Wood-Thermoplastic Composites Manufactured Using Beetle-Killed Spruce from Alaska

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The primary objectives of the study were to characterize the critical properties of wood flour produced using highly deteriorated beetle-killed spruce for wood-plastic composite (WPC) production and evaluate important mechanical and physical properties of WPC extruded using an industry standard formulation. Chemical composition analysis indicated no significant differences in wood constituents between highly deteriorated and sound wood. Preliminary investigation with Fourier transform infrared spectroscopy (FTIR), however, indicated partial degradation or depolymerization of carbohydrate components in highly deteriorated wood compared to sound wood from green trees; effects of these changes could be seen in cell collapse and poor interaction between thermoplastic matrix and deteriorated wood fiber. Physical and mechanical properties of extruded WPCs manufactured from highly deteriorated material were comparable to WPC properties produced using pine wood flour that served as a control material. POLYM. ENG. SCI., 49:129–136, 2009. **© 2008 Society of Plastics Engineers**

INTRODUCTION

In Alaska, the most damaging natural occurrence to the forests south of the Alaska Range, is the spruce bark beetle (SBB), which attacks white (*Picea glauca* (Moench) Voss), sitka (*Picea sitchensis* (Bong) Carriere), and Lutz (*Picea lutzii* Little) spruce [1–4]. Beetle infestation kills the trees and further exposes them to fungal decay and weathering over time, thus damaging the wood. In this article, the authors examine if this damaged wood can be used in producing wood-plastic composites (WPCs).

Currently valued at nearly $1 billion (USD) annually, WPC products a growing market segment with an average annual growth of 25% a year since 1998 [5]. WPCs are primarily used in the auto industry and also the construction industry (specifically as residential decking and moulding). The primary components of a WPC are wood in a particulate form and thermoplastic resins. An extruded WPC is comprised of 50–70% wood flour by weight. Presently, maple and pine are the most common wood flours utilized in WPC extrusion. Polyethylene (PE), polypropylene (PP), and polyvinyl chloride (PVC) are the most commonly used thermoplastics for WPC manufacturing. Besides wood and thermoplastic resins, many other additives are also used in WPC manufacturing to affect processing and performance [6–8]. Profile extrusion is the most common WPC manufacturing process. Performance of WPC depends on the inherent properties of individual components of the formulation, the type of additives, the interactions between the individual components, the processing parameters, and the end use environment. Generally, as the percent of wood filler is increased, there is an increase in the stiffness of the thermoplastic composite. Because of higher percentages of plastic, WPCs absorb less moisture and the rate of absorption is much slower than that of solid wood. The wood filler in WPCs makes them more thermally stable. These attributes make WPCs more dimensionally stable and resistant to fungal decay.

As the beetle infestation continues to spread and trees already dead continue to deteriorate, opportunities to produce primary wood products decline rapidly [9, 10]. However, WPCs, which use wood flour as a feedstock, could potentially use wood flour from beetle-damaged/killed trees, with limited concern for the integrity of the solid wood source.

The primary goal of the study was to characterize raw material properties critical for WPC production and to evaluate important mechanical and physical properties of WPC extruded using an industry standard formulation. A secondary goal was to determine if height location of deteriorated wood within the tree had any significant

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effect on raw material and WPC properties evaluated. Specific subobjectives to achieve the goals of this study were to characterize chemical and thermal analysis of the highly deteriorated wood flour produced from beetle-killed spruce; describe the particle size distribution of nominal 60-mesh wood flour generated from highly deteriorated wood; evaluate the physical and mechanical properties of extruded WPC material using highly deteriorated wood; and investigate the effect of location of highly deteriorated wood source by tree height (butt, middle, and top log) on wood flour and WPC properties.

EXPERIMENTAL DETAILS

Three geographic locations containing 10 live and five dead, highly deteriorated trees were chosen from three different sites on the Kenai Peninsula, Alaska. Highly deteriorated trees were defined and evaluated based on visual characteristics. Three 2.4 m (8 ft) logs were cut from each tree: one from the base (butt log) of the tree, one from below the 10 cm (4 in) top (top log), and the third equidistant between the two (middle log). Each log was labeled with the tree number and height position within the tree. While keeping the butt, middle, and top logs separate, they were all processed into wood flour to extrude WPC test specimens.

