SELECTED VOLATILE ORGANIC COMPOUND EMISSIONS AND PERFORMANCE OF ORIENTED STRANDBOARD FROM EXTRACTED SOUTHERN PINE

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Abstract. The impact of a hot water extraction procedure on select volatile organic compound emissions during pressing, as well as on properties of oriented strandboard (OSB) was evaluated. Southern pine strands were extracted with hot water using a rotating digester at 160°C for 22.9 or 53.6 min. Weight loss for the two extraction conditions was 6.3 ± 0.1% (short time) and 9.3 ± 0.9% (long time). The extract contained a mixture of hemicelluloses, acetic acid, and lignin. OSB panels were manufactured both with and without adhesive. The emissions (phenol, methanol, acetaldehyde, and formaldehyde) without adhesive present decreased from 38.2 to 24.2 mg/kg (oven-dry wood) as a result of the high severity factor (HSF) extraction. When adhesive was used, emissions totaled 22.1, 17.0, and 15.6 mg/kg (oven-dry wood) for control, low severity factor, and HSF, respectively. Water sorption and thickness swell were significantly reduced in panels made from extracted strands. Flexural modulus of elasticity of extracted panels exhibited significant increases in both dry and wet conditions. The flexural modulus of rupture and internal bond were slightly reduced in the dry condition as weight loss increased. The extraction procedure shows promise for improving a variety of properties of OSB, including performance, reduced environmental impact, and generation of a valuable chemical feedstock byproduct.

Keywords: Hemicelluloses, hot water extraction, weight loss, physical and mechanical properties, volatile organic compound, VOC.

INTRODUCTION

Over the last 30 yr, several programs and standards have regulated emissions produced during the manufacture of composite wood products. Emissions include particulate matter, carbon monoxide, nitrogen oxides, volatile organic compounds (VOCs), and hazardous air pollutants.

The manufacture of oriented strandboard (OSB) involves the production of VOC emissions from energy production, wood drying, resin blending, board pressing, and product storage (NCASI 1989, 1999; Larsen et al 1992; Carlson et al 1995). Larsen et al (1992) studied the combustion

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products from a wood waste boiler, finding concentrations of formaldehyde up to 80 ppm and up to 350 ppm (volume basis) of total hydrocarbons. The same study found that wood drying produced formaldehyde emissions up to 1 g/kg_{od} (1 g/kg oven-dry wood) and nonmethane organic emissions from 0.3 to 5.7 g/kg_{od}.

Recently, VOC emissions from press stacks have been the focus of much research (Carlson et al 1995; Barry and Corneau 1999; NCASI 1999; Wang and Gardner 1999). Carlson et al (1995) studied the VOC emissions from OSB during hot pressing, finding that formaldehyde emissions increased with increasing press temperature, mat moisture content, resin solids (phenol-formaldehyde), and press time. They also found that the use of polymeric diphenylmethane diisocyanate (pMDI) did reduce emissions compared with wood alone. Wood species significantly influences the amount of VOC emissions, because differing species exhibit varying amounts of volatile and semivolatile extractive compounds as well as different types and percentages of hemicelluloses (Fengel and Wegener 1984; Sjöström 1993; Rice and Erich 2008).

VOCs are primarily produced through the decomposition of hemicelluloses (Carlson et al 1995) which, of the three major components of wood (cellulose, hemicelluloses, and lignin) is most easily extracted from wood (Paredes et al 2008, 2009). The objective of this research is to present a potential process of hemicellulose extraction before OSB manufacture with the objectives of reducing VOC emissions and determining effects on physical and mechanical properties. The mass loss resulting from the extraction process was calculated and a sugar analysis of the extracted compounds was conducted. Finally, selected physical and mechanical properties of the manufactured panels were determined.

MATERIALS AND METHODS

Materials

Industrial southern pine (*Pinus* spp.) strands were donated by Huber Engineered Woods, Commerce, GA. The strands were sampled after the mill core dryer. A majority of fines were removed using an Acrowood Trillium and Diamond Roll combination screen. Typical strand geometries were 101 $\pm$ 3 mm long, 10-40 mm wide, and 0.7 $\pm$ 0.1 mm thick.

