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Bonding Wood Veneer with Biobased Poly(Lactic Acid) Thermoplastic Polyesters: Potential Applications for Consolidated Wood Veneer and Overlay Products

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Abstract: This study reports on the use of poly(lactic acid) (PLA) as a renewable thermoplastic adhesive for laminated panels using birch, spruce, and pine veneers. Consolidated panels were prepared from veneer and PLA foils by hot-pressing from 140 to 180 °C to achieve minimum bondline temperatures. Evaluation of panel properties revealed that the PLA-bonded panels met minimum tensile strength and internal bond strength performance criteria. However, the adhesion interface which developed within individual bondlines varied with distinctions between hardwood and softwood species and PLA grades. Birch samples developed greater bondline strength with a higher pressing temperature using semi-crystalline PLA, whereas higher temperatures produced a poorer performance with the use of amorphous PLA. Panels formed with spruce or pine veneers had lower bondline performance and were also similarly distinguished by their pressing temperature and PLA grade. Furthermore, the potential for PLA-bonded laminated panels was demonstrated by cold water soak testing. Samples exhibiting relatively greater bondline adhesion had wet tensile strength values comparable to those tested in dry state. Our study outcomes suggest the potential for PLA bonding of veneers and panel overlays with the added benefits of being renewable and a no added formaldehyde system.

Keywords: poly(lactic acid); PLA; PLA bondlines; wood adhesives; plywood; wood veneer laminates; wood plastic composites

1. Introduction

Globally, the momentum for change toward sustainable, renewable, and recyclable products is growing rapidly across many industrial and market sectors. This desire transcends the packaging and composite sectors where environmental indices, recyclability, and legislative regulations are the principal drivers for change [1,2]. In wood-based panel and composite applications, emissions and indoor air quality along with environmental product declarations are at the forefront of product developments in the furniture and construction sectors [3–6]. While formaldehyde use and emissions have featured prominently in these developments, the substitution of petrochemical components in wood binders is also topical. Such activities are creating opportunities for new adhesives and approaches to bonding wood along with adaptations to more traditional approaches.

For several decades, the use of polyolefin plastics with wood fiber has been an active research area including commercial applications as wood plastic composite (WPC) materials [7,8] and examples of

use as hot melts in veneer products [9]. Biobased and biodegradable polyesters such as poly(lactic acid) (PLA) and more broadly poly(hydroxy alkanoates) (PHAs) are viewed as replacements for polyolefin plastics in these applications [10]. To date, PLA has been extensively evaluated in food, beverage packaging and various consumer product applications [1]. As a thermoplastic polymer, PLA can be thermally processed by compounding, extrusion, and injection molding in a similar way to other industrial and consumer plastics [11]. Moreover, PLA can be compounded with wood fiber to form a range of products suitable for a wide range of WPC applications which include emerging uses in three-dimensional (3D) and four-dimensional (4D) printed forms [12–14]. Poly(lactic acid) use has been determined to offer greater bonding potential with wood than comparable polyolefin plastics [15,16]. When processing PLA with wood the bonding strength formed is additionally dependent on PLA polymer properties, the processing temperatures employed, and the wood species used [17]. Moreover, qualitative studies have revealed that the development of interfacial bonding between PLA and wood was associated with PLA migration away from the bondline at the surface to the wood bulk resulting in physical interlocking between the wood microstructure and the PLA polymer matrix [18]. However, although the potential for PLA use in veneer overlay and laminated products has been demonstrated [15,17–19], these fundamental approaches are yet to be explored at any scale.

In the current study, the manufacture of consolidated veneer products was evaluated using PLA in foil sheet form. Laboratory-scale laminated veneer lumber was produced employing semi-crystalline and amorphous PLA grades with different wood species and processing temperatures. Both hardwood and softwood veneers were processed into multi-layered laminates via a traditional hot-pressing approach with the use of two polymer grades to distinguish contributions of PLA melt and thermoplastic behaviors to adhesion performance. The resulting wood panel products were evaluated for their mechanical performance and water resistance properties through testing individual PLA bondlines, to understand the impacts of employing different PLA polymer grades and processing requirements for adaptation to larger-scale implementation and production.

2. Materials and Methods

2.1. Materials

The wood veneers were European white birch (B. pendula, 1.60 mm thickness), spruce (Picea abies, 1.60 mm thickness), and radiata pine (Pinus radiata D. Don, 2.65 mm thickness) with each cut into sheets with dimensions of 150 × 300 mm^2. Prior to pressing, the veneers were equilibrated by conditioning at a temperature of 20 °C and 30% relative humidity (RH). Two polymer grades of poly(lactic acid) were obtained from Natureworks LLC (USA) and were 3052D and 4060D, semi-crystalline and amorphous grades, respectively.

