THERMOPLASTIC ENCAPSULATION OF WOOD STRAND

COMPOSITE USING A TIE-LAYER

By

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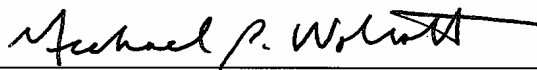To the Faculty of Washington State University:

The members of the Committee appointed to examine the thesis of STEVEN GERARD MICHAEL find it satisfactory and recommend that it be accepted.

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THERMOPLASTIC ENCAPSULATION OF WOOD STRAND COMPOSITE

USING A TIE-LAYER

Abstract

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One of the challenges in using wood or wood composites for exterior applications is durability. The approach taken in this study to increase the durability of structural composites for exterior applications was to apply an encapsulating thermoplastic to the surface. This study had two main objectives, to determine the optimum processing parameters for the maximizing the bond strength, and to evaluate the interfacial bond strength of the encapsulating thermoplastic. The optimum processing parameters were determined to be a platen temperature of 180ºC, a pressing pressure of 1035 kPa (150 psi) and a press time of 300 seconds. High density polyethylene (HDPE) was chosen to be used in this study because it is an environmentally friendly and an economical thermoplastic. It has been shown in previous studies (Wolcott and Englund 1999), however, that HDPE does not bond well to wood substrates. So to increase the bond strength of the HDPE to the wood substrate, a tie-layer was included in the specimen lay-up. To determine the bond strength two tests were conducted, a 90º peel test and a block shear test. To test the durability of the thermoplastic barrier layer an accelerated aging test was performed before the bond strength was tested. It was found that the tie-layers increased the bond strength by approximately 35 percent.

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CHAPTER ONE – INTRODUCTION

1.1 Introduction

One of the challenges in using wood or wood composites for exterior applications is durability. Some of the measures of durability are resistance to decay, weathering and moisture. In the past to overcome these environmental obstacles, a wood member for exterior applications was pressure treated with heavy metals. Before 2004 the most common of these treatments was chromated copper arsenate (CCA), which protects from both fungal decay as well as insect attack. However, because of the toxicity of the arsenic, the industry voluntarily banned the use of CCA for residential use (Stilwell and Gorny 1997). Some other common treatments included alkaline copper quaternary (ACQ) and copper azole (CA). Heavy concentrations of copper create the potential for galvanic corrosion of fasteners, connection hardware and flashing.

With increasing environmental concerns and regulations, there is a drive toward non-toxic methods of protecting wood against fungal decay and moisture. Wood composites, such as wood-plastic composites (WPCs), have been developed recently for exterior applications. WPCs are durable due in part to the plastic matrix which restricts moisture uptake which in turn limits the fungal decay. The plastic also creates a barrier to insect attacks (Morrell et al. 2006). However, there are some drawbacks to using WPCs. The biggest drawbacks are low stiffness of the WPC lumber and poor time dependent properties, such as creep. Most applications of the WPC lumber are for decking and railing systems. There are a wide range of engineered wood composites that are designed for structural applications; including LVL (laminated veneer lumber), PSL (parallel strand lumber), and OSL (oriented strand lumber). However, for these materials to be used in exterior applications, especially in direct contact with the ground, they

would need to be treated with preservative chemicals. A relatively begnign treatment that can be used to treat the structural wood composites, without corrosion problems, is a borate-based compound. The drawback to using borate-based compounds is that they are water soluble and can leach over time.

Therefore, we need a method to protect structural wood composites from moisture as well as contain the preservative chemicals and prevent leaching. In a study by Scheffer and Morrell (1997), specimens were placed inside a polyethylene bag (a "boot") and set in sifted forest soil. The specimens were kept in the soil for two years during which time the soil was kept moist. Even though the bags were not bonded to the specimens, they still showed marked improvement to the durability of the specimens to fungal decay.

The technique that will be examined in this study is a thermoplastic encapsulation (bonding of thermoplastic to the wood substrate) of wood composites to extend their durability. However, thermoplastics such as high density polyethylene (HDPE) do not bond well to wood substrates (Wolcott and Englund 1999). The way that WPCs overcome this obstacle is by breaking the wood down to wood flour so that the plastic can encapsulate the wood particles within the wood-plastic matrix. For solid wood and wood composites manufactured with larger size constituents such as veneers, veneer clippings, or strands, however, an effective tie-layer is necessary to both mechanically interlock as well as chemically bond the plastic to the wood substrate (Dai et al. 2004).

1.2 Objectives

The primary objective of this study is to evaluate the efficiency of a tie-layer for bonding HDPE to borate-treated Parallam® and solid sawn lumber. Specific goals to attain this objective are to:

1. Determine appropriate processing parameters, specifically temperature, pressure, and press time, for effectively bonding a thermoplastic barrier layer to the wood substrate.

2. Evaluate the bond strength between the thermoplastic barrier layer and the treated Parallam® and solid sawn lumber under initial and accelerated aging conditions.

1.3 Significance and Rationale

Thermoplastic encapsulation would improve the durability of wood composites by creating a barrier to moisture. Moisture infiltration and presence in wood and wood-based composites is a leading cause for further decay and degradation. By encapsulating wood composites, it would allow the use of less toxic and corrosive chemicals to preserve the wood without leaching. Since most polyolefins, such as HDPE, do not bond well to a polar material, such as wood, a tie-layer will be used to improve the bond. Most tie-layers are modified co-polymers that have a hydroxyl group graphed to the backbone of the polymer chain. This hydroxyl group then creates a hydrogen bond with the wood surface.

Many of the structural wood composites that are used in construction have very good mechanical properties, but since they are susceptible to environmental attack they are either pressure treated with chemicals or used in protected applications. One such composite is

Parallam$^{®}$. Parallam$^{®}$ was chosen for this study because it has "macro-pores", which will allow for greater mechanical interlocking of the thermoplastic and wood. This study will look at the effects of wood species and chemical treatment on the bond strength. For the Parallam$^{®}$ two wood species will be investigated: Douglas fir and southern pine. The solid wood, which is Douglas fir only, is used to compare the bond strength to that of Parallam$^{®}$. The solid wood was evaluated in both the radial and tangential grain directions to see if grain direction had any effect. All of the specimens in this study were treated with boric oxide (B_2O_3). Boric oxide is a water soluble chemical treatment that acts as a biocide. Other studies have found that modification to the wood substrate can have a negative effect on the bond strength (Kolosick et al. 1993). Encapsulating boric oxide treated material will further help in preventing the preservative from leaching over time. However, it is critical to understand the effects of boric oxide treatment on bond quality between the thermoplastic barrier layer and the wood substrate.

1.4 Structure of Thesis

This thesis is organized into two papers preceded by this introduction. The second chapter discusses determination of effective processing parameters to bond HDPE barrier layer to the wood substrate. To determine these parameters only one type of tie-layer and wood substrate were used. The third chapter deals with the evaluation of the bond strength using 90º peel and block shear tests before and after subjecting to accelerated aging. The final chapter summarizes the findings of the thesis and draws conclusions.

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CHAPTER TWO – PROCESSING PARAMETERS FOR BONDING THERMOPLASTIC TO PARALLAM®

2.1 Introduction

Wood and wood composites are more susceptible to decay and degradation when used in exterior applications. Several factors determine the rate at which they deteriorate including moisture and temperature conditions, presence of oxygen, and toxicity of the food source. Any one of these factors can be manipulated or eliminated to impart more fungal decay and insect attack resistance to wood materials, and thus prolong their durability. Two possible solutions for increasing the durability of wood are to pressure treat the material with heavy metals and to mill the wood into flour and manufacture wood-plastic composites (WPCs). Pressure treating the wood with heavy metals will take away the food source, and WPCs provide better moisture protection. However, both of these methods have drawbacks. Pressure treating the wood with high levels of metals like copper, can corrode fasteners and require more expensive galvanized or stainless steel fasteners (Zelinka and Rammer 2006). It may also be environmentally harmful in the long run. Another disadvantage to pressure treating is that not all wood materials receive the treatment equally. Some refractory species, such as Douglas fir, need to be incised to allow the treatment to penetrate into the wood. Incising decreases the strength and stiffness of the wood members.

The disadvantage of using WPCs is their relatively low moduli of elasticity. It has been shown that WPCs have good weathering properties (Wolcott and Englund 1999), but because they have low moduli of elasticity, compared to lumber and other structural wood-based composites, they are not well suited for structural applications. WPCs are a mixture of wood in

particulate form (wood flour) and thermoplastic matrix such as polyethylene, polypropylene or polyvinyl acetate, commonly extruded or injection molded with other additives. The hydrophobic nature of the thermoplastic makes them a great choice for exterior applications. The ratio of wood fiber to thermoplastic matrix will generally determine the physical and mechanical properties of the WPCs. One of the hurdles in bonding polyolefins to wood is poor adhesion between these two dissimilar materials, non-polar and polar. Bonding between the matrix and the wood particles is primarily due to mechanical interlocking. Gacitua and Wolcott (2008) found in their study that wood species with high interfacial areas had increased mechanical interlocking. However, in some cases the extrusion process can collapse cell walls effectively limiting the potential interfacial area.

Therefore, a method for improving the interfacial adhesion between thermoplastic resin, such as high density polyethylene (HDPE), and wood or wood composite surface is needed to combine the hydrophobic thermoplastic with structural wood composites manufactured using thermoset resins. The goal of the thermoplastic resin is to assist in extending the durability of these wood composites in exterior applications.

In this study, the use of a tie-layer between thermoplastic and wood-based substrate was examined to strengthen the bond between these two dissimilar materials and provide a barrier to moisture infiltration. A tie-layer is generally a thermoplastic polymer that is modified by grafting reactive functional groups, such as hydroxyl or maleic anhydride groups, onto its backbone. In theory, these functional groups enhance chemical bonding between tie-layer and the wood substrate. In a study conducted by Dai et al. (2004), a tie-layer was used to bond commingled E-glass/polypropylene yarns to an oak composite panel. It was found that bonding the E-glass/polypropylene yarns to the wood panel increased the bending strength of the

composite, implying that the tie-layer effectively bonded the E-glass/polypropylene yarns to the wood panel.

The ultimate goal of this project is to determine if it is possible to encapsulate a structural composite, specifically Parallam®, by hot-pressing a thermoplastic barrier layer onto the surface to improve its durability in exterior applications. Some possible advantages of effectively encapsulating a wood composite member with a thermoplastic barrier layer would be the slowing of moisture infiltration, as well as inhibiting the leaching of benign water-soluble biocides, such as borates. The study presented in this chapter will concentrate on understanding the influence of processing factors during hot-pressing of thermoplastic material, specifically HDPE film, to Parallam®.

2.2 Objective

The objective of this study was to determine optimum processing parameters, specifically temperature, pressure, and press time, for the maximizing the bond strength between a thermoplastic barrier layer and a wood composite substrate, Parallam®. Specific goals to attain this objective are to:

1. Evaluate the influence of temperature, pressure, and press time on the bond strength of the thermoplastic barrier layer to the wood substrate by using a 90º peel test.

2. Numerically optimize processing parameters to maximize the peel strength of the thermoplastic barrier layer from the wood substrate.

2.3 Materials and Methods

2.3.1 Materials

For this study Douglas fir Parallam® was used for the wood substrate because of its refractory nature and as it also is a commercially significant engineered wood product in the Northwest. The composite specimens were cut into 76 mm (3 in) wide by 229 mm (9 in) long by 25 mm (1 in) thick blocks from an 89 mm (3.5 in) wide by 241 mm (9.5 in) thick beam. Parallam® beams were all chemically treated with boric oxide (B_2O_3) by Pacific Wood Preserving of Oregon at a retention level of 0.28 pcf.

The thermoplastic that was used for the barrier layer in this study, HDPE, was acquired from Equistar (LB010000) in a powder form. The tie-layer used to bond the thermoplastic barrier layer to the wood composite was a styrene-butadiene polymer, which will be referred to in this paper as tie-layer B and was provided by BASF in a pellet form. The tie-layer B is a proprietary polymer, but the properties of the HDPE are shown in Table 2-1. For ease of application in this study both tie-layer and thermoplastic barrier were converted into a film form, with an approximate thickness of about 0.5 mm (20 mil), for specimen preparation. They were extruded using a Leistritz 18 mm co-rotating extruder that was starve fed. Thermal transitions of both HDPE and tie-layer B were characterized using a differential scanning calorimetry (DSC) (Figures 2-1 and 2-2 respectively). HDPE has a distinct melt temperature of 130ºC. Tie-layer B, however, does not have a true melting point but softens gradually which would indicate that the tie-layer is an amorphous polymer, such as a thermoplastic elastomer.

Table 2-1: Properites of HDPE.

