static bending), bonding strength (shear strength), and severe delamination were observed in rubberwood glulam beams treated with CCA and CCB and bonded with epoxy adhesive [196]. The poor mechanical and bonding quality of the beams were linked to the changes in the surface properties of wood (due to wood-adhesive interactions) after treatment. However, other researchers in their work on pine glulam treated with CCA and CCB reported no negative impact of PT on the mechanical properties of the glulam beams [197]. Thus, there was no universal agreement in the literature on how PT changes glulam's mechanical properties and/or bonding quality, which necessitate further research. Therefore, the compatibility between the treatment, adhesive, and wood species should be considered in glulam manufacturing as this will determine the quality of the final product.

2.6.1.3 LVL

The manufacturing process of LVL through surface treatment and possible formaldehyde-based adhesive interactions with wood enhances its durability in comparison to other MT products. Singh et al. [149] compared the resistance of untreated LVL and CLT subjected to the same biodeterioration conditions. The untreated CLT showed severe decay after 56 weeks of exposure, while slight traces of mycelium growth was only observed in LVL; the performance of LVL was attributed to the durability impacted during fabrication. Few studies explored the relationships between the adhesive, PT, and bonding quality of LVL. An important attribute of LVL is the thinner layers, which contributes to better preservative penetration as opposed to other MT products. Some researchers treated LVL made from red maple with copper-azole, MCA, and ACQ preservatives and found no effect of treatment on its bonding performance [162,169]. The positive attributes of LVL, including ease of treatment, inherent durability, and good bonding quality, make it worthy of consideration as a structural material. However, the limited studies on the role of PT, glue line interference, and manufacturing parameters on the quality of LVL might limit the confidence in its use. Therefore, more studies focusing on understanding the complex interactions between these factors and how they affect the durability and in-service performance of LVL are needed.

2.6.2 Effect of TM on MT properties

2.6.2.1 CLT

Few studies have been conducted on the impact of thermal or steam treatment on the properties of CLT where treatment improved the dimensional stability of the panels [198-199]. Gereke et al. [200] stated that heat treatment improves the swelling, warping, and internal stress in CLT. However, they pointed out the reduction in some mechanical properties following the heat treatment. One solution could be to fabricate CLT from a combination of treated and untreated wood to compensate for the reduction in mechanical properties; though, it resulted in higher stress and deformation [200]. Some authors also investigated the impact of steam treatment on CLT and found that manufacturing parameters such as the pressing time, adhesive amount and pressure significantly affected the properties of CLT [201]. They reported that based on the conditioning, the shear strength and the wood failure percentage could vary in the range of 1.3 MPa to 4.7 MPa and 76% to 92%, respectively.

Currently, the literature lacks comprehensive studies discussing the impact of thermal treatment on the gluing quality, wettability, and bonding strength of CLT. Thermal treatment improves the durability and dimensional stability of MT structure elements. However, the impact of such a treatment on the mechanical properties of the fabricated products can be hardly generalized and depends on different factors such as the treatment process, temperature range, holding time as well as the manufacturing process such as the gluing rate, pressing condition, etc. Therefore, further research should be performed to better explain the reduction or improvement in the mechanical properties of MT products following thermal treatment. Understanding the role of different manufacturing parameters along with optimizing the fabrication process can result in durable products being used in load-bearing applications. Still, one of the main challenges of thermally modified MT products is the lower MC and hydroxyl groups that lead to challenges with wettability and bonding strength or delamination. Thus, poor bonding quality resulting from thermal treatment requires more attention. The impact of wood machining and surface treatment methods such as sanding on the surface roughness and wettability and its possible impact on the bonding quality between the wood and adhesive should be further studied.

2.6.2.2 Glulam

The effect of heat/hydrothermal treatment on the stability and mechanical performance of glulam has been studied in the literature. Mirzaei et al. [202] showed that hydrothermal treatment improved the stability of glulam made from poplar resulting in lower moisture and water absorption of glulam beams. Likewise, Mirzaei et al. [203] showed that hygrothermal treatment decreased the moisture-induced stresses as well as relevant moisture gradients in the cross-section of glulam. They also reported an improvement in the mechanical properties, i.e., modulus of elasticity (MOE) and modulus of rupture (MOR) of glulam following the hydrothermal treatment. While the MOE and MOR of glulam made with poplar were lower than those of poplar wood, the treatment improved bending properties [202]. Yue et al. [204] reported that thermal treatment improved the MOE of glulam beam made of fast-growing poplar, but it reduced the MOR. They discussed that while the glulam made from thermally treated poplar can be used in construction, they should be checked for MOR and utilized in a limited range of structural applications. Different trends in the mechanical behavior of wood following thermal treatment were reported in the literature [24]. The impact of the parameters involved in the thermal treatment such as the treatment temperature, holding time, etc. on the mechanical properties of the fabricated MT products should be further studied.

Mirzaei et al. [203] discussed that hydrothermal treatment resulted in brittle failure in glulam beams under bending test. The poor mechanical performance of glulam made from thermally modified timber could be linked to its brittle behavior [181]. The brittleness of wood following TM [205] necessitates studying the seismic behavior of MT structures made from thermally modified timber.

Despite the abovementioned improvements in properties of glulam following heat treatment, it decreased the bonding shear strength and increased the delamination rate in glulam and this trend was positively correlated with the treatment temperature. Mirzaei et al. [202] explained that different factors could be responsible for the reduction in the bonding strength of glulam made of hydrothermally treated wood. One factor is the reduction of the hydroxyl groups in hydro-thermally treated wood [206], which weakens the bonding between the wood cell and adhesive. Another factor is the lower MC of treated wood that may result in insufficient penetration of adhesive in the wood during the manufacturing process of glulam. Since the adhesive penetration is impacted by the MC of wood [207], this can negatively contribute to lowering the bonding strength in fabricating MT from thermally modified wood. Finally, the bonding strength is greatly affected by the wettability and surface roughness of wood. Mirzaei et al. [202] reported a reduction in the wettability and roughness of hydrothermally treated poplar that may not allow for proper mechanical interlocking between the wood cells and adhesive and thus leads to poor bonding performance in the treated product.

2.6.2.3 LVL

The impact of thermal treatment on the mechanical properties of thermally modified LVL has been reported. Saražin et al. [208] reported a 19% reduction in the MOR of LVL made from thermally modified beech veneer while the MOE did not significantly change. Kol and Seker [209] also revealed that thermal treatment at 212°C for 2h reduced the MOR and hardness by 31.85% and 25.44%, respectively. Similar results where heat treatment reduced the MOR of LVL were found in the literature [175,210]. Similarly, a 31% reduction in the MOE of LVL made from heat-treated veneer at 180 °C was reported by Nazerian and Ghalehno [210]. Aro et al. [178] reported that MOE of LVL and laminated strand lumber (LSL) remained almost unchanged by thermal treatment up to 180°C while there was a noticeable reduction in the tensile strength of the products. Such a reduction in the tensile strength following heat treatment up to 210°C is reported for other veneer-based products such as plywood [211].

Percin and Altunok [212] discussed that reinforcing the LVL by carbon fiber can enhance the mechanical properties of LVL made from thermally modified beech veneer. Altinok et al. [213] also reported that increasing the heat treatment temperature in the range of 100°C-150°C can enhance the bending strength of LVL. The screw withdrawal strength of thermally treated LVL was reported to improve at 140°C but then decreased at higher treatment temperature [214]. It is clear from the presented results that the change in the mentioned mechanical properties of LVL can hardly be generalized and is affected by many factors such as the treatment cycle, heat treatment time and temperature, etc. Therefore, the interactions of these factors with the bonding strength and mechanical properties of LVL need to be understood.

2.6.3 Effect of CM on MT properties

Chemical modifications, like preservative and thermal treatments have been used to improve the durability of wood and wood products. Several authors have also shown improved mechanical properties, including, hardness, compression and bending strength as well as dimensional stability for chemically modified wood, thus, making it suitable for structural applications [215-217]. While these studies have lauded its effectiveness in enhancing wood's properties including durability, it has not yet been adopted in the MT industry. The research outcomes from chemically modified wood are encouraging for the MT industry, since MT products, particularly, glulam are used in beams and columns which are loaded in bending and compression. Additionally, one of the major challenges with CLT in service are moisture stresses which are created due to differential layer shrinkages [218-219]. Therefore, the use of chemically modified wood with increased dimensional stability could create opportunities to find a lasting solution to this problem.

Further, there are several CM methods (such as acetylation and furfurylation) with each of them having different effects on wood. Acetylation improves considerably the dimensional stability and fungal resistance [215,217], which are desirable traits in MT products. However, this technique (Acetylation) also reduces the water absorption of wood alongside other property changes, which could significantly affect the bonding quality of wood [220]. Similarly, furfurylation reduces the equilibrium MC (EMC) of wood, improves compressive strength, reduces water expansion and shrinkage by almost 50% compared to untreated wood, and also improves corrosion resistance [217]. Yet, there are doubts concerning the curing efficiency and brittleness of the modified wood.

It is consensus that MT products will benefit from using CM methods, yet only a few studies have studied this technology, established the difficulties associated with using them and proffer adequate solution. One of such studies by Crawford et al. [215] shows that apart from the increased mechanical properties, glulam beams made from acetylated wood and tested in four-point bending failed mostly in tension at the bottom of the beams which means that there were no adhesive failure and acetylation did not have a negative impact on adhesion of laminar. This was however, only visually observed and authors suggested a more robust test be carried out to adequately evaluate bond failure in modified wood. Bongers et al. [220] also evaluated the bonding performance of non-load bearing laminated acetylated wood, evaluating the results based on chemical composition, ultra-structure, and other properties altered by the acetylation process. They concluded that in non-load bearing applications, acetylated wood gives good performance with PUR adhesives even upon exposing them to severe delamination cycles. Additionally, while laboratory-made specimens with PUR achieved good results in terms of wood failure, commercially produced specimens did not perform well. Future studies should consider the treatability of wood species to be used in fabricating MT made from chemically modified wood, in that only well-treated species (for instance southern yellow pine and radiata pine) could exhibit the above-mentioned positive properties. MT products are mostly utilized in load-bearing conditions which necessitate studying the impacts of CM on the various properties of commercially manufactured MT products. Therefore, there is an urgent need to fill the literature gap as the MT industry continues to evolve and further product developments continue to take place.

2.7 Conclusions

The growing interest in MT utilization in buildings necessitates focusing on its durability and protection against moisture intrusion and decay. The research community has started studying the protection of mass timber products using PT as well as wood modification. This paper reviewed wood protection and modification methods and discussed the potential of applying these methods on MT. Health and environmental concerns of PT may highlight other methods. TM is green and effective but may negatively impact the mechanical performance of MT. The high cost is a challenging factor for the wide applications of chemically modified wood in MT. The impact of different protection strategies on the durability, mechanical, and gluing performance of MT should be further assessed. Studying the durability of MT is influenced by the size of the structure, type of the wood species used, service climate condition, fungal species and type of protection, and time and condition of incubation. More research is needed to account for these factors and provide standard protection protocols. Other than the protection and treatment methods, moisture monitoring and control is the key element of durability assessment of MT. Attention should be given to unified data collection and developing monitoring protocols in MT buildings. Identifying the high-risk locations, tools for data acquisition and processing, and correlating the collected big data to the meaningful durability features in the building are critical tasks. Future research can help in providing standardized methods for MT protection and monitoring its durability. Prospection of MT affects its physical and mechanical properties including surface wettability, glue penetration, or bonding strength. Such an impact depends on different manufacturing parameters that require further research to be well understood. Optimizing the treatment process and MT manufacturing may result in having a well-protected and durable product while not compromising the key mechanical properties that are of crucial importance in load-bearing applications.

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CHAPTER III

PRESERVATIVES PENETRATION AND RETENTION IN POST-TREATED CROSS-LAMINATED TIMBER PANELS WITH DIFFERENT LAYUP AND THICKNESS

Ayanleye, S., Quin, F., Zhang, X., Lim, H. & Shmulsky, R. (2023). Preservatives Penetration and Retention in Post-treated Cross-Laminated Timber Panels with Different Layup and Thickness. *Journal of Building Engineering*, 67, 106009.
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3.1 Abstract

Cross-laminated timber (CLT), a prefabricated multilayer engineered wood product, is a promising construction material for mid-and-high rise buildings due to its good mechanical properties, renewability, and low-carbon footprint. However, the vulnerability of CLT to biodeterioration limits its broad applications. Preservative treatment is an effective method of increasing the service life of structural wood products, while the treatment of CLT has not been widely studied. Herein, we fabricated 3- and 5-ply CLT panels and treated them with Cu-based preservatives. The effects of panel layup (lengthwise and crosswise) and thickness (3-and 5-layer) on the impregnation quality (i.e., Cu penetration and retention) are investigated using a color-based indicator approach and X-ray fluorescence spectroscopy. Generally, the Cu penetration ratio was mostly above 90%, especially for the top layers (1st layer in 3-ply and 1st and 2nd layers in 5-ply) of the treated CLT panels. Similarly, higher Cu retention values were observed in the 1st layer of the 3- and 5-layer CLT panels. Further, there was a similarity in the Cu penetration of the 2nd layer in 3- and 5-ply CLT, indicating the preservative diffusion across adjacent layers is negligible in the preservative treatment of lumber-based wood composites.

Overall, copper-azole type C (CA-C) exhibited better treatability than micronized copper azole (MCA) for CLT panels, resulting in higher Cu penetration and retention across the panels. Also, we found the lengthwise orientation preferable in fabricating preservative-treated CLT, this is due to the complete panel protection of the 1st and 2nd layers at all panel locations. Thus, our results show the feasibility of successfully treating CLT, especially when CA-C preservative and lengthwise orientation are utilized in fabricating the panels.