2.2. Poly(Lactic Acid) (PLA) Foil Preparation

Poly(lactic acid) beads were vacuum dried (40 °C) overnight before extrusion. Foils were prepared from both grades of PLA employing film extrusion. The PLA was processed at 200 °C in a laboratory extruder, and then calendared to achieve the extruded film as PLA foil. The PLA foils produced were in two nominal thicknesses of 0.3 and 0.5 mm. Foils were stored in sealed bags and cut to 150 × 300 mm^2 dimensions prior to hot-pressing.

2.3. Hot Pressing PLA Laminated Veneer Panels

Different laminate assembles were used according to the veneer thickness and number of laminae. For both birch and spruce, typically 5-ply laminated veneer lumber assemblies with a thickness of 8 mm were produced using veneers interspersed with layered PLA foils. For radiata pine veneer, 3-ply laminates were produced to a similar thickness as those formed with birch and spruce veneer. Using a 0.5 mm foil thickness provided a PLA application rate of ca. 430 g/m^2, whereas 0.3 mm foils provided a ca. 265 g/m^2 application rate. A simple press schedule was employed, pressing to

stops using a maximum pressure of 45 kN at three different temperatures of 140, 160, and 180 °C. After pressing, consolidated laminates were removed from the press, and then immediately cooled to ambient temperature maintaining a modest (5 kN) compression. Total hot-pressing and cooling times were dependent on pressing temperature and achieving a minimum inner core bondline temperature (Table 1). Consolidated panels were recovered, trimmed to size removing any PLA squeeze out, and then stored at 23 °C and 50% RH to equilibrate for at least 5 days.

Internal panel temperature profiles were obtained for each pressing temperature. A thermocouple was inserted into the core bondline of each laminate assembly and the temperature was measured until this core bondline equilibrated to the platen temperature. Temperature profiles were used to refine the total press schedule time and cooling step at each pressing temperature (Table 1).

Table 1. Nominal hot-pressing and cooling times for birch and spruce 5-ply laminate assemblies at different pressing temperatures and poly(lactic acid) (PLA) foil thicknesses.

Pressing Temperature (°C)	Hot-Pressing Time (s)	Cooling Time (s)	PLA Foil Thickness (mm)
140	740	800	0.3
160	920	950	0.3, 0.5
180	725	1020	0.3, 0.5

Note: 3-ply radiata pine laminates use same hot-press and cooling schedules.

2.4. Tensile Testing

Tensile testing was generally conducted to ASTM D906-98(2017) and EN314-1(2014) Standards criteria. Specimens (25 × 100 mm^2) were cut from panel samples, and then additionally cross cut for testing individual bondlines and, for 5-ply laminates, identified either as the surface, outer, or core, inner bondlines. For 3-ply laminates, specimens were identified and tested as core, inner bondlines. Specimens were equilibrated at 23 °C and 50% RH for at least 72 h before testing.

Shear strength testing was conducted on an Instron 5566 universal test machine equipped with a video extensometer in tensile mode. Crossbeam speeds of 0.5 mm/min (up to 0.4% strain), and then 50 mm/min were used reporting failing load, calculated tensile modulus, and, where stated, bondline wood failure. At least 5 specimens were tested per veneer species, PLA grade, foil thickness, and pressing temperature combination for each bondline. Testing results were statistically compared using SAS JMP 14.2.0 (see Supplementary Materials). Additionally, to reduce the impact of clamping pressures, pneumatic grip pressures were varied with 0.4 MPa for dry-tested birch and spruce specimens and reduced to 0.2 MPa for water soaked and radiata pine (dry and water soaked) specimens.

For cold water soaking prior to tensile testing, test specimens were immersed in water (20 °C) for 24 h. Specimens were recovered from the water, blotted with paper to remove excess water, and then tensile tested in their wet state using the above procedure. For those samples tested after hot water soaking, water immersion was first undertaken at 60 °C, for 6 h. After soaking, the samples were transferred into cold water (20 °C, <30 min), removed, blotted, and tested in a wet state, as above.

2.5. Internal Bond Strength Testing

Test specimens (50 × 50 mm^2) were cut from panel samples. These were adhered between internal bond strength (IB) test blocks using a hot-melt polyurethane-based adhesive. Glued IB specimens were, then, allowed to equilibrate at 23 °C and 50% RH prior to testing. Internal bond strength testing was conducted on an Instron 5566 universal test machine at a rate of 0.5 mm/min. At least 5 specimens were tested per veneer species, PLA type, and pressing temperature combination. Testing results were statistically compared using SAS JMP 14.2.0 and not significantly different means were grouped.