A Huntsman pMDI resin was used, having a viscosity of 33.4 Pa·s. The viscosity of the resin was determined on a Brookfield DV-I+ viscometer, model LV using a #3 (LV) spindle at 60 rpm at a resin temperature of 22.2°C. Hexion EW-45 emulsified wax (45% solids) was also used.

Extraction Process

Two hot water extraction (HWE) procedures, one short (low severity factor [LSF]) and one long (high severity factor [HSF]), were used using a custom-built rotating extractor (digester) spinning at 1 rpm. The strands, in 6.2-kg batches, were placed inside a vessel filled with fresh water with a water/wood ratio of 4:1 by weight. Moisture in the wood was included in the ratio calculation. The vessel was heated from room temperature to 160°C in 2.0 h (preheating) followed by constant temperature exposure times of 22.9 min or 53.6 min for the LSF and HSF conditions, respectively (Fig 1).

A total of 22 extraction runs were made, 7 LSF and 15 HSF. The severity factor (SF) can be expressed as a function of extraction time and temperature:

$$SF = log \int_0^t \exp \frac{[T_r - T_b]}{14.75} dt \qquad (1)$$

where t is residence time (min), T_r is reaction temperature (°C), and T_b is base temperature (100°C). The equation assumes that the overall process is hydrolytic and the overall conversion is pseudo first order (Overend and Chornet 1987; Mosier et al 2002). SF ranges from 0 (unextracted wood) to 4. Using Eq 1, including preheating, extraction, and cooling time, an SF of 3.29 was calculated for LSF and 3.59 for HSF.

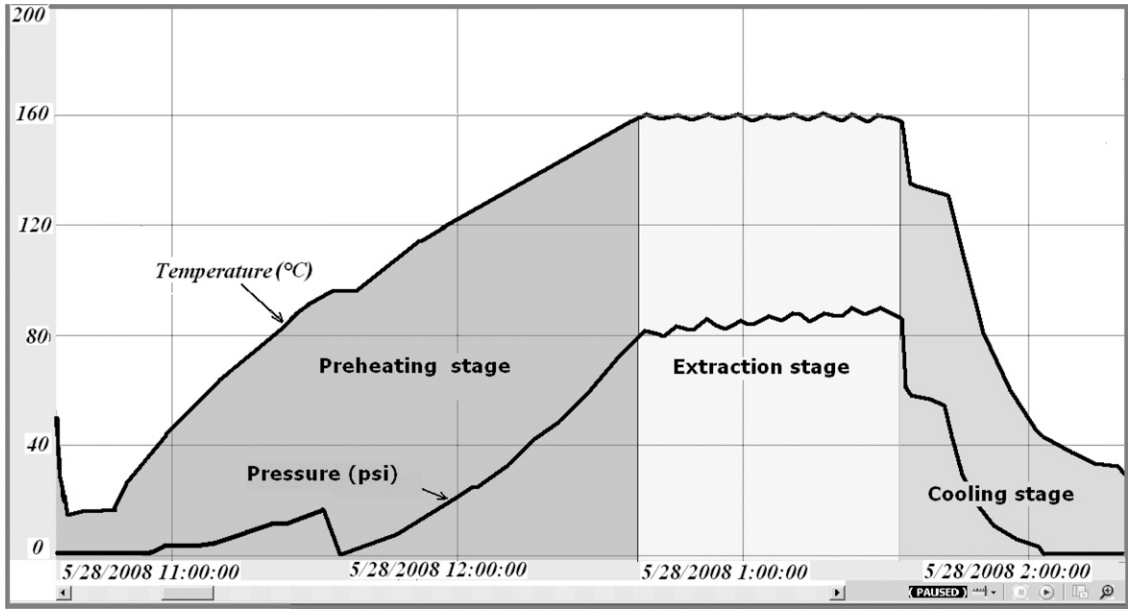

Figure 1. Temperature and pressure profile during hot water extraction process.