Material	Melt Index (g/10 min)	Density (g/cm^3)	Tensile Strength @ Break (psi)
HDPE	0.5	0.953	3,960
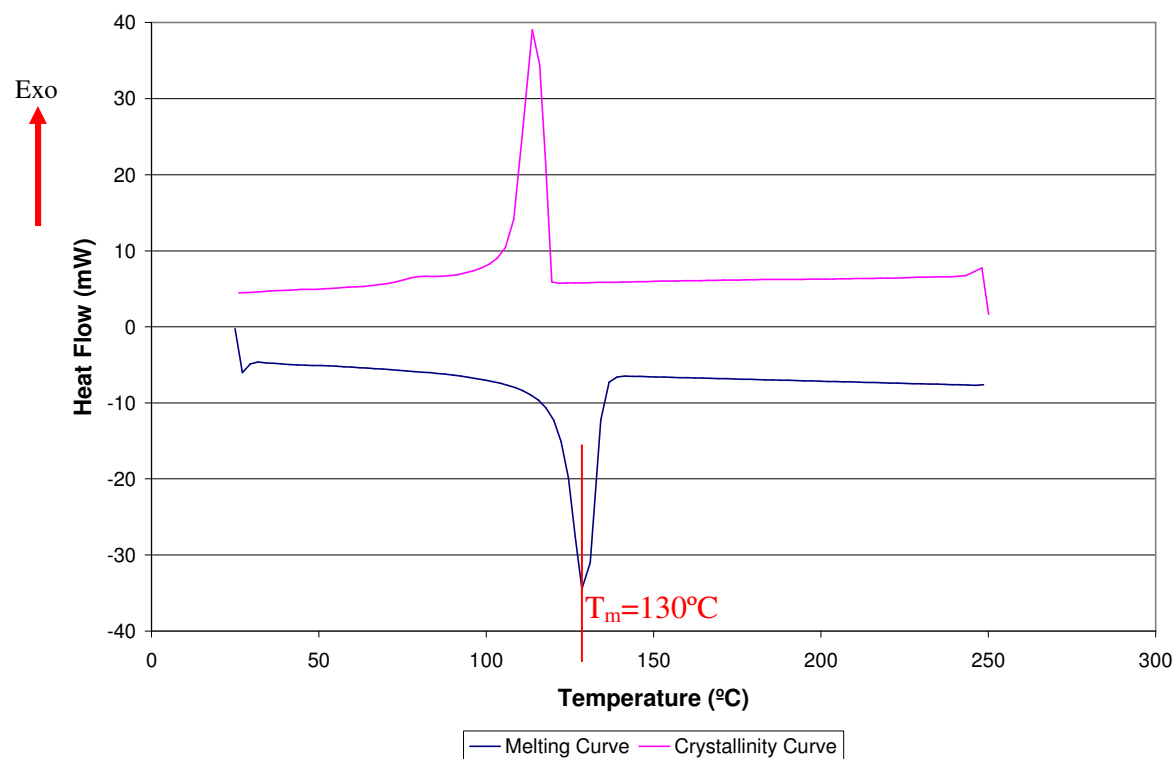
Figure 2-1: DSC curve for HDPE: melt temperature = 130ºC
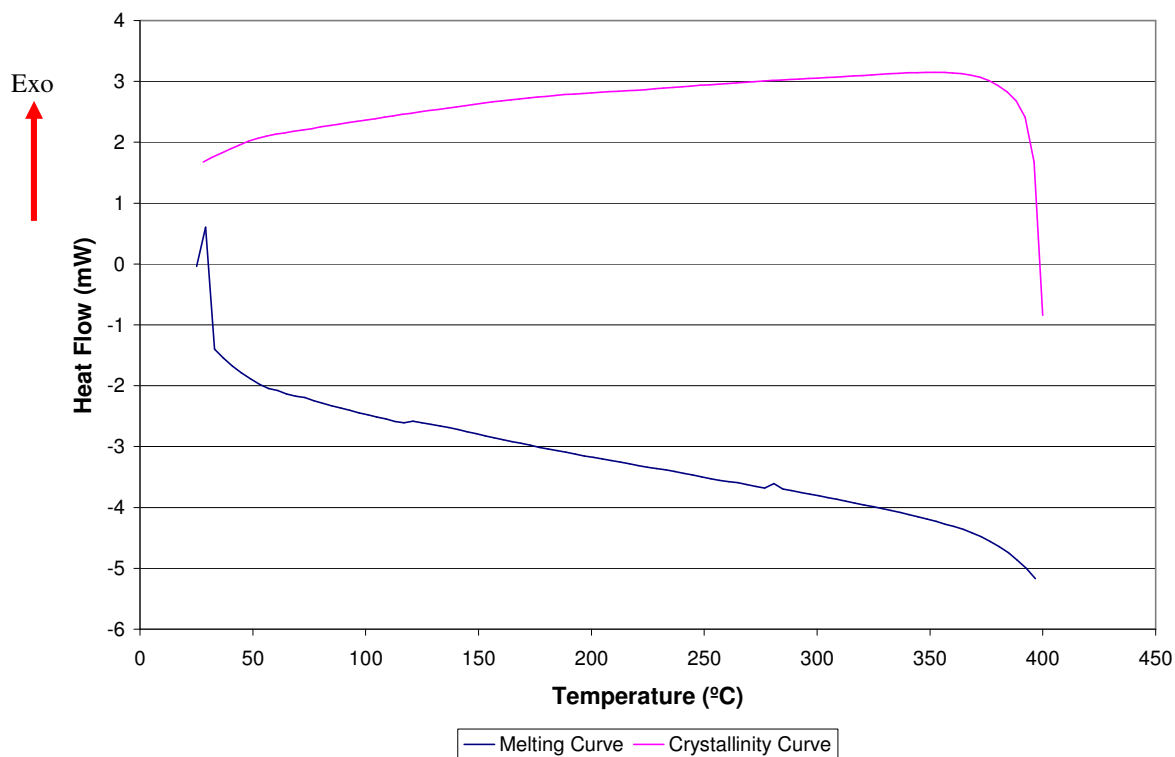
Figure 2-2: DSC curve for tie-layer B: NO discernable melt temperature due to its amorphous nature.

2.3.2 Specimen Preparation

After careful consideration, it was decided that three levels for each of the three processing factors would be used. The three temperature levels were 160ºC, 180ºC, and 200ºC. The three pressures used were 1035 kPa (150 psi), 1380 kPa (200 psi), and 1725 kPa (250 psi). Press times used were 120 seconds, 300 seconds, and 600 seconds. Three replicates were produced for each combination of factors. Table 2-2 shows the experimental design with a total of 81 specimens. All of the specimens were pressed using a hydraulic 0.914 m^2 oil-heated press in combination with the Pressman™ control system.

Table 2-2: Experimental Design

Pressure (kPa)	Temperature (ºC)			Press Time (sec)
	160	180	200	
1035	3	3	3	
1380	3	3	3	120
1725	3	3	3	
1035	3	3	3	
1380	3	3	3	300
1725	3	3	3	
1035	3	3	3	
1380	3	3	3	600
1725	3	3	3	

A problem that was anticipated was the inhibition of the melt front if the temperature of the wood substrate was lower than the melting temperature of the thermoplastic layers. Therefore, to determine the amount of time that it would take to raise the temperature of the wood substrate just below the surface, a thermocouple was placed approximately 1.5 mm (1/16 in) below the surface. The specimen was then placed on a caul sheet and placed into the hot-

press with a gap of 27 mm (1.0625 in) between the platens, and the platens had a temperature of 160ºC or 180ºC. At each of these corresponding temperatures, it took 600 seconds for the thermocouple to read 148ºC and 162ºC, respectively (Figure 2-3). From the graph it was concluded that it would take approximately 300 seconds to reach a temperature above the melting temperature of the HDPE (130ºC) just below the surface of the wood substrate at each of the platen temperatures.
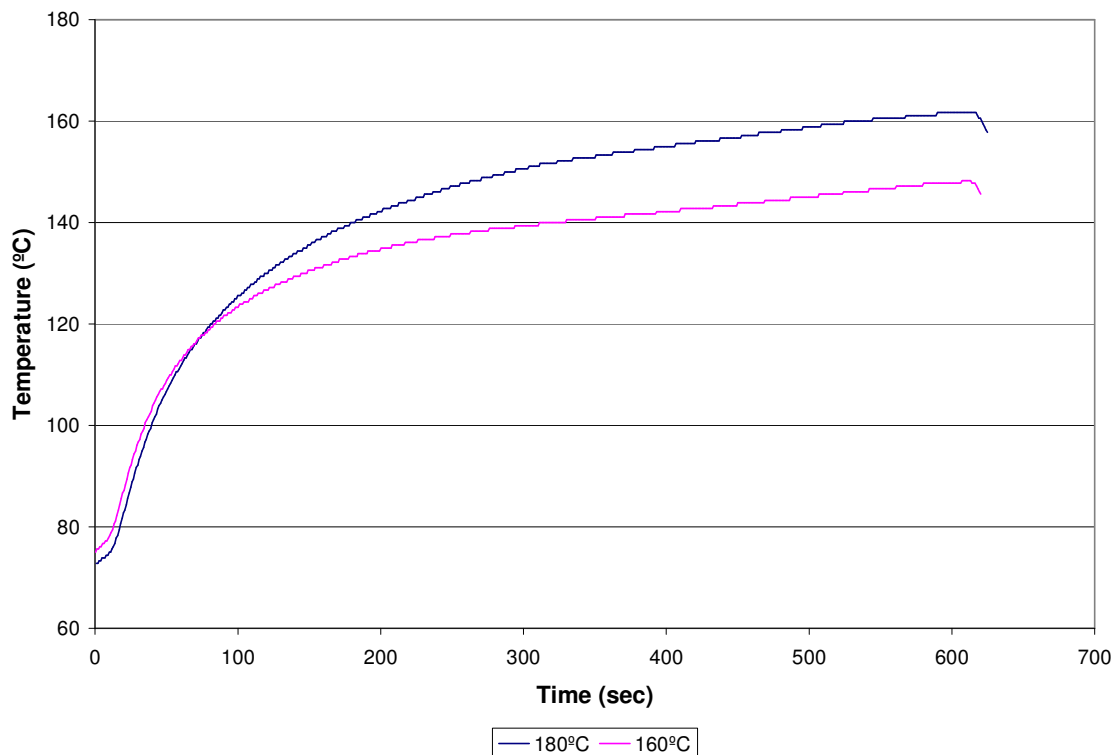
Figure 2-3: Temperature profile just below the surface of the specimen

Prior to hot-pressing, specimens were conditioned by oven drying in a Precision Quincy Corporation forced air oven at 105ºC until the specimens mass stabilized, which took approximately 7 days. Once the specimens were oven dry, a thin barrier layer was applied to one end and placed on a caul sheet. This thin barrier was used to prevent the plastic from bonding to

the wood so that a tab was created for a 90° peel test. This barrier was a vacuum bagging material from Air Tech International, Inc., and it was found that the barrier material was able to withstand the processing temperatures as well as inhibit outer thermoplastic and inner tie-layers from bonding to the wood substrate.

The specimen was then placed into the hot-press heated to the desired platen temperature. The gap between the platens was reduced to 27 mm (1.0625 in) to allow the wood to heat up for approximately 300 seconds (enough time for the first few millimeters to reach the melting temperature of the plastic). Upon completion of heating the specimen surface, a single tie-layer and HDPE film was placed onto the heated surface of the specimen. Figure 2-4 shows the specimen lay-up with the barrier, tie-layer and HDPE barrier layer. After placing a release sheet and a caul sheet on top of the layered surface, specimens were hot-pressed following a pressing schedule as shown in Figure 2-5.
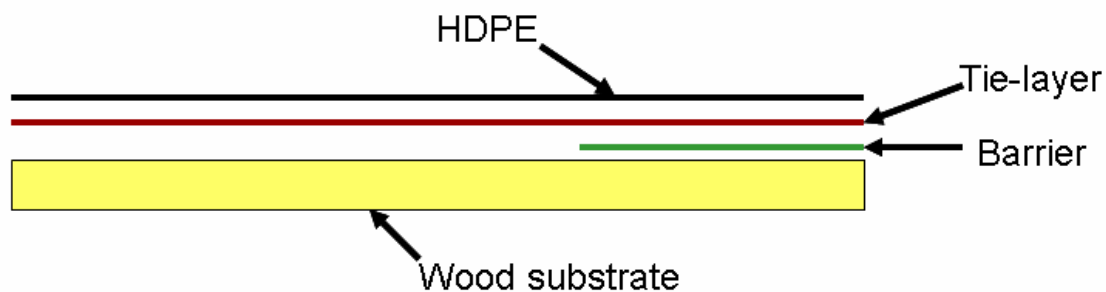
Figure 2-4: Specimen lay-up
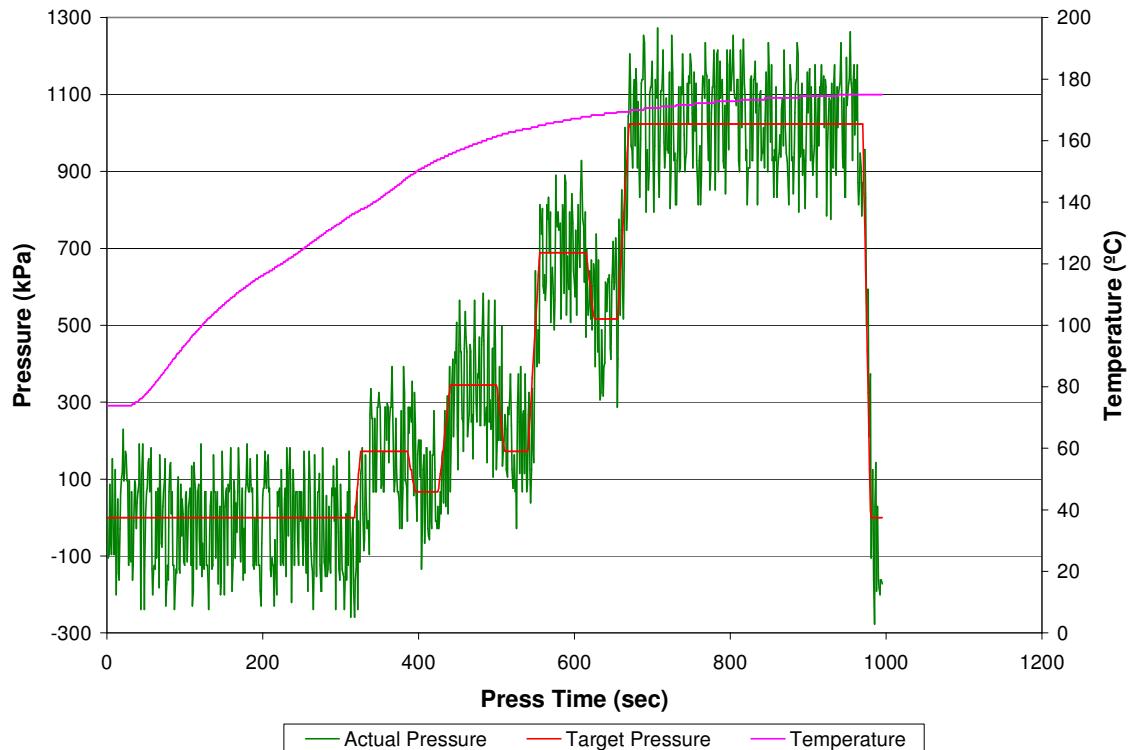
Figure 2-5: Pressing schedule used to bond thermoplastic barrier layer to wood substrate with tie-layer B. Platen temperature was maintained at 180ºC, and final pressure targeted was 1035 kPa that was maintained for 300 seconds.

The pressing cycle included a stepwise pressure control that slowly worked the plastics into the wood substrate. After hot-pressing the thermoplastic layers onto the specimens, they were trimmed down to a 51 mm (2 in) wide by 229 mm (9 in) long by 25 mm (1 in) thick block for testing.