Keywords: Cross-laminated timber, Copper-azole type C (CA-C), micronized copper-azole (MCA), penetration, retention

3.2 Introduction

Global warming, non-renewable energy, resource depletion, and environmental pollution are major challenges for human beings in the 21st century. In the United States, the CO_2 emission and energy consumption of buildings occupy 38% of total CO_2 emission and 70% of the entire energy consumption, respectively [1]. Large portions of CO_2 emission and energy consumption of buildings are associated with the manufacturing of building materials such as concrete and steel. To date, the market for building materials is still dominated by non-sustainable reinforced concretes [2], which limits the development of green buildings. According to Wang *et al*. [3] and Liang *et al*. [4], concrete and steel structures embodies and consume approximately 12% and 20% more energy than timber-based structures. The use of laminated-timber systems can significantly reduce the energy consumption of buildings [5]. Therefore, the utilization of sustainable materials for green building development is urgent. Recently, cross-laminated timber (CLT) has been developed as a new generation of sustainable engineered wood product for buildings and constructions [5]. CLT consists of several layers (at least 3) of structural lumber that are stacked orthogonally and bonded together by adhesives [6, 7]. Because of the renewability, good dimensional stability, high design flexibility for buildings, quick assembly/installation, good thermal and acoustic insulation performance, and high specific strength [8, 9], CLT is rapidly occupying the green building market worldwide. However, CLT is mostly fabricated from less durable softwoods, making it susceptible to fungi decay and termite attack upon exposure to high humidity conditions and ground environment [5, 10]. This limited the broad application of CLT for constructions and buildings, especially in tropical regions where the buildings are prone to high humidity and insect infestation [11]. Therefore, there is a critical need to develop high-durable CLT for use in constructions with harsh biodeterioration environments.

Preservative treatment is an effective and efficient method that increase the durability of wood products and prevent microorganisms and termite attacks [12]. Copper-based preservatives are widely used for wood protection because of their excellent protection performance and cost-effectiveness [13-15]. Copper-azole type C (CA-C) and micronized copper azole (MCA) are the representative copper-based preservatives in the market and are commonly used for lumber treatment [16]. The fixation of copper-based chemicals, characterized as Cu distribution and retention, is an indicator of the impregnation quality and determines the durability of treated wood products [17, 18]. Cu distribution and retention are affected by wood species, moisture content, thickness, wood grain, etc. A series of standard treatment protocols have been established for lumber and logs from several wood species for the manufacturing of preservative-treated products for different applications [19, 20]. However, CLT is composed of laminated lumbers that are glued by adhesives and its dimensions are much greater than conventional lumber products. Thus, the treatability of CLT panels through the conventional lumber treatment processes is unknown. Moreover, the adhesive layers between lumbers may block the preservative diffusion path during the treatment [21], thereby affecting the penetration and retention of Cu in CLT panels. Furthermore, the evaluation of the performance of treated CLT through proper sampling procedure is still unclear, which raises additional concerns.

There are two main approaches, that is, pre- and post-treatment in treating CLT panels with Cu-based preservatives. The former involves the manufacturing of CLT using preservative-treated laminations while the latter deals with the injection of preservative chemicals into already fabricated CLT panels [5]. Few studies were found on the fabrication of preservative-treated

CLT; however, these studies mostly focused on the CLT's mechanical properties and bonding performance with little emphasis on the penetration and retention of preservatives in the treated panels [10, 11]. For instance, Lim *et al.* [10] and Cai *et al*. [22] studied the influence of Cu retention levels on the bonding performance of pre-treated CLT, where delamination rates increased with retention levels based on different adhesive systems. Adnan *et al.* [9] also found no significant effect of preservative treatment on the block shear strength of pre-treated CLT panels. To our best knowledge, no studies were found that investigated the treatability of prefabricated CLT panels. To fill this research gap, this article treated prefabricated CLT panels with Cu-based preservatives and evaluated Cu penetration and retention. CLT panels were fabricated using SYP lumbers according to our already established procedures [10]. The panels were then treated with two commercial Cu-based preservatives (i.e., CA-C and MCA) through a standard pressure treatment protocol of SYP lumbers. The Cu penetration and retention in the post-treated CLT panels were investigated by Cr color indicator and X-ray fluorescence spectroscopy, respectively. We studied the influence of panel layup and thickness of CLT, as well as the types of Cu-based preservatives on penetration and retention. Finally, the distribution of Cu-based preservatives in the post-treated CLT panels was elucidated. The overall goal of this research is to determine the best preservative system and layup pattern for treating CLT and to further understand the chosen Cu-based preservatives' penetration paths.

3.3 Materials and Methods

3.3.1 Materials

Visually graded select structural southern yellow pine (SYP) 2 × 6 (38 mm × 140 mm) lumber (2.4 m long) was supplied by Shuqualak Lumber Company (Shuqualak, MS, USA). To minimize the effect of lumber variability on the distribution of preservatives in the treated CLT, defect-free flat-grained SYP lumbers in a certain density range were manually selected and used for CLT manufacturing. The selection of these SYP lumbers was done following the ASTM D2559 standard [23]. The moisture content (MC) of the SYP lumber was measured with an electronic moisture meter upon arrival at the testing lab. The lumber was stored indoors for at least four weeks before CLT assembly. The lumber with pith was excluded in the selection since heartwood is harder to penetrate than sapwood. Also, lumber with compression wood was eliminated from the study. One-component polyurethane adhesive (PUR HB X602) and primer (LOCTITE PR 3105) were supplied by Henkel corporation (Rocky Hill, Connecticut, USA). The primer was mixed with tap water in the ratio of 1:9 (10% and 90% volume of primer and water, respectively) before use. The commercially available CA-C and MCA preservatives were used for CLT treatment. According to the American Wood Protection Association (AWPA) Standards, the active ingredients of CA-C are 96.1% copper, 1.95% propiconazole, and 1.95% tebuconazole [24], and MCA contains 96.1% copper and 3.9% tebuconazole as active ingredients [25]. The efficacy of these preservatives (i.e., CA-C and MCA) are evaluated following the relevant standards such as AWPA A69-12 and A9-16 [26, 27] and thus utilized in this study. A comprehensive list of the common standards for evaluating treated wood products can be found elsewhere [5].

3.3.2 CLT Manufacturing

Prior to CLT manufacturing, SYP lumbers were conditioned indoors to MC of 12 ± 3% as recommended in the CLT Handbook [28]. The supplied lumbers were weighed with an electronic floor scale and divided into different weight groups to exclude the effect of lumber density on CLT's properties (e.g., bonding strength and preservative treatability). The lumber was classified into four weight groups, including, group A (weight < 6 kg), group B (weight of 6-7 kg), group C (weight of 7-8 kg), and group D (weight > 8 kg). Only the lumber in groups B and C was used in CLT fabrication. The lumbers were planed to a thickness of 35.6 mm and a width of 134.6 mm. The lumbers were then cut into 762 mm and 508 mm in length for defect-free laminations. The MC and oven-dry specific gravity ($SG_{oven\text{-}dry}$) of the laminations (**Table 3.1**) were determined following the ASTM D2395 and ASTM D4442 standards [29, 30].

Table 3.1 Summary statistics of MCs and SGs of lumbers used in CLT fabrication

		MC (%)			$SG_{oven\text{-}dry}$		
		Mean	SD[a]	COV[b]	Mean	SD	COV
3-layer CLT	Control	10.77	1.21	11.19	0.52	0.02	4.68
	CA-C	11.31	1.17	10.36	0.49	0.04	9.18
	MCA	12.44	1.38	11.13	0.48	0.04	7.54
5-layer CLT	Control	10.70	0.82	7.70	0.50	0.04	7.02
	CA-C	13.98	1.10	7.88	0.46	0.03	7.33
	MCA	13.19	1.18	8.93	0.46	0.03	6.21

Note: [a]SD means standard deviation, [b]COV means coefficient of variation.

The layers parallel to the longer side of the panels were composed of three laminations (1.4" × 5.3" × 26.6"), while the cross layers were composed of five laminations (1.4" × 5.3" × 15.9"). The laminations were used for CLT construction within 8 h of planning. After that, the laminations were spray coated with wood primer (spreading rate of 20 g/m^2) and cured for 10 mins, followed by applying PUR adhesive with a spreading rate of 180 g/m^2 following the adhesive product specifications [31]. The CLT panels were fabricated by bonding the laminas face to face into the final panel dimensions of 403.9 mm × 675.6 mm × 104.1 mm (3-ply) and 403.9 mm × 675.6 mm × 175.3 mm (5-ply) using a laboratory hydraulic press (Dieffenbacher North America, Inc) under a clamping pressure of 100 psi (0.69 MPa) for 3 h. Two configurations of 3-layered (3-ply) CLT panel and one configuration of 5-layered (5-ply) CLT panel were manufactured. The panels were labeled as *X*-ply-*Y*, where *X* indicates the number (3 or 5) of lumber layers, and *Y* reveals the lumber direction (P or C) at the top layer of the CLT panel. Specifically, “P” indicates that the longitudinal direction of the first layer of lumber is parallel to the panel’s length direction (or lengthwise configuration). In contrast, “C” indicates that the longitudinal direction of the first layer of lumber is crosswise to the panel’s length direction (or crosswise configuration). **Figure** 3.1 illustrates all three configurations of the fabricated CLT panels. Eight CLT panel replicates each were constructed for 3-ply-P and 3-ply-C, and four CLT panel replicates for 5-ply-P, making a total of twenty CLT panels constructed for all panel configurations. The panels were constructed using the parameters described above. Afterward, the panels were stored indoors until they were sent for treatment at the treating facilities.

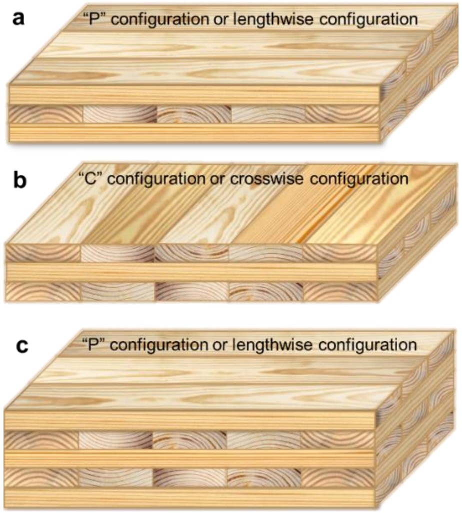

Figure 3.1 Schematic illustrates the fabricated CLT panels with different configurations, (a) 3-ply-P, (b) 3-ply-C, and (c) 5-ply-P.

3.4 Preservative treatment of CLT

The prefabricated CLT panels were treated with CA-C and MCA preservatives. Four panels each were chosen from 3-ply-P and 3-ply-C for CA-C and MCA treatment, respectively, while two panels each were chosen from 5-ply-P CA-C and MCA treated panels. 3-ply-P and 3-ply-C were used to compare the influence of panel layup while 3-ply-P and 5-ply-P were used to compare the effect of panel thickness on the penetration and retention of preservatives in the treated CLT panels. The CA-C and MCA treatments were carried out at a commercial treating facility (Deforest Wood Preserving, Bolton, MS) and a research facility (Koppers Performance Chemicals, Griffin, GA), respectively. Both treatments followed a treating schedule for 2"-thick lumber through a full-cell process, and the target Cu retention for lumber is 2.4 kg/m^3. While the CA-C panels were treated with 2"-thick lumber, the research facility (Koppers) treated CLT panels only. The target Cu retention was confirmed for the CA-C and MCA-treated panels. This retention level is designed for UC4A (ground contact or freshwater) applications as specified by the AWPA U1-18 [32]. For CA-C treatment, the treating parameters are as follows: initial vacuum 18 in, Hg hold for 3 min, next 155 psi (1.07 MPa) of pressure hold for 11 min, and final vacuum 20 in. Hg holds for 58 min to remove the excess preservative liquids. For MCA treatment, the treating parameters are as follows: initial vacuum 18 in. Hg holds for 5 min, next 150 psi (1.03 MPa) of pressure hold for 15 min, and final vacuum 26 in. Hg holds for 15 min. After treatment, the CA-C and MCA treated panels were allowed to air dry for 1 and 7 d, respectively, before being transported back to our laboratory. Then, the treated panels were stored under a covered shed with fans and air-dried for three months to an MC level of ~ 20%. **Figure** 3.2 shows a pictorial representation of some treated (3- and 5-ply) CLT panels.

Figure 3.2 A pictorial representation of some 3-ply and 5-ply treated CLT panels.

3.5 Evaluation of Preservative Penetration and Retention

3.5.1 Sampling Method

Wood is an anisotropic material that has three principal planes namely, longitudinal (L_L), radial (L_R), and tangential (L_T) directions. The penetration and retention of preservatives vary across the principal planes of wood and are thus taken into consideration in the evaluation of treated wood products. The longitudinal direction has the highest permeability for liquids and gases because of the alignment of hollow wood cells in this direction [33]. Thus, preservative penetration is most effective in the L_L direction, followed by the L_R direction (due to the presence of ray cells) and lastly in the L_T direction of wood [19]. The experiment was designed to examine the influence of panel layup and the number of layers (i.e., thickness) on preservatives' penetration and retention by considering the shortest lengths along the three penetration paths from the panels' closest boundary planes, as shown in **Figure** 3.3: L_L, L_R, and L_T. CLT panels were cut into 15 blocks, in which the length and width of blocks were 127 mm, while the height of the blocks was the same as the thickness of the panel (**Figure** 3.3). Then, the length and width of blocks were further trimmed to 101.6 mm by a band saw in the panel's width direction. The specimen locations were classified as Corner (CR), Center (C), and Edge (E) (**Figure** 3.3). And the specimen's laminations were classified as the layer numbers, as shown in **Figure** 3.3 for a 5-ply CLT panel. For the Cu penetration, six blocks each were evaluated from the sixteen and four panels constructed for 3-ply and 5-ply CLT, respectively. Also, we tested six blocks each from a total of six panels (1 panel each from 3-ply-P CA-C, 3-ply-P MCA, 3-ply-C CA-C, 3-ply-C MCA, 5-ply-P CA-C, 5-ply-P MCA) for the Cu retention determination. For each CLT panel, we evaluated the Cu penetration and retention of 6 specimens regardless of the number of layers, as illustrated in **Figure** 3.3.