3. Results

3.1. Press Cycle Development

Hot-pressed panels were prepared from wood veneers and PLA foils at three different pressing temperatures of 140, 160, and 180 °C (Table 1, Supporting Information). Panel core bondline temperature profiles obtained using birch, spruce, and pine veneers across these pressing temperatures are shown in Figure 1. Generally, an initial, rapid temperature increase was observed on press closure, typical of conventional plywood manufacture. At ca. 100 °C, the panel core temperature began to plateau before increasing to become equivalent to the platen temperature with extended time (>700 sec). Variation in these core temperature profiles were evident with different pressing pressures, as well as number and thickness of laminate veneers used (Supplementary Materials). Nonetheless, the heating profiles were very similar for both spruce and birch (1.6 mm) veneers, and for both amorphous or semi-crystalline forms of PLA and the two PLA foil thicknesses (0.3 and 0.5 mm). With pine, a thicker veneer (2.65 mm), a relatively broader heat profile was evident for these 3-ply laminates. In this case, greater water content in the developing panel likely hindered the rapid temperature increase above 100 °C seen with the other veneer (1.6 mm) types. However, after water evaporation, the conductive heat transfer continued, achieving a similar core temperature profile to birch and spruce.

Across the different hot-press temperatures and heating rates, as shown in Figure 1, a common hot-press schedule for 5-ply laminates was developed (Table 1). This schedule was based on achieving a requisite, minimum inner PLA bondline temperature [17], together with accommodating any variations in veneer or foil thicknesses. Importantly, the schedule avoided any significant thermal degradation of these materials. Furthermore, given that the panel hot-pressing concluded with PLA in a near or molten state [11], cooling of the hot-pressed panel under compression was necessary to cool down the PLA bondlines below 60 °C and ensure the integrity of the resulting panels. This post-pressing step was consistent with previous small-scale manufacture of PLA laminates [15,17], as well as wood-plastic composite manufacture [7]. As with the hot-pressing cycle, the duration of this cooling panel compression step varied with platen temperature (Table 1). Overall, the panel processing time was arguably impractical for a production environment, but necessary in this fundamental study to distinguish influences of pressing temperature and PLA properties on the resulting laminated panel performance.

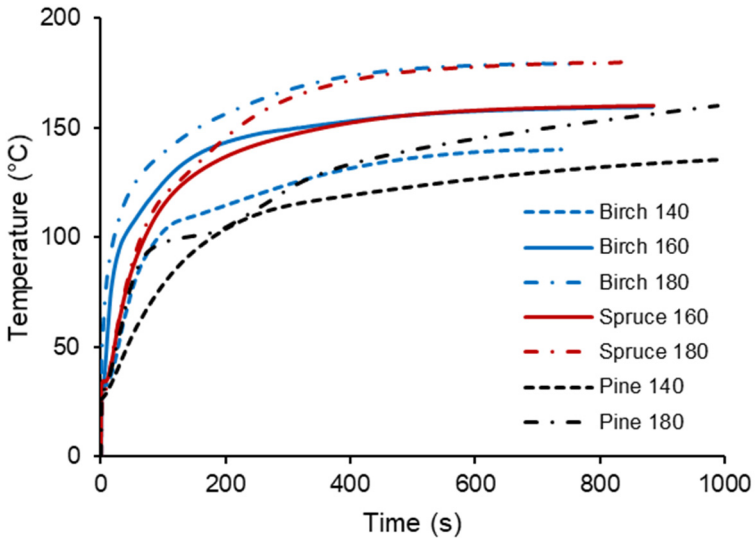

Figure 1. Representative core temperature profiles for consolidated panels with different veneer types and pressing temperatures using PLA foils (0.3 mm).

3.2. Wood-PLA Laminate Performance

Laminated panels were evaluated by standard tensile shear testing to establish any differences in bondline strength with pressing temperature, as well as distinctions between inner and outer bondlines across the wood species and the PLA grade used (Table 2, Supplementary Materials). With the exception of spruce veneer pressed with amorphous PLA, the outer bondline strength was significantly greater than core, inner bondlines of laminated panels pressed at a lower temperature. Using birch veneer and semi-crystalline PLA, the outer bondline strength significantly increased (p-value: 0.04) from 6.9 to 8.0 MPa as pressing temperature increased from 140 to 180 °C. There was no significant difference in tensile strength for inner bondlines which ranged from only 5.3 MPa (140 °C) to 5.8 MPa (180 °C). The use of amorphous PLA with birch veneer also gave significantly greater outer bondline strength (4.5 to 8.7 MPa) than the inner bondlines (5.2 to 6.5 MPa) for the 140 and 160 °C pressing temperatures, but, in contrast to semi-crystalline PLA use, relatively lower tensile strength values were found when pressing at higher temperatures. For example, greater inner bondline strength was found using amorphous PLA at 140 °C (8.7 MPa) as compared with the semi-crystalline (6.5 MPa), whereas at 180 °C, the amorphous PLA inner bondline strength decreased to 4.5 MPa as compared with the semi-crystalline PLA value of 5.8 MPa. Given the contrasting tensile strength values, individual bondlines were assessed by microscopy (Figure 2). Visual comparisons of bondlines revealed thinner bondlines at higher press temperatures. This was a result of lower melt viscosity during heating [11,18], but concurrently, microscopy also revealed open spaces and gaps that increased between the veneers for both PLA grades undergoing higher pressing temperatures.

Table 2. Summary of tensile testing results for inner and outer bondlines of laminated panels tested dry.

