

Commentary

# Barriers to the Effective Adhesion of High-Density Hardwood Timbers for Glue-Laminated Beams in Australia

William Leggate <sup>1,2,\*</sup>, Robert L. McGavin <sup>1,2</sup>, Andrew Outhwaite <sup>2</sup>, Benoit P. Gilbert <sup>1,2</sup> and Shanmuganathan Gunalan <sup>1</sup>

<sup>1</sup> School of Engineering and Built Environment, Griffith University, Gold Coast, QLD 4222, Australia; robbie.mcgavin@daf.qld.gov.au (R.L.M.); benoit.gilbert@daf.qld.gov.au (B.P.G.); s.gunalan@griffith.edu.au (S.G.)

<sup>2</sup> Queensland Department of Agriculture and Fisheries, Horticulture and Forestry Science, Salisbury Research Facility, Salisbury, QLD 4107, Australia; andrew.outhwaite@daf.qld.gov.au

\* Correspondence: william.leggate@daf.qld.gov.au; Tel.: +61-456132642

**Abstract:** A number of international timbers of high commercial importance are extremely difficult to glue, which is significantly hindering access to global market opportunities for engineered wood products, especially for heavily demanded structural products. Some particularly problematic timbers in Australia are the dominant commercial hardwood species, including spotted gum (*Corymbia* spp.) and Darwin stringybark (*Eucalyptus tetradonta*). These species are renowned for their very high mechanical properties, natural durability and attractive aesthetic appeal. However, they are notoriously difficult to glue, especially for sawn laminate-based engineered wood products, such as structural glue-laminated beams. Despite considerable effort and testing of diverse internationally established best-practice approaches to improve adhesion, glue-laminated beam samples of these timbers still frequently fail to meet the requirements of the relevant standard, mainly due to excessive glue line delamination. This paper discusses the key barriers to effective adhesion of these high-density timbers and particularly emphasises the necessity of achieving greater adhesive penetration. Greater adhesive penetration is required to enhance mechanical interlocking, entanglement and molecular interactions between the adhesive and the wood to achieve stronger and more durable bonds. Potential solutions to enhance adhesive penetration, as well as to improve gluability in general, are discussed in terms of their likelihood to satisfactorily prevent delamination and the potential to be applied at an industrial scale. This new fundamental understanding will assist the development of solutions, allowing industry to commercialise newly engineered wood products made from high-density timbers.

**Keywords:** wood adhesion; hardwood; adhesive penetration; adhesives; high-density timber



**Citation:** Leggate, W.; McGavin, R.L.; Outhwaite, A.; Gilbert, B.P.; Gunalan, S. Barriers to the Effective Adhesion of High-Density Hardwood Timbers for Glue-Laminated Beams in Australia. *Forests* **2022**, *13*, 1038. <https://doi.org/10.3390/f13071038>

Academic Editor: Petar Antov

Received: 16 May 2022

Accepted: 28 June 2022

Published: 1 July 2022

**Publisher's Note:** MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



**Copyright:** © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

## 1. Introduction

The growing demand for engineered wood products (EWPs) internationally creates lucrative market opportunities but also poses major technical challenges for many high-density hardwood timbers from Australia. Many of these timbers, such as spotted gum (*Corymbia* spp.) and Darwin stringybark (*Eucalyptus tetradonta*), with air-dry densities often exceeding 1000 kg/m<sup>3</sup>, have highly attractive traits such as extremely desirable mechanical properties (e.g., strength, stiffness, hardness etc.), high natural durability, along with attractive aesthetic appeal. This results in them being widely sought after to produce high-value EWPs such as glue-laminated (glulam) beams. However, these same characteristics also create barriers to successful wood adhesion that especially impedes the efficient manufacture of EWPs.

Globally, glulam is becoming more popular as a building product. The diminishing supply of suitable large-dimension, high-strength sawn timbers from Australia's native

forests is influencing the increasing demand in Australia for these product types [1]. Historically, post-and-beam sized (i.e., glulam-dimensioned) timbers were produced as solid (non-glued) sections from large, high-quality native forest sawlogs. However, given the transition in Australia's log supply to smaller-sized logs, and a lower quality plantation and native forest resource, it is now more difficult to produce large solid timber sections [1]. Instead, producing glulam allows the possibility to use more readily available smaller dimensions and lower quality sawn timber by laminating them together to make large cross-sectional EWPs. Indeed, these modern, alternative products offer some key advantages such as reduced variability in their mechanical properties and improved stability [2]. However, to make these products successfully, a key challenge to overcome is successful and repeatable wood adhesion.

Spotted gum and Darwin stringybark are two very important commercial high-density timbers from northern Australia's native forests. They are traditionally used in a wide range of applications such as heavy engineering construction, building framework, landscaping, poles, flooring and decking. However, attempts to manufacture structural glulam from these timbers have encountered major problems in gluing with glulam samples often failing to comply with Australian standard requirements (AS/NZS 1328.1:1998 [3]) due to excessive glue line delamination.

For these two hardwood species, structural glulam manufacture is currently a challenge with all common adhesive types used for glulam, which in Australia mainly includes resorcinol formaldehyde (RF) and one-component moisture-curing polyurethanes (1C-PUR) [4–8]. Most of the relevant research on these high-density timbers from Australia has focused on high molecular weight adhesives, such as RF and PUR because these are two of the dominant adhesives used in structural glulam internationally, as well as in Australia [4–11]. These adhesives are also considered to provide the strongest and most durable bonds for structural, weather-exposed engineered wood products. To date, alternative adhesives to RF and PUR have not been successful with these Australian high-density timbers for structural glulam [9]. Additionally, there are constraints on which adhesives can be used for structural glulam in Australia—they generally need to be cold-setting and accepted by the relevant standards for weather-exposed, structural glulam. The particular RF and PUR adhesive formulations referred to in this paper have been developed by international adhesive suppliers and provided as adhesive formulations best suited to high-density Australian timber types.

Considerable research has been undertaken by the Queensland Department of Agriculture and Fisheries (DAF) to develop a manufacturing protocol that results in a reliable and repeatable glue bond with these high-density timbers for glulam production [4–11]. These studies have included trials testing the latest international best-practice approaches with guidance from international adhesive companies to improve the adhesion of refractory species: including different surface machining methods (e.g., face milling, sanding, planing), chemical pre-treatments (including surface wipes and washes), adhesive primers, coupling agents, adhesion promoters, hydrophobic agents to promote dimensional stability, surface incisions, different adhesive formulations, surfactants, varying timber moisture content, different adhesive spread rates, elevated temperature curing, different press times and press conditions [4–11]. Studies to improve the adhesion of high-density and extractives rich hardwood timbers from Australia have also been reported with other alternative approaches such as flame treatments in combination with DCM (dichloromethane extraction) and PEI (polyethylene imine) graft chemicals [12], plasma treatments [13] and CSIRO Surf-Bond [14].