Analysis of Liquid Extract

Weight loss of the strands from the extraction process was determined for each run by freeze drying the extracted liquid at −42°C and 1.3-1.9 Pa of vacuum (Paredes et al 2008). The carbohydrate composition of the liquid extract is an important parameter for evaluating VOC emissions and OSB properties because the majority of VOC emissions come from the carbohydrates. High-performance liquid chromatography (HPLC) was used to analyze sugar, lignin, and acetic acid because of the simplicity of sample preparation and ease of adopting a routine (Kaar et al 1991; Lundqvist et al 2003). Sugar analysis was conducted using a single specimen on an HPLC-Shimadzu consisting of a pump, manual injector, refractive index detector, and two independent columns: 1) BioRad Aminex HPX-87H, at 60°C and 0.6 mL/min, using 5 mmol of H_2SO_4; and 2) BioRad Aminex HPX-87P, operated at 80°C and 0.6 mL/min using deionized water. Analysis of lignin and acetic acid were taken from the first column and glucose, xylose, mannose, and arabinose from the second.

Oriented Strandboard Manufacture

Control (unextracted) OSB strands were conditioned in a dehumidification kiln at 32.2°C and 33% RH for 6 da until constant weight was attained for an average 6.2 ± 0.1% MC. Extracted OSB strands were conditioned using the same dehumidification kiln at 32.2°C and 35% RH, again for 6 da, until constant weight was attained for averages of 6.3 ± 0.1% and 6.0 ± 0.1% for LSF and HSF conditions, respectively. Enough strands (39.7 kg) to produce three 19- × 864- × 864-mm panels were placed within a Coil spinning disk atomizing resin blender. pMDI adhesive and emulsified wax were added at a loading of 4 and 1% (solids content based on oven-dry wood), respectively. Resin was applied using a disk speed of 12,000 rpm and drum speed of 20 rpm with resin pumped to the blender at a feed rate of 200 mL/min. The E-wax was applied using a Spraying Systems air atomizer at a feed rate of 120 mL/min for a total blend time of approximately 7 min. Randomly oriented panels were formed manually on a 1.2-mm-thick steel caul plate that was sprayed with a thermosetting

mold release (Stoner E497). A 19-mm-thick aluminum picture frame VOCs collection caul, which also served as a mechanical stop, was placed around the mat. A matching steel caul plate was placed on the mat.

The mats were placed in a 1630-t capacity, 1.22- × 2.44-m hydraulic hot press (Erie Mill and Press) for panel manufacture. The press platens were heated to 204°C using two Mokon hot oil heaters, each with two zones. The PLC-controlled press collected the temperature of each platen, position, hydraulic pressure at 1-s intervals. Core temperature and vapor pressure were measured using a probe inserted into the geometric center of mat. The same press schedule was used for each run (Table 1).

Table 1. Press cycle for manufacture of oriented strandboard panels.

Stage	Ramp time (s)	Dwell time (s)	Mat thickness (mm)
Close	45	1	100.0-19.0
Cook	01	210	19.0
Degas 1	15	1	19.3
Degas 2	15	1	19.6
Degas 3	15	1	19.8
Caul evacuation	45	1	19.8

Volatile Organic Compound Collection

The VOC collection system had two basic parts: a gasketed picture frame caul and a VOC vapor collection apparatus (Fig 2). This collection system was an enhanced version of that used by Carlson et al (1995) and Jiang et al (2002).

The gasketed aluminum caul frame had interior dimensions of 1.04-2.26 m. The mat size, 0.81 m wide × 1.22 m long × 19 mm thick, was chosen for sufficient air space between the mat and the caul to provide for unobstructed flow of VOCs around the mat and to/from the intake/exhaust ports. The final target density of the panels was 640.7 kg/m^3 at 6% MC. Emissions were collected in sequential order as shown in Fig 2, where flasks 1 and 3 had 800 mL of deionized water and flask 2 had 550 mL of methylene chloride (MCL). Immediately after the press cycle had