Wood Flour Preparation and Analysis

Highly deteriorated logs were chipped using a Sumner Iron Works Chipper, and then hammer milled to a 60-mesh particle size using a Bliss Industries Hammermill with a screen size of 1.1684 mm (0.046 in). Wood flour was characterized for particle size distribution using a Ro-Tap sieve analyzer and then analyzed for thermal decomposition using thermal gravimetric analysis (TGA). Thermal analysis of wood flour was conducted using a Simultaneous Thermal Analyzer (STA) G25-3 from Rheometric Scientific. The pans used in the instrument were 2 mm aluminum crucibles and the sample size for each run was <10 mg. All wood flour was dried for 24 h at 103°C (217°F) and then subjected to a two-step heating program. The material was equilibrated at 30°C (86°F) for 5 min and then heated to 600°C (1112°F) at 10°C/min (18°F/min). All tests were performed in a nitrogen atmosphere with a constant flow rate of 90 ml/min. The weight loss with respect to time was monitored as the temperature was ramped. Thermal analysis results will establish the upper limits of processing temperatures during the extrusion process and will identify any changes in thermal degradation due to insect infestation and weathering over time.

Chemical analysis using wet chemistry process was performed on a subsample of wood flour to establish the constituent makeup of each deterioration level. Any differences found in chemical composition could be used as an explanatory variable in final WPC properties. Because of limited funds, only one sample per tree height and deterioration level was randomly selected (six specimens) and analyzed for chemical composition at Integrated Paper Services, Inc. (IPS), Wisconsin, USA. It is noted in past studies that chemical degradation, if not very severe, would not be evident in the constituent makeup (i.e., changes in polysaccharides and lignin percentages as detected through chemical analysis) [11]. Therefore, a preliminary study was also conducted where three samples from each deterioration level were analyzed for chemical composition using Fourier Transform Infrared (FTIR) spectroscopy and compared to the wet chemistry values. FTIR is more sensitive to functional groups and changes in wood chemistry that result from depolymerization of wood constituents during the wood decay process [12–14]. Spectra were collected using a ThermoNicolet Avatar 370 spectrometer from the Thermo Electron Corporation, using an attenuated total reflection (ATR) attachment and mode. The spectra were mathematically ATR corrected using the software program Omnic 7.0. The spectra for each specimen were taken as an average of 64 scans at a resolution of 4 cm^{-1}.

Wood Plastic Composite Manufacture and Testing

The wood flour was dried to less than 2% moisture content and extruded using the ExtrusionTek Milacron Twin Screw Extruder (TC-86 Extruder) at a rate of 32 rpm or ~454 kg/h (1000 lb/h) of extruded material. All the wood flour processed through the hammermill with a screen size of 1.1684 mm (0.046 in) was used in extruding WPC specimens. Average temperatures in the extruder barrel and die were maintained at 171°C (340°F) and 177°C (351°F). The WPC formulation consisted of 58% wood flour, 32% high density polyethylene (HDPE) (Petrothene® LB010000; 0.5 g/10 min Melt Index; 0.953 g/cc density), 3% Zinc Serrate (lubricant), 5% Talc, and 2% Zinc Borate (biocide). Four batches of WPC material were extruded using flour from the highly deteriorated trees; one from each of the three tree height levels (butt, middle, and top) and a fourth batch containing equal components of wood flour from all three tree height levels. A control formulation consisting of 60-mesh pine flour, a primary wood species used in commercial WPC manufacturing, was also extruded.

Six WPC specimens were prepared from each formulation and tested as per ASTM D 7031-04 [15] to determine modulus of elasticity (MOE), modulus of rupture (MOR), strain at break point, and density. Three specimens 10 cm × 10 cm × 0.64 cm (4 in × 4 in × 0.25 in) from each formulation were evaluated for water absorption (WA) and thickness swell (TS) properties as per ASTM D 7031-04 and ASTM D 1037-99 [16] guidelines at intervals of 24, 48, 168, 336, and 672 h. For the remaining three specimens, preparation for WA and TS was modified from that described in ASTM standards to test under more severe conditions. WPC specimens that were 102 mm × 102 mm (4 in × 4 in) in cross-section and 6.35 mm