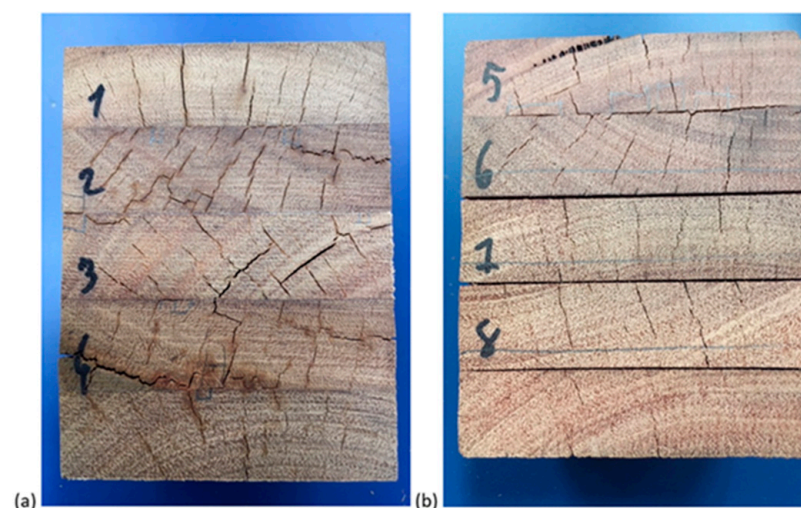
However, despite this intensive research effort and some notable improvements with the use of surface machining approaches, such as face milling, a commercially acceptable gluing protocol that can reliably and consistently produce compliant structural bonds has not yet been established. Studies so far have confirmed the extreme difficulties in gluing these timbers, which have been mainly attributed to their high dimensional

movement, extractive content, low wettability, minimal permeability and poor adhesive penetration [4–8,12,13].

Finding gluing solutions to facilitate the manufacture of structural glulam from high-density timbers in Australia is a major industry priority for opening new market opportunities, improving business resilience and increasing international competitiveness. The primary aim of this paper is to detail a new understanding that can contribute to the development of optimal adhesion protocols for these timbers. This paper discusses the latest knowledge and understanding of key barriers to effective adhesion of high-density hardwood timbers from Australia and provides direction for future research to improve the wood adhesion of these timbers for structural glulam applications. These solutions are discussed in terms of their likelihood to satisfactorily prevent delamination and the potential to be applied at an industrial scale.

## 2. The Delamination Problem Encountered with Gluing High-Density Hardwood Timbers

In Australia, acceptable adhesive bond performance for structural glulam qualification testing is based on glue line delamination performance in accordance with AS/NZS 1328.1:1998 [3]. When targeting structural products, glued samples are tested in accordance with Test Method A of Appendix C of the standard, which allows for qualification for use as a Service Class 3 (most stringent class where timber is directly exposed to sun and/or rain) product. Glulam samples are subjected to wet and dry cycling and then assessed for glue line delamination. The wet and dry cycles of AS/NZS 1328.1:1998 [3] involve submersing glulam samples in water in a pressure cylinder and then a vacuum (70 to 85 kPa) is applied for 5 min followed by a 1 h pressure (500 to 600 kPa) cycle. This vacuum and pressure cycle is conducted twice. The samples are then dried at a temperature of 65 °C, a relative humidity (RH) of 15%, and an air velocity of 2.4 m/s for 21 h. The wetting and drying process is repeated to provide two full cycles. High-density hardwood timbers such as spotted gum and Darwin stringybark frequently fail with glue line delamination exceeding the requirements of the standard. Figure 1 illustrates the differences between a glulam sample that met the requirements of the standard (Figure 1a) and a sample that failed due to excessive glue line delamination (Figure 1b), in accordance with the requirements of AS/NZS 1328.1:1998 [3].



**Figure 1.** (a) Glulam sample that has passed the AS/NZS 1328.1:1998 [3] requirements; (b) Glulam sample that has failed the AS/NZS 1328.1:1998 [3] requirements, evidenced by the widespread delamination of all glue lines [8].

### 3. Causes of Adhesion Difficulties with High-Density Hardwood Timbers

High-density timbers have been shown in many studies to be usually more problematic to glue [4–8,12–21]. The key reasons that have been identified for these gluing problems are summarised below and discussed hereafter in detail. They:

- Usually have elevated strength and mechanical resistance, which results in a lower rate of wood failure versus adhesive failure, and apply greater force on the bondline;
- Are often more difficult to bring in to close contact during product assembly;
- Often have a high content of extractives that can interfere with the gluing process;
- Commonly have low wettability;
- Tend to exhibit higher dimensional movement;
- Have lower porosity and permeability, resulting in minimal adhesive penetration.

All of these factors can negatively influence adhesive bond performance, especially when glulam is subjected to the severe stresses imposed by the wet and dry cyclic conditions of the test method [3]. Many of these issues are not just confined to high-density hardwood timbers. For example, the high-density latewood content of southern pine (*Pinus elliottii* (PEE), *Pinus caribaea* (PCH), and PEE × PCH: the hybrid between these two species) timber produced in Australia can exceed 900 kgs/m<sup>3</sup>, and also its high resin content introduces adhesive challenges compared to other common softwood timbers [4,5,17]. The problems encountered with gluing higher density softwood become particularly pertinent when considering that it is the high density, stiffer wood that is preferred for structural glulam [4,5,17].

#### 3.1. Elevated Strength and Mechanical Resistance

A major issue that poses problems for successfully bonding high-density timbers is that their elevated mechanical resistance leads to a lower rate of wood failure [22]. Structural adhesives are generally assumed to be stronger than the substrate, and bond quality tests (including AS/NZS 1328.1:1998 [3]) often judge performance on the percentage of wood versus adhesive failure. However, for high-density hardwoods, high wood failure can be difficult to achieve because of their high strength. In addition, they impart a greater force on the bondline.

#### 3.2. Close Contact during Product Assembly

Hovanec [23] notes that because dense woods tend to be stronger, more dense substrates are generally harder to bring into contact with each other when assembling the glue joint—essentially overcoming any board distortion or deformation in the pressing process. Poor assembly negatively impacts bond strength due to variability in glue line pressure and the resultant glue lines being too thin or too thick [21,23].

#### 3.3. High Content of Extractives and Low Wettability

High-density timbers from Australia are also usually higher in wood extractives that contribute to their favorable natural durability characteristics but can interfere with the gluing process [4–8,12,13,15,16]. Widsten et al. [15] found that spotted gum (*Corymbia maculata*) exhibited relatively poor adhesion when compared to radiata pine (*Pinus radiata*) and seven other *Eucalyptus* species included in the study. According to Widsten et al. [15], the poor bonding of spotted gum was linked to its high density and high phenolic and lipophilic extractives content. Ramos [13] provided a detailed discussion on the negative impacts of extractives on wood adhesion, emphasising that extractives have major effects on wood gluing. In particular, they affect the wettability of wood, which is commonly used as a measure of wood's suitability for gluing [13]. Leggate et al. [4], Widtsen et al. [15] and Burch [24] have demonstrated the lower wettability of spotted gum compared to many other timber types. Consequently, the removal of extractives from wood has been shown to improve the wettability and gluing of wood [12,13,25,26]. Hse and Kuo [13,27] outlined several ways in which extractives may reduce glue bond strength in wood products:

- Heavy deposits of extractives may block reaction sites and prevent the anchorage of glue;
- There is a chemical incompatibility between the adhesives and extractives;
- Extractives influence the wettability and polarity of wood surfaces;
- Extractives affect the curing and setting characteristics of adhesives.