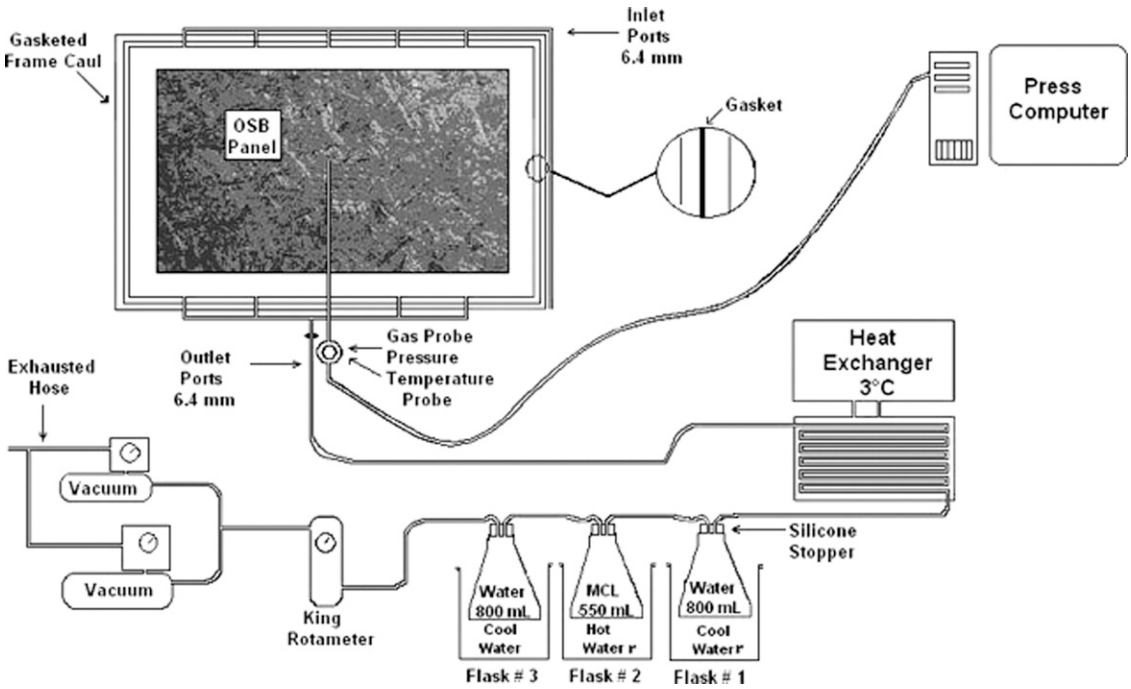

Figure 2. The volatile organic compound collection system.

finished, the VOC collection was stopped. All flasks were then weighed, volume recorded, and the contents poured into a 4-L separation funnel. The separation funnel was agitated by hand and then allowed to stand for 30 min. MCL (300 mL) was added to the separation funnel, which was again agitated and allowed to stand for an additional 30 min. A 20-mL aliquot was obtained from the MCL and water solutions of each panel and immediately refrigerated.

VOC analysis was done by gas chromatography/mass spectrometry (GC/MS) conducted on an Agilent 6890 equipped with an Agilent 5973 mass spectrometer. Three replicates were obtained from each treatment. Phenol content was analyzed from the MCL fraction while all other compounds, methanol, formaldehyde, and acetaldehyde, were analyzed from the water fraction using a 2, 4-dinitrophenyl hydrazine.

Physical and Mechanical Property Determination

Panels were cut into specimens according to the plan shown in Fig 3 for flexural tests, APA test S-6 (adhesive performance, APA 2001), internal bond (IB), water sorption, and thickness swell. All specimens were conditioned at 21 ± 2°C and