Sample		**Dry, Outer**		**Dry, Inner**		***p*-Value**
PLA Grade	Pressing Temperature (°C)	Tensile Strength (MPa)		Tensile Strength (MPa)		-
		Average	Standard Deviation	Average	Standard Deviation	-
Birch						
Amorphous	140	8.7	0.3	6.5	1.3	0.04 *
	160	7.2	0.7	5.4	0.6	0.002 *
	180	4.5	0.4	5.6	1.1	0.004 *
Semi-crystalline	140	6.9	0.5	5.3	0.8	0.008 *
	160	6.2	0.4	5.9	1	0.61
	180	8	0.7	5.8	1.8	0.054
Spruce						
Amorphous	140	5.5	0.4	5.3	2.1	0.88
	160	6.5	1.3	4.7	0.7	0.03 *
	180	4.3	0.8	4.3	0.4	0.99
Semi-crystalline	140	5.6	1.2	3.7	1.1	0.03 *
	160	5.5	0.4	3.3	0.8	0.002 *
	180	5	1.2	4.5	0.4	0.45
Pinus Radiata						
Amorphous	140	-	-	3.4	0.4	-
	160	-	-	5.3	0.8	-

Note: Testing used at least 5 replicates per individual bondline variable; * indicate statistically significant differences.

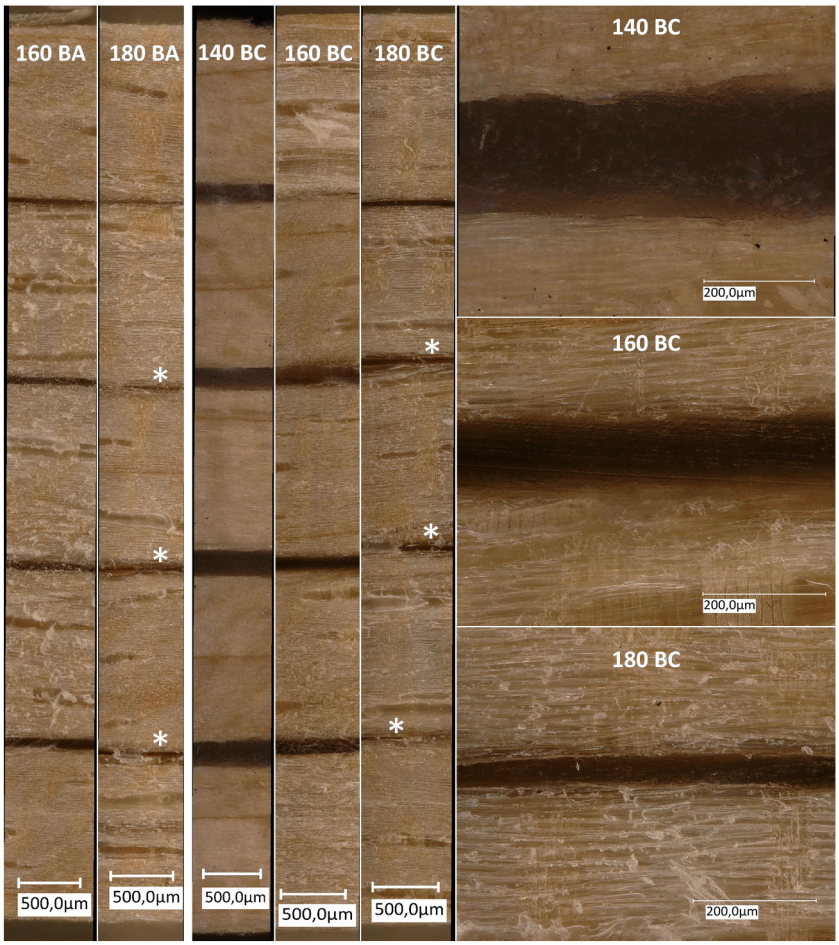

Figure 2. Comparison of birch laminated panel samples produced with amorphous (BA) and semi-crystalline (BC) PLA grades. Asterisks (*) indicate areas of unbonded veneer surfaces.

For laminated panels produced with spruce veneer and the semi-crystalline PLA, there were differences in outer bondline strength values which ranged between 4.3 and 6.5 MPa across the three pressing temperatures (Table 2 and Figure 3). In contrast, for spruce veneer pressed with the amorphous PLA, the bondline strength was significantly lower at 180 °C pressing temperature as compared with 160 and 140 °C (p-values 0.01 and <0.001, Supplementary Materials). For inner bondline strength values, semi-crystalline PLA gave significantly lower tensile strength values at 140 and 160 °C as compared with birch veneer use (p-value <0.04). Amorphous PLA foils with spruce veneer similarly produced significantly lower inner bondline strength at 140 and 160 °C (4.7 and 5.3 MPa, respectively) with no significant difference between the amorphous and semi-crystalline PLA use at 180 °C. This relatively similar performance of spruce samples across pressing temperatures and PLA grades was not unexpected, given that using this softwood species has similarly produced lower bonding strength previously with PLA and other synthetic polymers [15,17,18].