2.3.3 Testing Methodology

A 90º peel test, following ASTM D 6862 test standard, was conducted as a measure of bond efficiency and how it is affected by the investigated processing factors. Speed of testing was 254 mm/min. Specimens were tested within 24 hours of pressing. The test setup and a typical specimen are shown in Figure 2-6.

Tab for peeling
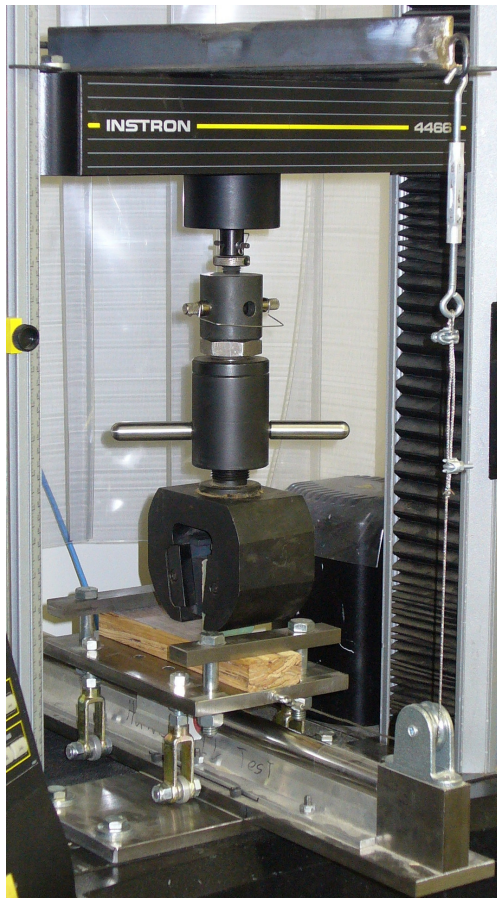
Figure 2-6: 90º peel test setup and specimen

Peel strength was calculated using Equation 1. Average recorded load was determined by taking

the mean load over the entire peel length (Figure 2-7).

$$\text{Peel Strength} = \frac{\text{Average Recorded Load}}{\text{Peel Width}}$$ [1]

2.4 Results and Discussion

Results of peel testing are shown in Tables 2-3, 2-4, and 2-5.

Table 2-3: Peel strength values for platen temperature of 160ºC

	160ºC								
	1035 kPa			1380 kPa			1725 kPa		
	120 s	300 s	600 s	120 s	300 s	600 s	120 s	300 s	600 s
	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength
	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)
# of Specimens	2	6	3	2	4	2	1	2	3
Mean	3.21	1.71	1.99	2.08	2.29	2.43	2.81	2.04	1.38
SD	N/A	0.21	1.06	N/A	0.87	N/A	N/A	N/A	0.35
COV (%)	N/A	12.51	52.93	N/A	37.93	N/A	N/A	N/A	25.66
Minimum	3.06	1.37	0.99	1.67	1.50	2.22	2.81	1.77	0.97
Maximum	5.63	1.94	3.09	2.49	3.87	2.64	2.81	2.31	1.59

Table 2-4: Peel strength values for platen temperature of 180ºC

	180ºC								
	1035 kPa			1380 kPa			1725 kPa		
	120 s	300 s	600 s	120 s	300 s	600 s	120 s	300 s	600 s
	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength
	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)
# of Specimens	3	4	2	2	4	2	2	3	2
Mean	1.96	1.99	1.32	2.90	2.77	2.73	2.57	1.88	2.92
SD	0.33	1.01	N/A	N/A	0.56	N/A	N/A	0.24	N/A
COV (%)	16.79	50.73	N/A	N/A	20.34	N/A	N/A	12.64	N/A
Minimum	1.63	1.80	0.82	2.06	2.06	2.59	2.00	1.66	2.07
Maximum	2.29	4.18	1.81	3.73	3.24	6.61	3.15	2.13	4.53

Table 2-5: Peel strength values for platen temperature of 200ºC

	200ºC								
	1035 kPa			1380 kPa			1725 kPa		
	120 s	300 s	600 s	120 s	300 s	600 s	120 s	300 s	600 s
	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength
	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)
# of Specimens	2	8	2	2	2	0	1	2	1
Mean	2.73	2.98	4.85	1.47	2.77	N/A	4.21	2.89	5.80
SD	N/A	1.26	N/A	N/A	N/A	N/A	N/A	N/A	N/A
COV (%)	N/A	42.22	N/A	N/A	N/A	N/A	N/A	N/A	N/A
Minimum	2.43	1.92	3.51	1.19	1.51	0.00	4.21	2.64	5.80
Maximum	3.03	5.55	6.19	1.76	4.03	0.00	4.21	3.13	5.80

Two primary modes of failure were observed during the 90º peel test: the plastic would peel off of the wood or the plastic would tear and eventually fail. The preferred mode of failure was to peel the plastic with some signs of wood failure (an effective or good peel), but only a small number of specimens fell into this category. For a majority of the specimens, the thermoplastic barrier layer tore at some point in the peeling process (the specimens that completely failed were not included in the results). It is speculated that the macro-voids that were present on the surface of the Parallam® were being filled in with the HDPE which made the surrounding plastic thinner and more susceptible to plastic tearing. Another place that the plastic would tear was at the tab interface. When starting to peel the plastic from the wood, if the bond to the wood was greater than the yielding strength of the plastic then the tab had the tendency to tear off (undesirable failure that did not yield useful information).

Figure 2-7 shows a typical graph of the peel load compared to the peel length for a peel width of 2 inches for a specimen that failed at the wood interface (plastic did not tear). The peaks and valleys were created by the wood-plastic bond being stressed and then broken. As

mentioned before, from Equation 1 the average peel strength was found by averaging the peel

load over the entire peel length for a given width.
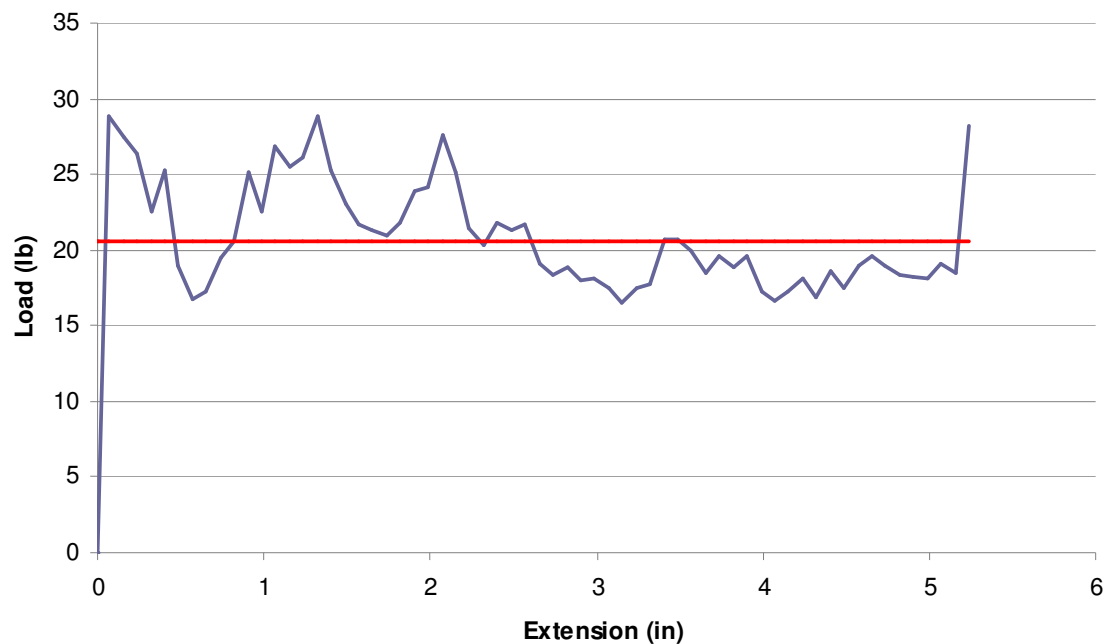
Figure 2-7: Typical 2" wide peeling graph with mean load shown in red.

Similar studies in the past (Mahlberg et al. 2001, Kolosick et al. 1993) that looked at

thermoplastic bonding to wood substrates yielded lower peel strengths than the process

implemented in this study. Mahlberg et al. (2001) looked at the effect of chemical modification

of wood using a 90º peel test to determine bond strength and found values that ranged from 0.16

N/mm to 0.64 N/mm. The processing parameters that were used were a temperature of 165ºC, a

pressing time of 120 seconds, and pressures of 300 kPa (43 psi) and 900 kPa (130 psi). Kolosick

et al. (1993) used a maleated polypropylene (PP) wax modifier to laminate wood substrates with

PP. They also used a 90º peel test to determine the bond strength and found peel strength values

that ranged from 0.04 N/mm to 0.1 N/mm. The processing parameters used in their study were a

temperature of 175ºC, a pressure of 275 kPa (40 psi), and pressing times of 180 seconds and 360 seconds. In comparison the peel strengths in this study ranged from 0.82 N/mm to 6.61 N/mm with similar pressing temperatures and times.

Upon completion of the specimen preparation using hot-pressing at 200ºC, visual inspection of the specimens indicated what is assumed to be a thermal degradation of HDPE or tie-layer B as shown in Figure 2-8. As it was prevalent in all specimens pressed at this temperature, it was concluded that hot-pressing at 200ºC should probably be avoided. However, the results of hot-pressing at 200ºC are also presented in this study.

Figure 2-8: Signs of HDPE degradation on 200ºC specimen

Figure 2-9 compares the performance of specimens hot-pressed at different levels of the processing factors examined in this study. In general, the variation in peel strength seems to be larger at 160ºC and 180ºC especially at the lowest and highest pressures (1035 and 1725 kPa). Also the difference in peel strengths between the 300 seconds and 600 seconds press times was small on the average for all pressures and temperatures. As visual inspection of specimens at 200ºC indicated discoloration of the thermoplastic barrier layer, probably because of degradation, it was decided that high pressing temperature should be avoided. As the intention of the researchers is to minimize energy required during this operation, it is probably also practical to avoid high temperatures, pressing times, and pressures during the bonding process.

This would be especially true if the incremental gain in peel strength with increasing temperature and time is not significant. It is also critical that pressing pressures are relatively low as it is not the intention in this process to further densify the composite or damage it due to excessive compressive forces in the transverse direction. As per TrusJoist (2003), an allowable design of 5170 kPa (750 psi) is specified for the compression perpendicular to grain. Past studies (Mahlberg et al. 2001, Kolosick et al. 1993) as discussed earlier, did not exceed pressures of 900 kPa (140 psi) and had considerably lower peel strengths. In this study, to achieve good penetration of thermoplastic encapsulate into the macro-voids of the composite, higher pressures were applied. Of course, the ultimate goal would be to achieve adequate peel strengths at lower pressure since it requires more energy to apply higher pressures.

The trend in Figure 2-9 shows that at 300 seconds, the peel strength peaked for both processing temperatures at 1380 kPa. Statistical analysis was not conducted as the number of specimens was limited due to difficulty with achieving acceptable failure modes during the peel test.
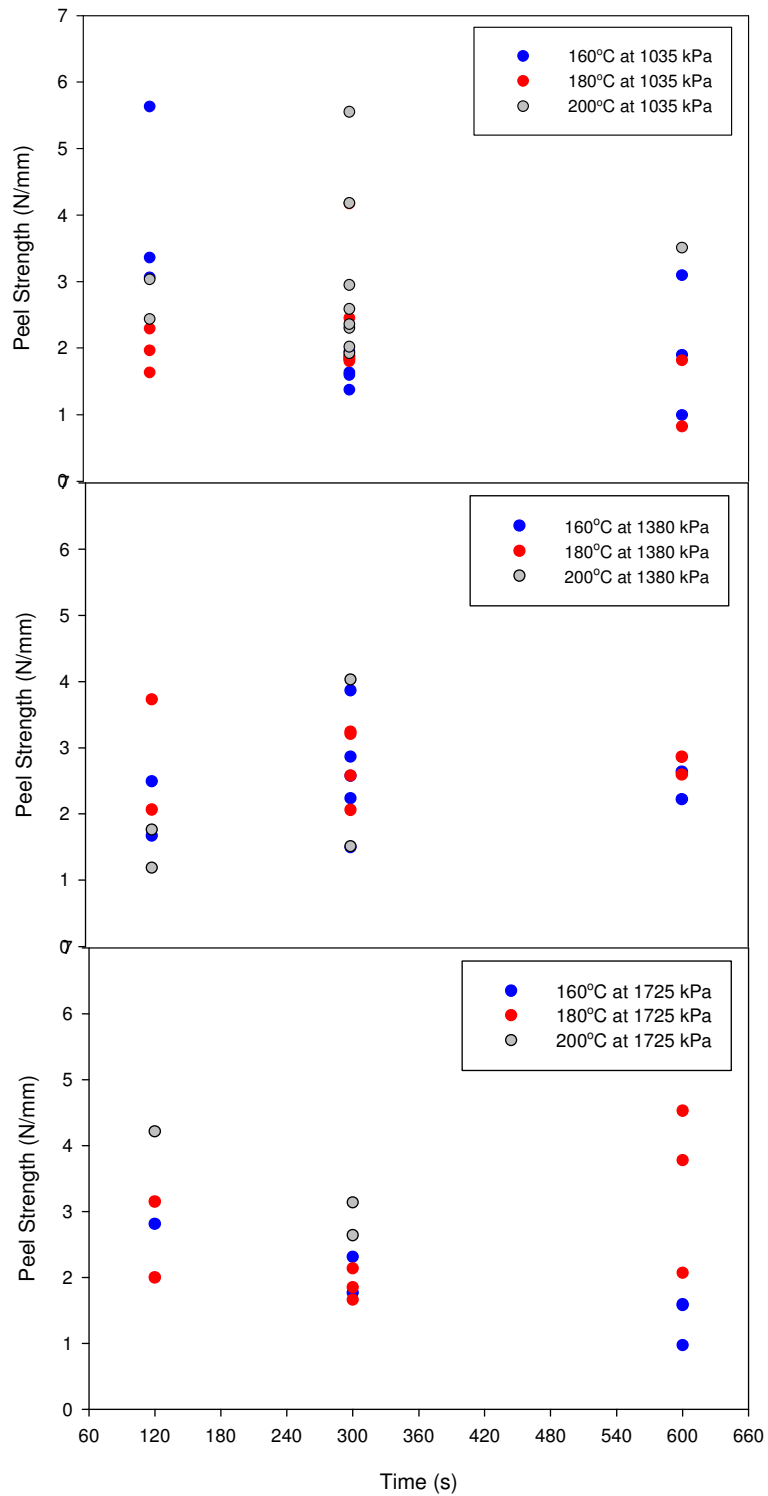
Figure 2-9: Comparison of peel strengths at varying levels of processing factors examined.