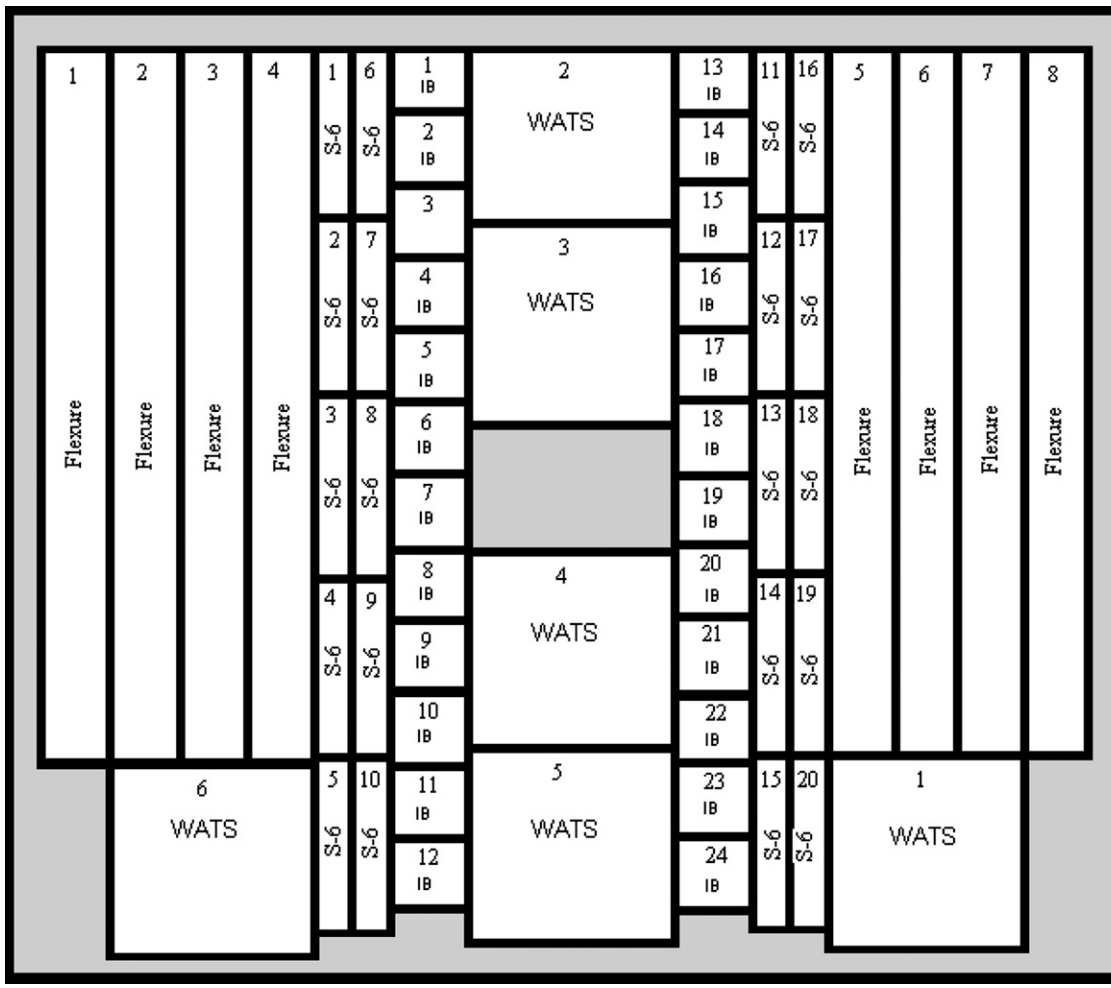

Figure 3. Testing cut-up plan (not to scale).

65 ± 5% RH until constant weight was attained. One-half of the specimens for the mechanical tests and all of the specimens for physical testing were water-soaked for 24 h at 20 ± 2°C following ASTM D1037-06A (ASTM 2006). A 50- × 50-mm specimen was cut from both ends of flexural specimens to determine the vertical density profile (VDP) using a QMS density profilometer.

RESULTS AND DISCUSSION

Hot Water Extraction

HWE of southern pine (SP) strands exhibited similar behavior to other species such as red maple (*Acer rubrum*) strands and loblolly pine (*Pinus taeda*) chips in that weight loss was proportional to extraction time (Yoon et al 2006; Paredes et al 2008, 2009). The influence of SF on weight loss is summarized in Table 2. The weight loss was significantly different ($p = 0.0001$) for the LSF and HSF conditions based on a one-way analysis of variance (ANOVA). It has been reported that other factors such as species, softwood vs hardwood, and type of extraction system (open or closed) also influence weight loss (Hill 2006). In general, under hygrothermal treatment, softwoods have less mass loss than hardwoods (Hill 2006). This is mainly because hardwoods are less thermally stable than softwoods and of differences in the hemicellulosic content and composition (Fengel and Wegener 1984). It is important to note that the weight loss values were less than expected from fresh material because of the prior high-temperature drying of the industrial material.