From Table 2, it is evident that the difference between outer and inner bondline strength was dependent on the veneer, pressing temperature, and PLA polymer grade combination. Regardless of pressing temperature, all amorphous PLA-birch samples had higher outer bondline strength than inner bondlines. Using the semi-crystalline PLA, this difference was only significant for the 140 °C pressing temperature (p-value 0.01, Supplementary Materials). However, for spruce veneer, the effect of the pressing temperature on the bondline strength was more apparent for samples produced with semi-crystalline PLA. At 140 and 160 °C, the outer bondline strength was greater than the inner bondline strength with p-values of 0.03 and 0.01, respectively. Interestingly, amorphous PLA-spruce samples

only differed in outer and inner bondline strength at 160 °C pressing temperature. Inner bondlines of semi-crystalline PLA-spruce samples had comparable performance across pressing temperatures (140 °C, 3.7 MPa; 160 °C, 3.3 MPa; 180 °C, 4.5 MPa). At 180 °C pressing temperature, the inner bondline strength was only affected by the wood veneer choice when amorphous PLA was used.

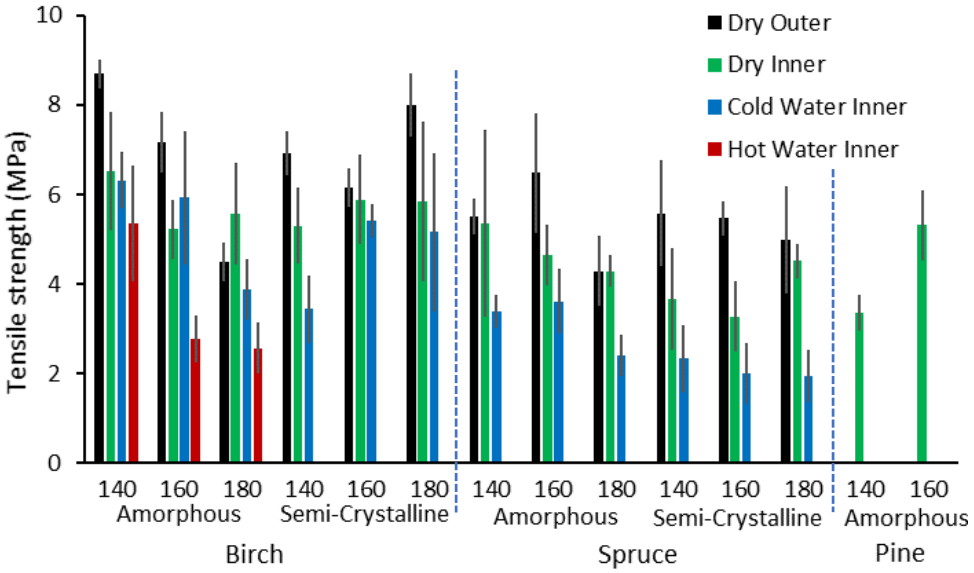

Figure 3. Comparison of tensile bondline strength for laminated panel samples tested dry and after 24 h of cold water or 6 h of hot water soaking.

Pressing with radiata pine veneer (2.65 mm) was undertaken to determine any impacts on using this softwood species and as a thicker veneer. On the basis of the panel temperature profiles (Figure 1) and results for birch and spruce, hot-pressing was undertaken at only 140 and 160 °C. The inner bondline strength (5.3 MPa, Table 2) formed between pine veneers and amorphous PLA at 160 °C was comparable to that of spruce pressed at any of the three tested temperatures (*p*-values at 140 °C, 0.03; at 160 °C, 0.01; at 180 °C, 0.01, Supplementary Materials). At 140 °C, the pine inner bondline strength (3.4 MPa) was lower than the comparable spruce sample (*p*-value: 0.03). This suggests an increased pressing temperature (160 °C) was beneficial with pine veneer providing a further contrast to results found for birch and spruce use.

Evaluation of 0.3 and 0.5 mm PLA foil thicknesses was undertaken at 160 and 180 °C hot-pressing temperatures, and there were no significant differences in bondline strength values found (Table 3). Testing revealed the use of 0.5 mm amorphous foils at a PLA application rate of ca. 430 g/m^2 with birch veneer gave higher bondline strength than using 0.3 mm foils (ca. 265 g/m^2) at both 160 and 180 °C. In the case of spruce veneer, a thicker PLA foil contributed to a lower average bondline strength, but, as above, this was not statistically different at either temperature. Similarly, the use of 0.3 and 0.5 mm PLA foils did not impact bondline strength formed between pine veneers. Overall, it can be concluded that there was no distinction using the 0.3 mm and 0.5 mm PLA foils, which was considered to be the practical range of PLA thicknesses most likely to be used in lamination and panel assembly procedures [11].