To glean more information on the effects of processing factors with limited specimens

that failed in an acceptable manner, further analysis was conducted following a Box-Behnken

design. The Box-Behnken design generated a response surface using a design of experiment statistical package, Design-Expert® (Stat-Ease 2006). Response surface methods are useful in determining how a response (peel strength in this study), is affected by a set of factors over a region of interest. A reason for choosing this design is that it requires only three levels of each quantitative factor (Breyfogle III 1992, Stat-Ease 2005). The response surface at three pressing times with varying pressure and temperature over the design space in this study is shown in Figure 2-10. Response surface analysis indicates that peel strength tends to peak between values of 1035 kPa and 1380 kPa in all cases. Figure 2-11 shows the response surface at 1035 kPa and 1380 kPa pressures over changing temperature and time. Analysis indicates that at lower pressing temperatures, there is a decrease in peel strength with increase in pressing time. Highest peel strengths were achieved at peak pressing temperature and time. However, this was coupled with large variation in data. Using Design-Expert® software, numerical optimization was conducted by setting goals which included minimization of time, temperature and pressure while maximizing peel strength to determine optimal conditions for achieving consistent and maximum possible peel strength values. In setting goals, several combinations of importance levels for the three processing variables could be assigned. We chose to assign an importance level of three to temperature, five to pressure, two to pressing time, and five to peel strength over a scale of five. Results indicate optimum hot-pressing temperature, pressure and time to be 176°C, 1140 kPa and 296 seconds. Visual inspection also showed that barrier layer thickness was consistently uniform in the case of specimens that were pressed for 300 seconds. Additionally, visual inspection also indicated that quality of barrier layer in terms of uniformity in thickness and smoothness was similar for specimens pressed at either 1035 kPa or 1380 kPa. Observing the peel strength values (average, minimum and maximum values) in Table 2-4 of

specimens pressed for 300 seconds at 180ºC also indicates that there may not be a significant

practical difference between peel strength values when pressed at 1035 kPa and 1380 kPa.

Therefore, it seems reasonable to choose the lower pressure value for bonding thermoplastic

barrier layer when the optimum value is shown to be 1140 kPa, especially if it is desired to

minimize energy requirements during the pressing process.

Press Time = 120 s

Press Time = 300 s

Press Time = 600 s
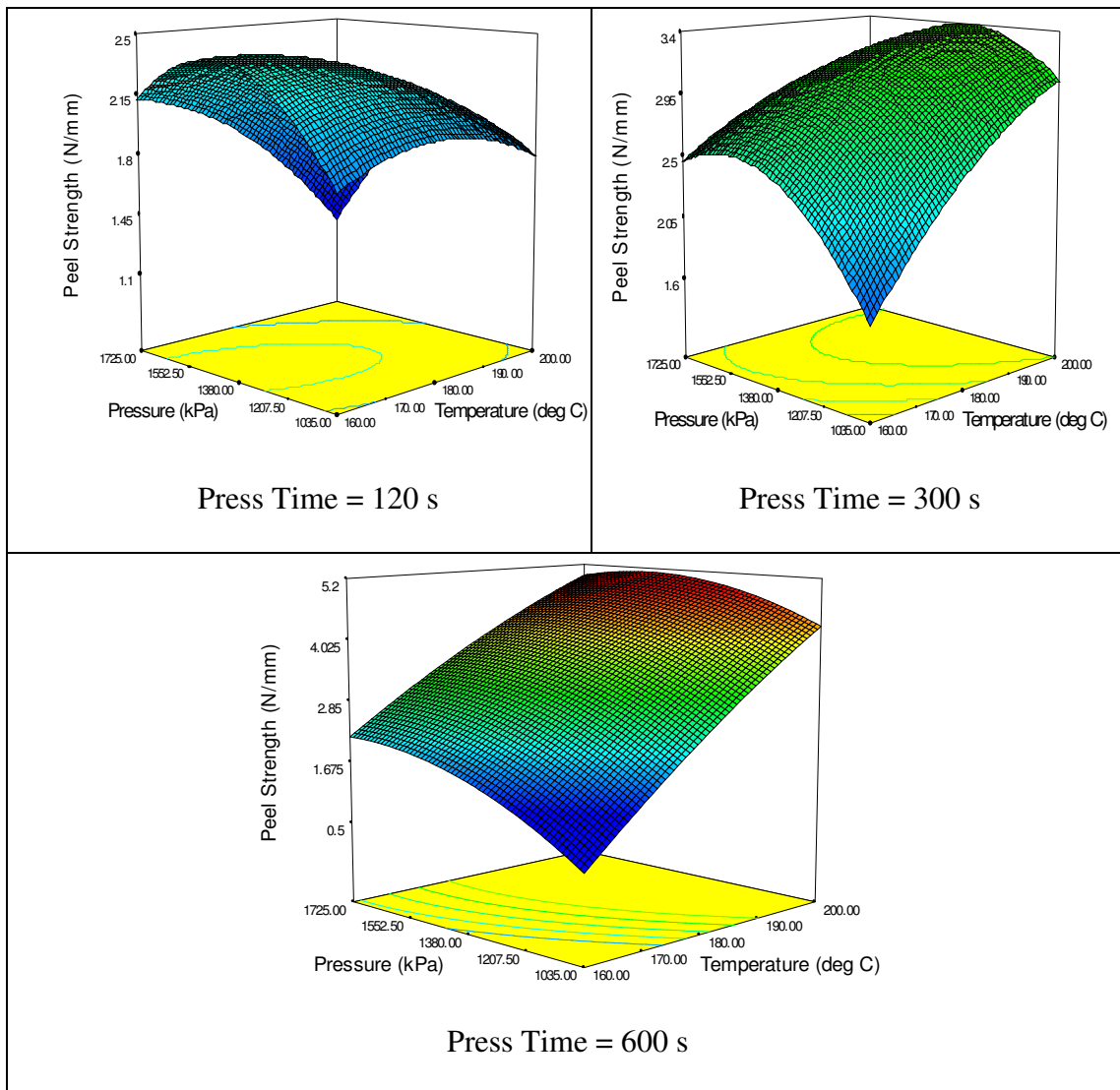
Figure 2-10: Response surface analysis at three pressing times.
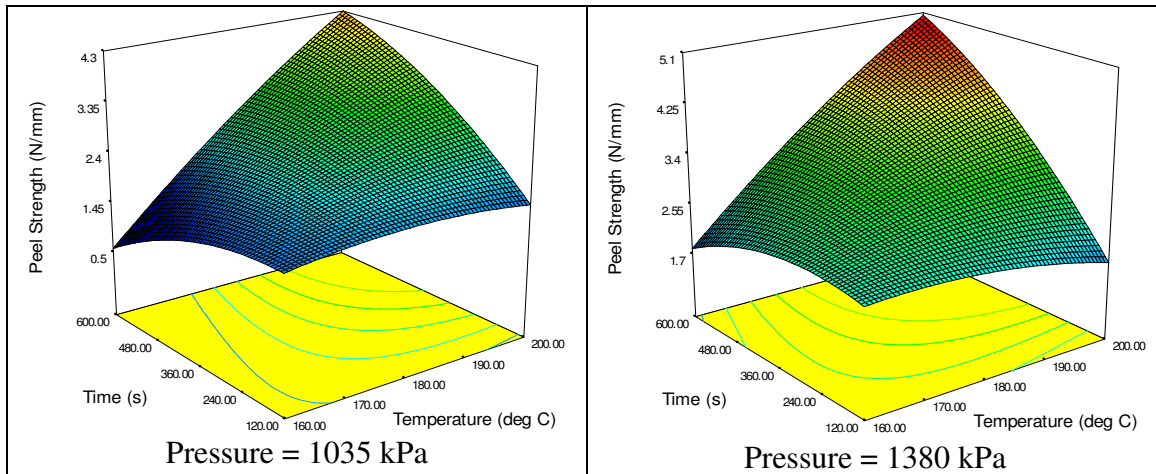
Figure 2-11: Variation in peel strength over time and temperature in the region of interest at constant pressures of 1035 kPa and 1380 kPa.

2.5 Summary and Conclusions

In this study, it was found that using a styrene-butadiene polymer tie-layer (tie-layer B) increased the bond strength between HDPE and boric oxide treated Douglas fir Parallam[®]. Due to the quality of the bond and thinning of HDPE around macro-voids in the composite surface, an ideal failure (peeling of HDPE from wood substrate with some wood failure) was not achieved for all specimens. Therefore, based on visual inspection, peel strength values from limited number of specimens that had acceptable peel failures, and response surface analysis, it was determined that a temperature, pressure and press time combination of 176ºC, 1140 kPa, and 296 seconds produced an acceptable and consistent peel strength. For ease of control, temperature and time will be rounded to 180ºC and 300 seconds for further studying the effects of using different tie-layers to bond a moisture barrier layer to several wood substrates. As it is the intention to lower the energy requirements, pressing pressure will be maintained at 1035 kPa.

2.6 References

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CHAPTER THREE – EFFECT OF TIE-LAYER ON THE BOND STRENGTH BETWEEN THERMOPLASTIC AND PARALLAM[®]

3.1 Introduction

Wood materials are used for a number of residential applications including exterior uses such as decks. With the advent of wood-plastic composites (WPCs), the decking and guard rail materials have become more durable and easier to maintain. However, the substructures of the decks are still typically solid sawn wood materials that need to be chemically treated to improve their durability. Many of the copper rich preservative chemicals that are used, though, can be corrosive to fasteners and flashings. A more benign preservative treatment using borates is available to extend the durability of these substructure members, however, borates are known to leach out over time. So, a potential solution investigated in this study is the use of thermoplastics to encapsulate a structural wood composite, such as Parallam[®], to slow moisture uptake into the composite, as well as to keep borates, from leaching out of the composites. However, thermoplastics, such as polyethylene or polypropylene do not adhere to wood due to difference in polarity of the materials. This study will examine the use of tie-layers to encourage chemical bonding in addition to mechanical bonding between the thermoplastic barrier layer and the wood substrate. Within the past decade a number of studies have been done to investigate the possible uses of tie-layers in the improvement of the bond between thermoplastics and wood substrates.

Dai et al. (2004) used a thin tie-layer to bond E-glass/polypropylene (PP) yarns to an oak substrate. The study looked at the ability to bond E-glass/PP yarns to a wood surface for the purpose of reinforcing it in bending (i.e. a floor system). Examining the failure mode of the

composite in lap shear specimens, it was observed that a cohesive failure of the composite was common in areas that were weakened by voids. The number of voids could be reduced by increasing the consolidation time and the pressing pressure. This study found that a higher heating temperature and pressure would improve the bonding strength. In another study by Kumar and Ramani (2000), a tie-layer is again used to bond a unidirectional continuous glass fiber reinforced polypropylene (UCGPP) composite to an oak substrate. Effects of changing moisture on composite modulus using dynamic mechanical analysis (DMA) were investigated. It was found that under desorption/sorption cyclic conditions the wood-UCGPP composite underwent significant modulus changes due to the differential shrinkage/swelling of the oak compared to the UCGPP composite. However, the tie-layer did increase the storage modulus of the wood-UCGPP composite compared to the neat wood.

Other studies in the past (Mahlberg et al. 2001, Kolosick et al. 1993), however, have used surface modification of the wood substrate instead of a tie-layer to increase the adhesion between wood substrates and thermoplastics. Mahlberg et al. (2001) used succinic and phthalic anhydride as modifiers to laminate birch veneers with polypropylene (PP). A 90º peel test was implemented to determine the force required to peel the PP laminate from the veneer surface. The peel results ranged from 0.29 N/mm to 0.64 N/mm for the phthalic anhydride treated specimens, and from 0.38 N/mm to 0.63 N/mm for the succinic anhydride treated specimens. Kolosick et al. (1993) used maleated polypropylene wax as a modifier to laminate birch and aspen veneers with PP. They also used a 90º peel test to determine the force required to peel the PP laminate from the veneer surface. Results indicated that the peel strength with the surface modification was lower than the untreated specimens, in most cases. The untreated specimens

had a peel strength that ranged from 0.08 N/mm to 0.1 N/mm. The treated specimens had varying degrees of treatment, but had a range of peel strengths from 0.04 N/mm to 0.1 N/mm.

In this study, based on the knowledge gained from the previously mentioned studies and the results of the study reported in Chapter 2, it was decided to use two commercially available tie-layers to bond a thermoplastic resin to Parallam® as well as a solid wood substrate using hot-pressing.   The best suited parameters for hot-pressing a thermoplastic barrier layer into a wood composite, as reported in Chapter 2, are a platen temperature of 180ºC, a pressing pressure of 1035 kPa, and press time of 300 seconds. In this study, two bond strength tests and an accelerated aging test were used to study the efficacy of tie-layers in improving the bond strength and investigate the effects of weathering on the bonding as a measure of durability in exterior applications.

3.2 Objective

The objective of this study was to characterize the improvement in bond strength between thermoplastic barrier layer and borate-treated Parallam® and solid sawn lumber due to the use of a tie-layer between the two materials. The following two tasks were performed to characterize the effectiveness of bond strength:

1. Evaluate the efficacy of tie-layers in improving the bond strength between thermoplastic barrier layer and wood substrates, both solid sawn lumber and Parallam®, using 90º peel and shear block tests.

2. Investigate the durability of bond using a cyclic wet/dry accelerated aging test.

3.3 Materials and Methods

3.3.1 Materials

For the purposes of comparing the differences in bond strength between thermoplastic and wood substrates, southern pine Parallam®, Douglas fir Parallam® as well as Douglas fir solid lumber was used. To examine the effect of differences in wood surface, strength of bond between the barrier layer and the two surfaces of the solid wood substrate (namely radial and tangential), were evaluated. Douglas fir and southern pine were used in this study as they are the most common wood species that are commercially used for construction in the United States. Parallam® beams as well as solid lumber were chemically treated with boric oxide (B_2O_3) by Pacific Wood Preserving of Oregon at a retention level of 0.28 pcf. The composite specimens of dimensions 76 mm (3 in) wide by 229 mm (9 in) long by 25 mm (1 in) thick were cut from 89 mm (3.5 in) wide by 241 mm (9.5 in) thick beams. Similar size specimens were also prepared from treated lumber.