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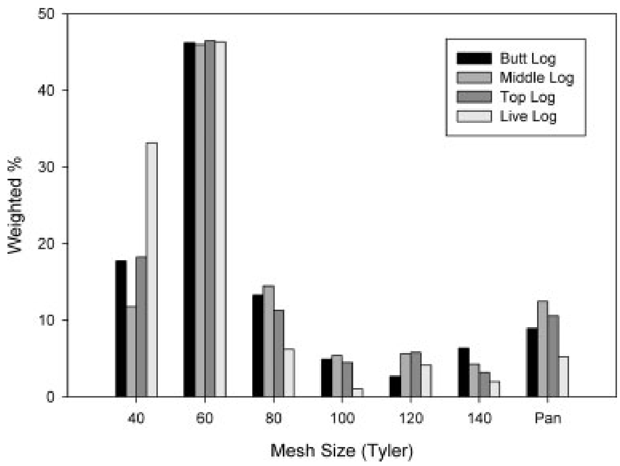

FIG. 1. Particle size distribution of spruce wood flour by log position using 1.1684 mm (0.046") screen.

(0.25 in) thick were subjected to WA and TS tests instead of using the full cross section of the as-extruded WPC material. This modified specimen size results in a more severe test than that recommended by ASTM as it could potentially expose more wood fiber to moisture due to disruption of encapsulation during the planing (surfacing) process.

RESULTS AND DISCUSSION

Wood Flour Particle Size Analysis

Viscosity of the formulation during WPC production and interaction between wood flour and the thermoplastic are partly determined by the particle size of the wood flour used. As mechanical interlocking between wood fiber and thermoplastic is the primary method of interaction between the two WPC constituents, particle size also relates to the final structural qualities. Wood particles are used as inexpensive fillers in WPCs and are known to contribute to WPC stiffness. Stark and Rowlands [17] note that the aspect ratio of wood particles increases as mesh size used in wood flour preparation increases. WPC stiffness improves significantly as aspect ratio of wood particles increases. Besides WPC stiffness, wood flour mesh size also affects the impact energy with larger particles leading to higher stress concentrations. Since wood flour was generated starting with highly deteriorated spruce in this study, it was of interest to understand the array of particle sizes produced when a 60-mesh screen was used during hammer milling. It is hypothesized that compared to wood flour from live spruce wood, highly deteriorated spruce wood may lead to different distribution in particle size as breakage of fiber could be influenced by deterioration of fibers. Figure 1 shows the particle size distribution by log position for highly deteriorated wood and of wood flour generated from live spruce trees. Close observation indicates a greater percent of finer particles (∼40%) in case of wood flour from highly deteriorated spruce than that of wood flour from live spruce (∼25%). All the wood flour that passed through 40- and 60-mesh is considered fine in this case. Wood degradation due to weathering and decay fungi and moisture loss are probable reasons for this generation of higher percentage of finer particles.

Thermogravimetric Analysis

Thermal decomposition tests were administered on wood flour from the three log positions (bottom, middle, top) using thermogravimetric analysis (TGA) to determine an optimum processing temperature in the production of the WPC. In all three log locations, the temperature at which thermal decomposition began to occur was around 225°C (437°F) and maximum thermal decomposition occurred at 361°C (682°F) similar to commercially sold pine wood flour (Fig. 2). This was well above the processing temperatures used in the extruder during production, which was around 171°C (340°F) in both the barrel and screw chambers. No trends in thermal decomposition behavior were detected due to log location.

Chemical Analysis

Chemical analysis was performed to determine if decay fungi had altered the proportions of cellulose, hemicellulose,