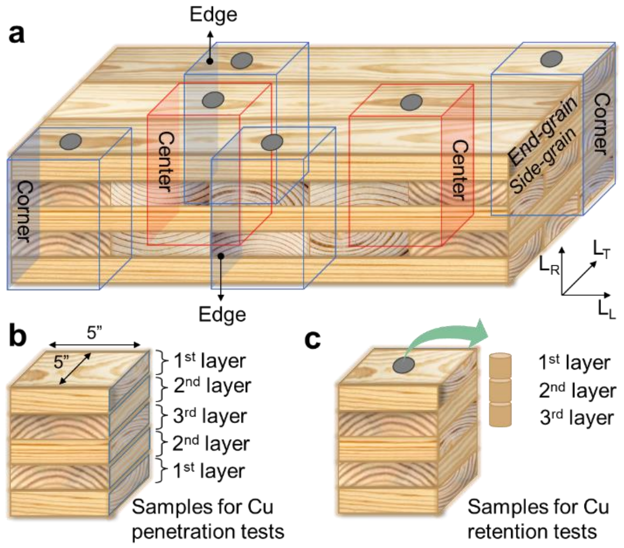

Figure 3.3 (a) Sampling method for Cu penetration and retention tests, in which the panel's length, width, and thickness directions are termed as L_L, L_T, and L_R-direction, respectively. Six samples were cut from each panel for Cu penetration test. The blue and red colored faces are chrome azurol sprayed surfaces, which are classified into three groups, corner, center and edge, respectively. The black circle marked places are where samples were taken for Cu retention tests. (b) A typical example of the Cu penetration sampling method. (c) A typical example of the Cu retention sampling method.

3.5.2 Evaluation Method

The preservative penetration was investigated by spraying the CLT blocks with chrome azurol solution (CAS), following the procedures listed in the AWPA A69-12 standard [26]. Specifically, surfaces of the CLT blocks' x-directions (blue and red color labeled surfaces in **Figure** 3.3) were sprayed with CAS for Cu penetration evaluation. After spraying with CAS, the surfaces turned to a blue color, indicating the presence of Cu. The sprayed surfaces were air-dried for 24 h, and then digital images of the dried (sprayed) surfaces were recorded by a Flatbed Scanner (CanoScan LiDE 400). The images were processed using ImageJ software (National

Institutes of Health, Bethesda, MD, USA) and the blue-colored areas were marked for the calculation of the Cu penetration ratio for the lumber located at different locations in the panel.

For the copper retention determination, the particle samples were obtained from six blocks (black circles marked blocks in **Figure** 3.3) of each panel using a sharp increment borer. For each location, the samples were collected throughout the entire panel thickness. Then, the particles were milled to 20 mesh fine powders using a Wiley mill followed by oven-dying them at 103^{o}C for 24 h. The dried powders were shipped to Timber Products Inspection (Georgia, US) and the Cu retention was tested by X-ray spectroscopy according to the AWPA A9-16 standard [27].

3.6 Statistical Analysis

The study was designed to evaluate the effects of the panel layup (lengthwise and crosswise) and thickness (3- and 5-layers) on the penetration and retention of preservatives in CLT panels. Data were tested for normality and homogeneity of variance using the Shapiro-Wilk test and Levene's test, respectively. In the scenarios where the assumptions were not met, the data were transformed and tested again. If after transformation, the data could not be normalized, the Kruskal-Wallis H test, a non-parametric equivalent of ANOVA was implemented to analyze the significance of the main effects [10]. Also, Dunn's test for multiple comparisons was used to compare several groups of observations when the effect analyzed was significant. All statistical analyses were performed at a 5% significance level and done using IBM SPSS Statistics, Version 28 (IBM Corporation, New York, NY, USA).

3.7 Results and Discussion

3.7.1 Preservative penetration in the treated CLTs

In this study, the Cu penetration ratios of lumber specimens obtained from different locations of CLT were determined to elaborate on Cu diffusion in the treated panels. A comprehensive summary of the average Cu penetration ratio is presented (**Table** 3.2), along with the shortest distances (L_L, L_R, and L_T) from the panels' boundary planes to the examined specimens' surface. Overall, we found higher Cu penetration (≥ 90%) for the 1st layer, irrespective of the panel layup and thickness (number of layers) of the treated panels. For the 2nd layer, the Cu penetration was mostly above 85.0%, especially for 3- and 5-ply CA-C treated panels. According to the AWPA T1-16 standard for the evaluation of Cu penetration in post-treated wood composites, 85.0% of the Cu penetration ratio in the top 0.6 in. (15.3 mm) thickness of the composite panel will be sufficient [34].

As shown in **Table** 3.2, the Cu penetration ratios of the 1st layer (thickness is 35.6 mm) for the 3- and 5-ply CLT were mostly above 90%, indicating the possibility of treating prefabricated CLT with adequate Cu penetration. In 3- and 5-ply CLT, a higher Cu penetration ratio was observed at the panel corner, for the 1st and 2nd layers of CA-C and MCA-treated panels (**Table** 3.2). This is attributable to more exposure of the corner specimens to preservatives compared to other panel locations. Generally, the Cu penetration of the 1st laminations was higher in the treated (3- and 5-ply) panels. This could be because more than one of the L_L, L_R, and L_T is zero, which results in the better treatability of the 1st laminations. While the Cu penetration of 2nd layer in 3-ply-P panels was mostly above 85%, we found a low Cu penetration ratio for 2nd layer center specimens of 3-ply-P MCA panels. This was sought to be attributed to the Cu penetration evaluation method and mode of fabrication of CLT, since the Cu penetration of 2nd layer was measured on side-grain rather than end-grain specified in the AWPA standard. To evaluate this possibility, we sampled two 3-ply-P MCA center specimens with Cu penetration of 45.4% and 45.5% at the side grain and end-grain, respectively (see **Figure** 3.9 in supporting information). This indicates that the evaluating method (i.e., either end-grain or side-grain evaluation) had no significant effect on the Cu impregnation in lumber. Therefore, the reduced Cu penetration might be due to less exposure of center specimens to preservatives as well as the low solubility of MCA.

Additionally, we investigated the effect of wood anatomical features (such as annual ring width) on the treatability of the CLT specimens. While we observed wide annual rings in some specimens, others had a narrow gap in their annual rings. However, we found a comparable Cu penetration ratio in specimens with wide and narrow rings in 3-ply CA-C and MCA treated panels. For instance, two sampled corner specimens with wide and narrow rings had Cu penetration of 100 and 98%, respectively (**Fig.** 3.10 a and b). Similarly, we recorded Cu penetration ratios of 95 and 97% for some corner specimens with wide and narrow rings in MCA-treated panels (**Fig.** 3.10 c and d). These observations further demonstrate that the influence of wood anatomical features on CLT's treatability is negligible when lumber with similar density is used for CLT manufacturing.

Table 3.2 Summary of Cu penetration ratio in the 3- and 5-ply CLT panels

Specimen	Location	Layer #	Penetration ratio				
			Average (%)	S.D.	L_L (mm)	L_T (mm)	L_R (mm)
3-ply-P CA-C	Corner	1st	96.0	5.7	0	0	0
		2nd	96.0	4.9	0	0	35.6
	Center	1st	95.1	7.1	134.6	134.6	0
		2nd	88.2	21.0	134.6	134.6	35.6
	Edge	1st	90.1	14.8	201.9	0	0
		2nd	97.8	3.5	0	201.9	35.6
3-ply-P MCA	Corner	1st	96.0	8.2	0	0	0
		2nd	83.6	17.2	0	0	35.6
	Center	1st	97.3	3.9	134.6	134.6	0
		2nd	69.1	29.4	134.6	134.6	35.6
	Edge	1st	89.6	12.8	201.9	0	0
		2nd	88.9	9.3	0	201.9	35.6
3-ply-C CA-C	Corner	1st	97.8	4.8	0	0	0
		2nd	96.5	3.8	0	0	35.6
	Center	1st	94.8	9.9	134.6	134.6	0
		2nd	75.4	23.8	134.6	134.6	35.6
	Edge	1st	96.6	5.8	0	201.9	0
		2nd	56.8	37.1	201.9	0	35.6
3-ply-C MCA	Corner	1st	97.6	6.6	0	0	0
		2nd	94.4	6.7	0	0	35.6
	Center	1st	93.6	10.2	134.6	134.6	0
		2nd	76.0	19.6	134.6	134.6	35.6
	Edge	1st	97.6	6.5	0	201.9	0
		2nd	60.4	36.8	201.9	0	35.6
5-ply-P CA-C	Corner	1st	100	0	0	0	0
		2nd	99.5	1.1	0	0	35.6
		3rd	100	0	0	0	71.1
	Center	1st	98.3	2.7	134.6	134.6	0
		2nd	93.3	13.6	134.6	134.6	35.6
		3rd	83.9	10.7	134.6	134.6	71.1
	Edge	1st	83.8	13.4	201.9	0	0
		2nd	98.4	3.5	0	201.9	35.6
		3rd	40.8	39.4	201.9	0	71.1
5-ply-P MCA	Corner	1st	99.6	0.7	0	0	0
		2nd	82.7	17.8	0	0	35.6
		3rd	98.8	2.5	0	0	71.1
	Center	1st	96.8	3.8	134.6	134.6	0
		2nd	71.9	28.2	134.6	134.6	35.6
		3rd	87.8	9.7	134.6	134.6	71.1
	Edge	1st	94.5	7.9	201.9	0	0
		2nd	85.1	25.0	0	201.9	35.6
		3rd	60.8	36.5	201.9	0	71.1

According to the statistical analysis, there was no significant difference in the Cu penetration of 3-ply CA-C and MCA treated panels for both layups (**Figure** 3.4). However, 3-ply-P CA-C panels were slightly better than 3-ply-P MCA panels as illustrated in **Figure** 3.4. For CA-C treated panels, the 2nd layer center specimen had higher Cu penetration and was statistically superior to MCA-treated panels. The same observation was noted in 5-ply-P (CA-C and MCA) treated panels, where CA-C outperformed MCA-treated panels. Although the Cu penetration of 5-ply-P CA-C and MCA panels were statistically similar, there was an increased Cu penetration in CA-C compared to MCA-treated panels. As an example, the Cu penetration of 2nd layer in 5-ply CA-C treated panels was statistically better than that of MCA-treated panels (at all panel locations) (**Figure** 3.5). Based on these results, it can be concluded that CA-C treated panels had better Cu impregnation in both 3- and 5-ply CLT in comparison to MCA-treated panels. This agrees with other reports where CA-C treated specimens had better preservative impregnation compared to MCA-treated specimens [20, 35]. As earlier discussed, the higher Cu penetration in CA-C treated panels could result from high solubility of the preservative which resulted in better impregnation in the treated CLTs. Therefore, the use of CA-C is recommended in the preservative treatment of prefabricated CLT panels.

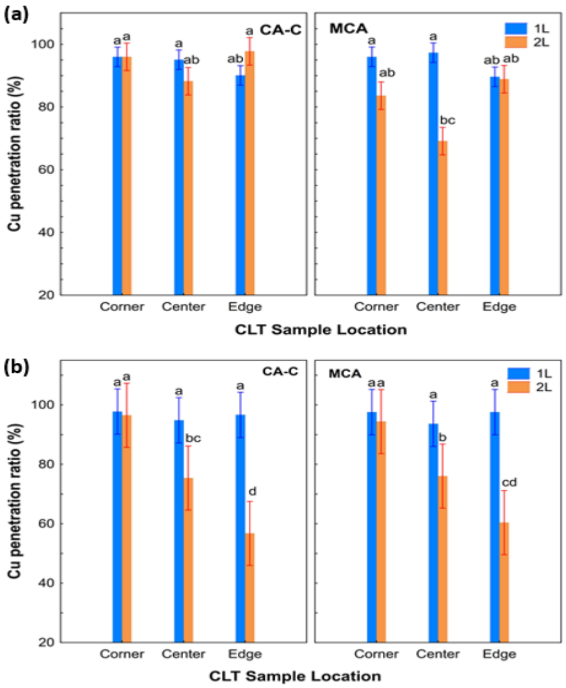

Figure 3.4 Preservative penetration ratio for (a) 3-ply-P CA-C and MCA b) 3-ply-C CA-C and MCA (bars represent standard error; different letters above bars indicate significant differences at $P < 0.05$. Dunn's multiple comparison test was used to compare the Cu penetration of (a) and (b).

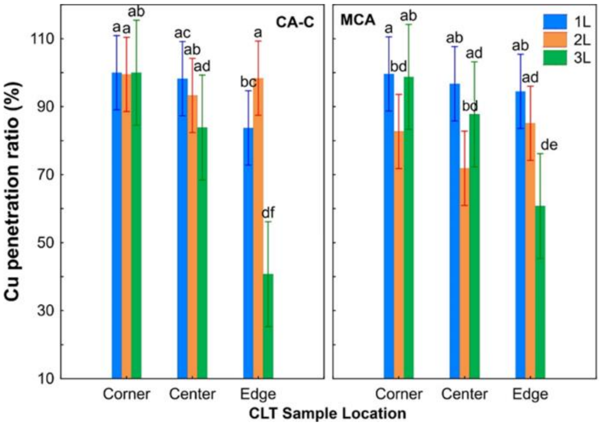

Figure 3.5 Preservative penetration ratio for 5-ply-P CA-C and MCA (bars represent standard error; different letters above bars indicate significant differences at $P < 0.05$. Dunn's multiple comparison test was used to compare the Cu penetration of 5-ply-P CA-C and MCA panels).

3.7.1.1 Effect of panel layup on the preservative penetration in the treated CLTs

The effect of panel layup that is, lengthwise and crosswise orientations on Cu penetration was elucidated for the 3-layer CLT panels. First, we observed a similarity in the Cu penetration ratio of both layups in CA-C and MCA-treated panels. For instance, the Cu penetration ratios of 95.1% and 94.8% were recorded for 1st layers of CA-C-P and CA-C-C, respectively. To elaborate, although the penetration along the L_L of the 1st layer is shorter in 3-ply-C compared to 3-ply-P, the influence of the L_L on the Cu penetration ratio was not significant. This was also confirmed by the Kruskal Wallis H test, where no significant difference was detected in the Cu penetration ratio of 3-ply-P and 3-ply-C in CA-C treated panels (**Figure** 3.6). While there was no statistical significance between the two layups used for CA-C panel treatment, 3-ply-P displayed slightly better impregnation than 3-ply-C, particularly for the 2nd layer of the panels (**Figure**. 3.6).