Results from internal bond strength evaluations revealed laminated panels formed with PLA contrast achieved minimum performance criteria (Figure 4). While birch samples prepared with amorphous PLA were distinguished by the nature and variability in IB sample failures. Sample failures included both inner and outer bondlines and the wood substrate, contributing to a range of individual IB values for each sample. However, there were no significant differences in these IB values (ca. 1.6 MPa) across the three pressing temperatures. For semi-crystalline PLA-birch samples, the 140 °C sample had significantly lower IB (0.98 MPa) as compared with hot-pressing at either 160 °C

(*p*-value <0.001) or 180 °C (>1.3 MPa, *p*-value 0.007). With spruce veneer, IB testing revealed samples prepared with amorphous PLA to have comparable strength (1.6 MPa) to the corresponding birch samples. Spruce veneer samples pressed at 180 °C had a significantly lower IB (1.25 MPa) as compared with the 140 °C (1.57 MPa and *p*-value 0.02) and 160 °C (1.56 MPa and *p*-value 0.002). Spruce samples prepared with semi-crystalline PLA all showed lower IB strength (ca. 1.0 MPa), which was independent of pressing temperature and comparable to the 140 °C birch-semi-crystalline PLA sample. Overall, across all samples, IB testing provided minimum strength values with average values generally greater than ca. 1 MPa. It was noted that sample failures were variable, reflecting the different bondline strengths of individual bondlines established in tensile shear testing (Table 2 and Figure 3). It is this distinction that accounts for the similarity in IB values which contrast shear strength evaluations where testing involved individual bondlines (Table 2).

Table 3. Tensile strength values for samples formed with different amorphous PLA foil thickness compared with all samples produced in this study.

Species	Pressing Temperature (°C)	PLA Foil Thickness (mm)	Tensile Strength (MPa)				*p*-Value
			Samples per Foil Thickness		All Samples		
			Average	Standard Deviation	Average	Standard Deviation	
Birch	160	0.5	5.47	0.28	5.4	0.6	0.91
		0.3	5.38	0.93			
	180	0.5	5.7	1.97	5.6	1.1	0.78
		0.3	5.19	1.04			
Spruce	160	0.5	4.35	0.39	4.7	0.7	0.43
		0.3	4.85	0.84			
	180	0.5	4.08	0.14	4.3	0.4	0.29
		0.3	4.42	0.41			
Radiata pine	160	0.5	5.21	0.83	5.3	0.8	0.80
		0.3	5.45	1.06			

Note: Testing used at least 5 specimens per sample.

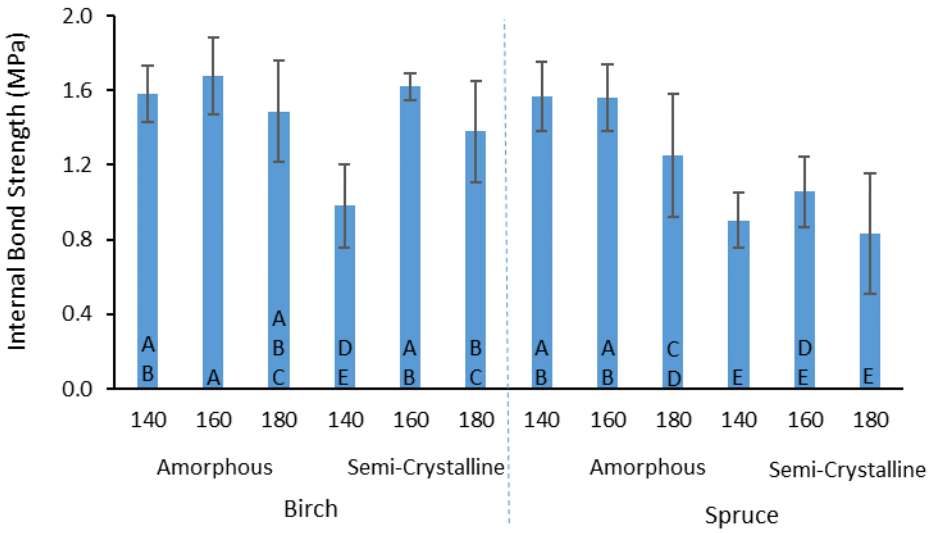

Figure 4. Comparison of internal bond testing results for laminated panel samples, including grouping of results. Levels not connected by the same letter are significantly different.