Carbohydrates

A variety of carbohydrates and acetic acid and lignin were extracted from SP strands (Table 3).

Table 2. Influence of severity factor on weight loss.

Treatment	Number of extractions	Extraction time (min)	Severity factor	Weight loss (%)
Low severity factor	7	23	3.29 ± 0.054	6.3 ± 0.1
High severity factor	15	54	3.69 ± 0.04	9.3 ± 0.9

The production of acids occurs when wood is heated in the presence of water, resulting in a cellular breakdown and/or solubilization of hemicelluloses, lignin, water of constitution, and volatile extractives (Hill 2006; Borrega and Kärenlampi 2008). As anticipated, hemicelluloses were more readily extracted.

The formation of acetic acid during hygrothermal treatment in softwood is because of cleavage of acetyl groups that are located at either the C-2 or C-3 positions of glucose and mannose units of galactoglucomannan, which is 5-10% of the wood weight, with an average of one acetyl group for every four backbone units (Sjöström 1993). The quantity of extracted sugars increased as the SF increased with the exception of arabinose. This reduction is likely from hydrolysis of the galactoglucomannan under these acidic conditions.

Volatile Organic Emissions

In conducting statistical analyses on the results of select VOC emissions, a randomized complete block design (RCBD) was used with the treatments (LSF and HSF) comprising two blocks. This blocking was to determine the influence of resin on select VOC emissions. The first randomized block (no resin [NR]) had two treatments (control and HSF) with three replicates, while the second randomized block (with resin [WR]) had three treatments (control, LSF, and HSF) with three replicates (Table 4). Physical and mechanical properties were measured only from the

Table 3. Materials removed by the hot water extraction process based on oven-dry mass of wood.

Components	Low severity factor (g/kg)	High severity factor (g/kg)
Glucose	8.29	12.83
Xylose	11.97	17.11
Galactose	4.23	6.19
Arabinose	9.14	6.41
Mannose	22.41	32.36
Total sugars	56.04	74.90
Acetic acid	4.01	8.89
Lignin	3.42	8.67
Total weight loss	63.47	92.46

Table 4. Influence of pMDI (resin) and hot water extraction on the amount of select volatile organic compound (VOC) emissions based on oven-dry mass of wood.

Source		Phenol	Methanol	Acetaldehyde	Formaldehyde	Total select VOCs
			(mg/kg)			
No resin	Control	0.0^{NS}	20.0 ± 3.7^{a}	4.7 ± 0.7^{a}	13.3 ± 1.6^{NS}	38.0 ± 2.0
	HSF	0.0^{NS}	9.5 ± 3.0^{a}	1.4 ± 0.4^{a}	13.1 ± 2.8^{NS}	24.0 ± 2.1
With resin	Control	0.0^{NS}	10.2 ± 2.2^{NS}	3.5 ± 0.2^{a}	8.2 ± 0.7^{a}	21.9 ± 1.0
	LSF	0.0^{NS}	10.2 ± 1.8^{NS}	0.7 ± 0.1^{a}	5.9 ± 0.8^{a}	16.8 ± 0.9
	HSF	0.0^{NS}	7.5 ± 1.0^{NS}	0.9 ± 0.1^{a}	7.0 ± 0.8^{a}	15.4 ± 0.6

[a] Bonferroni multiple comparisons at the 95% confidence level
[NS], not significant at the 0.05 probability level.
pMDI, polymeric diphenylmethane diisocyanate; HSF, high severity factor; LSF, low severity factor.

second block. Select VOC emissions were analyzed using ANOVA. In the second randomized block (WR), phenol emissions were not detected by GC/MS for the control or LSF or HSF. Unlike the NR block, methanol emissions were not significantly influenced by extraction condition. The acetaldehyde and formaldehyde emissions, however, were significantly reduced by hot water extraction (HWE).

The addition of pMDI adhesive significantly reduced methanol and acetaldehyde emissions for the nonextracted controls, similar to results found by Carlson et al (1995). Use of extracted strands in the presence of pMDI reduced acetaldehyde and formaldehyde, but not methanol. This reduction was because of the extraction of acetic and formic acid from hemicelluloses during HWE (Table 3).