The migration of extractives to wood surfaces during drying and ageing of wood is thought to worsen the conflict between extractives and successful bonding [12,13,15,27]. Peeling and gluing trials on various eucalypts also highlight the harmful effect that migration of extractives has on the gluing of plywood [13,28]. Extractives can also affect the rheological properties of adhesives and retard their gelation [13,29,30]. Onishi and Goto [13,31] found that extractives isolated from Marri (*Eucalyptus calophylla* R.Br. ex Lindl.) increased the gelation time of a UF resin. Extractives from *Acacia mangium* timber have also been shown to interfere with the chemical cure of RF adhesives [32]. Extractives may also impede adhesive penetration into the lignocellulosic matrix [12]. Ramos [13] especially highlighted the particular problems posed by waxes in wood. Waxes, another type of wood extractive, are hydrophobic in nature, and when present on wood surfaces, they can reduce the wettability of wood and impair glue bonding [13,29]. Ramos [13] reported that the presence of wax on wood surfaces of certain eucalypt species, for example tallowwood (*E. microcorys*), is known to interfere with glue bonding.

To overcome the negative influence of extractives and waxes, it is common industry practice to prepare the wood surface (usually by planing) immediately before glue application to reduce the amount of extractives and waxes that remain on the wood surface after drying. However, attempts to reduce the detrimental effect of extractives and waxes on wettability and gluability in timbers such as spotted gum and Darwin stringybark have largely failed to result in satisfactory bond quality. This has included varied approaches such as surface machining immediately before adhesive application, trialing novel surface machining approaches such as face milling, chemical pre-treatments such as surface wipes and washes, the addition of surfactants to adhesives and elevated temperature curing [4–11]. In addition, even though Leggate et al. [4–8] were able to markedly improve the surface wettability of spotted gum through face milling and the use of surfactants, these improvements did not translate to producing acceptable bond quality results according to AS 1328.1:1998 [3]. It is argued that the main reason for the lack of success with these approaches has been due to adhesive penetration in all cases being insufficient to provide a reinforced zone surrounding the bondline that can withstand the high shrinkage and swelling forces that result from the test method.

### 3.4. Higher Dimensional Movement

Higher density timbers tend to exhibit greater dimensional movement with changing moisture content [13,16,18]. Table 1 illustrates the higher shrinkage rate (from unseasoned to 12% moisture content) and unit movement values (with each 1% change of moisture content) of spotted gum and Darwin stringybark compared to two other lower-density commercial timbers commonly used in glulam. When compared to other species, this higher dimensional movement combined with a higher modulus of elasticity of the timber likely creates a greater propensity for the glulam to delaminate due to high stresses developing at the glue-line and causing the sawn laminates to separate [13,16,18,22]. However, attempts to dimensionally stabilise spotted gum and Darwin stringybark timber through the use of hydrophobic agents and coatings have not yet been successful in achieving acceptable glue line delamination results in accordance with AS/NZS 1328.1:1998 [3,9].

**Table 1.** Comparison of shrinkage and unit movement <sup>1</sup> between different timbers used for glulam <sup>2</sup>.

|   | Air-Dry Density (kgs/m <sup>3</sup> ) | Tangential Shrinkage (%) | Radial Shrinkage (%) | Tangential Unit Movement (%) | Radial Unit Movement (%) |
|---|---------------------------------------|--------------------------|----------------------|------------------------------|--------------------------|
| Spotted gum ( <i>Corymbia</i> spp.)                 | 1010                                  | 6.10                     | 4.30                 | 0.38                         | 0.32                     |
| Darwin stringybark ( <i>Eucalyptus tetradonta</i> ) | 1090                                  | 4.90                     | 3.80                 | 0.38                         | 0.31                     |
| Radiata pine ( <i>Pinus radiata</i> )               | 550                                   | 5.00                     | 3.00                 | 0.27                         | 0.20                     |
| Spruce ( <i>Picea abies</i> )                       | 470                                   | 5.00                     | 1.90                 | 0.33                         | 0.16                     |

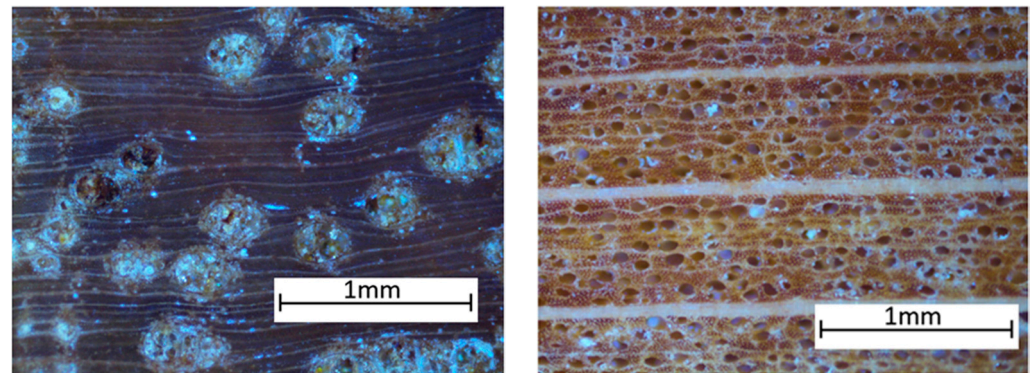
<sup>1</sup> (Percentage of dimensional change for each 1% moisture content change between about 3% and the fibre saturation point for the particular species) <sup>2</sup> (Data sourced from [33–35]).

### 3.5. Lower Porosity and Permeability

Another major cause of adhesion difficulties in gluing high-density timbers is low porosity and low permeability. The low porosity and permeability can limit adhesive penetration. Ramos [13] highlighted the adhesion problems that are caused by the high density, commenting that dense wood species are more difficult to glue than lower-density woods because they contain thicker cell walls, smaller cell lumens and few connective openings such as pits. These characteristics restrict the glue from penetrating into the wood, which contributes to weaker bonding [13,15,18,19]. Ramos [13] also commented that although impermeable materials such as metals, plastics and glass can be successfully bonded without adhesives penetrating into the substrate, some degree of adhesive penetration into wood is necessary because its outermost surfaces are usually contaminated or weakened during processing.

The penetration of an adhesive into the wood structure is believed to have a strong influence on bond strength, durability and performance [18,36–48]. Adhesive penetration into wood can occur in various forms, including gross penetration and penetration into cell walls. Gross penetration is the movement of the adhesive into the cell lumens as well as into inter-cellular voids, whereas cell wall penetration is the diffusion or infiltration of the adhesive into the cell walls [20,41,48]. However, not all adhesives can penetrate cell walls, with current data suggesting that cell wall penetration is achieved only by in-situ polymerised adhesives with low molecular weight fractions, e.g., RF and PRF [20]. There is no experimental evidence to demonstrate that pre-polymerised adhesives, e.g., PUR can penetrate cell walls [20,48]. If adhesive penetration into wood is restricted, it usually results in insufficient mechanical interlocking in adhesive bonding [15,19]. Vick [19] especially highlighted the importance of adhesive penetration in producing the most durable adhesive bonds stating that effective mechanical interlocking can only take place if the adhesive penetrates beyond the surface debris and damaged cells into sound wood, at least two to six cells deep. Deeper penetration of the adhesive into the wood microstructure increases the surface area of contact between adhesive and wood for more effective mechanical interlocking and molecular interactions [19]. The reinforced zone surrounding the glue line also increases the likelihood of failure during testing to be positioned well clear of the glue line, often at the boundary layer between the adhesive reinforced wood and natural timber.