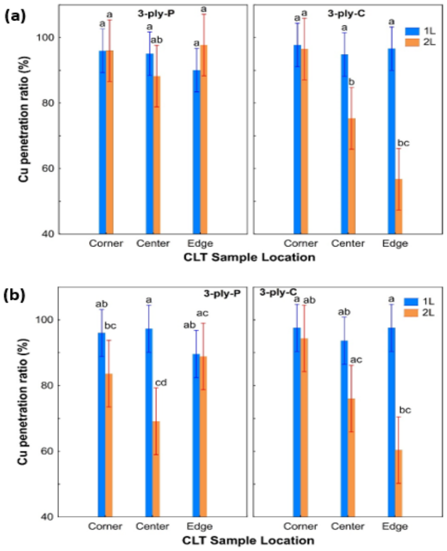

Figure 3.6 Preservative penetration ratio for 3-ply-P and 3-ply-C in (a) CA-C (b) MCA (bars represent standard error; different letters above bars indicate significant differences at $P < 0.05$. Dunn's multiple comparison test was used to compare the Cu penetration of 3-ply-P and 3-ply-C in (a) and (b).

Moreover, there was a decline in the Cu penetration of 1st layer edge specimen in 3-ply-P CA-C panels. This could be due to the long diffusion along the L_L of the treated specimens. The influence of L_L on Cu penetration was also noted for 2nd layer edge specimen where there was a decrease in the penetration ratio of 3-ply-C compared to 3-ply-P panels (**Figure**. 3.6). These results indicate that the penetration along the L_L has more influence on the Cu penetration ratio compared to that in the L_R direction. Craniun *et al*. [19] and Rasouli *et al*. [36] found that the penetration of preservatives was five to ten times better in the L_L than in the L_R of the treated specimens.

While we found high Cu penetration in 3-ply-P of MCA-treated CLT, the 2nd layer corner and center specimens were slightly better in 3-ply-C compared to 3-ply-P panels (**Figure**. 3.6). A possible explanation for the slight improvement in the Cu penetration of 3-ply-C is the better treatability of MCA-treated specimens in the tangential direction of the wood. Overall, 3-ply-P panels had Cu penetration mostly above 80.0% at all panel locations for the 1st and 2nd layers as opposed to 3-ply-C with penetration values as low as 56.8%. In addition, the Kruskal Wallis H test revealed that Cu penetration in 3-ply-P was significantly greater than that in 3-ply-C panels, especially in CA-C treated panels. Thus, the lengthwise orientation might be desirable in fabricating post-preservative treated CLT panels due to the anticipated complete panel protection.

3.7.1.2 Effect of panel thickness on the preservative penetration in the treated CLTs

The penetration depth of preservatives is deemed important in determining the efficacy of wood treatment [18]. To investigate the effect of panel thickness on preservative impregnation in CLT, the same number of observations was obtained for the 2nd layer specimens in 3- and 5-layer CLT. For instance, eight observations were each made at the 2nd layer of 3-ply-P and 5-ply-

P of CA-C treated panels. Therefore, we compared the Cu penetration of the 2nd layer in 3-ply-P and 5-ply-P panels to elucidate the effect of panel thickness (number of layers) on Cu impregnation. For instance, the Cu penetration of 2nd layer at the corner, center, and edge were 96.0, 88.2, and 97.8% in 3-ply-P CA-C treated panels, respectively. And they were 99.5, 93.3 and 98.4% in 5-ply-P CA-C treated panels, respectively. Similarly, the penetration ratios of the 2nd layer corner, center, and edge specimens were 83.6, 69.1, and 88.9% and 82.7, 71.9, and 85.1% in 3- and 5-ply MCA-treated panels, respectively. In 3-ply-P, the 2nd layer was sandwiched between top layers that are almost fully impregnated while the 2nd layer is in between the top layers (with over 90% penetration ratio) and 3rd layer with less penetration ratio (40%) in 5-ply-P panels. However, similar Cu penetration ratio was observed in 2nd layer of 3-ply-P and 5-ply-P CA-C and MCA-treated panels as supported by the statistical analysis (**Figure** 3.7).

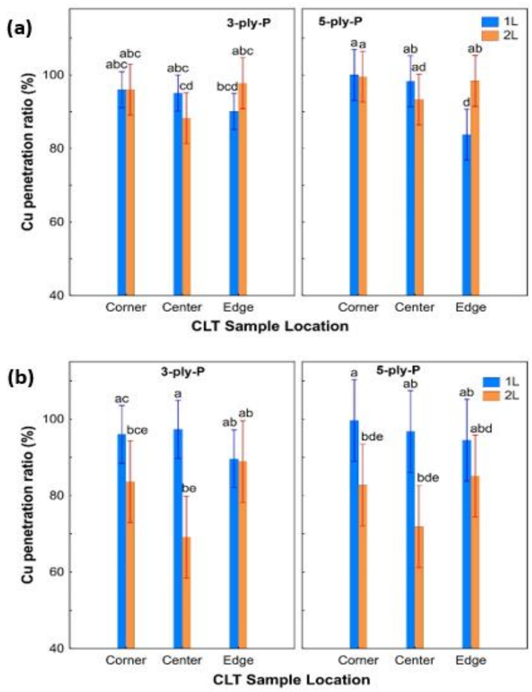

Figure 3.7 Preservative penetration ratio of 2nd layer in 3-ply-P and 5-ply-P in (a) CA-C (b) MCA (bars represent standard error; different letters above bars indicate significant differences at $P < 0.05$. Dunn's multiple comparison test was used to compare the Cu penetration of 3-ply-P and 5-ply-P in (a) and (b).

A similar observation was noted for the 2nd layer penetration of 3-ply-C and 3rd layer of 5-ply-P panels. To illustrate, the Cu penetration at the corner, center, and edge of 3-ply-C were 94.4, 76.0, and 60.4%, respectively, which are similar to that of 5-ply-P of MCA-panels (i.e., 98.8, 87.8, and 60.8%, respectively). The 2nd layer (middle layer) of 3-ply-C showed a Cu penetration ratio over 90%, which is similar to the 3rd layer (middle layer) of 5-ply-P that has almost complete impregnation. However, the 2nd layer of 5-ply-P exhibited less penetration ratio than its 3rd layer in MCA-treated panels, no matter the sample location, for instance, the corner (**Figure** 3.8, left) or the center (**Figure** 3.8, right) of the CLT panel.

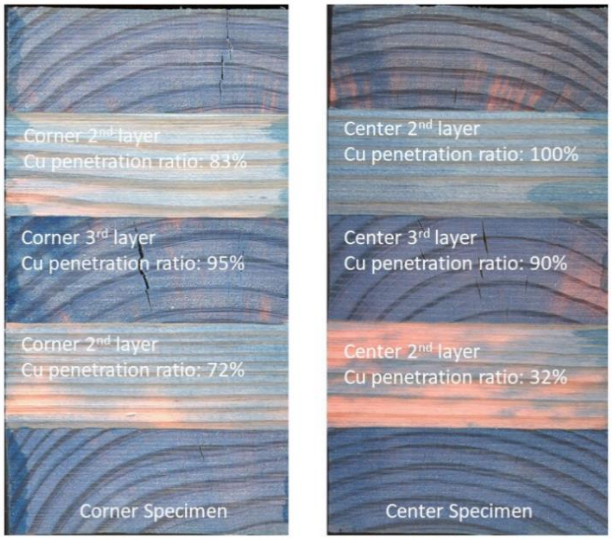

Figure 3.8 Photographs of 5-ply-P MCA-treated panel show that there was higher Cu penetration in 3rd layer compared to 2nd layer for both corner (left) and center (right) specimens.

Moreover, we found no statistical difference in the Cu penetration of 2nd layer of 3-ply-C and 3rd layer of 5-ply-P in CA-C treated panels (**Figure** 3.11). Also, no difference was detected in the Cu penetration of 2nd layer in 3-ply-P and 3rd layer in 5-ply-P of MCA-treated panels (**Figure** 3.12). These observations indicate that preservative penetration in CLT can be predicted based on L_L and L_T values. Also, the results suggest that the Cu diffusion in CLT panel's thickness direction across adjacent layers is prohibited (because of the glue layer), and this diffusion can be disregarded in the preservative treatment of lumber-based wood composites [37].

3.7.2 Preservative retention in the treated CLTs

Cu retention, expressed as kg/m^3, which is the amount of preservative solution retained by wood after treatment is an indicator of impregnation quality in treated wood products [38, 39] and thus utilized to evaluate the Cu retention of the CLT panels. For the Cu retention evaluation, we sampled 48 specimens (6 locations × 2 CLT layers × 2 layups × 2 preservatives) in 3-ply CLT while we collected 36 specimens (6 locations × 3 CLT layers × 2 preservatives) in 5-ply CLT, for a total of 84 specimens. The influence of the CLT sample location (corner, center, and edge) on Cu retention was also investigated. Overall, the panel corner has the highest Cu retention values for the 1st and 2nd layers in 3- and 5-ply CA-C treated panels. Specifically, the Cu retention values of 2.29 and 2.26 kg/m^3 were recorded for 1st layer at the corner and center of 3-ply-P CA-C panels (**Table** 3.3). Similarly, the 1st layer corner and center specimens have Cu retention values of 2.80 and 2.48 kg/m^3, respectively, in 5-ply-P CA-C panels (**Table** 3.3). This is expected because the corner specimens have more surfaces directly exposed to preservatives than center specimens upon treatment, thus more preservatives can be diffused and fixed in the wood cells. Moreover, we found a decline in the Cu retention value of 2nd layer center specimen in 3- and 5-ply CA-C treated panels. The low Cu retention at the center is not surprising since no surface was directly exposed to preservatives. In contrast to CA-C treated panels, the corner has the lowest Cu retention for 1st layer in 3- and 5-ply MCA-treated panels. For instance, the Cu retention of 1st layer at the corner, center, and edge were 3.20, 3.52, and 3.26 kg/m^{3,} respectively, in 3-ply-P, And they were 2.51, 2.90, and 2.99 kg/m^3, respectively, in 5-ply-P panels (**Table** 3.3). The low Cu concentration at the panel corner is attributable to the slow fixation of MCA (resulting from its low solubility); which might have affected the uptake of preservatives in the CLT panels [40, 41].

Table 3.3 Cu retention (kg/m^3) in the treated 3- and 5-ply CLT panels.

Location	Layer	3-ply-P CA-C	3-ply-C CA-C	3-ply-P MCA	3-ply-C MCA-CS	5-ply-P CA-C	5-ply-P MCA
Corner	1st	2.29	2.84	3.20	2.94	2.80	2.51
	2nd	2.90	2.14	2.52	1.62	1.95	1.37
	3rd	-	-	-	-	2.43	2.03
Center	1st	2.26	2.34	3.52	2.38	2.48	2.90
	2nd	2.35	1.63	0.59	2.32	1.58	1.03
	3rd	-	-	-	-	1.22	0.82
Edge	1st	2.28	2.64	3.26	3.16	2.15	2.99
	2nd	2.79	0.61	2.11	0.63	1.97	1.70
	3rd	-	-	-	-	2.39	0.98

N.B: According to the AWPA UI-18 standard, the minimum retention value specified for wood used in ground contact or fresh-water applications is 2.4 kg/m^3 [32].

Further, there was higher Cu retention for 1st layer compared to 2nd layer in the 3- and 5-ply CLT panels. This is not unexpected considering that the 1st layers have at least one more surface directly exposed to preservatives compared to the 2nd layers. Another possible explanation is the effect of adhesive layers on the uptake of preservative solutions in glued wood products. Kuka *et al*. [42] found higher Cu concentration for the 1st layer compared to 2nd and 3rd layers of glued pine specimens, which was attributed the filtration effect of the glue-line that hindered the preservative diffusion and resulted in the reduced Cu retention of the 2nd and 3rd layers. Additionally, the tangential-longitudinal plane of 1st layer was directly exposed to preservatives compared to the radial-longitudinal plane exposed in the 2nd layer which could have influenced the Cu retention in the treated CLTs. For instance, we found higher Cu retention for 1st layer of 3-ply-P MCA compared to CA-C panels (**Table** 3.3). A similar trend was also observed in 5-ply panels, especially for the center and edge specimens. The reason for this is still unclear but it could be that MCA treats better in the tangential direction through the radial-longitudinal plane of wood specimens. Thus, more studies should be conducted to better understand this mechanism.

Moreover, the recommended retention value of 2.4 kg/m^3 specified for sawn timber products used in ground contact or freshwater applications was used as a reference for the studied CLT panels. Although the regions away from the CLT panel surfaces (i.e., the 2nd and 3rd layers) showed lower retention levels, the regions near the CLT panel surfaces (the 1st layer, which has higher biodeterioration risks when panels are used for ground contacts) achieved retention values greater than 2.4 kg/m^3. Thus, we expect that the treated CLT panels are durable when used in water or soil-contacted environments. Nevertheless, further studies will be needed to evaluate the durability performance of these treated CLT panels.

3.7.2.1 Effect of panel layup on the preservative retention in the treated CLTs

The influence of panel layup on the preservative retention in CLT was also investigated. We found that the Cu retention of 1st layer was higher in 3-ply-C compared to 3-ply-P of CA-C treated panels. For instance, the Cu retentions of 1st layer corner specimens were 2.84 kg/m^3 and 2.29 kg/m^3 in 3-ply-C CA-C and 3-ply-P CA-C, respectively. A similar trend was observed at the center and the edge of the 3-ply CA-C treated panels. The better Cu retention in 3-ply-C is probably due to the shorter L_L which could have contributed to more uptake of preservatives. This agrees with other findings where a reduction in Cu retention was noted with increasing L_L in treated wood specimens [43]. The effect of L_L on Cu retention was also evident for the 2nd layer where there was an increased Cu retention in 3-ply-P than 3-ply-C of CA-C panels. Contrastingly, the Cu retention of the 1st layer was higher in 3-ply-P than 3-ply-C of MCA at all panel locations. For instance, the Cu retention of 1st layer corner specimen of 3-ply-P MCA was

3.20 kg/m^3 compared to 2.94 kg/m^3 in 3-ply-C MCA panels. Likewise, the center specimens have Cu retention values of 3.52 kg/m^3 and 2.38 kg/m^3 in 3-ply-P MCA and 3-ply-C MCA, respectively.