Total select VOC emissions, the sum of phenol, acetaldehyde, methanol, and formaldehyde, was 38.2 and 24.2 mg/kg_{od} for the control and HSF without resin, respectively. The 37% overall VOC reduction from extraction confirms that the removal of hemicelluloses and acetic acid plays an important role in VOC emissions (Carlson et al 1995). Total select VOC emissions in panels manufactured with resin was 22.1, 17.0, and 15.6 mg/kg wood for control, LSF, and HSF, respectively. In addition to the reduction from HWE, the use of pMDI resin caused a reduction in select VOC emissions by 29%.

Physical Properties

Most physical and mechanical properties are known to be highly correlated with density. No

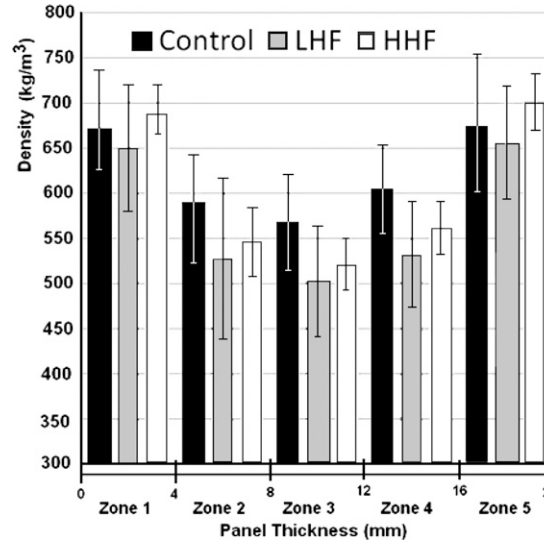

Figure 4. Vertical density profile of oriented strandboard panels for five zones through the panel thickness.

significant difference ($p = 0.2423$) in density was found among the treatments (based on average panel density). However, a density gradient is developed during pressing from differential heat transfer, moisture movement, wood stress relaxation, wood consolidation, and resin curing (Winistorfer et al 2000). There is a strong relationship between VDP and panel properties such as dimensional stability and bending strength that are of critical importance to OSB researchers and manufacturers (Winistorfer et al 2000). The VDP testing verified that for control, LSF, and HSF treatments, there were no significant differences among treatments at the same depth zone (Fig 4).

Equilibrium moisture content of OSB manufactured from extracted strands was significantly lower than the control, which has also been found by others (Schneider and Rusche 1973; Paredes et al 2008, 2009).

The wettability of HWE wood decreases because of the reduction of hydroxyl content (Pétrissans et al 2003) from the extraction of the hemicelluloses (Hill 2006). Table 5 shows that the thickness swell of the extracted material was improved (up to 61%). Paredes et al (2008) found a similar reduction in thickness swell as a result of hemicellulose extraction. In terms of field performance, thickness swell is a critical value for OSB panels and such reduction would have significant value.

OSB panels typically absorb water faster than plywood and lumber (Zhang et al 2007). Higher levels of resin and wax are often used reduce the sorption of moisture to mitigate decay and lessen effects on mechanical properties (Bowyer et al 2007; Zhang et al 2007). Table 5 shows that the sorption after 24-h water soak for the HSF panels was significantly lower than both the control and LSF panels.

Sorption for the control specimens after a 24-h water soak was within the range found by Gu et al (2005). OSB is a material with high variation in structure and composition, and for this reason, water is not only in the cell wall and lumens, but also between flakes. The reduction of water sorption in HWE panels is because of reduced swelling (less interflake voids) (Paredes et al 2008), lower starting equilibrium moisture content, and less hemicelluloses (Paredes et al 2009). Therefore, extracted OSB panels had better water resistance properties, although cell wall porosity was increased (Paredes et al 2009).

Mechanical Properties

Mechanical results are presented in Tables 6 and 7. The experimental statistical design used a

Table 5. Physical properties of oriented strandboard.