The thermoplastic that was used as a barrier layer in this study was high density polyethylene (HDPE), and was acquired from Equistar (LB010000) in a powder form. The two tie-layers evaluated were a maleic anhydride modified HDPE tie-layer (referred to as tie-layer A) and a styrene-butadiene polymer tie-layer (referred to as tie-layer B). Tie-layer A was provided by Dow chemical (Amplify GR 205) in a pellet form and tie-layer B was provided by BASF in a pellet form. The tie-layer B is a proprietary polymer, but the properties of HDPE and tie-layer A are shown in Table 3-1.

Table 3-1: Thermoplastic properties for HDPE and tie-layer A.

Material	Melt Index (g/10 min)	Density (g/cm^3)	Tensile Strength @ Break (psi)	MAH Graft Level (wt% MAH)
HDPE	0.5	0.953	3,960	-
Tie-layer A	2.0	0.960	2,300	> 1.0

Since the plastics were obtained in either a powder or pellet form, they were extruded into a film

using a Leistritz 18 mm co-rotating extruder for ease of manufacturing. The approximate

thickness of the films was about 0.5 mm (20 mil). Thermal transitions of the thermoplastic

barrier layer and the two tie-layers were characterized using a differential scanning calorimetry

(DSC) to determine the corresponding melt temperatures for checking the platen temperature

settings during the hot-pressing process (Figures 3-1, 3-2 and 3-3 respectively).
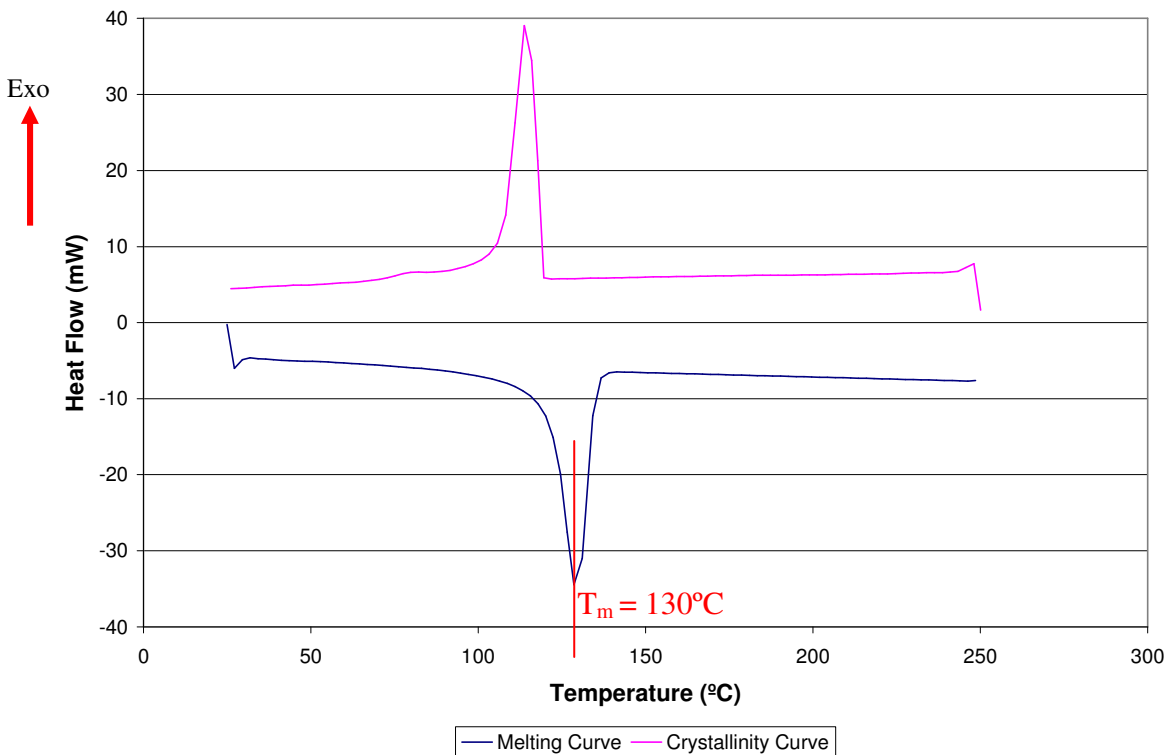
Figure 3-1: DSC curve for HDPE: melt temperature = 130ºC
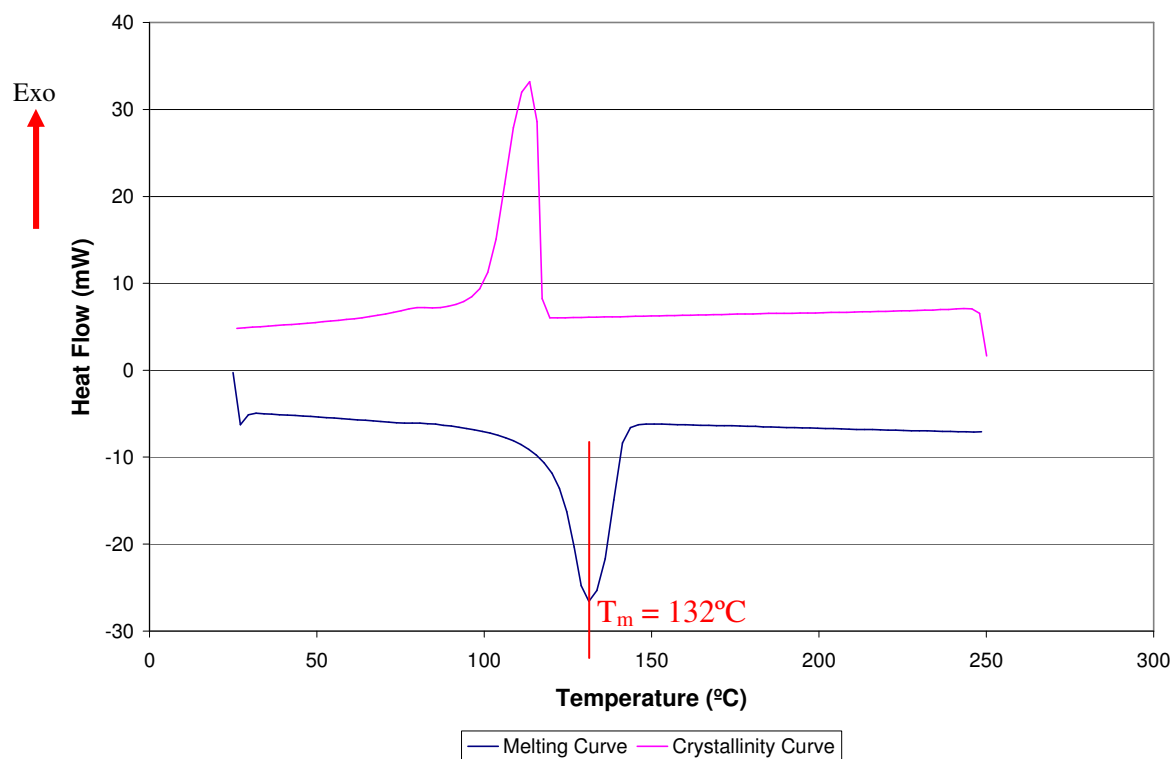
Figure 3-2: DSC curve for tie-layer A: melt temperature = 132ºC
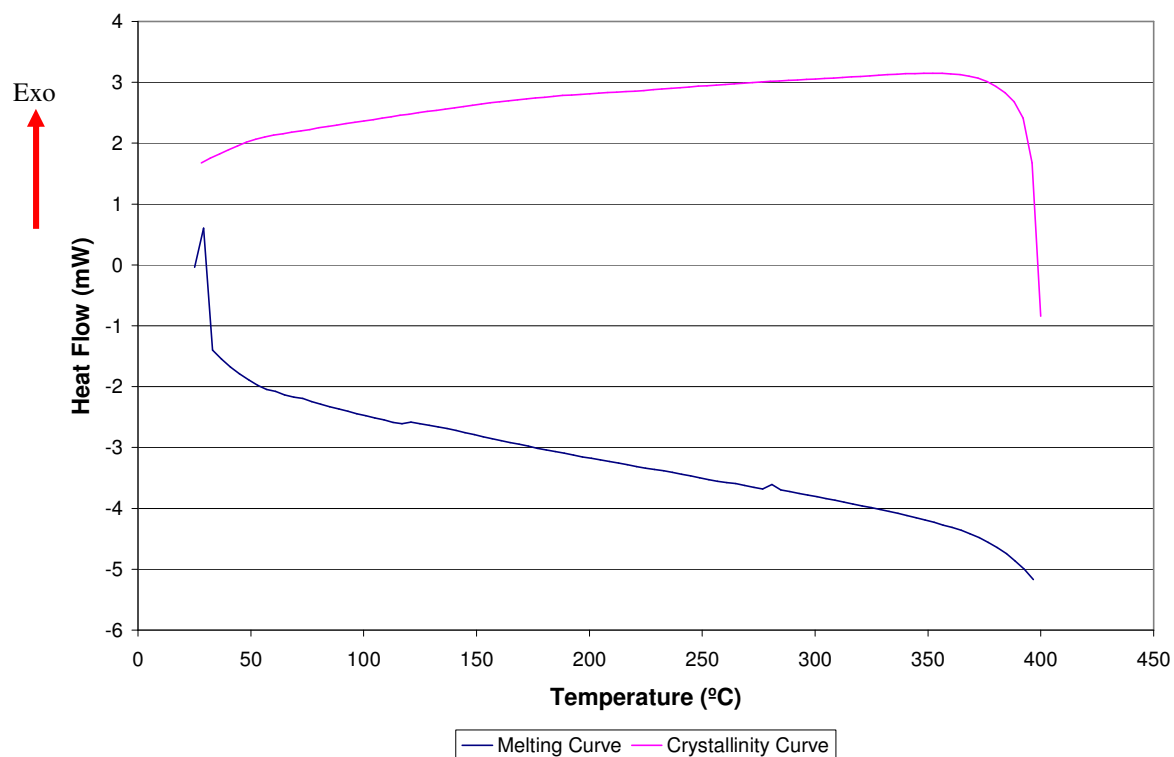
Figure 3-3: DSC curve for tie-layer B: NO discernable melt temperature due to its amorphous nature.

The HDPE and tie-layer A have distinct melt temperatures of 130ºC and 132ºC respectively. Tie-layer B, however, does not have a true melting point but softens gradually which would indicate that the tie-layer is an amorphous polymer material, such as a thermoplastic elastomer.

A thin barrier was used to prevent the plastic from bonding to the wood to generate a tab for a 90º peel test. This barrier was a vacuum bagging material from Air Tech International, Inc. and was able to withstand the processing temperatures.

3.3.2 Specimen Preparation

All of the specimens were hot-pressed using a hydraulic 0.914 m^2 oil-heated platen press in combination with the Pressman™ control system. To start the pressing process the specimens were placed in a Precision Quincy Corporation forced air oven at 105ºC until the specimens mass stabilized, this took approximately 7 days. Once the specimens were oven dry, a barrier layer was applied to one end and placed on a caul sheet. It was then positioned into the hot-press at a platen temperature of 180ºC with a gap of 27 mm (1.063 in) between the top and bottom platens, and was held for approximately 300 seconds. The purpose of this pre-pressing cycle was to heat the first few millimeters of the wood substrate to the melting temperature of the plastic. When the pre-pressing cycle was completed, the tie-layer film (for those specimens where a tie-layer was used) and the HDPE film were placed on top of the specimen (Figure 3-4) and pressed at a temperature of 180ºC, pressure of 1035 kPa, and pressing time of 300 seconds. A top caul sheet was placed on the specimens during pressing with a release sheet between the specimens and the caul sheet. Next the specimens were positioned back into the hot-press and followed a pressing

schedule as that shown in Figure 3-5. The pressing cycle included a stepwise pressure control that slowly worked the plastics into the wood substrate.
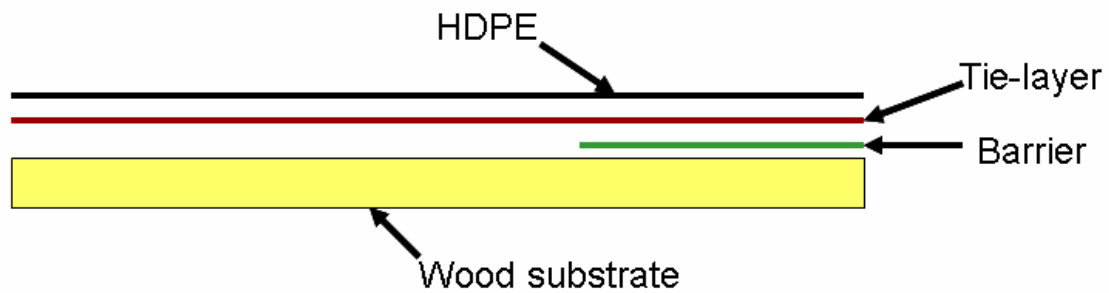
Figure 3-4: Specimen lay-up
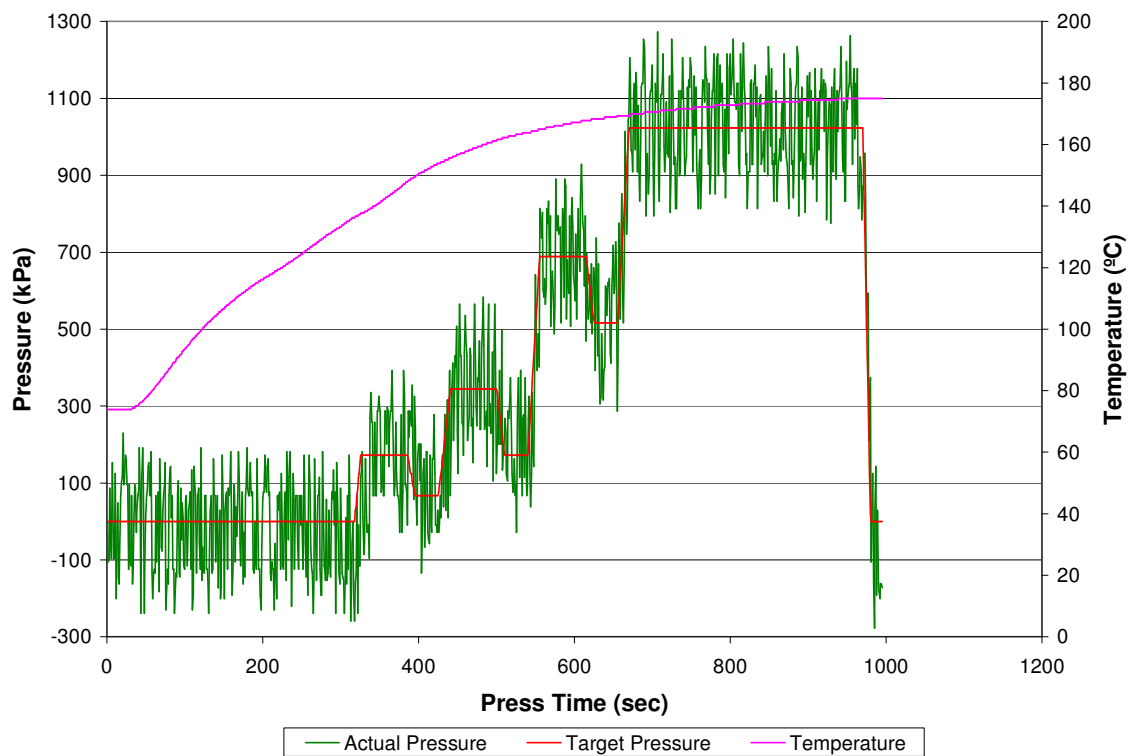
Figure 3-5: Pressing schedule used to bond thermoplastic barrier layer to wood substrate with or without a tie-layer. Platen temperature was maintained at 180ºC, and final pressure targeted was 1035 kPa that was maintained for 300 seconds.