3.3. Bondline Performance on Water Soaking

To understand any potential water resistance of the PLA bondlines, both cold and hot water soaking performance of wood-PLA laminates was evaluated (Table 4). After 24 h of soaking, birch samples formed with semi-crystalline PLA at 160 and 180 °C had inner bondline wet strength comparable to strength values attained in a dry state at 5.4 and 5.9 MPa (*p*-value 0.06) and 5.2 MPa/5.8 MPa (*p*-value 0.17), respectively. For the 140 °C sample, water soaking led to significantly lower bond strength (3.4 MPa) than when tested dry (5.3 MPa, *p*-value 0.002). Testing of the amorphous PLA-birch samples after water soaking found the wet strength for the 160 °C sample (5.9 MPa) was comparable to dry testing (*p*-value 0.73), whereas the 140 and 180 °C birch samples had significantly lower wet strength, 6.3 and 3.9 MPa, respectively, (*p*-values 0.02 and 0.01). Water soaking laminate panels formed with spruce veneer resulted in significantly lower bondline strengths (1.9–3.6 MPa) for both PLA grades and all pressing temperatures. Across these spruce samples, water soaking revealed greater performance of the 160 °C sample formed with amorphous PLA as compared with semi-crystalline PLA or pressing at 180 °C, consistent with trends in dry testing performance.

Table 4. Summary of tensile testing results for inner bondlines of laminated panels, tested dry and tested after 24 h of cold water soaking.

Sample		**Dry**		**Cold Water Soaking**		***p*-Value**
PLA Grade	Pressing Temperature (°C)	Tensile Strength (MPa)		Tensile Strength (MPa)		-
		Average	Standard Deviation	Average	Standard Deviation	-
Birch						
Amorphous	140	6.5	1.3	6.3	0.6	0.02 *
	160	5.4	0.6	5.9	1.5	0.73
	180	5.6	1.1	3.9	0.7	0.005 *
Semi-Crystalline	140	5.3	0.8	3.4	0.8	<0.001 *
	160	5.9	1	5.4	0.4	0.06
	180	5.8	1.8	5.2	1.8	0.17
Spruce						
Amorphous	140	5.3	2.1	3.4	0.4	0.001 *
	160	4.7	0.7	3.6	0.7	0.006 *
	180	4.3	0.4	2.4	0.5	<0.001 *
Semi-Crystalline	140	3.7	1.1	2.3	0.7	0.002 *
	160	3.3	0.8	2	0.7	0.001 *
	180	4.5	0.4	1.9	0.6	<0.001 *
Pinus Radiata						
Amorphous	140	3.4	0.4	-	-	-
	160	5.3	0.8	-	-	-

Note: Testing used at least 5 replicates per individual bondline variable; * indicate statistically significant differences.

With hot water soaking (60 °C), the bondline strength was generally found to be lower for the selected birch samples tested (Figure 2). With these amorphous PLA-birch samples, hot water soaking led to significantly lower inner bondline strength for the 160 and 180 °C pressed samples. This distinction in bondline strength after hot water soaking differed from that of the 24 h cold water soak results which revealed both samples to have similar tensile strength values in both dry and wet states (Table 2). For the 140 °C sample, after hot water soaking, the inner bondline strength was similar to that in the dry state, which was also the case after 24 h of cold water soaking.

4. Discussion

Overall, satisfactory bondline performance was found for the laboratory-scale PLA-bonded panels produced in this study. Generally, this performance met the requirements of standards, as well as matched results from smaller-scale, fundamental evaluations undertaken previously with a range of wood species and processing temperatures [15,17,18]. As shown in Table 1 and Figure 3, tensile shear strength and IB values met minimum criteria for interior grade performance of veneer products, as well as that required for bonding of overlays to wood-based panels.

Across testing results and study findings, there were key distinctions in the performance of bondlines attributed to pressing temperature, PLA polymer characteristics, and the wood species used. The impact of polymer properties was most evident using birch veneer with contrasts found between the two PLA grades. With semi-crystalline PLA, greater bondline performance was achieved pressing above 160 °C consistent with exceeding the PLA melt temperature enabling PLA to flow several wood cells from the bondline (Figure 2) and development of the PLA-wood adhesion interface as previously seen for PLA bondlines [11,17]. In contrast, the use of amorphous PLA resulted in lower bondline performance at higher temperatures which could be attributed to lowered PLA viscosity promoting greater PLA movement into the wood microstructure and away from bondlines, in particular, the outer bondlines, as demonstrated previously [17,18]. This greater mobility of PLA away from the bondline is akin to over-penetration of liquid resins in traditional plywood manufacture [20]. The formation of gaps and unbonded areas between the veneer surfaces at higher temperatures (Figure 2) was also attributed to this mobility. Nonetheless, semi-crystalline PLA still seemed to be able to generate higher tensile strengths due to a greater functionality and compatibility of this polymer with wood [15,16]. Arguably, a relatively higher tensile strength achieved with 0.5 mm PLA foil for amorphous PLA-birch samples (Table 3) was likely due to greater PLA retained at the PLA-wood interface than achieved with the use of 0.3 mm foil. Wood ultrastructure and porosity along with wood chemistry were determinates [15–18] for distinctions between birch, spruce, and pine veneer use. The lower performance of both softwood species was primarily due to a reduced extent of physical interlocking achievable at the PLA-wood adhesion interface demonstrated previously [18].