Treatment	Density (kg/m^3)	Equilibrium moisture content (%)	24-h thickness swell (%)	24-h water sorption (%)
Control	591 ± 22^{NS}	10.3 ± 0.1^{a}	9.9 ± 1.2^{a}	29.2 ± 3.4^{a}
Low severity factor	596 ± 32^{NS}	7.3 ± 0.1^{a}	6.0 ± 1.4^{a}	26.0 ± 4.9^{a}
High severity factor	602 ± 30^{NS}	6.8 ± 0.1^{a}	3.8 ± 0.8^{a}	20.0 ± 1.9^{a}

[a] Bonferroni multiple comparisons at the 95.0% confidence level
[NS], not significant at the 95% probability level.

Table 6. Oriented strandboard flexural properties.

	Modulus of rupture (MPa)		Stress at the proportional limit (MPa)		Modulus of elasticity (GPa)	
Treatment	Dry	Wet	Dry	Wet	Dry	Wet
Control	29.8 ± 5.4^{NS}	15.9 ± 2.2^{a}	18.3 ± 2.1^{NS}	9.5 ± 0.7^{a}	3.46 ± 0.21^{a}	1.65 ± 0.19^{a}
Low severity factor	27.6 ± 3.9^{NS}	22.7 ± 3.8^{a}	17.8 ± 0.9^{NS}	13.2 ± 0.9^{a}	4.05 ± 0.20^{a}	2.53 ± 0.25^{a}
High severity factor	26.5 ± 4.9^{NS}	22.7 ± 4.9^{a}	16.7 ± 2.3^{NS}	13.0 ± 2.0^{a}	4.14 ± 0.22^{a}	2.67 ± 0.34^{a}

[a] Bonferroni multiple comparisons at the 95% confidence level
[NS], not significant at the 0.05 probability level.

Table 7. Oriented strandboard internal bond (IB) and APA adhesive performance S-6 results.

	IB (MPa)		S-6 (MPa)	
Treatment	Dry	Wet	Dry	Wet
Control	0.72 ± 0.14^{NS}	0.22 ± 0.06^{a}	34.5 ± 5.1^{NS}	14.2 ± 3.1^{a}
Low severity factor	0.69 ± 0.16^{NS}	0.35 ± 0.11^{a}	31.4 ± 6.3^{NS}	18.1 ± 3.1^{a}
High severity factor	0.62 ± 0.17^{NS}	0.37 ± 0.10^{a}	30.7 ± 4.3^{NS}	18.3 ± 4.1^{a}

[a] Bonferroni multiple comparisons at the 95.0% confidence level
[NS], not significant at the 95% probability level.

RCBD, blocking dry and wet conditions. These two blocks had three treatments each: control, LSF, and HSF. Dry bending strength (MOR) and dry bending stress at the proportional limit (SPL) were not significantly different among the treatments while the MOR and SPL of extracted panels in the wet condition were significantly higher than the control, 42.6 and 36.9%, respectively. Dry MOE was significantly different among the treatments with a maximum increase of 19.7% at HSF vs control, whereas the wet MOE also differed significantly among the treatments with a maximum increase of 61.2% at HSF vs control.

Bond strength was evaluated using both IB and the APA S-6 protocols. Under dry conditions, the slight decrease in bond quality was not statistically significant. In contrast, bond strength performance in the wet condition was significantly improved by the hot water extraction process.

In summary, mechanical properties in the wet condition showed consistently higher increases for the extracted material than the controls. This is presumably from lower swelling of the extracted wood (Table 5), thereby reducing swelling stresses and bond breakage relative to controls.

CONCLUSIONS

This study has identified that emissions of methanol and acetaldehyde can be reduced by conducting a hot water extraction of the strands before hot pressing. Reductions in formaldehyde emissions were also detected when a pMDI adhesive was used. Oriented strandboard panels manufactured from the hot water-extracted strands exhibited significant decreases in equilibrium moisture content, 24-h thickness swell, and water absorption. At equal panel densities, the flexural modulus and stress at the proportional limit increased at both dry and wet conditions for extracted material. The flexural strength (MOR) and IB of panels from extracted material decreased with extraction level in the dry condition but maintained or improved performance when tested in the wet state.

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