Three different lay-ups on wood substrate were used:

1. HDPE only (no tie-layer)
2. Tie-layer A and HDPE
3. Tie-layer B and HDPE

Eight specimens were produced for each of the lay-ups, wood species and aging condition (described later). Table 3-2 shows the experimental design with a total of 192 specimens.

Table 3-2: **Experimental design of comparative study**

	Parallam®		Douglas Fir Solid Wood	
Treatment	Douglas Fir	Southern Pine	Radial Surface	Tangential Surface
HDPE	8 initial 8 aged	8 initial 8 aged	8 initial 8 aged	8 initial 8 aged
HDPE - tie-layer A	8 initial 8 aged	8 initial 8 aged	8 initial 8 aged	8 initial 8 aged
HDPE - tie-layer B	8 initial 8 aged	8 initial 8 aged	8 initial 8 aged	8 initial 8 aged

After the specimens were finished pressing, they were trimmed down to 51 mm (2 in) wide by 229 mm (9 in) long by 25 mm (1 in) thick blocks for the 90º peel test.

Problems were encountered with 90° peel test of specimens bonded with tie-layer A, which will be discussed in the results section. Therefore, block shear tests, in lieu of 90° peel tests, were conducted for specimens bonded with tie-layer A to overcome problems with missing data due to HDPE barrier layer failure during peel testing.

To prepare shear block specimens for block shear tests, specimens were cut into 50 mm (2 in) wide by 50 mm (2 in) long by 19 mm (¾ in) thick blocks after undergoing the same hot-pressing schedule as described before to bond HDPE to wood substrate using tie-layer A. Then, plastic surfaces of two of these blocks were bonded together by reheating the surfaces with an infrared lamp. The blocks were placed approximately 100 mm (4 in) below the lamp and allowed to heat until the surface temperature reached around 140ºC to 150ºC (Figure 3-6).
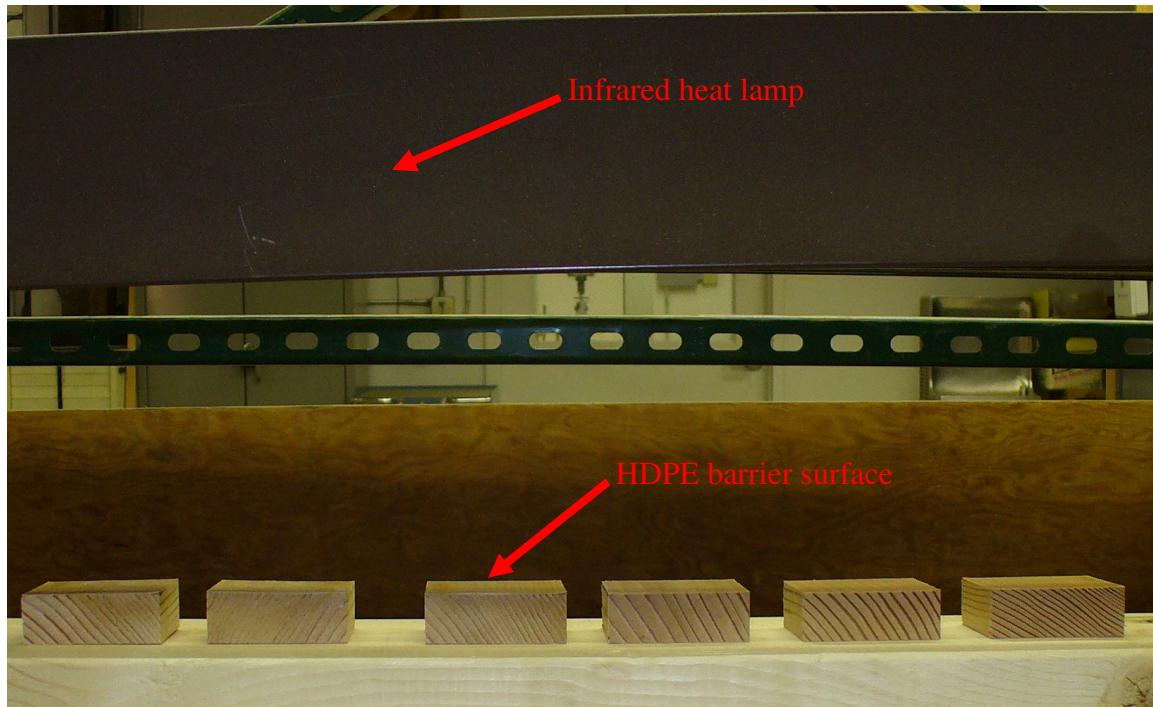
Figure 3-6: Heating of thermoplastic barrier surfaces of hot-pressed specimens using an infrared lamp to fabricate shear block specimens.

Within this temperature range the entire surface of the small blocks was "wetted" (the plastic had softened). Once the plastic was re-melted, the two surfaces were placed together and cold pressed at approximately 700 kPa (~100 psi) for 120 seconds. The specimens were then cut to size according to the testing standard (Figure 3-7).
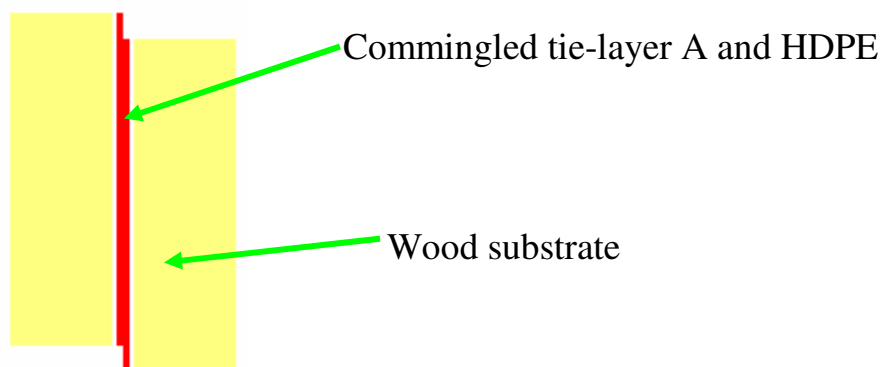
Figure 3-7: Shear block specimen lay-up

3.3.3 Testing Methodology

A 90º peel test (ASTM D 6862) and a block shear test (ASTM D 905) were used to determine the bond strength of the wood-plastic interface. Testing speeds for peel test and block shear test were 254 mm/min and 5 mm/min, respectively. The specimens were tested within 24 hours of pressing, as well as in an accelerated aged condition. The aging test used was ASTM D 1101, which is a test method of the integrity of adhesive joints in structural laminated wood products for exterior use. This test method involves two vacuum-pressure cycles while the specimens are completely submerged in water, and are then dried for 22 hours to complete one full cycle. This weathering test requires a total of two full cycles. Results of bond strength using tie-layers for different wood substrates were compared to those bonded without any tie-layers. However, as two different testing methods were used for evaluating the tie-layers, they were not compared to each other directly.

3.4 Results and Discussion

3.4.1 Tie-layer B – Peel Test Results

As was the case in establishing suitable hot-pressing parameters in Chapter 2, there were difficulties obtaining a consistently good peel for both the Parallam® and solid wood specimens. Only about a third of the specimens had a "good" peel, from which a peel strength value was determined. If the barrier layer started to tear, it was not valid as it did not reflect the peel strength but more of the yield strength of the thermoplastic/tie-layer as it would tear. As mentioned in Chapter 2, it was observed that the plastic was filling the voids on the surface of the wood substrate, thus causing localized thinning of the thermoplastic barrier layer on the

substrate surface. These thinner portions of the plastic tended to tear instead of peeling. Limited

results were obtained from two of the three treatment combinations: HDPE only (no tie-layer)

and HDPE-tie-layer B. These results are tabulated in Tables 3-3 and 3-4. In the specimen

identification, the first abbreviation is for the species in the case of Parallam® or surface

orientation in the case of solid wood (DF – Douglas fir, SP – southern pine, RAD – radial

surface, and TAN – tangential surface). The second abbreviation is for the treatment type (N

indicating HDPE only with no tie-layer and B indicating tie-layer B). The last abbreviation is for

the conditioning of the specimen (no abbreviation means that the specimen was tested within 24

hours of pressing, and WT for after accelerated aging test). Peel strength results are also

graphically represented in Figure 3-8.

Table 3-3: Peel strength values for Parallam®

	DF-N	DF-N-WT	DF-B	DF-B-WT	SP-N	SP-N-WT	SP-B	SP-B-WT
	Parallam®							
	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength
	(MPa)	(MPa)	(MPa)	(MPa)	(MPa)	(MPa)	(MPa)	(MPa)
# of Specimens	2	3	2	2	2	5	2	6
Mean	0.83	1.18	2.26	1.10	1.11	0.78	2.65	0.40
SD	N/A	0.24	N/A	N/A	N/A	0.17	N/A	0.18
COV (%)	N/A	20.3	N/A	N/A	N/A	21.2	N/A	45.4
Minimum	0.68	0.90	1.94	0.66	0.99	0.64	2.32	0.17
Maximum	0.98	1.32	2.58	1.53	1.22	1.05	2.97	0.65

Table 3-4: Peel strength values for Douglas-fir solid wood

	RAD-N	RAD-N-WT	RAD-B	RAD-B-WT	TAN-N	TAN-N-WT	TAN-B	TAN-B-WT
Solid Wood								
	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength
	(MPa)	(MPa)	(MPa)	(MPa)	(MPa)	(MPa)	(MPa)	(MPa)
# of Specimens	6	7	3	4	5	7	6	1
Mean	0.84	0.50	0.99	0.93	0.71	0.53	1.70	0.52
SD	0.28	0.13	0.65	0.36	0.22	0.06	0.57	N/A
COV (%)	33.7	25.3	65.5	39.1	31.2	12.0	33.7	N/A
Minimum	0.50	0.33	0.28	0.46	0.50	0.46	1.19	0.52
Maximum	1.23	0.69	1.55	1.29	1.01	0.64	2.57	0.52

Figure 3-8: Peel strength results.

In general, peel strength improved significantly with the use of tie-layer B for all substrates. This improvement was not as significant when bonded to the radial surface of Douglas-fir solid wood specimens (Figure 3-8). Results also indicate a drop in peel strengths after undergoing the accelerated aging test; however, these strengths are still significantly higher than those reported in Mahlberg et al. (2001) and Kolosick et al. (1993). Aging of specimens had a severe negative effect on the bond strength of tie-layer B specimens. Figure 3-9 shows the separation of tie-layer B and HDPE from the surface of the wood, which led to lower peel strengths. The white area in Figure 3-9 is the area of delamination.

Figure 3-9: Delamination between HDPE barrier layer and wood composite specimens bonded with tie-layer B after aging test.

Before the aging treatment, tie-layer B increased the adhesion of the HDPE to the wood substrate. There is a strength difference between the two species of the Parallam® substrate. The southern pine specimens yielded slightly higher peel strengths than the Douglas fir specimens, which is partly due to the open nature of the southern pine wood to accept treatment. Douglas-fir is generally considered to be refractory in nature due to a common occurrence of pit aspiration during seasoning of lumber (Meyer 1971, Islam et al. 2008).

When comparing the solid wood peel strengths, we can see that the tie-layer B specimens have higher peel strengths than the specimens without a tie-layer. This was more significant on the tangential surface than the radial. Comparing the different treatments we see that the specimens that do not have a tie-layer performed better on the radial specimens. This is partially due to the ability of the HDPE to penetrate through the radial surface due to presence of pits between ray parenchyma and longitudinal cells in Douglas fir (Summitt and Sliker 1980). However, when comparing the specimens with tie-layer B, the tangential specimens showed greater peel strengths. In both cases, aging had a detrimental affect on peel strength. Further tests are necessary to reliably conclude differences in wood surfaces and affects of aging as there was only one valid specimen for the aged condition for tie-layer B on the tangential wood substrate.

The statistical significance of wood substrate and use of tie-layer B was determined using an analysis of variance (ANOVA). It should be noted that in some groups, only two valid peel test values were available. Results indicate a significant difference in the effects across all group types. Further comparison of means based on Duncan's multiple range test was preformed at a significance level of 0.05 ($\alpha = 0.05$). Table 3-5 shows the comparison of means results where means with the same letter are not significantly different at an α-level of 0.05. Comparison of means results confirm the observations made earlier.

Table 3-5: Duncan's multiple range test for peel strength of specimens with and without tie-layer B (means with the same letter are not significantly different at α-level of 0.05).