Besides, the highest Cu retention for the 1st and 2nd layers were obtained at the center of 3-ply-P and 3-ply-C, respectively, in MCA-treated panels. These observations revealed that the preservative retention increased with increasing diffusion length in MCA-treated panels and might be linked to the preservative chemistry of micronized Cu formulations. Additionally, the Kruskal Wallis test and ANOVA were implemented to evaluate the effect of panel layup on Cu retention in CA-C and MCA panels, respectively. While the statistical analysis revealed no significant difference between the layup patterns ($p = 0.337$) for CA-C and ($p = 0.553$) for MCA as shown in **Table** 3.4 in supporting information. The Cu retention in 3-ply-P was generally better than 3-ply-C, especially for 1st and 2nd layers (**Table** 3.3). Thus, the lengthwise layup is recommended in the fabrication of preservative-treated CLT, especially when adequate preservative retention is desired. However, more studies should be conducted to evaluate the biodeterioration resistance of post-treated commercial-sized CLT panels with the lengthwise layup that favors preservative treatment based on the penetration depth and retention.

3.7.2.2 Effect of panel thickness on the preservative retention in the treated CLTs

The influence of panel thickness on the Cu retention gradient in the treated panels was also investigated. We compared the Cu retention values of the 1st and 2nd layers in 3- and 5-ply (CA-C and MCA) treated panels. In 3-ply CA-C panels, there was adequate preservative retention in the 1st and 2nd layers of the panels. To illustrate, the Cu retention values of 2.29 kg/m^3 and 2.90 kg/m^3 at the corner and 2.28 kg/m^3 and 2.79 kg/m^3 at the edge were recorded for the 1st and 2nd layers of 3-ply-P CA-C panels. Also, the Cu retention of the 1st and 2nd layers were

2.80 kg/m^3 and 1.95 kg/m^3 and 2.15 kg/m^3 and 1.97 kg/m^3 at the corner and edge of 5-ply-P CA-C panels. While there was a similarity in the Cu retention of the 1st layer in 3- and 5-ply CLT, we noticed a declining trend in the Cu retention of the 2nd layer in 5-ply-P compared to 3-ply-P panels. To illustrate, there was higher Cu retention at the 2nd (core, 2.90 kg/m^3) layer of 3-ply-P CA-C panels compared to the 2nd layer (equivalent to the core layer in 3-ply-P, 1.95 kg/m^3) of 5-ply-P CA-C panels. A similar trend was observed in MCA-treated panels. It could be that the panel thickness influenced the preservative uptake of the 2nd layer (assuming Cu retention occurred thickness-wise) and thus affects the Cu retention of the 2nd layer in 5-ply-P CLT panels. Although the statistical analysis showed no effect of panel thickness on preservative retention ($p = 0.150$ and 0.262 in CA-C and MCA panels, respectively (**Table** 3.5). The Cu retention trend of the 2nd layer in 3- and 5-ply CA-C and MCA-treated panels indicates that the panel thickness might be an influencing factor in the Cu retention of CLT.

3.8 Conclusions

The presented findings show the feasibility of successfully treating prefabricated cross-laminated timber panels with copper-azole preservatives. In this study, CA-C and MCA-treated CLT panels fabricated with different layups (i.e., lengthwise and crosswise) and layers (i.e., 3- and 5) were tested for penetration and retention of preservatives. Based on the investigations, the main conclusions from this study are:

- ❖ The Cu penetration in the treated 3- and 5-ply CLT panels was mostly above 85% for all lumber, especially for the 1st layer lumber (e.g., ≥90% was observed), thereby meeting the penetration requirement specified in AWPA T1-16 standard for post-treated wood composites.

- Lumber layup influenced the Cu penetration and retention in the treated CLT panels. The lengthwise layup had better treatability for the 1st and 2nd layers and thus recommended in the fabrication of preservative-treated CLT.
- CA-C exhibited better treatability than MCA for CLT panels, resulting in higher Cu penetration and retention across the CLT panels. Thus, CA-C preservative is suggested for the treatment of prefabricated CLT panels.
- The penetration along the longitudinal (L_L) direction and number of surfaces directly exposed to preservatives influenced the Cu penetration. Cu penetrations in laminations with no direct exposure to preservatives rely on the L_L values. Thus, the L_L values of the inner (core) layers of CLT should be minimized for better treatment.

- While the Cu retention of the 1st layer was above the recommended value for sawn lumber used in ground contact or freshwater applications, the low retention values of the 2nd layer could improve when their L_L values are minimized.

3.9 Acknowledgements

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3.10 Supporting Information (SI)

Table 3.4 Statistical analysis for the effect of panel layup on preservative retention

	Preservative	
	CA-C	MCA
Levene’s test		
$F_{(1,10)}$ value	3.484	0.075
p-value	0.092	0.790
One-way ANOVA (retention)		
F value	-	0.377
p-value	-	0.553^{ns}
Kruskal-Wallis H test (retention)		
test statistic	-	12.0
p-value	-	0.337^{ns}

Note: ns Not significant at $p > 0.05$

* Result is significant ($p < 0.05$)

Table 3.5 Statistical analysis for the effect of panel thickness on preservative retention

	Preservative	
	CA-C	MCA
Levene’s test		
$F_{(1,10)}$ value	0.523	0.071
p-value	0.486	0.795
Kruskal-Wallis H test		
test statistic	9.0	11.0
p-value	0.150^{ns}	0.262^{ns}

Note: ns Not significant at $p > 0.05$
* Result is significant ($p < 0.05$)

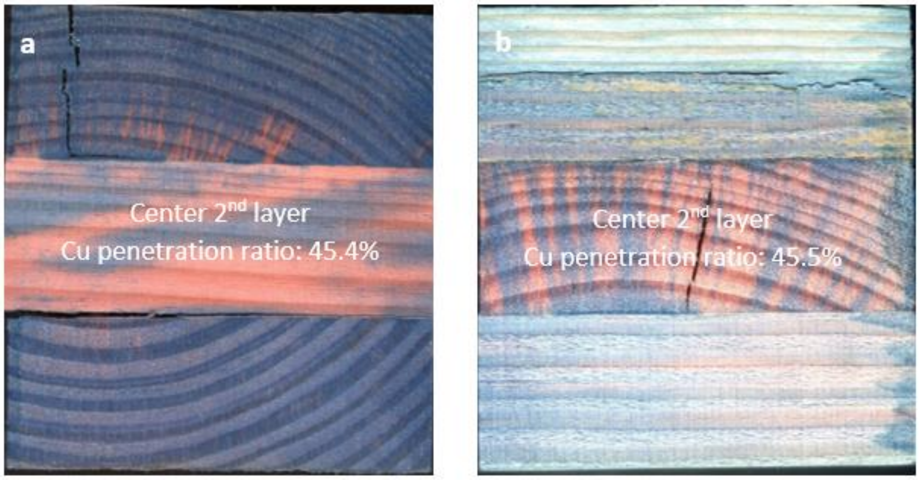

Figure 3.9 A typical illustration of 2nd layer penetration from the side (a) and end-grain (b) of 3-ply-P MCA treated panel.

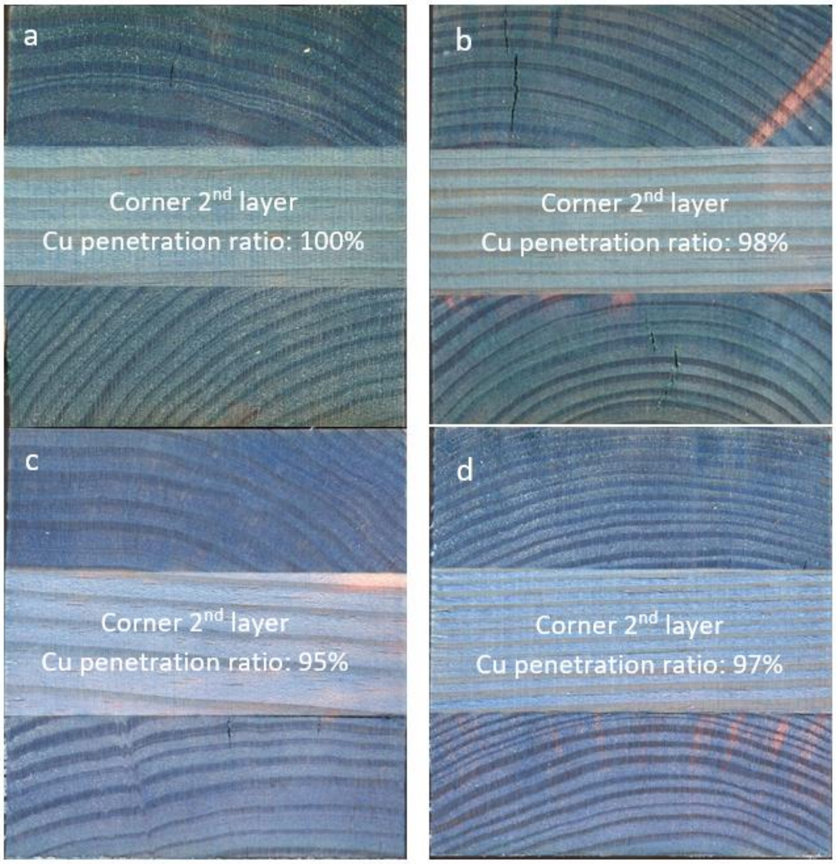

Figure 3.10 A typical illustration of 2nd layer penetration of specimens with wide and narrow annual rings in (a, b) for 3-ply-C CA-C and (c, d) for 3-ply-C MCA treated panel.

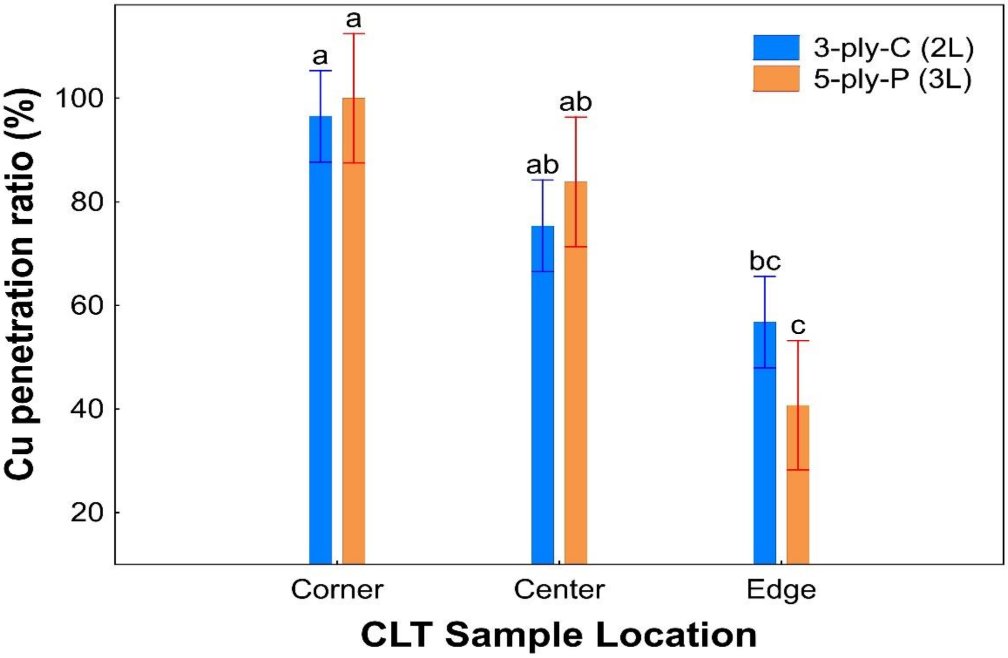

Figure 3.11 Penetration ratio of 2nd layer of 3-ply-C and 3rd layer of 5-ply-P in CA-C treated panels.

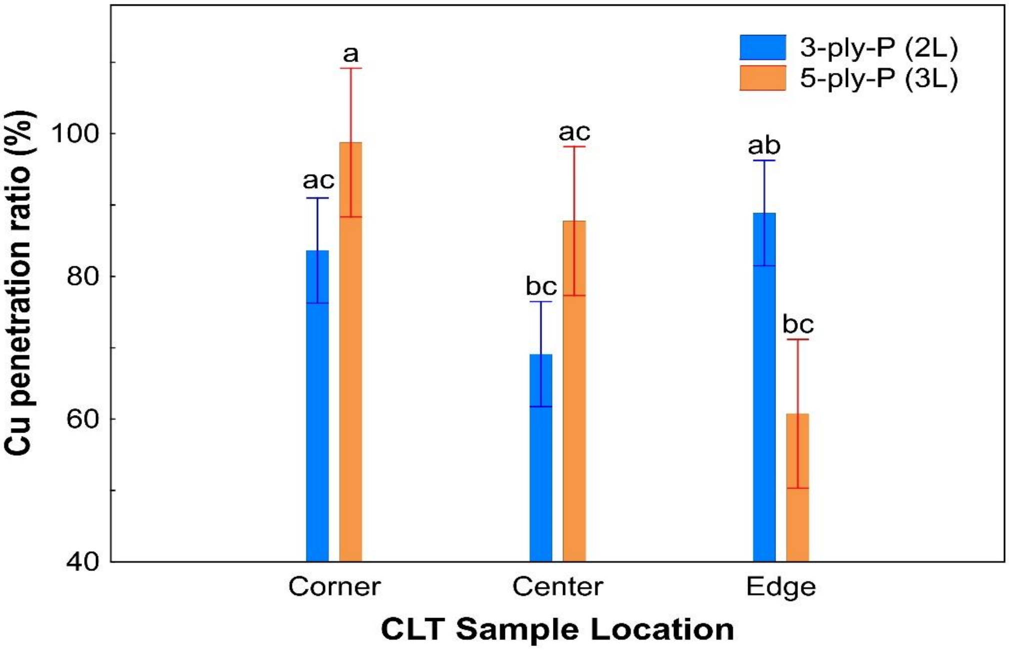

Figure 3.12 Penetration ratio of 2nd layer of 3-ply-P and 3rd layer of 5-ply-P in MCA-treated panels.

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CHAPTER IV

DEVELOPMENT OF LIGNIN-REINFORCED POLYURETHANE ADHESIVE FOR GLUED LAMINATED TIMBER

Ayanleye, S., Quin, F., Zhang, X., & Shmulsky, R. (2023). Development of lignin-reinforced polyurethane adhesive for glued laminated timber. (Submitted to Journal of Adhesion Science and Technology).