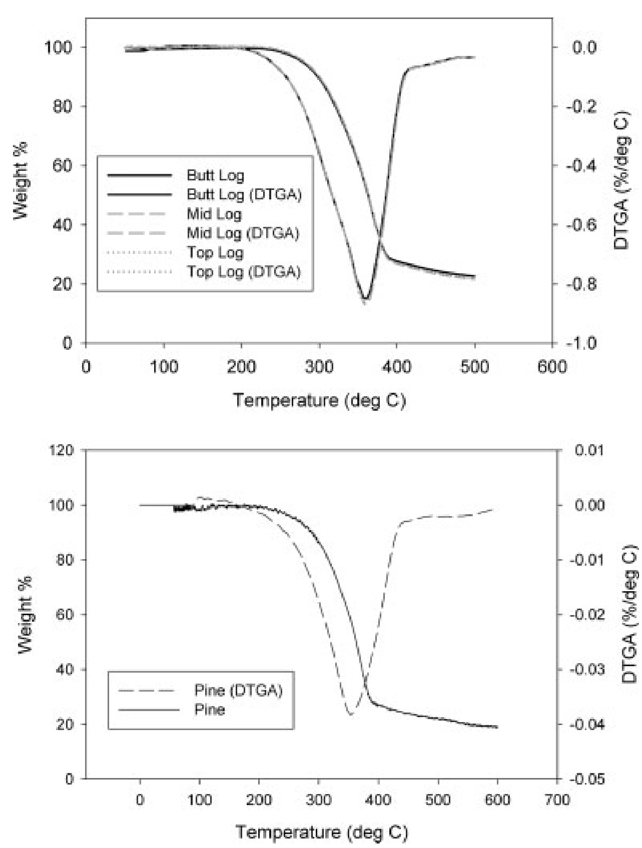

FIG. 2. Thermal degradation analysis of pine and highly deteriorated spruce wood flour.

TABLE 1. Carbohydrate, lignin, and extractive content vs. tree position for highly deteriorated and live spruce from Alaska's Kenai Peninsula.

Chemical constituent	Tree Condition					
	Highly deteriorated			Live		
	Log Position					
	Butt	Middle	Top	Butt	Middle	Top
Hemicellulose (%)	21.2	20.6	21.8	18.0	18.5	22.0
Cellulose (%)	42.9	41.5	43.0	37.0	37.6	43.7
Lignin (%)	28.5	28.5	28.7	28.1	27.3	27.0
Extractive (%)	1.46	2.37	2.07	2.07	2.93	2.13

and lignin in the beetle-killed wood and whether these proportions varied with height position in the tree. Table 1 presents the percentage of chemical constituents in the highly deteriorated wood and the analysis of wood from live trees. Only minor changes in proportions of chemical constituents were observed. Brown-rot fungi are known to attack softwoods where they mainly decompose the polysaccharides and cause only a small loss in the lignin [11]. Often, in the initial stages of decay, polysaccharides are depolymerized to low-molecular-weight and soluble molecules; thus, chemical analysis would not necessarily indicate a change in the chemical composition. Previous work on Kraft pulp yield, which can be sensitive to changes in chemical composition of carbohydrates in particular, showed no significant difference when comparing yields from live trees and beetle-killed trees that had been dead for 50 years [18].

FTIR spectroscopy was used on wood flour from both highly deteriorated and live logs to distinguish if any chemical changes occurred as a result of deterioration. The FTIR spectra for live and highly deteriorated beetle-killed spruce are shown in Fig. 3. Within the fingerprint region (1800 cm^{-1} to 600 cm^{-1}), several peaks that indicate changes in wood constituents resulting from the decay process are identified [13, 14, 19–22]. Further examination of the FTIR spectra (Fig. 4a, b, and c) indicate that the band peaks at 793 cm^{-1}, 898 cm^{-1}, 1318 cm^{-1}, 1368 cm^{-1}, 1422 cm^{-1}, and 1733 cm^{-1}, assigned to the carbohydrates such as cellulose ring vibrations, became weaker with deterioration of beetle-killed spruce wood. The disappearance of these bands is evident of qualitative changes in lignin and carbohydrate components in highly deteriorated wood. In the functional group region (4000 cm^{-1} to 1800 cm^{-1}) there are two prominent peaks shown, first being a strong hydrogen bonded O—H stretching absorption at 3350 cm^{-1} (Fig. 3) and second at 2895 cm^{-1} (Fig. 4d) indicating C—H stretching absorption. Bands that exhibit visible variations between live and highly deteriorated wood are 3350 cm^{-1} and 2895 cm^{-1}. Whereas white rot fungi degrade cellulose, hemicellulose, and lignin components of wood, brown rot fungi are preferential degraders of the carbohydrate components of wood and are responsible for extensive depolymerization of cellulose early in the decay process. Initial analysis using FTIR spectroscopy confirms qualitative changes in carbohydrate components in highly deteriorated beetle-killed spruce wood.