Figure 2 illustrates the starkly contrasting wood structures of spotted gum, a very impermeable timber, and European beech (*Fagus sylvatica*), a more permeable hardwood. Spotted gum is characterised by a very dense and closed wood structure with very few cellular openings and also an abundance of tyloses and extractives that block the vessels. In contrast, European beech has a more open wood structure.



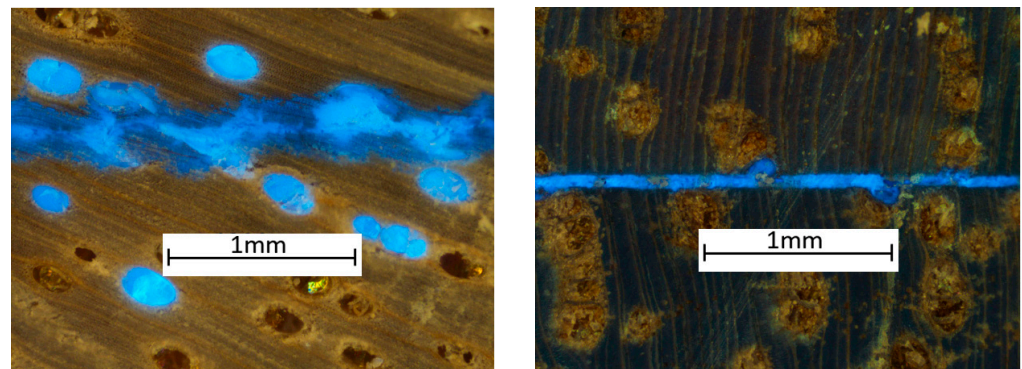
**Figure 2.** Contrasting wood structures (cross-sections) at 50× magnification of spotted gum (left) and European beech (right).

Studies by Leggate et al. [4,6] have highlighted the very low permeabilities of spotted gum and Darwin stringybark. The liquid permeability of spotted gum timber was so low that it was unable to be measured. Redman et al. [49] also reported extremely low permeability in spotted gum, highlighting its much lower porosity compared to other common commercial wood species. Spotted gum and Darwin stringybark are characterised by vessels in the heartwood containing abundant tyloses and extractive deposition (Figure 2), which impedes the movement of gases and liquids [4,7,50,51]. Wood permeability strongly influences gluability mainly through its influence on wettability and adhesive penetration. Wood permeability is one of the main controlling factors influencing the depth of adhesive penetration [23,24,52].

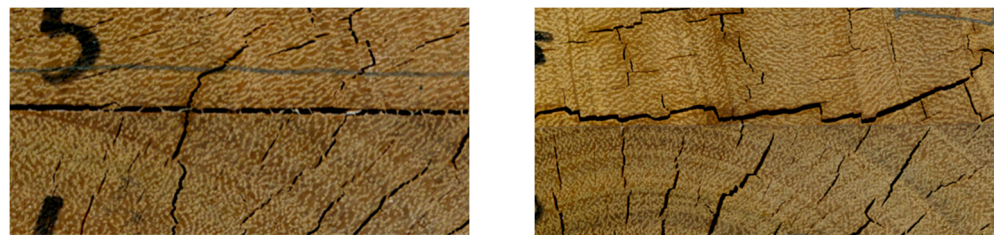
#### 4. The Root Cause of Poor Adhesive Bond Quality in These High-Density Timbers

It is asserted that the most important contributor to the typically poor adhesive bond performance in timbers such as spotted gum and Darwin stringybark is inadequate adhesive penetration regardless of which adhesive is used and their different forms of penetration (e.g., gross penetration only with PUR and gross and cell wall penetration with RF). The minimal adhesive penetration in these species is linked to the extremely low wood permeability discussed above. Leggate et al. [7] demonstrated the much lower adhesive penetration in spotted gum and Darwin stringybark timber compared to southern pine. The poor penetration in spotted gum is also contrasted strongly with that observed in other lower-density timbers that are much easier to glue such as radiata pine (*Pinus radiata*) and shining gum (*Eucalyptus nitens*). Figure 3 illustrates the marked difference in the 1C-PUR adhesive penetration in spotted gum versus shining gum. The shining gum timber shown in Figure 3 produced glulam samples that met the adhesive bond requirements of the AS/NZS 1328.1:1998 standard [3]. Figure 3 shows that in shining gum, the adhesive penetrated much deeper and more uniformly and into all wood cell types—including the fibres, rays and vessels, whereas in spotted gum, there was minimal penetration of adhesive away from the glue line, and it was mainly isolated to occasional vessels.

It is suggested that because the adhesive penetration is so low in timbers such as spotted gum, there is inadequate anchorage of the adhesive, inadequate meshing, interlocking or entanglement and insufficient molecular interactions between the wood and the adhesive to withstand the high shrinkage and swelling forces being exerted on the glue line, leading to delamination. When the adhesive penetration is so restricted, and the glulam sample is subjected to wet and dry cycling, the fibre failure in the wood will be very shallow, mainly confined to the surface fibres very close to the glue line—and classified as delamination according to the standards (Figure 4). Conversely, when the adhesive penetration is deeper and more uniform and the glulam sample subjected to wet and dry cycling, wood fibre failure will likely occur distant from the glue line (often at the boundary of adhesive reinforced wood and natural wood), and it will not be classified as delamination (Figure 4).



**Figure 3.** Contrasting adhesive (1C-PUR) penetration at 50× magnification in shining gum (**left**) and spotted gum (**right**).



**Figure 4.** Delamination due to very shallow adhesive penetration (**left**) and wood fibre failure distant from the glue line (**right**) and not classified as delamination—due to deeper adhesive penetration in spotted gum glulam.

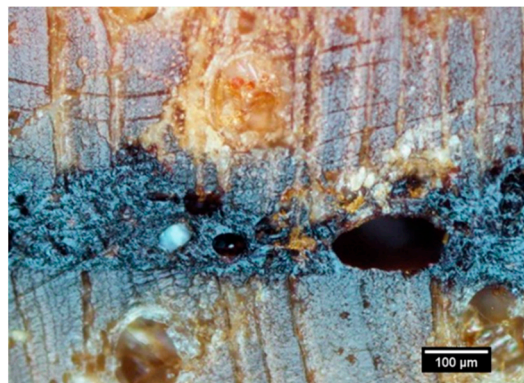
Although studies by Leggate et al. [4–8] have demonstrated the beneficial effect of face milling compared to planing in improving the adhesive bond performance of high-density timbers from Australia for structural glulam, semi-commercial scale trials with face milling have shown that acceptable delamination results are not being consistently achieved [8,10,11]. It is suggested that the main reason for that is that although alternative surface machining approaches such as face milling improve bond performance through increasing surface roughness, fibrillation and reduced sub-surface damage or compaction etc. [4–8], adhesive penetration is still too minimal to adequately reinforce the bond lines resulting in unacceptable delamination.

As mentioned earlier, apart from alternative surface machining, many other approaches have been tested to improve the adhesive bond performance of high-density timbers from Australia, particularly targeting extractive removal, increasing timber wettability and reducing dimensional movement. It is argued that the main reason for the limited success with all of these approaches is that they all fail to improve adhesive penetration even though other factors that influence gluability have been enhanced. The low permeability not only restricts the penetration of adhesives, but it also restricts movement into the wood of other liquids such as surface washes, water sprays and adhesion-promoting agents such as primers and hydrophobic coatings. This can severely limit the efficacy of these approaches, which have been shown to be beneficial when applied to other more permeable timbers.