Matching pressing temperature with PLA grade and wood species can achieve satisfactory adhesion at the PLA-wood interface (Table 2). This performance was also comparable to both urea formaldehyde resin and phenolic resin interior grade adhesive performance [20]. Most PLA-laminate samples achieved minimum tensile strength (>4 MPa) and IB values (>0.8 MPa) typically expected of these formaldehyde-based adhesives in laminated veneer and overlay products. These findings also mirror those from a recent, unrelated study applying PLA in emulsified forms to bond laboratory plywood [19].

Cold water soak testing also demonstrated the potential water resistance of the PLA-wood bond. For those better performing combinations of PLA grade and temperature for each wood species, the potential water resistance of bondlines was evident with comparable water soak tensile strength values as those tested dry. However, those samples which had exhibited lower bondline performance in their dry state also showed relatively poorer water resistance. This was particularly evident for spruce samples or the birch samples produced at high temperature with amorphous PLA or the semi-crystalline PLA sample pressed at 140 °C. Hot water (60 °C) soaking generally gave poorer water soak performance. This distinction is attributed to PLA bondlines being held above the polymer glass transition temperature (Tg), a critical temperature previously determined for PLA adhesion and composite performance [17]. With PLA in a softened state above its Tg, it is likely unable to resist the dimensional change of wood on water immersion. With this in mind, it is prudent to undertake further, long-term cold water soak testing to further understand the potential exhibited by PLA-bonded wood veneer products produced in this study and those applying PLA in other forms [19]. Nonetheless, the better performing amorphous PLA-birch bondline formed at 140 °C was found to retain strength in a wet state after hot water soaking (Figure 3), demonstrating the potential for PLA bonded laminates.

In this study, unrealistic hot-press cycle times for industrial transfer were applied to ensure PLA bondlines attained minimum temperatures matching fundamental PLA polymer properties such as melt rheology to relate to PLA-wood bondline performance. There was also a desire to develop a simple, practical approach toward implementing PLA laminate bonding at scale, and avoid introducing water or complex preheating steps. To realize PLA foil applications in veneer consolidation, this study revealed, in practice, that a combination of PLA grade, wood species, veneer moisture, and internal panel heating rate (Figure 1) would be required to achieve minimum performance of consolidated veneer products. It was evident that higher temperatures beyond conventional plywood hot-pressing regimes would be required to significantly shorten press times approaching those of traditional wood-based panel manufacturing. We view application of PLA in foil sheet form to be beneficial for reducing pressing time, because this does not contribute water to the panel during the pressing process. Conventionally dried veneer (<2%) would further assist achieving internal panel temperatures and avoid steam generation, as evident in Figure 1. With this combination of veneer preparation, platen temperature and PLA grade selection, appropriate press schedules can be developed which rapidly heat the consolidating panel and maximize internal panel temperature to produce significantly shorter press cycle times than shown in Figure 1. This can be adjusted for the number of plys and panel thickness to approach press cycles similar to traditional veneer laminate manufacturing (Supplementary Materials). We have also shown the potential of applying PLA bonding for veneer overlays. With the added benefits of being renewable and a no added formaldehyde system, bonding with a thermoplastic ester such as PLA could have additional advantages over traditional adhesives such as post-forming modifications.

5. Conclusions

This study has successfully evaluated the impacts of pressing temperature and PLA grade on the production and performance of PLA-laminated veneer wood panels. Satisfactory bondline adhesion performance was achieved with birch, spruce, and radiata pine veneer as evidenced with both panel tensile strength (>4 MPa) and internal bond strength (>1 MPa) testing. These panel properties were dependent on internal panel temperatures developed during hot-pressing which variously influenced the performance of outer and inner bondlines. Satisfactory bondline adhesion was developed using platen temperatures ranging between 140 and 180 °C with both birch and spruce veneer, with this further distinguished by using either amorphous or semi-crystalline PLA forms. For those better performing PLA laminated panels, similar bondline strength values were achieved after 24 h cold water soaking as with those tested dry. This was also the case for the 140 °C amorphous PLA-bonded birch panel after 6 h of hot water soaking. This wet strength performance is indicative of the adhesive bond integrity which can be formed with PLA when using optimal combinations of veneer species, PLA grade, and pressing temperature. Overall, our study outcomes have demonstrated the potential for PLA substitution into traditional veneer and overlay laminate production.

Supplementary Materials: The following are available online at http://www.mdpi.com/2079-6439/8/8/50/s1, Table S1: Sample acronyms used, Table S2: Inner bondline statistics: Student t-test least-significant-difference (LSD) threshold matrix, Table S3: Outer bondline statistics: Student t-test least-significant-difference (LSD) threshold matrix, Table S4: Comparison of inner and outer bondline strength, Table S5: Bondline wet strength statistics: Student t-test least-significant-difference (LSD) threshold matrix, Table S6: Comparison of bondline dry and wet strength, Table S7: Internal bond strength statistics: Student t-test least-significant-difference (LSD) threshold matrix, Figure S1: Examples of middle bondline temperature profiles of consolidated 7-ply panels formed spruce veneer at different pressing temperatures.

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