4.1 Abstract

Glue-laminated timber (Glulam), a structural engineered wood product, is mostly manufactured using structural adhesives including one-component polyurethane (PUR). Nevertheless, the concerns associated with PUR such as unsatisfactory gap-filling properties and lower resistance to delamination create opportunities for newer adhesives with enhanced bonding performance in outdoor environments. In this study, the use of fractionated lignin as a modifier in a polyurethane-based adhesive system was explored. Herein, the effect of lignin content (1, 2, and 3%) on the block shear strength (BSS), wood failure percentage (WFP), and delamination were evaluated. The results indicated that all the lignin-reinforced PUR specimens showed better adhesion performance compared to the controls (without lignin). Markedly, the lignin-PUR adhesive formulation containing 1% lignin addition exhibited superior adhesion properties than those with 2% and 3% lignin content. The statistical analyses also revealed that the lignin content influenced the BSS and WFP of the glulam specimens. Moreover, the lignin-modified PUR specimens showed increased delamination resistance and met the requirements for delamination stipulated in the ASTM D2559 standard, regardless of the lignin content. The lowest percent delamination (0.07%) was obtained from specimens bonded with 1% lignin. The presented data suggest that specimens bonded with 1% of lignin provided better bonding strength compared to other lignin-filled specimens. Thus, this study demonstrated the technical feasibility of fabricating glulam with enhanced adhesion performance using lignin-modified PUR adhesive.

Keywords: Glue-laminated timber, Polyurethane adhesive, Fractionated lignin, Block shear strength, Delamination

4.2 Introduction

The annual global market for wood adhesives is rapidly growing and is projected to be worth about USD 21 billion by 2024. This growth is due to the high demand for wood-based composites and advancement in gluing technologies [1]. In the past decades, several adhesives including, melamine-formaldehyde adhesive (MF), phenol-resorcinol formaldehyde (PRF), and urea-formaldehyde adhesive (UF) have been used for gluing wood products [2]. However, the formaldehyde emissions associated with these adhesives and the resulting environmental impacts led to the search for environment-friendly adhesive systems such as polyurethane adhesive (PUR) [1, 3]. PUR adhesives are synthesized from the reaction between isocyanate groups and polyol, forming urethane linkage and contributing to enhanced bond strength and durability of the laminated products [4]. The advantages of PUR adhesives such as improved strength, little or no formaldehyde emission, fast cure, and high moisture tolerance make them a suitable choice in the fabrication of engineered wood products [5-7]. Despite the advantages of PUR, the sources of polyol used in its production are mostly petroleum-based which is not sustainable. Amongst the renewable sources of polyol (starch, polysaccharides, lignin, etc.), lignin is a promising alternative for fossil-based polyols in the formulation of PUR adhesives because of its high hydroxyl content [3].

Lignin is one of the most abundant aromatic biopolymers on earth. It is found in woody biomass and other agricultural residues. It is relatively inexpensive and has versatile properties with approximately 150 billion tons extracted annually [8]. While a large amount of lignin is produced every year, approximately 5% is utilized in the production of value-added products such as resins, adhesives, and other binders [9, 10]. Recently, lignin has been incorporated into PUR adhesive matrix for adhesive formula modification [4]. However, technical lignin suffers from structural heterogeneity, making it less compatible with conventional petroleum-based resins [11]. Several studies have employed a fractionation method to obtain more homogeneous lignin with better compatibility with commercial resins and used it for polymer synthesis [12 - 15]. Fractionation is done using an organic solvent such as ethyl acetate, ethanol, methanol, and acetone to obtain lignin with specific properties (e.g. molecular weight distribution and improved polydispersity) that can be incorporated in PUR for improved bonding quality [15 - 18]. Thus, the development of lignin-reinforced PUR adhesive with improved properties would be useful in the fabrication of mass timber (MT) such as cross-laminated timber (CLT) and glue-laminated timber (Glulam). Glulam is a popular MT made from gluing sawn lumber with the grains running parallel to each lamination using structural adhesive. The benefits of glulam compared to conventional lumber includes larger size, improved strength properties, and renewability, making it suitable for tall wood buildings [19]. As a result of the importance of glulam in load-bearing applications, the bonding quality has been deemed crucial to ensure the structural integrity and safe use of the final products [20].

Moreover, the use of lignin as a modifier or substitute agent for PUR adhesives has been reported to increase the overall bonding performance of glued wood specimens [4, 21]. For instance, Garcia *et al*. [21] reported up to 50% increase in the shear strength of lignin-reinforced one-component PUR (1C-PUR) for bonding beech wood specimens. They attribute the enhanced bonding performance to the increased glue line delamination resistance as a result of lignin addition. Also, the addition of hydrophobic lignin particles seems to improve the water-resistance of the glue layer, thereby decreasing the water-induced adhesion failure. This finding is similar to other reports with improved shear strength, thermal stability, and delamination of glued wood specimens utilizing lignin-modified adhesives [3, 6, 22]. Therefore, the use of lignin as the reinforcement material for PUR adhesive could be a solution for improving the bonding strength of glulam used in structural applications. Although there are some studies on the bonding of wood using 1C-PUR; there are no studies that have investigated the effect of lignin-reinforced PUR on the gluing performance of southern yellow pine (SYP) lumber, which is the focus of this study. Thus, the overarching goal of this study was to investigate the effect of ethanol-extracted lignin (at different wt%) in PUR adhesive formulation by assessing the adhesion performance of the glued laminated timber specimens.

4.3 Materials and Methods

4.3.1 Materials

Visually graded No.2 SYP 2 × 6 (38 mm × 140 mm) lumber (2.4 m long) was procured from a local vendor. Defect-free and flat-grained lumber pieces (shook) were cross-cut to minimize the effect of lumber defects. The lumber with pith and with compression wood were not included in this study. The selection of the SYP lumber specimens was done in accordance with ASTM D2559 standard [23]. All the pieces of shook were randomized to spread the variability among the treatment variables and replications. Then, they were sorted by weight and separated into two classes, control and treatment. Each class received the full assortment of weight-sorted stock. The moisture content (MC) of the SYP lumber was taken with an electronic moisture meter upon arrival at the testing lab. The lumber was then stored indoors for approximately four weeks before glulam manufacturing. Commercial PUR adhesive (PUR HB X602) and primer (LOCTITE PR 3105) were supplied by Henkel Corporation (Rocky Hill, Connecticut, USA). The primer was mixed with tap water in a ratio of 1:9 (10% and 90% volume of primer and water, respectively).

4.3.2 Preparation of Ethanol-extracted Lignin

Lignin fractionation was performed in a 2000 mL beaker containing 70g of Biochoice^{TM} and 500 mL of ethanol, with continuous stirring at room temperature for 2 - 3 hours. The soluble fraction was extracted through rotary evaporation and dried at 100 ℃ for 24 h to obtain oven-dried ethanol-extracted lignin. The insoluble fraction was then captured through filtration (filter paper pore size 15-20 μm). The evaporated ethanol was recovered and utilized for the next cycle of lignin fractionation; each cycle was recycling approximately 50% of the ethanol. Ethanol-extracted lignin was used to make lignin-reinforced PUR adhesive and was then immediately applied to the test pieces. Four sets of adhesives were prepared which includes the commercial PUR without lignin (control), PU-1 (0.1 g of Lignin + 4 mL of acetone + 10 g of primer), PU-2 (0.2 g of Lignin + 4 mL of acetone + 10 g of primer) and PU-3 (0.3 g of Lignin + 4 mL of acetone + 10 g of primer). To ensure good dispersion of lignin in PUR, the ethanol-extracted lignin was first dissolved in 4 mL of acetone, followed by direct addition of 10 g of wood primer (LOCTITE PR 3105) into the solution with mechanical blending. Then, the primer was applied to the test pieces before applying the PUR adhesive.

4.3.3 Glulam Fabrication

Prior to Glulam assembly, SYP lumber was moisture conditioned to an MC of 12 ± 3% per ASTM D2559 standard [23]. The SYP lumber was then sorted based on the weights using an electronic floor scale. Only the lumber with the weight of 6 - 7 kg was used in the manufacturing of glulam, this was done to eliminate the influence of lumber density on glulam's properties (e.g. bonding strength). The SYP boards were sawn to 19 mm (radial, thickness) × 134.6 mm (tangential, width) × 304 mm (length, longitudinal) dimensions and conditioned at 20 ± 2 ℃ and 65% relative humidity to an MC of 12 ± 3% after board preparation and before bonding. The MC and specific gravity (SG_{EMC}) of the laminations (**Table** 4.1) were determined following the ASTM D2395 and ASTM D4442 standards [24, 25].

Table 4.1 Summary statistics of MCs and SGs of lumber used in Glulam fabrication

		MC (%)			SG_{EMC}		
		Mean	SD[a]	COV[b]	Mean	SD	COV
2-layer Glulam	Control	12.99	0.70	5.41	0.55	0.03	6.31
	PU-1	13.53	0.27	2.03	0.52	0.03	6.29
	PU-2	12.45	0.49	3.94	0.47	0.03	5.89
	PU-3	13.96	0.86	6.15	0.46	0.02	5.32

Note: [a]SD means standard deviation, [b]COV means coefficient of variation.

In this study, 2-layer glued blocks of 38 mm (radial, thickness) × 134.6 mm (tangential, width) × 304 mm (length, longitudinal) were produced by face gluing two pieces of lumber parallel to each other using a laboratory hydraulic press (Dieffenbacher North America, Inc) under a clamping pressure of 100 psi (0.69 MPa) for 3 h (**Figure** 4.1). Prior to lamination, SYP lumber were separated into two groups, namely A and B. The lumber in group A were used for the 1st lamination while those in group B were used for the 2nd lamination. The laminations were then used to manufacture glulam within 8 h of planing. According to the adhesive manufacturer's instructions, the wood primer was sprayed (spreading rate of 20 g/m^2) on the specimens' surface and allowed to cure for 10 mins before applying the PUR and lignin-reinforced PUR adhesive at a spreading rate of 180 g/m^2 [26]. Five glulam replicates each was produced for the control and lignin-reinforced samples (1, 2, and 3%) for a total of 20 adhesive-bonded glulam samples. Then, the laminated blocks were conditioned at 65 ± 5% RH and 20 ± 2 °C for approximately 2 weeks before cutting into shear block and delamination specimens.

Figure 4.1 Manufacturing of glulam using a laboratory hydraulic press

4.3.4 Block Shear and Wood Failure Percentage (WFP) Tests

For the bond evaluation tests, forty-four (44) SYP laminates (19 mm × 134.6 mm × 304 mm) were glued together to produce 22 (2-layer) adhesive-bonded samples. Then, eight (8) test joints were cut into 40 specimens with dimensions of 38 mm × 50.8 mm × 50.8 mm for the block shear strength (BSS) test. For the delamination test, three 38 mm × 127 mm × 76 mm per test joint were cut from 14 glued blocks, thus creating a population of 40 specimens (**Figure** 4.2). The sample preparation and tests for the block shear and delamination were done following ASTM D905 and ASTM D2559 standards [23, 27]. 10 replicates were prepared for each lignin loading (i.e., 1, 2, and 3 %) and 10 control samples bonded with commercial PUR for the block shear and delamination tests. A total of 80 specimens were obtained from the glued assemblies for the bond evaluation tests.

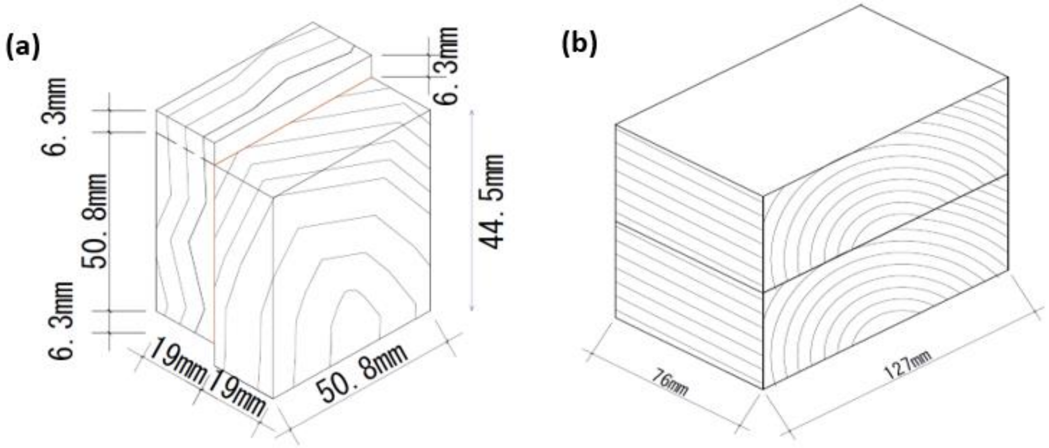

Figure 4.2 Schematics of the block shear (a) and delamination (b) specimens.

For the block shear test, 10 replicates each for control and three types of adhesive treatments were manufactured and tested. In accordance with ASTM D905-08, each block shear specimen was further processed by removing ~6.3 mm at adjacent ends in the longitudinal direction of the specimen [27]; thus, the actual dimension of bond line was 38 mm × 50.8 mm. For each condition, 10 stair-stepped glulam specimens with glue-bond areas of 38 mm × 50.8 mm were prepared in accordance with ASTM D2559 Standard, making a total of 10 bond areas (10 specimens × 1 bond area). The block shear specimen was cut so that the grain of the laminations was parallel to the loading direction and then tested using the Tinus Olsen machine (**Figure** 4.3). The block shear strength (BSS) of the individual bond line was calculated using Equation. 1.

$$\tau BS = \frac{\mathrm{F}}{\mathrm{b} \times t} \tag{4.1}$$

where τBS = BSS (MPa), F = failure load (N), b = sample width (mm), and t = sample thickness (mm).

Figure 4.3 Device for the block shear test

For the wood failure percentage (WFP) estimation, the block shear specimens were loaded until complete failure and separation of the 2-layer glued blocks. Thereafter, the sheared area was captured using a Flatbed Scanner (CanoScan LiDE 400) and the images were processed for the mapping and measurements of the failure area using ImageJ software (National Institutes of Health, Bethesda, MD, USA). The WFP was calculated by dividing the wood failure area by the sheared area of the specimen. This approach for WFP estimation was adopted by other researchers [28, 29].