A high frequency of air bubbles or voids in the glue line has also been shown with spotted gum and Darwin stringybark with both 1C-PUR and also RF adhesives [7] (Figure 5). Voids in the cured adhesive in bond lines are expected to reduce the bond strength and durability, and minimising or eliminating air bubbles is essential to achieving the strongest bond [7,53–55]. The presence of these voids may be associated with the low permeability of these timbers, which could result in air or gas entrapment in the glue line. However, as previously discussed by Leggate et al. [7], voids in cured adhesives can be caused by a number of factors, which can vary depending on the adhesive type and process. Possible sources of voids include air entrapment in the adhesive during the mixing and application



process; chemical reactions and emissions, e.g., the reaction of PUR adhesives with water, which generates carbon dioxide gas bubbles; fillers, which may have micro-air bubbles attached to their surface; and excessive shrinkage of the adhesive during hardening.



**Figure 5.** Voids in RF adhesive in spotted gum glulam [7].

### 5. Strategies to Increase Adhesive Penetration to Improve Bond Performance in High-Density Timbers

Many different options have been tested internationally for a variety of wood processing or utilisation purposes to improve wood permeability, and these can be divided into four categories: namely, chemical, biological, mechanical and physical treatments.

Chemical treatments to improve permeability include:

- Enzymatic treatments that break down or dissolve the pits between cells [56–58];
- Wood swelling agents that swell and open up the wood to allow better penetration of liquids into the wood structure [59–61]. However, if the particular swelling agent occupies all the available free cell lumen and cell wall volume, this approach may not be successful;
- Resin and extractive dispersing, dissolving or removal agents—resins and extractives can block the movement of liquids into the wood via the resin/extractive itself, blocking the cell lumens, other interstitial spaces or pits (via encrustations) or else the resin/extractive combining with the penetrating liquid (e.g., adhesives or preservatives) and this complex obstructing liquid flow [57,62–65];
- Surfactants and wetting agents, which can improve penetration and wetting by reducing liquid surface tension but also extractive dissolution [66–68]. If non-polar materials are blocking the surface, these need to be dissolved before the wood itself becomes accessible to the adhesive;
- Altering the characteristics of the adhesive to improve penetration—e.g., changes to viscosity, reactivity, molecular weight of elements, surface tension [42–45,57];
- Using gaseous or supercritical fluids [69,70]. This is a method that is sometimes used to improve wood preservative penetration, and it is unlikely to be compatible with a wood gluing process. It is also a method that would involve high capital costs.

With the exception of the gaseous or supercritical fluid approach, the other chemical treatment methods outlined above should be prioritised in future efforts to increase adhesive penetration in high-density timbers. Chemical pre-treatments of timber surfaces and/or modifications to adhesive formulations are likely to be relatively simple to incorporate into wood adhesion processes in glulam manufacturing operations compared to many of the other biological, mechanical and physical methods discussed in this section.

Biological treatments include techniques such as using microorganisms such as fungi and bacteria to open pits in the wood or create other changes to the wood, thus increasing fluid flow [71]. Whilst technically possible, this approach is likely to be very time-consuming, expensive and result in considerable inconsistency and variability in adhesive penetration and, therefore, not suitable for industrial gluing applications.

Mechanical treatments include methods such as incising, static compression and compression rolling [71,72]. These methods are sometimes used prior to wood treatment: incising opens up the wood via the use of sharp knives, and compression rolling creates micro-cracks in the wood, thus improving fluid flow. Incision has already been shown to result in some improvements in adhesive bond performance with southern pine; however, this success has not yet been replicated with higher-density hardwood timbers [9,17]. However, it is recommended that a high priority for future research would be investigating different incision methods and configurations as well as alternative mechanical treatments such as rolling compression. Mechanical treatments are likely to be more readily implemented in manufacturing plants compared to some of the biological and physical penetration enhancing systems discussed in this section.

Physical treatments to increase wood permeability include microwave treatments, high-temperature modification of wood, plasma treatment, variations in drying, steaming and vacuum, pressure and diffusion treatments [71]. Shock waves and acoustic approaches have also been trialled with some success [73,74]. Many of these physical treatments are likely to be relatively expensive in terms of both capital and operating costs and also could be difficult to implement in industrial production processes; however, plasma treatments may be an option to further investigate, given some success in improving adhesion performance in higher density timbers in earlier studies [13].

As discussed above, research priority should be allocated to more practical, cost-effective methods that could be more readily introduced into wood processing and manufacturing operations. It is also important to note that given that wood adhesion is a surface phenomenon, in order to improve adhesive penetration, the enhancement of permeability only needs to be confined to the timber surface and not the entire timber cross-section.

## **6. Additional Strategies to Achieve a Better Adhesive Bond Performance in High-Density Timbers**

Given the extreme difficulties in achieving durable glue bonds with high-density Australian timbers for glulam, even when adopting the latest international best-practice techniques and technologies, it is recommended that in addition to improving adhesive penetration, other alternative options should also be explored to support bond line integrity. A focus on dimensional stabilisation of the glulam and lamella to minimise the shrinkage and swelling forces on the glue lines that lead to delamination could be considered. These dimensional stabilisation approaches may include hydrophobic coatings, water repellents and wood modification techniques (e.g., acetylation or thermal treatments). The use of hydrophobic coatings and water repellents is likely to be a cheaper and easier approach compared to wood modification methods; however, wood modification methods may be more effective. However, acetylation and/or a dimensional stabilisation approach using hydrophobic agents may create an additional conflict between the adhesive and the stabilisation solution. Frihart et al. [75] have shown that acetylation increases the strain on the bondline under wet conditions, and increasing the hydrophobicity of the timber usually does not facilitate gluability. However, with these high-density timbers, the benefits provided by dimensional stabilisation may outweigh other negative impacts on gluability. Frihart et al. [75] demonstrated that acetylated wood can be bonded with some adhesives such as RF that perform as well as or as better than the adhesive bond performance with unmodified wood. These timbers have a much higher modulus of elasticity than most other commercial timbers used internationally for glulam—this higher stiffness in combination with high shrinkage coefficients is associated with very high shrinkage and swelling forces being exerted on the glue lines [13,16]. Therefore, another important area for future research should be to measure and characterise these forces and develop appropriate manufacturing protocols to minimise their concentration at the bond line.

Research to date on Australian high-density timbers has mainly focused on testing two common adhesives used for structural glulam internationally—one-component polyurethanes (1C-PUR) and resorcinol formaldehyde (RF) [5–11]. Some limited trials

have also been undertaken with phenol resorcinol formaldehyde (PRF), emulsion polymer isocyanate (EPI), aqueous polymer isocyanate (API), epoxy resins and melamine urea formaldehyde (MUF) adhesives [9]. For spotted gum and Darwin stringybark structural glulam, the application of all these adhesive types has not been successful. Alternative structural adhesive types and formulation chemistries should be evaluated with these species, specifically targeting improved penetration. Indeed, special novel adhesive formulation chemistries may need to be developed to overcome the deficiencies identified in commercially available adhesives commonly used internationally for lower-density timbers.