Mechanical and Physical Properties

The mechanical properties of the extruded WPC test samples indicated that when compared to the pine controls, the MOE, MOR, strain at failure, and density of WPC from beetle-killed material were all close in value (Table 2). Very little variation in density was observed among all the specimens irrespective of the type of wood flour used (coefficient of variation was less than 0.5%). Comparison of means tests at α-level of 0.05 indicated no significant difference among the means of three tree height levels for density, MOE, and MOR, and strain at failure. However, MOE of WPC produced using highly deteriorated spruce wood flour were significantly lower from corresponding properties of WPC produced using pine wood flour irrespective of tree height level. Although these differences are statistically significant, they are not practically significant and could be controlled through formulation design, a flexible manufacturing feature of WPC technology. WPC specimens produced using pine flour yielded higher MOE values than those produced from highly deteriorated spruce flour, but lower MOR values.

Scanning electron microscope (SEM) micrographs of a highly deteriorated spruce fiber bundle in a matrix of HDPE are shown in Fig. 5a and b. A clear separation between wood fiber and thermoplastic matrix is evident showing poor interaction between wood and plastic. These figures also show collapsed wood fibers with very little penetration of HDPE into cell lumens. Figure 5c illustrates typical penetration of thermoplastic into cell lumens of pine flour. It is hypothesized that cell wall deterioration as a result of brown rot attack after insect infestation caused wood fibers of highly deteriorated spruce to collapse more readily from processing pressures

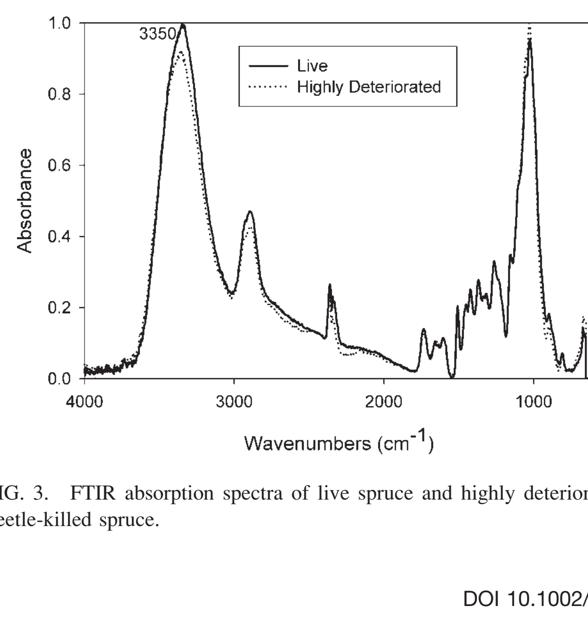

FIG. 3. FTIR absorption spectra of live spruce and highly deteriorated beetle-killed spruce.