4.3.5 Delamination Test

For the delamination test, 10 specimens each (control and 3 adhesive treatments) with a dimension of 38 mm × 127 mm × 76 mm were prepared, for a total of 40 specimens (**Figure** 4.2). The moisture-conditioned test blocks were weighed immediately to the nearest 1 g following their removal from the conditioning chamber. Prior to the delamination test, the condition of the blocks was noted by capturing at least 3 sides of the specimens using a Flatbed Scanner (CanoScan LiDE 400). All the specimens selected for the test had no visually apparent delamination. The delamination test was performed according to ASTM D2559 [23]. This procedure involves weighing down the specimens in the autoclave such that the end-grain is exposed to water at a temperature of 18 to 27 °C and completely submerged throughout the test period. An initial vacuum of 85 kPa was drawn and held for 5 min. Then, the vacuum was released, and a pressure of 75 ± 2 psi was applied for 1 h after which the pressure was released. The vacuum-pressure cycle was repeated two times making a two-cycle impregnating period of 130 min. Thereafter, the impregnated specimens were placed 51 mm apart in the Blue M Oven set at 65 ± 2 °C until dried to within 15% of the original weight. Once the specimen reached the desired target weight, they were visually inspected for delamination following the ASTM D2559 standard [23]. The delamination of bond lines was measured immediately after the specimens were removed from the oven. Delamination measurements were made by capturing the two sides of the tested specimens representing the end grain with the Flatbed Scanner followed by image analysis using Image J software. Delamination rates of the bond lines for each condition were calculated as the total delaminated bond line length divided by the total tested bond line length.

4.3.6 Fourier-transform infrared spectroscopy (FTIR)

Control and lignin-treated samples were characterized using Spectrum Two Spectrometer (PerkinElmer, Waltham, MA, USA). The FTIR sample preparation is similar to that described earlier (section 2.2). Briefly, 3 sets of lignin treatments (1, 2, and 3%) were prepared by dissolving lignin in acetone before direct addition to the primer. The resulting solution was then characterized using FTIR. The commercial wood primer (LOCTITE PR 3105) without lignin addition was retained as control. The Fourier-transform infrared spectroscopy (FTIR) spectra were recorded in a wavenumber range between 400 and 4000 cm^{-1} (at a resolution of 2 cm^{-1})

and 10 scans per sample. The spectra baselines were corrected by the “data tune-up” function using the Spectrum Quant software [30].

4.4 Statistical Analysis

The mean values and standard deviation were calculated for BSS, WFP, and delamination. Data were tested for normality and homogeneity of variance using the Shapiro-Wilk test and Levene’s test, respectively. If the assumptions were met, a one-way ANOVA was conducted to evaluate the effect of the lignin loading (1, 2, and 3%) on the bonding strength of the adhesive-bonded blocks. In the scenarios where the assumptions were not met, the data were transformed and tested again. If after transformation, the data could not be normalized, then Kruskal-Wallis H test, a non-parametric equivalent of ANOVA was implemented to analyze the significance of the main effects. Also, Dunn’s test for multiple comparisons was used to compare several groups of observations when the effect analyzed was significant. All of the statistical analyses were performed at the 5% significance level and done using IBM SPSS Statistics, Version 28 (IBM Corporation, New York, NY, USA).

4.5 Results and Discussion

4.5.1 Block shear and wood failure percentage tests

The descriptive statistics of the shear strength for the glulam specimens are presented in **Table** 4.2. The BSS values of the control specimens ranged from 1.47 - 8.28 MPa with an average of 6.06 MPa. We found higher BSS values for specimens bonded with lignin-reinforced PUR adhesive compared to the controls. For instance, the mean BSS value for PU-1 was 6.85 MPa with values ranging from 5.66 - 7.79 MPa and the BSS values for PU-2 was between 5.22 - 8.38 MPa with an average of 7.50 MPa. The highest BSS value was achieved with PU-3 with a mean value of 7.63 MPa. Similar BSS values have been reported for softwood glulam by other researchers [31, 32]. Overall, all specimens treated with lignin had greater BSS values irrespective of the wt% of lignin in the PUR adhesive matrix.

Table 4.2 Descriptive statistics of BSS and WFP for the Glulam Specimens

		BSS (Mpa)		WFP (%)		
	Sample Size (n)	Mean	COV	Mean	COV	No. < 75% WFP**
Control	10	6.06 [2.27]	37.50	68.23 [33.23]	48.70	2
PU-1	10	6.85 [0.73]	10.72	77.28 [11.96]	15.47	2
PU-2	10	7.50 [0.95]	12.62	63.46 [17.59]	27.72	7
PU-3	10	7.63 [1.58]	20.69	41.93 [28.08]	66.97	9

* The value in parenthesis is standard deviation; BSS: Block shear strength; WFP: Wood failure percentage; COV: Coefficient of variation ; ** Number of specimens with < 75% WFP

Additionally, the BSS data was compared with the requirements in ASTM D2559 standard. While the shear strength test was conducted in line with ASTM D2559, we obtained slightly lower BSS values than required by the standard. This could be due to the MC variation among the laminates, the MC of laminates in this study was between 12 - 15% compared to the MC of 12% reported in ASTM D2559 [23]. This could also be related to our choice of sorting the lumber by weight and then selecting the lowest weight pieces for the investigation. The highest BSS value was observed in PU-3 which could be attributed to the increased MC of the laminates. This is similar to other findings where MC was found to influence the BSS of glued specimens [33]. The increased BSS in high MC groups may be reasonable since PUR is a moisture-curing adhesive that performs better upon the availability of moisture below the FSP.

Further, the influence of wood properties (such as SG) on the BSS of glulam was investigated. We found that the groups (PU-2 and PU-3) with lower SG (mean = 0.46) had the highest BSS values. The better BSS values reported for low-density specimens might have resulted from the higher porosity and larger cell lumens that facilitate the penetration of adhesive and thus improved the bonding performance of the glued specimens [34-36]. However, assuming similar penetrations, it would seem that higher SG values would yield higher strength values. Statistical analysis also revealed that the effect of MC and SG on BSS was significant ($p < 0.05$), this indicates that the variation in MC and SG could have impacted the BSS of the glulam specimens.

A comprehensive summary of the WFP with the number of specimens that failed to meet the requirement specified in ASTM D2559 is shown (**Table** 4.2). As shown in **Table** 4.2, only PU-1 met the minimum average WFP of 75% specified in the standard. Even though two values in PU-1 fell short of the required WFP value, they were close to meeting the requirement (WFP values of 51.42 & 64.52%). Noteworthily, greater WFP values as high as 95.85% were noted in PU-1 group. Comparable to PU-1, we found moderately high WFP values ranging from 76.13 - 92.16 % with an average of 68.23% for the control specimens. Although some extremely low WFP values (2.81 & 8.88%) were recorded in the control group. The low values might be a result of starved glue joint at the edge of the bonded specimens. While the adhesive was applied uniformly on the specimens’ surface using a pointy nozzle and glue spreader, there might be areas with not enough adhesive which could have affected the bonding strength of the specimens. As reported in **Table** 4.2, the lowest WFP values were found in PU-2 and PU-3 with an average of 63.46 and 41.93%, respectively, while the highest mean WFP value of 77.28% was observed in PU-1 bonded specimens. Thus, it can be concluded that the addition of high amounts of lignin had a significant impact on the WFP of the glulam specimens.

Although a good bonding is usually characterized by a combined high shear strength and wood failure. The observed BSS values in this study showed no clear relationship with the WFP of the glulam specimens. This finding may be due to the complex relationship that exists between wood material characteristics, adhesives and wood-adhesive interactions. For instance, the ductility behavior of PUR under load reduces the stress concentration in the bond line which could lead to a decrease in the wood failure of glued specimens [37]. Similarly, Hass *et al*. [38] and [39] found no correlation between the BSS and WFP of adhesive-bonded specimens. They attributed this anomaly to the failure mechanism in the adhesive bonds. It is possible that the formation of adhesive bridges across the bond line slowed down the failure process, resulting in low WFP values in the bonded specimens. Further, the cohesive strength within the lumber might be greater than that of adhesion strength at the wood-adhesive interface, thus resulting in the decreased WFP values obtained in PU-2 and PU-3 bonded specimens. A better understanding of the adhesion and cohesion failures in PUR-bonded specimens as well as the influence of wood density average and variation on bond line quality will be necessary to explain the discrepancies in the BSS and WFP results of glued specimens.

4.5.2 Delamination test

The results from the delamination test for all scenarios (control, PU-1, PU-2 & PU-3) are summarized in **Table** 4.3. Generally, all the treatment groups (including the controls) had an average delamination rate of less than 1%, thereby meeting the delamination requirement for softwood specified in ASTM D2559 standard [23]. In this study, PU-1 showed the lowest percent delamination at 0.07% which demonstrates its ability to perform better when subjected to internal stresses caused by the differential swelling and shrinkage in wood. The delamination observed for other lignin-treated samples (PU-2 & PU-3) was slightly higher---presenting mean values of 0.14 and 0.38%, respectively (**Table** 4.3). However, the delamination rates were still less than 1% as required by the standard. The results obtained in this study agrees with those reported by Martins *et al*. [40] who reported mean values between 0.1 - 0.4% for some pine glulam bonded with PUR adhesive. Moreover, the results herein are higher than those reported in the literature [32, 41-42]. For instance, some Douglas-fir specimens bonded with PUR experienced severe delamination after a two-cycle boil test [41]. Similar findings were reported by Rajakaruna [42] where over 40% delamination was observed for PUR-bonded radiata pine. As shown in **Table** 4.3, the rate of delamination increased with increasing wt% of lignin, although, the control specimens showed the highest percent delamination (0.71%). This indicates that the wt% of lignin influenced the delamination results, and this will be further discussed in the later sections.

Table 4.3 Summary of delamination test results for the Glulam Specimens

	Bondline	Bondline delam (mm)	Delam rate (%)
Control	A1	181	0.71
	A2	0	0
PU-1	A1	17.98	0.07
	A2	0	0
PU-2	A1	10.01	0.04
	A2	26.00	0.10
PU-3	A1	52.96	0.21
	A2	43.97	0.17

A1 and A2 represent the two surfaces where the delamination measurement was taken; The total length of glue line on both end-grain surfaces of the specimens was 2540 mm.

4.6 Discussion

4.6.1 Effect of lignin addition on the Block shear and WFP of Glulam Specimens

This study demonstrated that the incorporation of lignin in the PUR-adhesive matrix improved the bonding strength of glulam. **Figure** 4.4 shows the FTIR spectra of the control and lignin-reinforced PUR specimens. The spectra of all lignin samples agrees with the typical absorption bands of lignin. The absorption bands detected at 1089 cm^{-1} and 1644 cm^{-1} are attributed to the C-O stretching of secondary alcohols and aliphatic ethers [43] and stretching vibrations of carbonyl groups in lignin [44]. In addition, the lignin-filled specimens show smaller peaks at 1238 cm^{-1} and 1375 cm^{-1}, corresponding to phenolic OH and aliphatic C-H in methyl groups [44, 45]. Markedly, some new peaks which corresponds to urethane-related linkages were detected such as C-N (1218 cm^{-1}) and C═O (1710 cm^{-1}), particularly for PU-2 and PU-3; these peaks probably facilitates the reaction between lignin and PUR adhesive [46, 47]. Generally, the BSS of glulam increased with increasing wt% of lignin as shown in **Table** 4.2. The statistical analysis also showed that the wt% of lignin has a significant influence on the BSS of the specimens (**Figure** 4.5). As can be seen in Fig. 4.5, the mean BSS of PU-3 was significantly greater than that of control ($p = 0.023$) and PU-1 ($p = 0.022$). While the mean BSS value of PU-3 was higher than that of PU-2, no significant difference was detected between the BSS of PU-2 and PU-3 ($p = 0.479$). Overall, all lignin-filled specimens had higher BSS values compared to the controls (**Table** 4.2). The increased BSS observed for lignin-reinforced specimens is attributable to the reinforcement effect of lignin on the shear strength of glulam. It could be that lignin addition enhanced the polymerization reaction between the wood and PUR adhesive resulting in higher cross-link density and thus improved the bonding strength [4]. The FTIR investigation also revealed an increase in intensity of peaks at 3350 - 3364 cm^{-1} for the lignin-reinforced specimens, which can be attributed to the formation of lignin-urethane linkages (**Figure** 4.4) Previous studies also confirmed lignin-related linkages such as N-H between lignin and isocyanate at absorption bands of 3200 - 3400 cm^{-1} [15, 46].

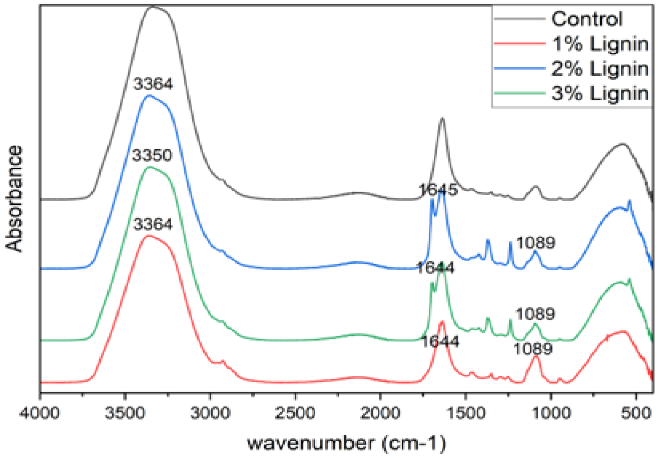

Figure 4.4 FTIR spectra of control (without lignin) and lignin-treated samples

Although the BSS increased with increasing wt% of lignin, the BSS values were slightly lower than required by ASTM D2559 standard. As earlier reported in section 3.1, the variation in MC and SG might have affected the BSS of the specimens. The low BSS values might also be due to the effect of factors such as pressure and PUR adhesive chemistry on the bonding strength. To illustrate, Wang *et al*. [48] reported an increased shear strength for some PUR-glued specimens when the pressure was increased from 0.28 MPa to 0.83 MPa. Similarly, Yusof *et al*. [49] recorded mean BSS value of 4.4 MPa when CLT specimens were glued at 0.9 Mpa. However, the BSS increased to 6.6 and 8.7 MPa when the specimens were glued at 1.2 and 1.5 MPa, respectively. While the PUR manufacturers recommended pressure of 0.69 MPa was used in this study, increasing the pressure might have probably improved the bonding strength of the glulam specimens. Another factor that might have influenced the BSS results is the adhesive chemistry. PUR are known to have higher molecular weight and lower polarity that could reduce the penetration of the adhesives into the wood structure and thus affect the bonding results [47, 50].