## 7. Conclusions

This paper combines the latest knowledge concerning the fundamental causes of adhesion problems in high-density timbers such as spotted gum and Darwin stringybark. Recent research by Leggate et al. [4–8] has demonstrated that the common approaches of adopting alternative surface machining methods to improve bond performance, while beneficial with spotted gum and Darwin stringybark, is not sufficient to overcome the significant challenge faced with gluing these wood species for industrial glulam manufacture. Similarly, the research has also shown that improving the surface wettability through immediate adhesive application after machining, preparing the surface with a range of chemical treatments or modifying the adhesive through the addition of surfactants has not resulted in any tangible improvements [4–11]. The research undertaken by Leggate et al. [4–8] has, however, clearly highlighted the critical importance of increasing adhesive penetration to improve adhesive bond quality in these species for applications such as structural glulam. With this new detailed understanding, future studies on gluing of high-density timbers from Australia for glulam need to prioritise the development of a cost-effective method to achieve satisfactory adhesive penetration. Without significant improvement in adhesive penetration, the application of other adhesion-promoting approaches is unlikely to succeed. There are a variety of mechanical, chemical and physical approaches that may provide the necessary solution for increasing adhesive penetration and, therefore, should be further explored in future research focused on developing a suitable manufacturing protocol that allows spotted gum and Darwin stringybark (and other high-density timbers) to be used in industrial glulam manufacturing. Commercially suitable approaches may be mechanical approaches such as incising or chemical-based adhesive penetration enhancing agents. Methods to improve dimensional stabilisation of the wood and new adhesive formulations may also provide some benefits in improving adhesive penetration and bond performance with these high-density timbers.

**Author Contributions:** Conceptualisation, W.L. and R.L.M.; methodology, W.L., R.L.M. and A.O.; investigation, W.L., A.O. and R.L.M.; writing—original draft preparation, W.L.; writing—review and editing, R.L.M., B.P.G., A.O. and S.G.; supervision, R.L.M., B.P.G. and S.G. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research received no external funding.

**Data Availability Statement:** Not applicable.

**Acknowledgments:** The support provided by the Queensland Government Department of Agriculture and Fisheries (DAF) through the provision of the unique facilities located at the Salisbury Research Facility is acknowledged as critical to facilitating studies of this nature.

**Conflicts of Interest:** The authors declare no conflict of interest.

## References

1. McGavin, R.L.; Leggate, W.; Dorries, J. *Increasing the Value of Under-Utilised Forest Resources Through the Development of Advanced Engineered Wood Products*; Forest and Wood Products Australia (FWPA) Project Number PNB 407-1516; FWPA: Melbourne, Australia, 2020.
2. Stark, N.M.; Cai, Z.; Carll, C. Wood-based composite materials: Panel products, glued-laminated timber, structural composite lumber and wood-nonwood composite materials. In *Wood Handbook—Wood as an Engineering Material*; Gen. Tech. Rep. FPL–GTR–282; U.S. Department of Agriculture, Forest Service, Forest Products Laboratory: Madison, WI, USA, 2010.

3. AS/NZS 1328.1:1998; Glued Laminated Structural Timber. Part 1: Performance Requirements and Minimum Production Requirements. Standards Australia: Sydney, Australia, 2011.
4. Leggate, W.; McGavin, R.L.; Miao, C.; Outhwaite, A.; Chandra, K.; Dorries, J.; Kumar, C.; Knackstedt, M. The Influence of Mechanical Surface Preparation Methods on Southern Pine and Spotted Gum Wood Properties: Wettability and Permeability. *BioResources* **2020**, *15*, 8554–8576. [[CrossRef](#)]
5. Leggate, W.; McGavin, R.L.; Outhwaite, A.; Kumar, C.; Faircloth, A.; Knackstedt, M. The Influence of Mechanical Surface Preparation Methods on the Bonding of Southern Pine and Spotted Gum: Tensile Shear Strength of Lap Joints. *BioResources* **2021**, *16*, 46–61. [[CrossRef](#)]
6. Leggate, W.; McGavin, R.L.; Outhwaite, A.; Dorries, J.; Robinson, R.; Kumar, C.; Faircloth, A.; Knackstedt, M. The Influence of Mechanical Surface Preparation Method, Adhesive Type and Curing Temperature on the Bonding of Darwin Stringybark. *BioResources* **2021**, *16*, 302–323. [[CrossRef](#)]
7. Leggate, W.; Shirmohammadi, M.; McGavin, R.; Outhwaite, A.; Knackstedt, M.; Brookhouse, M. Examination of Wood Adhesive Bonds via MicroCT: The Influence of Pre-Gluing Surface Machining Treatments for Southern Pine, Spotted gum and Darwin stringybark Timbers. *Bioresources* **2021**, *16*, 5058–5082. [[CrossRef](#)]
8. Leggate, W.; Outhwaite, A.; McGavin, R.; Gilbert, B.P.; Shanmuganathan, G. The Effects of the Addition of Surfactants and the Machining Method on the Adhesive Bond Quality of Spotted Gum Glue-Laminated Beams. *Bioresources* **2022**, *17*, 3413–3434. [[CrossRef](#)]
9. DAF; Queensland Department of Agriculture and Fisheries (DAF), Brisbane, Queensland, Australia. Unpublished work. 2022.
10. Outhwaite, A.; McGavin, R.L.; Leggate, W. *Adhesion System Development for Spotted Gum Solid Timber Engineered Wood Products*; Australian Centre for International Agricultural Research (ACIAR) Report; Project Number FST/2016/151; ACIAR: Canberra, Australia, 2020.
11. McGavin, R.L.; Outhwaite, A.; Leggate, W. *Adhesion System Development for Darwin Stringybark: Variable Scale Glulam Trials*; Advance Queensland Report; Advance Queensland Project (AQIP) Indigenous Employment Livelihoods, Mining; AQIP: Brisbane, Australia, 2020.
12. Gutowski, W.S.; Widtsen, P.; Li, S.; Cerra, T.; Molenaar, S.; Spicer, M. Improving the Adhesion Strength of Wood Cross-Lap Joints by Flame Treatment and Solvent Extraction. In Proceedings of the 59th Appita Annual Conference and Exhibition, Auckland, New Zealand, 16–19 May 2005.
13. Ramos, A.L. Improving the Gluability of Eucalypt Timber by Plasma Modification of Wood Surfaces. Master’s Thesis, The Australian National University, Canberra, Australia, 2001.
14. Zhang, X.; Li, S.; Belleville, B.; Ozarska, B.; Gutowski, M.; Kuys, B. *High-Tech Modular Building Components with High Contents of Australian Hardwoods*; Forest and Wood Products Australia (FWPA) Project Number PNA380-1516; FWPA: Melbourne, Australia, 2020; pp. 1–70.
15. Widtsen, P.; Gutowski, V.S.; Li, S.; Cerra, T.; Molenaar, S.; Spicer, M. Factors Influencing Timber Gluability With One-Part Polyurethanes—Studied with Nine Australian Timber Species. *Holzforschung* **2006**, *60*, 423–428. [[CrossRef](#)]
16. Li, S.; Belleville, B.; Gutowski, M.; Kuys, B.; Ozarska, B. Achieving Long-Term Adhesion and Bondline Durability with Difficult-to-Bond Australian Hardwood Species. In Proceedings of the 61st International Convention of Society of Wood Science and Technology, Nagoya, Japan, 5–9 November 2018.
17. Vella, R. Improving the Adhesion of High-Density Softwoods with Isocyanate Based Adhesives. Master’s Thesis, University of Queensland, Brisbane, Australia, 2020.
18. Marra, A.A. *Technology of Wood Bonding: Principles in Practice*, 1st ed.; Van-Nostrand Reinhold: New York, NY, USA, 1992; pp. 76–80.
19. Vick, C.B. Adhesive bonding of wood materials. In *Wood Handbook—Wood as an Engineering Material*; Gen. Tech. Rep. FPL–GTR–113; U.S. Department of Agriculture, Forest Service, Forest Products Laboratory: Madison, WI, USA, 1999.
20. Frihart, C.R. Adhesive groups and how they relate to the durability of bonded wood. *J. Adhes. Sci. Technol.* **2009**, *23*, 601–617. [[CrossRef](#)]
21. Frihart, C.R.; Hunt, C.G. Adhesives with wood material—Bond formation and performance. In *Wood Handbook—Wood as an Engineering Material*; Forest Products Laboratory: Madison, WI, USA, 2010; Chapter 10.
22. Hänsel, A.; Sandak, J.; Sandak, A.; Mai, J.; Niemz, P. Selected Previous Findings on the Factors Influencing the Gluing Quality of Solid Wood Products in Timber Construction and Possible Development: A Review. *Wood Mater. Sci. Eng.* **2022**, *17*, 230–241. [[CrossRef](#)]
23. Hovanec, D. Effect of Wood Characteristics on Adhesive Bond Quality of Yellow Poplar for Use in Cross-Laminated Timbers. Master’s Thesis, West Virginia University, Morgantown, WV, USA, 2015.
24. Burch, C.P. Adhesion Fundamentals in Spotted Gum (*Corymbia* spp.). Master’s Thesis, Virginia Polytechnic Institute and State University, Blacksburg, VA, USA, 2015.
25. Chen, C.M. Effect of Extractive Removal on Adhesion and Wettability of Some Tropical Woods. *For. Prod. J.* **1970**, *20*, 36–41.
26. Moredo, C.C., Jr.; Sakuno, T.; Kawada, T. The Improvement of Bond Strength Properties and Surface Characteristics of Resinous Woods. *J. Adhes.* **1996**, *59*, 183–195. [[CrossRef](#)]
27. Hse, C.Y.; Kuo, M.L. Influence of Extractives on Wood Gluing and Finishing: A Review. *For. Prod. J.* **1988**, *38*, 52–56.