Specimen Type	Mean Value (N/mm)	Duncan Grouping			
SP-B	2.64	A			
DF-B	2.26	A			
Tan-DF-B	1.53		B		
DF-N-WT	1.18		B	C	
SP-N	1.10		B	C	
DF-B-WT	1.10		B	C	
Rad-DF-B	0.95		B	C	D
DF-N	0.83			C	D
SP-N-WT	0.78			C	D
Rad-DF-N	0.66			C	D
Tan-DF-N	0.61			C	D
SP-B-WT	0.40				D

3.4.2 Tie-Layer A – Block Shear Test Results

When a 90° peel test was conducted on HDPE barrier layer bonded with tie-layer A, the plastic tab always failed. This could have been due to a variety of factors including surface plastic thinning around voids in composite wood substrate, excellent bond strength, or the brittle nature of tie-layer A. Once the tie-layer and HDPE had re-crystallized (cooled) after pressing, the tab became stiff and rigid and broke as it was bent to a 90° angle. Therefore, to evaluate the bond strength developed due to tie-layer A, a block shear test was performed on all specimens bonded with this tie-layer. A large majority of the shear blocks failed not at the wood-plastic interface, but in the wood structure itself, as shown in Figure 3-10. The lines indicate the wood area that failed in shear.

Figure 3-10: Wood failure after block shear test bonded with tie-layer A.

The results from the block shear tests are tabulated in Tables 3-6 and 3-7. For the specimen identification, all of the previously mentioned identifications apply except for the second abbreviation which is for the treatment type (A – tie-layer A and CON – control, which means block shear specimens cut from the substrate with no bond layer at the shear interface). For solid wood specimens, shear values from the Wood Handbook (USDA 1999) was taken as benchmark for comparison. In every case, the shear strength of specimens bonded with tie-layer yielded higher values than control specimens indicating that the bond formed with tie-layer A is as strong as the shear strength of the wood substrate (Parallam®). This establishes at least the lower limit of the bond strength that can be achieved with tie-layer A. Figure 3-11 graphically illustrates the shear strength results.

Table 3-6: Shear strength results for Parallam® specimens in both the initial and aged condition.

	Parallam®					
	DF-A	DF-A-WT	DF-CON	SP-A	SP-A-WT	SP-CON
	Shear Strength (MPa)	Shear Strength (MPa)	Shear Strength (MPa)	Shear Strength (MPa)	Shear Strength (MPa)	Shear Strength (MPa)
# of Specimens	12	12	24	12	12	24
Mean	6.5	5.6	5.9	8.2	7.4	7.5
SD	2.5	1.3	1.2	1.7	1.6	1.0
COV (%)	39.1	23.5	20.3	20.8	21.0	13.0
Minimum	2.9	2.3	2.8	6.1	5.0	5.5
Maximum	11.1	6.9	8.1	11.7	10.5	9.5

Table 3-7: Shear strength results for solid wood specimens in both the initial and aged conditions.

	Solid Wood			
	RAD-A	RAD-A-WT	TAN-A	TAN-A-WT
	Shear Strength (MPa)	Shear Strength (MPa)	Shear Strength (MPa)	Shear Strength (MPa)
# of Specimens	12	12	12	12
Mean	8.8	2.8	8.0	3.5
SD	1.5	1.5	3.7	2.5
COV (%)	16.6	52.9	46.4	70.7
Minimum	6.5	0.8	2.4	0.5
Maximum	10.6	5.8	15.7	8.0
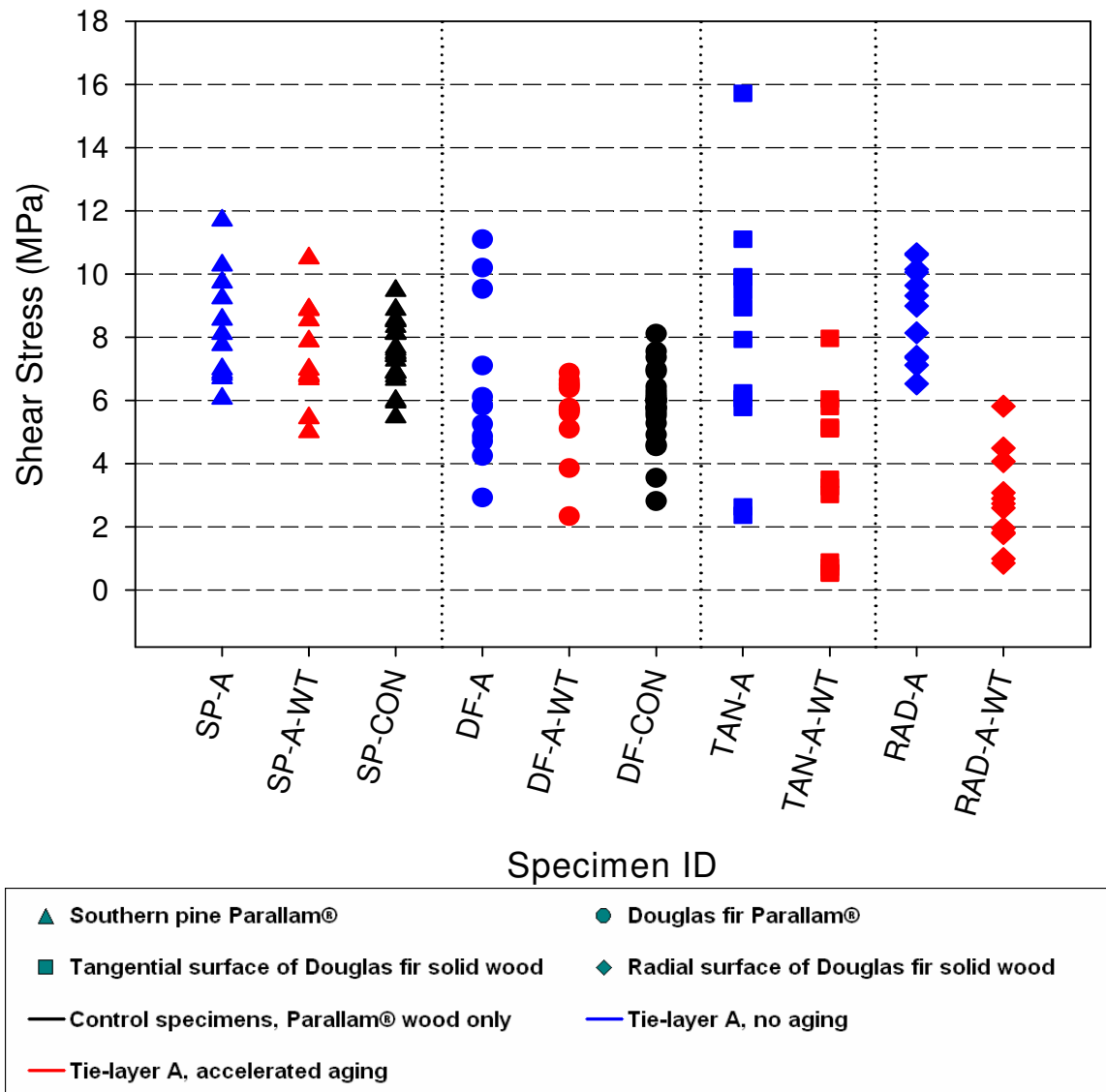
Figure 3-11: Shear strength results with tie-layer A.

As the results in Figure 3-11 indicate, shear strength of the tie-layer A bonded specimens did not change significantly even after being subjected to an aging test in accordance with ASTM D 1101. On the average, shear strength of Parallam® specimens of both species decreased by less than 15 percent after undergoing accelerated aging. Even with this decrease in shear strength, it was not significantly different than the shear strength of the substrate itself. As for the solid wood specimens, there was no control specimens tested. However, according to the

Wood Handbook (USDA 1999) the shear strength of Douglas fir is 7.8 MPa. Again when

compared to the treated specimens, those that were bonded with tie-layer A yielded higher shear

strength values leading to failure in the wood. However after the aging of the solid wood, the

shear strength was greatly reduced for both surfaces (by 68 percent for radial and 56 percent for

tangential). This was caused by the differential shrinking and swelling during the aging process.

It was observed during testing that nearly half of the specimens had some degree of interfacial

separation due to shrinking or swelling of the wood. Analysis of variance results on block shear

values once again indicated a significant difference between the treatments investigated. Table

3-8 shows the comparison of the means results where means with the same letter are not

significantly different at an α-level of 0.05.

Table 3-8: Duncan's multiple range test for shear strength of specimens with and without tie-layer A (means with the same letter are not significantly different at α-level of 0.05).

Specimen Type	Mean Value (MPa)	Duncan Grouping			
RAD-A	8.83	A			
SP-A	8.24	A			
TAN-A	8.01	A	B		
SP-CON	7.52	A	B		
SP-A-WT	7.43	A	B		
DF-A	6.48		B	C	
DF-CON	5.83			C	
DF-A-WT	5.58			C	
TAN-A-WT	3.55				D
RAD-A-WT	2.76				D

Due to time constraints, only a limited evaluation of the bond interface was conducted.

Scanning electron microscopy (SEM) was used to verify that the pressing process was causing

plastic to penetrate into the wood substrate. A Hitachi S-570 scanning electron microscope was

used (voltage 20kV, working distance 20 mm) to obtain the following micrograph. Figure 3-12

shows a SEM micrograph of a Douglas fir Parallam[®] specimen that was pressed with tie-layer B

under the following conditions: 180ºC – 1035 kPa – 300 seconds. The green line shows the

interface between the thermoplastic and wood substrate. The arrow points to an area that was

filled with thermoplastic; note that it is below the interface. Also visible is the filling of several

cells along the wood-plastic interface. Gacitua and Wolcott (2008) used a vacuum bagging

process that looked at the bonding between HDPE and neat wood without coupling agents. They

found that the mobility of the thermoplastic was greatly influenced by the anatomical features,

namely the collapse of cells in specific wood species.

Thermoplastic pool
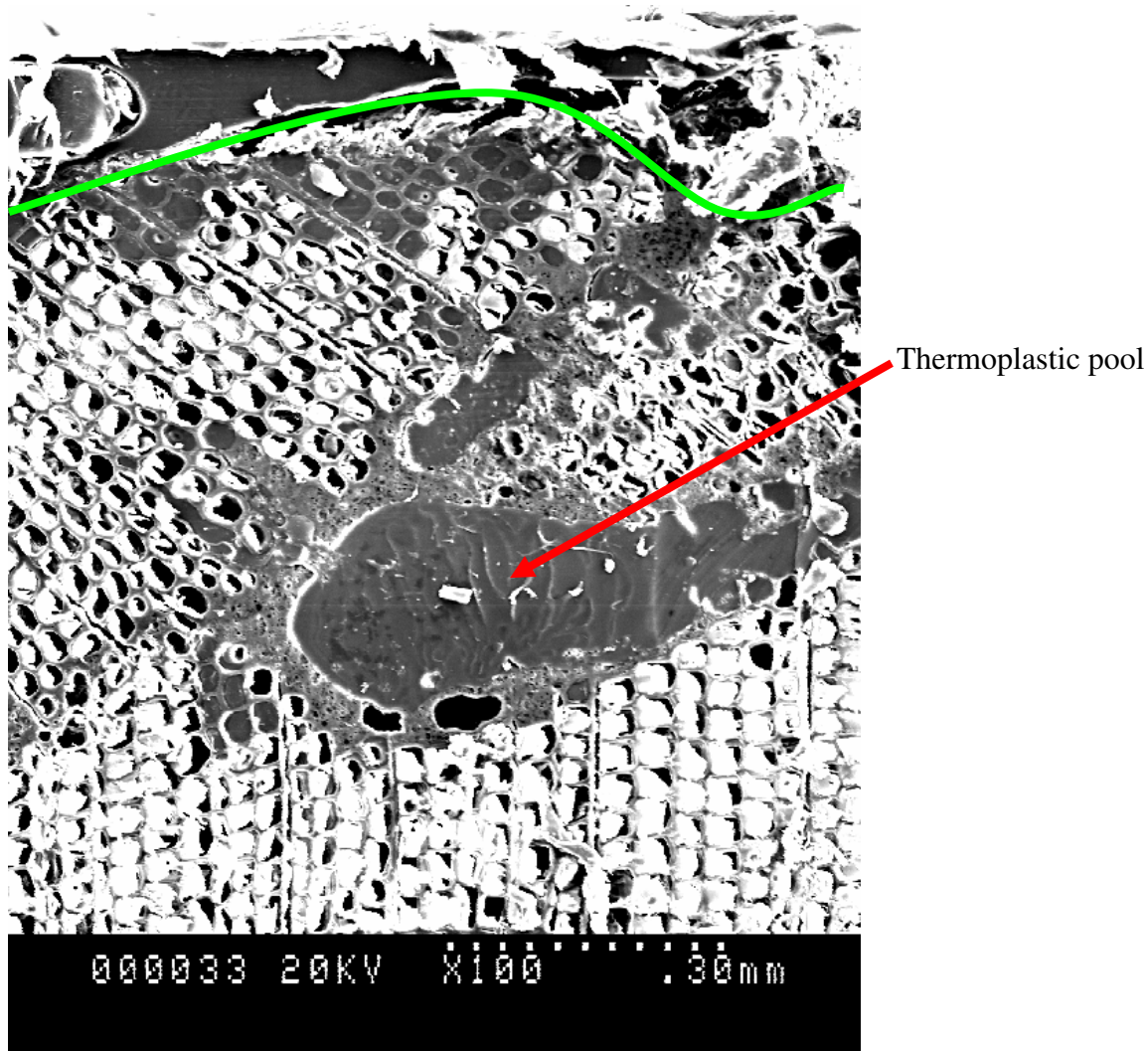
Figure 3-12: SEM micrograph of Douglas-fir Parallam® specimen showing penetration of thermoplastic into wood substrate.

3.5 Summary and Conclusions

This study looked at the ability of thermoplastics (HDPE) to bond to the substrate of a wood composite (Parallam®) with and without the use of tie-layers. The bond was tested using two test methods, a 90º peel test (ASTM D 6862) and a shear block test (ASTM D 905). The specimens were also tested after an accelerated aging process (ASTM D 1101) to simulate an exterior application.