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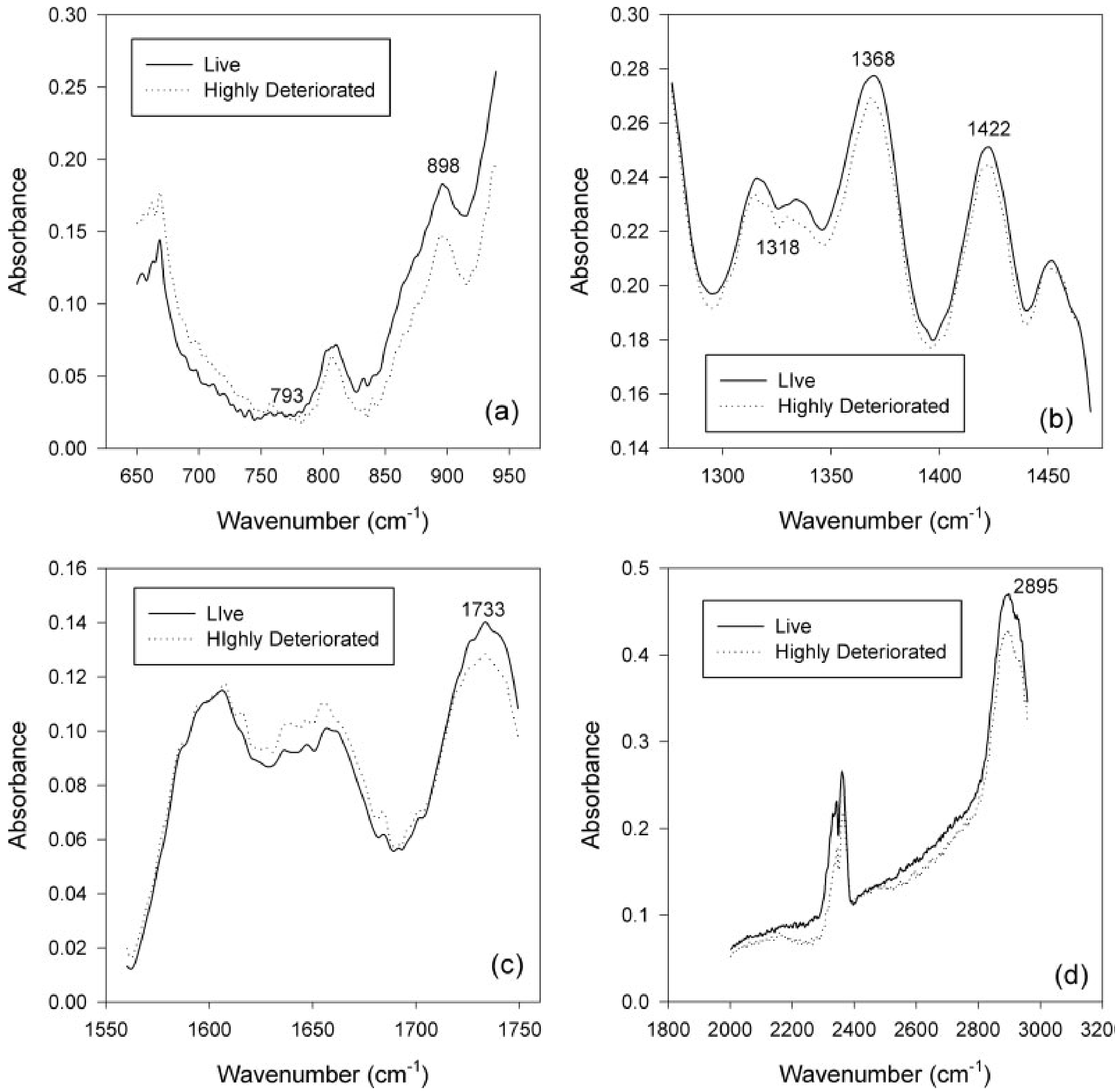

FIG. 4. Detail comparison of live and highly deteriorated beetle-killed spruce FTIR absorption spectra within different sub-regions of the spectra presented in Fig. 3.

TABLE 2. Mechanical properties vs. tree position for wood plastic composites manufactured from highly deteriorated spruce from Alaska's Kenai Peninsula and pine.

Position within tree	Avg. density, kg/m^3 (pcf)	COV[a] (%)	Avg. MOE[b], GPa (psi)	COV, (%)	Avg. MOR[c], MPa (psi)	COV, (%)	Strain at failure, (%)
Butt Log	1153 (72)	0.3	4.11 (595,750)	4.0	23.1 (3,350)	1.7	0.99
Middle Log	1153 (72)	0.2	4.13 (599,250)	2.6	23.1 (3,350)	2.4	1.00
Top Log	1153 (72)	0.3	4.13 (598,650)	1.5	23.0 (3,340)	1.7	1.00
All combined	1169 (73)	0.4	4.01 (581,450)	0.9	22.8 (3,300)	0.8	1.01
Pine (control)	1153 (72)	0.2	4.29 (621,800)	0.6	21.9 (3,180)	1.3	1.03

[a] Coefficient of variation.
[b] MOE, modulus of elasticity.
[c] MOR, modulus of rupture.