28. Wade, J. *Peeling and Gluing of New South Wales Eucalypts*; Technical Paper No. 55; Forestry Commission of New South Wales: New South Wales, Australia, 1991.
29. Plomley, K.F.; Hillis, W.E.; Hirst, K. The Influence of Wood Extractives on Glue-Wood Bond. I. The Effect of Kind and Amount of Commercial Tannins and Crude Wood Extracts on Phenolic Bonding. *Holzforschung* **1976**, *30*, 14–19. [CrossRef]
30. Yazaki, Y.; Collins, P.J.; Iwashima, T. Extractives from Blackbutt (*Eucalyptus pilularis*) Wood Which Affect Gluebond Quality of Phenolic Resins. *Holzforschung* **1993**, *47*, 412–418. [CrossRef]
31. Onishi, H.; Goto, T. Studies on the Wood gluing VIII. The Effects of Wood Extractives on the Gelation Time of Urea Formaldehyde Resin Adhesive. In *Study on the Gluability of Tropical Woods. II. Gelation Time of Urea Formaldehyde Resin Adhesive*; Yamagishi, Y., Kawai, N., Ono, S., Eds.; Bulletin of the Faculty of Agriculture; Shimane University: Matsue, Japan, 1971; pp. 61–65.
32. Alamsyah, E.M.; Yamada, M.; Taki, K. Bondability of Tropical Fast-Growing Tree Species 111: Curing Behaviour of Resorcinol Formaldehyde Resin Adhesive at Room Temperature and Effects of Extractives of *Acacia mangium* Wood on Bonding. *J. Wood Sci.* **2008**, *54*, 208–213. [CrossRef]
33. Wood Solutions. Available online: <https://www.woodsolutions.com.au/wood-species> (accessed on 16 March 2022).
34. QTimber. Available online: <https://qtimber.daf.qld.gov.au/browse-timbers> (accessed on 16 March 2022).
35. Hopewell, G. *Characteristics, Utilisation and Potential Markets for Cape York Peninsula Timbers*; Agency for Food and Fibre Sciences Report; Queensland Forestry Research Institute: Brisbane, Australia, 2001.
36. Collett, B.M. A Review of Surface and Interfacial Adhesion in Wood Science and Related Fields. *Wood Sci. Technol.* **1972**, *6*, 1–42. [CrossRef]
37. Frihart, C.R. Adhesive Interactions with Wood. In *Fundamentals of Composite Processing: Proceedings of a Workshop*; Winandy, J.E., Kamke, F.A., Eds.; General Technical Report FPL-GTR-149; US Department of Agriculture, Forest Service, Forest Products Laboratory: Madison, WI, USA, 2004.
38. Sernek, M.; Resnik, J.; Kamke, F.A. Penetration of Liquid Urea-Formaldehyde Adhesive into Beech Wood. *Wood Fiber Sci.* **1999**, *31*, 41–48.
39. Paris, J.L. Wood Adhesive Bondline Analyses with Micro X-ray Computed Tomography. Ph.D. Dissertation, Oregon State University, Corvallis, OR, USA, 2014.
40. Luedtke, J.; Amen, C.; van Ofen, A.; Lehringer, C. 1C-PUR-bonded Hardwoods for Engineered Wood Products: Influence of Selected Processing Parameters. *Eur. J. Wood Prod.* **2015**, *73*, 167–178. [CrossRef]
41. Kamke, F.A.; Lee, J.N. Adhesive Penetration in Wood—A Review. *Wood Fiber Sci.* **2007**, *39*, 205–220.
42. Gavrilovic-Grmusca, I.; Miljković, J.; Djiporović-Momčilović, M. Influence of the degree of condensation on the radial penetration of urea formaldehyde adhesives into Silver Fir (*Abies alba* Mill.) wood tissue. *J. Adhes. Sci. Technol.* **2010**, *24*, 1437–1453. [CrossRef]
43. Gavrilovic-Grmusca, I.; Dunky, M.; Miljković, J.; Djiporović-Momčilović, M. Radial penetration of urea-formaldehyde adhesive resins into beech (*Fagus Moesiaca*). *J. Adhes. Sci. Technol.* **2010**, *24*, 1753–1768. [CrossRef]
44. Gavrilovic-Grmusca, I.; Dunky, M.; Miljković, J.; Djiporović-Momčilović, M. Influence of the degree of condensation of urea-formaldehyde adhesives on the tangential penetration into beech and fir and on the shear strength of the adhesive joints. *Eur. J. Wood Wood Prod.* **2012**, *70*, 655–665. [CrossRef]
45. Gavrilovic-Grmusca, I.; Dunky, M.; Miljković, J.; Djiporović-Momčilović, M. Influence of the viscosity of UF resins on the radial and tangential penetration into poplar wood and on the shear strength of adhesive joints. *Holzforschung* **2012**, *66*, 849–856. [CrossRef]
46. Gavrilovic-Grmusca, I.; Dunky, M.; Miljković, J.; Djiporović-Momčilović, M.; Popović, M.; Popović, J. Influence of pressure on the radial and tangential penetration of adhesive resin into poplar and on the shear strength of adhesive joints. *Bioresources* **2016**, *11*, 2238–2255. [CrossRef]
47. Nuryawan, A.; Park, B.; Singh, A.P. Penetration of urea-formaldehyde resins with different formaldehyde/urea mole ratios into softwood tissues. *Wood Sci. Technol.* **2014**, *48*, 889–902. [CrossRef]
48. Jakes, J.E.; Frihart, C.R.; Hunt, C.G.; Yelle, D.J.; Plaza, N.Z.; Lorenz, L.F.; Ching, D.J. Integrating multiscale studies of adhesive penetration into wood. *For. Prod. J.* **2018**, *68*, 340–348. [CrossRef]
49. Redman, A.L.; Bailleres, H.; Turner, I.; Perré, P. Characterisation of wood water relationships and transverse anatomy and their relationship to drying degrade. *Wood Sci. Technol.* **2016**, *50*, 739–757. [CrossRef]
50. Dadswell, H.E. *The Anatomy of Eucalypt Woods*; Paper No. 66; Forest Products Laboratory, Division of Applied Chemistry Technological, Commonwealth Scientific and Industrial Research Organisation (CSIRO): Canberra, Australia, 1972.
51. Queensland Government. Wood Properties and Uses of Australian Timbers: Spotted Gum. 2021. Available online: <https://www.business.qld.gov.au/industries/farms-fishing-forestry/forests-wood/properties-timbers/spotted-gum> (accessed on 12 January 2021).
52. Kumar, R.N.; Pizzi, A. *Adhesives for Wood and Lignocellulosic Materials*, 1st ed.; John Wiley & Sons, Inc.: Hoboken, NJ, USA; Scrivener Publishing LLC: Beverly, MA, USA, 2019.
53. Sage, G.N.; Tiu, W.P. The Effect of Glue-line Voids and Inclusions on the Fatigue Strength of Bonded Joints in Composites. *Composites* **1982**, *13*, 228–232. [CrossRef]
54. Duhamel, G. Why You Have Voids in Your Cured Adhesive. 2016. Available online: <https://www.appli-tec.com/articles/voids-cured-adhesive/> (accessed on 11 January 2021).
55. Permabond. How to Eliminate Air from Adhesive and Remove Air from a Bond Joint. 2021. Available online: <https://www.permabond.com/resource-center/eliminate-air-adhesive-remove-air-bond-joint/> (accessed on 10 January 2021).