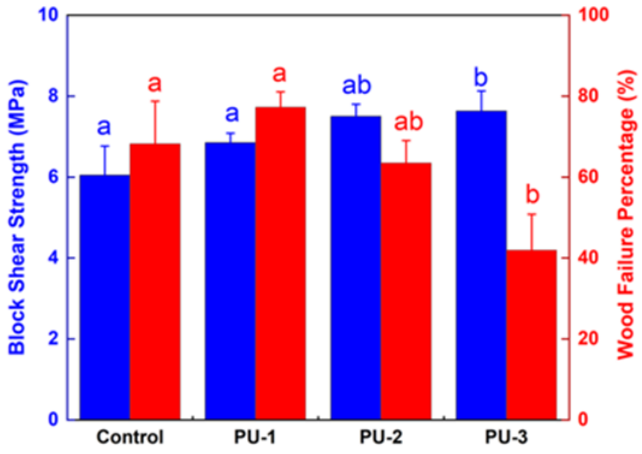

Figure 4.5 Mean BSS (primary axis) and WFP (secondary axis) of Lignin-reinforced Glulam Specimens (bars represent standard error; means followed by a common letter are not significantly different at $P < 0.05$). Dunn's multiple comparison test was used to compare the BSS and WFP values between the treatment groups.

Wood failure percentage is an indicator of bonding strength of glued specimens, with a higher WFP indicating better penetration of adhesive into wood products [51]. As per ASTM D2559, the required minimum average WFP value is 75%. As earlier reported and shown in **Table** 4.2, all the tested scenarios failed to meet this requirement with the exception of specimens bonded with PU-1. Although the mean WFP value of PU-1 was higher than that of PU-2, there was no statistical difference between the WFP of PU-1 and PU-2 specimens (**Figure** 4.5) Also, there was no difference in the WFP of control and PU-1 specimens. However, PU-3 had the lowest mean WFP value which was significantly different from PU-1 ($p = 0.005$) and control specimens (p = 0.005). These results indicate that the wt% of lignin influenced the WFP of the glued specimens. Moreover, the slight improvement in the WFP of PU-1 can be explained by the uniform dispersion of the lignin particles in PUR which thus contributed to the bonding strength. Garcia *et al*. [21] stated that the dispersion of lignin in a PUR adhesive system could help improve the bonding quality of bonded specimens, making it suitable for structural applications. In contrast to the BSS results, the WFP of glulam decreased as the wt% of lignin increases. It appears that the presence of high lignin content in the PUR-adhesive matrix affects the WFP of the glulam specimens. This is similar to other bond performance studies on engineered wood products. For instance, Pradyawong *et al*. [52] and Lubis *et al*. [53] found that the adhesion strength of lignin-based adhesive decreased with increasing wt% of lignin. The authors attributed their results to the reduction in adhesive viscosity following the addition of high amounts of lignin. The decreasing trend of bond line strength with increasing wt% of lignin might also be related to the distribution of lignin on the specimens' surface. The increased wt% of lignin possibly results in non-uniform dispersion of lignin particles in the adhesive matrix and thus affects the bonding of the glued specimens.

Overall, the lignin-modified specimens showed higher BSS and WFP values compared to the controls (especially when the lignin content is low). The better bonding results at reduced wt% of lignin could help save cost and increase the production of high-quality glulam used in structural applications. These results show that incorporating lignin in PUR-adhesive mix improved the bonding performance of glulam. Therefore, the rationale that lignin can improve the adhesion properties of PUR adhesive is supported by this study.

4.6.2 Effect of lignin addition on the Delamination of Glulam Specimens

The influence of lignin addition on the delamination resistance of PUR-bonded specimens was also investigated in this study. The PUR-bonded specimens were subjected to the delamination test as per ASTM D2559 which is considered more vigorous compared to other standards. In delamination test, specimens are exposed to repeated cycles of wetting and drying which leads to internal stresses that affect the bond line strength of glued specimens. Interestingly, all the lignin-reinforced and the control specimens passed the delamination test, despite the unsatisfactory wood failure observed in some specimens, particularly PU-2 and PU-3. Also, the slight delamination noted in a few specimens was mostly observed in a single glue line on one side of the glued specimens. All the lignin-reinforced specimens had better delamination resistance compared to the controls (without lignin). The increased delamination resistance in the lignin-reinforced specimens might have resulted from the hydrophobicity of lignin which limited the access of water into the bond line during the delamination test [21]. This effect thus reduces the internal stresses imposed on the bond line when specimens are subjected to repeated swelling and shrinkage cycles. Also, the slower curing time of PUR as a result of lignin addition could have contributed to increased delamination resistance in the lignin-reinforced specimens [54].

As shown in **Table** 4.3, the percent delamination of the specimens increased with increasing wt% of lignin. We found that PU-1 specimens displayed better delamination properties compared to PU-2 and PU-3 specimens. The increased delamination with increasing wt% of lignin is not completely understood but might be due to the poor dispersion of lignin particles in PU-2 and PU-3 bonded specimens. This probably makes the wood surface rougher and thus reduced the bond line quality of the glued specimens. An illustration of the delamination observed in the specimens and the effect of lignin on the results can be seen in

Figure 4.6. It can be observed from Fig. 4.6 that almost no delamination occurred for specimens bonded with PU-1 compared to other groups. These findings show a decrease in the percent delamination when commercial PUR-adhesive was reinforced with softwood kraft (Biochoice^{TM}) lignin, irrespective of the wt% of lignin. Based on these results, all the treatments (PUR + lignin) could be considered suitable for fabricating glulam used in structural applications.

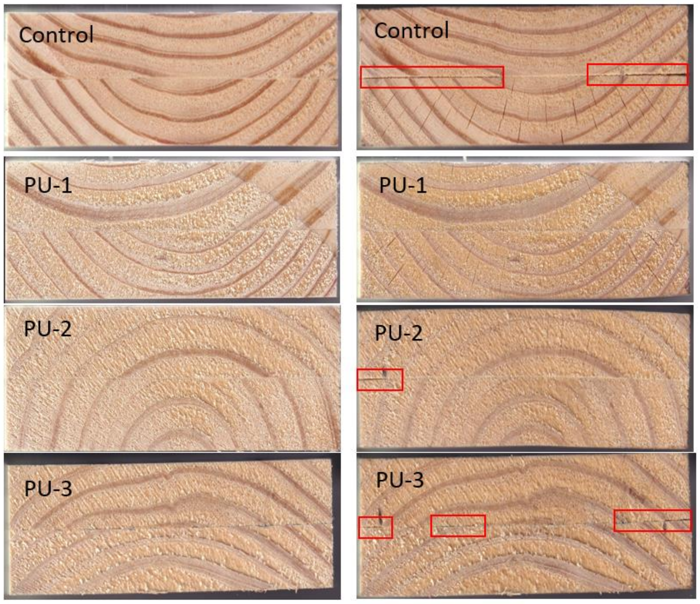

Figure 4.6 End views of glulam specimens before (left) and after (right) delamination test; the red lines indicates the delamination observed in the specimens.

4.7 Conclusion

This study shows the potential of lignin to enhance the adhesion properties of polyurethane adhesives. Herein, it was observed that the modification of PUR adhesive with lignin increased the bonding strength and reduced the delamination of glulam. Moreover, adequate values of BSS and WFP were obtained particularly for specimens bonded with PU-1. The evaluation of the bond line delamination showed that that majority of specimens bonded with lignin-reinforced PUR adhesive met the delamination requirements of 1% for softwood used in manufacturing glulam. An increase in lignin content decreased the adhesion properties of PUR which was attributed to the non-uniform distribution of lignin in PUR adhesive. Although satisfactory behavior was observed in the glue line shear and delamination tests for all the lignin-reinforced specimens. It is recommended that a wider range of wood density and increased bonding pressure be considered in further studies for better results. Based on the findings in this study, lignin can be used as a modifier in polyurethane-based adhesive system for improved bond line performance.

4.8 Acknowledgement

This research was conducted in cooperation with the USDA Forest Service. In accordance with Federal law and U.S. Department of Agriculture policy, this institution is prohibited from discrimination on the basis of race, color, national origin, sex, age or disability. (Not all prohibited bases apply to all programs.) to file a complaint of discrimination: write USDA, Director, Office of Civil Rights, Room 326-W, Whitten Building, 1400 Independence Avenue, SW, Washington, D.C. 20250-9410 or call (202) 720-5964 (voice and TDD). USDA is an equal opportunity provider and employer. This manuscript is #SB1100 of the Department of Sustainable Bioproducts, MSU. This publication is a contribution of the Forest and Wildlife Research Center, MSU. The authors would like to thank USDA Forest Products Laboratory and Henkel Corporation for providing the materials and Ms. Rathnayaka and Story for helping with the lignin extraction part of this project.

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CHAPTER V

CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

This study investigated the feasibility of successfully treating prefabricated CLT panels with copper-azole preservatives. To achieve this objective, a simple and effective evaluation method via color-indicator and image J. analysis were implemented to determine the impregnation quality via the penetration and retention of preservatives in the treated panels. In addition, an experimental programme was developed to examine the effect of lignin addition on the adhesion properties of polyurethane-based adhesives. Here, a comprehensive evaluation of the bonding quality was conducted via the shear strength, wood failure percentage, and delamination of the glulam specimens. Based on the investigations and presented results, the significant findings of this project includes:

1. The results showed that the Cu penetration in the treated 3- and 5-ply CLT panels was mostly above 85.0% especially for the 1st layer, thereby meeting the penetration requirement specified in AWPA T1-16 standard for post-treated wood composites.
2. Copper-azole type C (CA-C) exhibited better treatability than micronized copper azole (MCA) for CLT panels, resulting in higher Cu penetration and retention across the CLT panels. Thus, CA-C preservative is suggested for the treatment of large and commercial-sized CLT panels.

3. It was shown that the panel layup influenced the Cu penetration and retention in the treated CLT panels; the lengthwise orientation had better treatability for the 1st and 2nd layers and thus recommended in the fabrication of preservative-treated CLT.
4. The outcome of the investigations revealed that the penetration along the longitudinal direction (L_L) and the number of surfaces directly exposed to preservatives governed the Cu penetration in the treated CLT panels.
5. It can also be concluded that the low Cu retention observed for 2nd layer specimens in the treated panels could be improved by minimizing their L_L values.
6. The shear strength of glulam specimens bonded using lignin-reinforced PUR adhesive was generally higher than that of the control specimens. It was also shown that the shear strength increased with increasing wt% of lignin. In contrast, there was a reduction in the wood failure of glulam as the wt% of lignin was increased.
7. The results showed that all the lignin-modified and the control specimens passed the delamination requirement stipulated in ASTM D2559 for glued structural wood products used in outdoor exposure conditions.
8. While slight delamination was observed in PU-2 and PU-3 bonded glulam specimens, some groups particularly PU-1 specimens experienced negligible delamination.
9. Overall, the findings presented in this study shows the effectiveness of lignin as a modification or reinforcement agent in PUR adhesive formulation, especially when the lignin content is low. However, some factors such as bonding pressure, density and lignin dispersion in PUR adhesive are postulated to have affected the bonding performance of the glulam specimens. Thus, the consideration of these factors in lignin-reinforced PUR

adhesive formulation could provide valuable information for improved bond performance in glulam.

5.2 Recommendations for Future Research

1. The preservative-treated CLT research was designed with only four and two replicates for the 3- and 5-layer CLT, respectively. The sample size could be a limitation in making sound judgements/conclusions regarding the effect of the panel layup and thickness on preservative penetration and retention in CLT. Thus, future studies should consider a minimum of 10 replicates for the treatment combinations.
2. While the CLT laminations were pre-selected and grouped based on their density class, there were few samples with high-density which could have impacted the preservative impregnation in the CLT panels. Therefore, efforts should be made to include only lumber with a lower density range during the CLT fabrication process. Although there is need for precaution in selecting low-density lumber to avoid compromising the fabricated CLT strength properties.
3. In this study, the CLT panels were fabricated using dried lumber i.e., moisture content (MC) of 12-15%, however, it should be noted that the commercially available preservative-treated lumber usually has a higher moisture content (> 50%). Future research could utilize high-MC lumber in fabricating the CLT panels, to study the effect of MC levels on the CLT properties (such as bonding strength and preservative treatability). Additional studies could also be conducted to assess the influence of manufacturing parameters, including, bonding pressure and duration and glue spread rate on the impregnation quality of the CLT panels.

4. This research demonstrated the possibility of incorporating preservative technologies in the manufacturing of CLT. However, the study was conducted using small-dimension CLT panels. It would be interesting to evaluate the effects of panel layup and thickness on the treatment performance of commercial-sized CLT panels. This could provide valuable information and more definitive conclusions regarding the preservative treatability of southern pine CLT.
5. The effect of lignin-reinforced polyurethane adhesive on the bonding strength of glulam was investigated in this study. Although promising results were found, only 10 replications each per treatment were tested. It is recommended to expand the sample size as this could help to increase the statistical power of the tests. Also, the best performing adhesive could be implemented on large-sized glulam panels used in industrial applications.
6. In this study, the treatment/modification of the Biochoice^{TM} lignin was done by fractionation. While this method was used to obtain lignin with improved properties, it is suggested to explore other lignin modification techniques to obtain rich and high-quality lignin particles with specific properties (e.g., molecular weight, polydispersity).
7. This study showed an improvement in the bonding strength of glulam beams bonded using lignin-reinforced PUR adhesive. Even though the results are encouraging, more studies should be conducted to establish the optimal adhesive treatment (lignin + PUR) and manufacturing parameters. Afterwards, this approach should be tested on other wood-based composites such as cross-laminated timber.