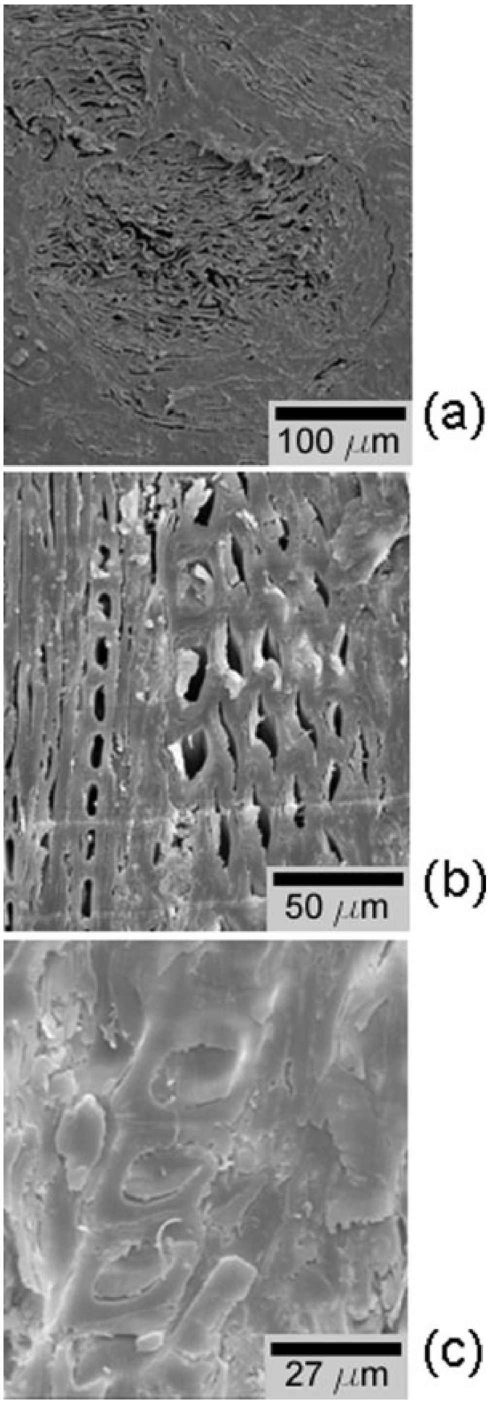

FIG. 5. Micrograph showing (a) cell collapse of deteriorated spruce fiber bundles, (b) poor penetration of HDPE matrix into fiber lumen of deteriorated spruce fiber, and (c) typical penetration of thermoplastic into cell lumens of pine fiber.

in the extrusion process. Even with poor interlocking between wood and thermoplastic matrix, WPC specimens produced with highly deteriorated wood exhibited reasonably good mechanical properties when compared to WPCs extruded with pine flour. Greatly increased surface area as a result of higher proportions of fines generated with deteriorated wood could have influenced nucleation of the matrix polymer.

Because of the hydrophilic nature of wood and intended use of WPC for exterior applications, water absorption testing can provide an indication of how the material will perform in such a setting. Figure 6 shows water absorption following 4 weeks of water submersion, where readings were taken at intervals (24 h, 48 h, 1 week, 2 weeks, 4 weeks) to help establish trends for WPC specimens extruded with highly deteriorated spruce and control pine flour. The WPC composed of strictly wood flour from the butt log of the tree exhibited the highest water absorption, with a 17.55% increase in moisture content after 4 weeks. A similar water uptake value (16.96%) was found for WPC specimens extruded with wood flour that is a combination of equal proportions of the three log locations (butt, middle, and top). The WPC manufactured using wood flour from the middle section and the top section of the tree exhibited increases of water absorption of 15.71% and 15.93%, respectively. Generally the greatest increase in water absorption occurred between 2 and 4 weeks. Water absorption of WPC specimens with control pine flour was greater than that of deteriorated spruce wood flour over time, but the two begin to converge at the end of four weeks indicating a slower water absorption rate for highly deteriorated spruce specimens. It is interesting to note that WPC specimens with pine flour began to plateau at around 18% which would be equivalent to 31% if normalized to a specimen composed of entirely pine wood flour (calculated with the assumption that only wood flour in WPC contributes to moisture uptake and thickness swell and not HDPE matrix). This value seems reasonable considering that fiber saturation point for all woods is ~30%. Results indicate that wood flour from deteriorated spruce wood tends to absorb water at a slower rate than pine wood flour. Collapse of spruce fiber bundles and greatly