The results from the peel tests (Figure 3-8) indicated that tie-layer B increased the bond strength of the HDPE barrier layer bonded to the wood substrates. The HDPE barrier layer without a tie-layer had an average peel strength of 0.97 N/mm, and the specimens with tie-layer B had an average peel strength of 2.45 N/mm, an increase of over 150 percent. However, there is no significant difference in the bond strengths between the two species, Douglas-fir and southern pine Parallam®. These peel strengths are significantly higher than what was found in previous studies, which showed a maximum peel strength of about 0.65 N/mm (Mahlberg et al. 2001). There does appear to be a difference in the bond strength, however, between the species after the accelerated aging process. The accelerated aging process that was used in this study followed ASTM D 1101, which is a test method for determining the integrity of adhesive joints in structural laminated wood products for exterior uses. The Douglas-fir Parallam® specimens with tie-layer B retained approximately 49 percent of their initial peel strength, however southern pine Parallam® specimens with no tie-layer retained approximately 71 percent of their initial peel strength. It was also noticed that tie-layer B did not survive the accelerated aging test very well (Figure 3-9). The southern pine Parallam® specimens with tie-layer B only retained approximately 15 percent of their initial peel strength.

The results from the shear block tests (Figure 3-10) indicated that the bond between tie-layer A and the wood substrate was stronger in shear than the wood substrate alone. The tie-layer A specimens had a very strong bond to the wood substrate not only on the Parallam® specimens, which had macro-pores to fill on the surface, but also the solid wood specimens. There was a significant difference in the shear strengths between the species. The southern pine Parallam® specimens had higher shear values (8.24 MPa) than the Douglas-fir Parallam® specimens (6.48 MPa). Accelerated aging of the block shear specimens did not significantly reduce the shear strength between HDPE barrier layer and Parallam® substrate. The solid wood on the other hand seemed to be greatly influenced by the differential shrinking/swelling of the wood.

Based on the results of this study, it can be concluded that it is possible to achieve a good and durable bond between a laminated structural wood composite and a thermoplastic barrier layer, such as HDPE, with the use of a tie-layer. In this study a maleic anhydride modified HDPE tie-layer showed the greatest ability to bond and to remain bonded to the Parallam® substrate, even after a severe aging test.

3.6 References

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CHAPTER FOUR – PROJECT SUMMARY AND CONCLUSIONS

Preservative chemicals are used to extend the service life of solid and composite wood structural materials. However, many of the chemicals have high concentrations of copper, which can lead to galvanic corrosion of connection hardware and flashing. Borate chemical treatments do not exhibit the problem, but have the disadvantage of leaching – hence limiting its use. Thermoplastic encapsulation is a promising way to improve the durability of wood composites by creating a barrier to moisture uptake and leaching of chemical treatments. Since most polyolefins, such as HDPE, do not bond well to a polar material, such as wood, a possible solution is to use a tie-layer to improve the bond. In this study, the efficacy of two tie-layers in bonding HDPE to treated wood substrate was examined along with determining the ideal hot-pressing parameters necessary to achieve a good bond. The two types of tie-layers evaluated were a maleic anhydride modified HDPE tie-layer (referred to as tie-layer A) and a styrene-butadiene polymer tie-layer (referred to as tie-layer B). Douglas fir and southern pine Parallam$^{®}$ and Douglas fir solid wood were the substrates investigated. All materials were pressure treated with boric oxide. Bonding to both the radial and tangential surfaces of solid wood specimens were examined.

In Chapter 2, the effects of temperature, pressure and press time, on the bond strength between thermoplastic barrier layer and the wood substrate was examined. Based on the general trends and to minimize the required energy during the hot-pressing process, it was judged that the best combination of the parameters was a platen temperature of 180ºC, a peak pressing pressure of 1035 kPa and a press time of 300 seconds.

Using the processing parameters obtained in Chapter 2, 90º peel and block shear tests were conducted in Chapter 3 to examine the efficacy of using two types of tie-layers to improve the bond between HDPE barrier layer and the wood substrates. The results of the peel tests were mixed; however, it was shown that tie-layer B did increase the peel strength when compared to specimens with no tie-layer in the initial condition. Although, the tie-layer B specimens that were aged using ASTM D 1101 preformed poorly when compared to the aged specimens that had no tie-layer. The tie-layer B aged specimens had a high percentage of the bond interface delaminating. In some cases the delamination occurred between the tie-layer and the wood substrate, but in other cases it occurred between the tie-layer and the HDPE.

Shear block tests were preformed on the tie-layer A specimens which showed very good adhesion properties. When compared to the control Parallam® shear blocks, the tie-layer A specimens had considerably higher shear strengths. A large majority of the shear blocks failed not at the wood-plastic interface, but in the wood structure itself.

From this study the best bond that was produced was the maleic anhydride modified HDPE tie-layer – HDPE plastic combination bonded to a southern pine Parallam® substrate. It is hard to compare the two tie-layers directly, because they were tested with two separate tests.

Major Conclusion

It has been shown that HDPE barrier layer can be successfully bonded to treated Parallam® using at least one of the tie-layers (A). The shear strength of the comingled thermoplastic – wood composite was higher or equal to the wood composite's shear strength. Also there was only a slight loss of shear strength after the aging process.

Recommendations

If this study is continued, there are a few things that might be considered. First, the processing parameters could be reexamined using more specimens and with more rigorous statistical analysis. Second, there was considerable trouble with the 90º peel test, the plastic layer was too thin to get a "good" peel. This can be addressed in a couple ways. First, the plastic could be built up on the surface either before pressing or with secondary processes after pressing. Another way to address this issue would be to direct the force of the press directly onto the plastic without allowing it to squeeze out over the edges of the specimen. The other option is to look at another test to determine the "quality" of the bond interface. A test that might give useful information would be the shear block test that was used for the tie-layer A specimens.

The major idea for this study was to encapsulate an entire Parallam® beam with a thermoplastic barrier layer. One alternative to encapsulate the entire beam is to use an autoclave, to control both temperature and pressure. To produce an encapsulated Parallam® beam, first wrap the beam with a tie-layer and barrier layer film (or have a commingled plastic film). Then place the wrapped beam into a vacuum bag and seal. This will allow for a vacuum to be drawn while the thermoplastic softens and the pressure builds. The vacuum will keep the thermoplastic from squeezing out. Consideration should also be given to suspending the beam to insure equal pressure from all sides. A second alternative is to explore using pultrusion.

Another issue to address is *in situ* coating of areas exposed when onsite work is done such as cutting to length or drilling holes for bolts or lag screws. The protective barrier only works if there are no "leaks" in the member. However, beams almost always need to be cut and/or drilled for bolt holes. So, some type of field treatment is needed, such as commercial sealants.

APPENDIX A - TABLES

Table A- 1: Peel strengths for platen temperature of 160ºC

160ºC								
1035 kPa			1380 kPa			1725 kPa		
120 s	300 s	600 s	120 s	300 s	600 s	120 s	300 s	600 s
Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength
(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)
3.06	1.59	1.89	1.67	1.50	2.64	2.81	2.31	0.97
3.36	1.87	0.99	2.49	2.23	2.22		1.77	1.58
	1.37	3.09		2.87				1.59
	1.83			2.57				
	1.63							
	1.94							

	1035 kPa			1380 kPa			1725 kPa		
	120 s	300 s	600 s	120 s	300 s	600 s	120 s	300 s	600 s
Mean	3.21	1.71	1.99	2.08	2.29	2.43	2.81	2.04	1.38
SD	N/A	0.21	1.06	N/A	0.87	N/A	N/A	N/A	0.35
COV (%)	N/A	12.51	52.93	N/A	37.93	N/A	N/A	N/A	25.66
Minimum	3.06	1.37	0.99	1.67	1.50	2.22	2.81	1.77	0.97
Maximum	5.63	1.94	3.09	2.49	3.87	2.64	2.81	2.31	1.59

Table A- 2: Peel strengths for platen temperature of 180ºC

180ºC								
1035 kPa			1380 kPa			1725 kPa		
120 s	300 s	600 s	120 s	300 s	600 s	120 s	300 s	600 s
Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength
(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)
2.29	1.80	0.82	2.06	3.24	2.59	3.15	2.13	3.78
1.96	1.84	1.81	3.73	3.20	2.86	2.00	1.85	2.07
1.63	1.88			2.58			1.66	
	2.45			2.06				

	1035 kPa			1380 kPa			1725 kPa		
	120 s	300 s	600 s	120 s	300 s	600 s	120 s	300 s	600 s
Mean	1.96	1.99	1.32	2.90	2.77	2.73	2.57	1.88	2.92
SD	0.33	1.01	N/A	N/A	0.56	N/A	N/A	0.24	N/A
COV (%)	16.79	50.73	N/A	N/A	20.34	N/A	N/A	12.64	N/A
Minimum	1.63	1.80	0.82	2.06	2.06	2.59	2.00	1.66	2.07
Maximum	2.29	4.18	1.81	3.73	3.24	6.61	3.15	2.13	4.53

Table A- 3: Peel strengths for platen temperature of 200ºC

	200ºC								
	1035 kPa			1380 kPa			1725 kPa		
	120 s	300 s	600 s	120 s	300 s	600 s	120 s	300 s	600 s
	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength	Peel strength
	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)
	3.03	2.95	6.19	1.76	4.03		4.21	2.64	5.80
	2.43	5.55	3.51	1.19	1.51			3.13	
		4.18							
		2.30							
		1.92							
		2.36							
		2.02							
		2.59							
Mean	2.73	2.98	4.85	1.47	2.77	0.00	4.21	2.89	5.80
SD	N/A	1.26	N/A	N/A	N/A	N/A	N/A	N/A	N/A
COV (%)	N/A	42.22	N/A	N/A	N/A	N/A	N/A	N/A	N/A
Minimum	2.43	1.92	3.51	1.19	1.51	0.00	4.21	2.64	5.80
Maximum	3.03	5.55	6.19	1.76	4.03	0.00	4.21	3.13	5.80

Table A- 4: Peel strengths for Parallam® specimens with no tie-layer and tie-layer B

	Parallam®							
	DF-N	DF-N-WT	DF-B	DF-B-WT	SP-N	SP-N-WT	SP-B	SP-B-WT
	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength
	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)
	0.98	1.31	2.58	0.66	1.22	0.67	2.32	0.23
	0.68	0.90	1.94	1.53	0.99	0.64	2.97	0.54
		1.32				1.05		0.65
						0.82		0.42
						0.73		0.17
								0.41
Mean	0.83	1.18	2.26	1.10	1.11	0.78	2.65	0.40
SD	N/A	0.24	N/A	N/A	N/A	0.17	N/A	0.18
COV (%)	N/A	20.3	N/A	N/A	N/A	21.2	N/A	45.4
Minimum	0.68	0.90	1.94	0.66	0.99	0.64	2.32	0.17
Maximum	0.98	1.32	2.58	1.53	1.22	1.05	2.97	0.65

Table A- 5: Peel strengths for solid wood specimens with no tie-layer and tie-layer B

Solid Wood							
RAD-N	RAD-N-WT	RAD-B	RAD-B-WT	TAN-N	TAN-N-WT	TAN-B	TAN-B-WT
Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength	Peel Strength
(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)	(N/mm)
0.50	0.69	1.55	0.84	1.01	0.47	1.27	0.52
1.10	0.49	1.14	1.12	0.87	0.58	2.57	
1.23	0.42	0.28	1.29	0.59	0.64	1.54	
0.91	0.58		0.46	0.57	0.56	1.36	
0.64	0.40			0.50	0.51	1.19	
0.69	0.33				0.51	2.25	
	0.58				0.46		
Mean 0.84	0.50	0.99	0.93	0.71	0.53	1.70	0.52
SD 0.28	0.13	0.65	0.36	0.22	0.06	0.57	N/A
COV (%) 33.7	25.3	65.5	39.1	31.2	12.0	33.7	N/A
Minimum 0.50	0.33	0.28	0.46	0.50	0.46	1.19	0.52
Maximum 1.23	0.69	1.55	1.29	1.01	0.64	2.57	0.52

Table A- 6: Block shear strengths for Parallam® specimen's with tie-layer A

Parallam®							
DF-A	DF-A-WT	DF-CONTROL		SP-A	SP-A-WT	SP-CONTROL	
Shear Strength	Shear Strength	Shear Strength		Shear Strength	Shear Strength	Shear Strength	
(MPa)	(MPa)	(MPa)		(MPa)	(MPa)	(MPa)	
2.9	5.7	2.8	5.6	6.7	8.9	8.6	7.5
5.2	3.9	6.0	6.3	6.8	6.8	7.4	8.1
11.1	6.5	3.5	5.5	9.7	8.5	6.9	6.0
4.9	6.9	6.9	5.7	7.0	6.8	8.3	6.9
7.1	6.4	4.9	4.5	8.6	6.8	7.6	5.5
10.2	6.5	8.1	6.4	6.9	7.0	5.9	8.5
5.8	5.6	6.0	5.3	7.8	8.9	7.7	8.5
6.1	2.3	6.0	7.4	8.1	6.7	6.8	9.5
5.9	5.8	7.0	5.8	10.3	10.5	8.1	8.1
9.5	5.7	6.2	4.6	6.1	7.9	7.3	6.9
4.7	5.1	5.8	7.6	11.7	5.0	7.4	8.9
4.2	6.6	5.9	6.1	9.2	5.4	7.5	6.7
Mean 6.5	5.6	5.9		8.2	7.4	7.5	
SD 2.5	1.3	1.2		1.7	1.6	1.0	
COV (%) 39.1	23.5	20.3		20.8	21.0	13.0	
Minimum 2.9	2.3	2.8		6.1	5.0	5.5	
Maximum 11.1	6.9	8.1		11.7	10.5	9.5	

Table A- 7: Block shear strengths for solid wood specimen's with tie-layer A

	Solid Wood			
	RAD-A	RAD-A-WT	TAN-A	TAN-A-WT
	Shear Strength (MPa)	Shear Strength (MPa)	Shear Strength (MPa)	Shear Strength (MPa)
	9.0	2.0	2.4	0.9
	9.6	2.9	11.1	0.5
	8.1	1.0	9.9	6.0
	6.5	1.8	6.1	3.5
	10.1	2.6	7.9	0.7
	10.6	1.8	8.9	5.2
	7.3	2.7	15.7	3.0
	9.3	4.5	2.6	3.3
	7.1	5.8	9.5	5.8
	10.6	4.1	6.2	0.6
	7.4	3.1	5.8	5.1
	10.1	0.8	9.9	8.0
Mean	8.8	2.8	8.0	3.5
SD	1.5	1.5	3.7	2.5
COV (%)	16.6	52.9	46.4	70.7
Minimum	6.5	0.8	2.4	0.5
Maximum	10.6	5.8	15.7	8.0