56. Nicholas, D.D.; Thomas, R.J. The Influence of Enzymes on the Structure and Permeability of Loblolly Pine. In Proceedings of the American Wood Preservers' Association, 22–24 April 1968; Volume 64, pp. 1–7.
57. Morrell, J.; Morris, P. Methods for Improving Preservative Penetration into Wood: A Review. In Proceedings of the International Group on Wood Preservation, IRG/WP 02-40227, Cardiff, UK, 12–17 May 2002.
58. Durmaz, S.; Yildiz, U.; Yildiz, S. Alkaline Enzyme Treatment of Spruce Wood to Increase Permeability. *Bioresources* **2015**, *10*, 4403–4410. [[CrossRef](#)]
59. Mantanis, G.I.; Young, R.A.; Rowell, R.M. swelling of wood. part ii. swelling in organic liquids. *Holzforschung* **1994**, *48*, 480–490. [[CrossRef](#)]
60. Archer, K.; Lebow, S. Chapter 9. Wood Preservation. In *Primary Wood Processing. Principles and Practice*, 2nd ed.; Walker, J., Ed.; Springer: Dordrecht, The Netherlands, 2006.
61. Thaler, N.; Lesar, B.; Humar, M. Performance of Selected Copper Amine Based Wood Preservative Supplemented with Wood Swelling Agents. *Wood Res.* **2012**, *57*, 453–462.
62. Rak, J.R. *Penetration by and Stability of Copper-Chrome-Arsenic Wood Preservatives*; Information Report OPX 87E; Environment Canada, Forest Service, Eastern Forest Products Laboratory: Gatineau, QC, Canada, 1975.
63. Gjovik, L.R. Treatability of Southern Pine, Douglas-fir and Engelmann Spruce Heartwood with Ammoniacal Copper Arsenate and Chromated Copper Arsenate. In Proceedings of the American Wood Preservers Association, Kansas City, USA, 17–20 April 1983; Volume 79, pp. 18–30.
64. Rhatigan, R.G.; Freitag, C.; El-Kasmi, S.; Morell, J.J. Preservative Treatment of Scots Pine and Norway spruce. *For. Prod. J.* **2004**, *54*, 91–94.
65. Rademacher, J. Improving Penetration of Copper in Micronized Copper Azole Pressure Treated Michigan Red Pine. Masters's Thesis, Forestry, Michigan State University, East Lansing, MI, USA, 2011.
66. Kumar, S.; Morrell, J.J. Penetration and Absorption of Different CCA compositions in Six Western Conifers. *For. Prod. J.* **1989**, *39*, 19–24.
67. Meijer, M.; Van de Velde, B.; Militz, H. Rheological Approach to the Capillary Penetration of Coating into Wood. *J. Coat. Technol.* **2001**, *73*, 914. [[CrossRef](#)]
68. Chen, D.; Berk, R.; Kubes, G. Effect of Different Wood Species on the Performance of Surfactants Used to Improve the Rate of Kraft Liquor Penetration into Chips. *Cellul. Chem. Technol.* **2010**, *45*, 51–56.
69. Kayihan, F. Method of Perfusing a Porous Workpiece with a Chemical Composition Using Cosolvents. U.S. Patent 5,094,892, 10 March 1992.
70. Acda, M.N.; Morrell, J.J.; Levien, K.L. Supercritical Fluid Impregnation of Selected Wood Species with Tebuconazole. *Wood Sci. Technol.* **2001**, *35*, 127–136. [[CrossRef](#)]
71. Wood, K.C.; Morrell, J.; Leggate, W. Enhancing the Durability of Low Durability Eucalyptus Plantation Species: A Review of Strategies. In Proceedings of the International Research Group on Wood Protection, IRG/WP 20-40910, Bled, Slovenia, 10–11 June 2020.
72. Kumar, C. Investigation of an Innovative Preservative Treatment of Green Timber Using Compression. *Wood Mater. Sci. Eng* **2021**. [[CrossRef](#)]
73. Itoh, S.; Rahman, G.M.S. Underwater Shock Wave Treatment of Wood for Improving the Fire Protecting Properties. *Trans. Mater. Res. Soc. Jpn.* **2010**, *35*, 537–541. [[CrossRef](#)]
74. Tanaka, T.; Avramidis, S.; Shida, S. A Preliminary Study on Ultrasonic Treatment Effect on Transverse Wood Permeability. *Madera. Cienc. Technol.* **2010**, *12*, 3–9. [[CrossRef](#)]
75. Frihart, C.R.; Brandon, R.; Beecher, J.F.; Ibach, R.E. Adhesives for achieving durable bonds with acetylated wood. *Polymers* **2017**, *9*, 731. [[CrossRef](#)]