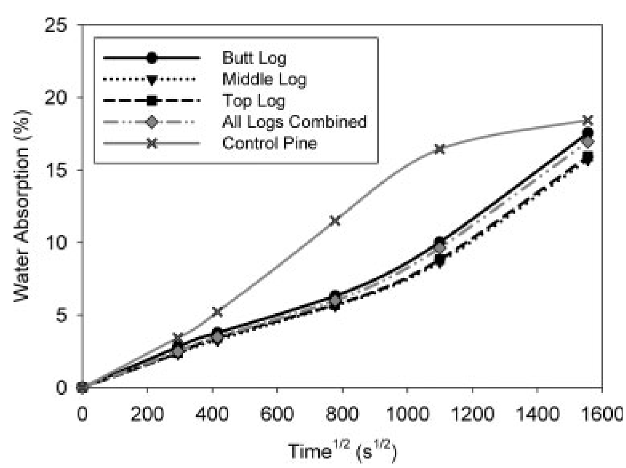

FIG. 6. Water absorption, by log position, of wood plastic-composites manufactured from highly deteriorated beetle-killed spruce and pine as a control.

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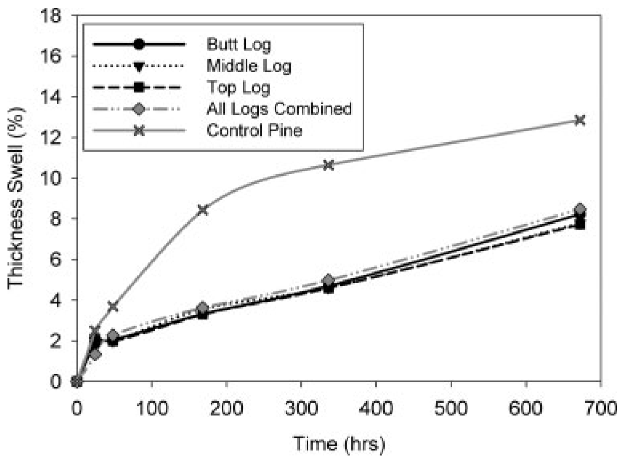

FIG. 7. Thickness swell, by log position, of wood plastic-composites manufactured from highly deteriorated beetle-killed spruce and pine as a control.

increased surface area, as a result of higher proportions of fines generated with deteriorated spruce wood, may have contributed to decreased moisture sorption because of better encapsulation by thermoplastic matrix. If immersed for a longer time period, it is possible that moisture absorption of WPC specimens with deteriorated spruce wood flour would also begin to plateau around fiber saturation point of spruce wood.

A similar trend as in water absorption is also evident with thickness swell data. The WPC made with equal parts wood flour from the three log positions (butt, middle, top) showed the greatest increase in thickness swell, with an 8.48% increase from its initial thickness (Fig. 7). The WPCs composed of wood flour from the butt, middle, and top showed a gradual decrease in thickness swell from the butt to the top, each showing an increase of 8.23%, 7.78%, and 7.72%, respectively, after 4 weeks of water immersion. The overall difference between the butt and the top was 0.51%. Once again, the cell wall of highly deteriorated spruce fibers indicated a significantly lower uptake in moisture compared to control pine flour. Collapse of fiber bundles and better thermoplastic encapsulation of finer deteriorated spruce wood flour could have contributed to this behavior. Past research [23] has shown that moisture absorption of wood flour-filled polypropylene composite decreased with decreasing particle size. Slower rates of water absorption and thickness swell results for shorter exposure durations of WPC specimens with deteriorated spruce wood flour point to a possibility of improving WPC moisture resistance by partial inclusion of highly deteriorated spruce flour in the formulation.

CONCLUSIONS

Results show that manufacture of WPC from highly deteriorated spruce trees is a viable opportunity for utilization of beetle-killed spruce trees. Chemical composition analysis did not show any significant differences in constituent make-up between highly deteriorated and live spruce wood; however, signs of break down of chemical constituents due to decay fungi attack was evident in the FTIR spectra. Wood flour produced from deteriorated spruce wood contained greater proportions of fines than control pine wood flour. Nevertheless, mechanical properties were comparable to WPC properties manufactured from pine wood flour, which served as the control. SEM micrographs show collapse in fiber cells and evidence of poor penetration of HDPE into cell lumens of spruce wood flour attributable to degradation of fiber quality in deteriorated trees. Moisture uptake and thickness swell rates of WPC specimens produced with highly deteriorated spruce flour were significantly lower than those with control pine flour possibly because of better barrier provided by thermoplastic encapsulation of finer particles and collapse of fiber bundles during extrusion process. Further research on interaction between highly deteriorated spruce fiber and thermoplastic matrix is necessary to understand this physical behavior.

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