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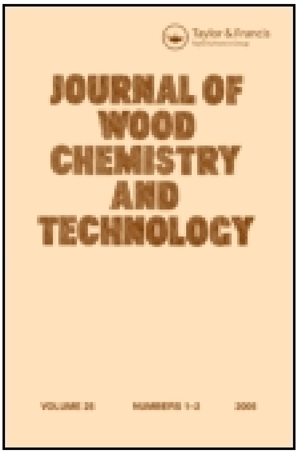

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Effect of Fiber Origin, Proportion, and Chemical Composition on the Mechanical and Physical Properties of Wood-Plastic Composites

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Abstract: *This study assessed the potential of wood residues as fiber sources for wood-plastic composites (WPC) and examined the impact for intrinsic fiber properties on strength development. Sawmill sawdust, underused wood species, bark, composite panel, and pulp and paper sludge residues were sampled. Fibers were characterized for cellulose content, ash content, and fiber aspect ratio. WPC samples were formed by twin-screw extrusion compounding, followed by injection molding at three fiber proportions. WPC mechanical properties, water uptake, and water swelling increased with increasing fiber proportion, whereas tenacity decreased. WPC made with residues had lower mechanical and physical properties than those made with clean wood, with some exceptions. Kraft sludge produced one of the best WPC formulations in terms of thickness swell, water swelling, tensile strength, and impact energy. Deinking sludge produced the toughest and the most dimensionally stable WPC. Panel industry residues formed roughly similar WPC to those made with clean wood. Bark led to poorest WPC in terms of mechanical properties. High correlation coefficients were found between cellulose content, wood content, and all WPC properties except impact energy. However, the correlations between aspect ratio and the WPC were insignificant.*

Keywords Wood-plastic composite, recycling, wood residues, mechanical properties, physical properties, chemical composition

Introduction

Since the emergence of wood-plastic composites (WPC), interest in them has grown, and market forecasts predict a substantial increase in demand in the next five years, mainly in Asia.[1] These new markets and new materials have created promising opportunities for the forest product industry.

Although global demand for wood, wood products, and environmentally responsible products is increasing, forest resources are already used almost at their full potential. In addition to the problem of fiber availability, new environmental concerns and waste disposal laws are pushing the wood processing industry to find new uses for residues. The

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province of Quebec in eastern Canada will soon prohibit the disposal of wood and paper waste in landfills and by incineration.[2] Recycling wood processing residues for WPC would optimize the use of harvested trees, increase the entire value chain, and reduce the environmental footprint.

Some studies have addressed this issue. Lightsey et al.[3] characterized polyethylene and polystyrene filled with residue from screened chips from Kraft pulp mills. The strength and tensile modulus of composites containing wood residue were slightly greater than for commercial wood flour-filled polymers. These results were explained by the more fibrous nature of the residue. However, its high ash content reduced the effective reinforcement. Boeglin et al.[4] found that WPC made with mixed wood waste and recycled plastic had higher dimensional stability but moderate mechanical properties compared to traditional wood-based composites such as particleboard. Georgopoulos et al.[5] used fibrous residues from eucalyptus wood, ground corncob, and brewery's spent grain as fillers in thermoplastic polymers. They showed that loading the polymer with natural fibers decreased the mechanical strength and increased the modulus, with high brittleness. Panthapulakkal and Sain[6] compared wood flour and milled newsprint as filler in polypropylene and found that newsprint led to slightly superior tensile strength and impact strength and similar modulus of elasticity. Slama[7] assessed sawed residues from a medium-density fiberboard (MDF) plant. The residue was fibrous, contained cured urea-formaldehyde resin, and produced more hydrophobic and dimensionally stable WPC than a control made with virgin fibers from the same plant. Mechanical properties of the composites made with the residue were similar to the control panel. It was suggested that the resin had beneficial effects on the composite properties, but that the shorter fiber length of the residue adversely affected mechanical properties. Several studies[8-11] have assessed the potential of bark as a fiber source for WPC. The results largely concur that composites made with bark particles have lower water absorption and lower mechanical properties than composites made with wood particles. Bark produced WPC with higher elongation to break and sometimes higher toughness than those made with wood. These three studies suggest that the low mechanical performance of bark WPC is due to low reactivity with MAPE-coupling agents and low intrinsic resistance compared to wood. Other plausible explanations for these differences include variation in chemical composition, fiber surface properties, crystallinity, thermal degradation, and fiber specific gravity. Bark is generally described as a filler, not as a reinforcement. There is also substantial potential for recycling residues from natural non-wood fibers such as agricultural residues.[6]

The objective of the present study was to compare the potential of some conventional wood residues for WPC production.

Experimental

Material Collection and Preparation

Samples of 10 common wood residues were obtained from industrial wood mills in the province of Quebec in eastern Canada. In this paper, the term "fibers" is used for all materials used as filler or reinforcement in the polymer matrix (e.g., fibers, particles, wood, bark). The fibers are classified into four residue groups, as presented in Table 1. Sludge is the residue obtained after thickening of the pulp and paper mill waste waters.

Prior to composite processing, fibers were air-dried to a dry basis moisture content of about 10%, milled with a Wiley Laboratory Mill Model 4 mounted with a 2-mm opening

Table 1
Description of the fibers used in wood-plastic composite (WPC) formulations

Name	Description
CLEAN WOOD GROUP: Sawmill sawdust and underused wood species	
Aspen wood	Trembling aspen (*pupulus tremuloides*) from eastern Canada. Underused species in the north of the province of Quebec, Canada.
Birch wood	White birch (*betula papyrifera*) from eastern Canada. Underused species in the province of Quebec, Canada.
Spruce wood	Sawdust from an eastern Canadian softwood sawmill using about 90% black spruce wood, with some jack pine and balsam fir.
BARK GROUP	
Aspen bark	Bark from aspen trees (see above).
Spruce bark	Bark from spruce trees (see above).
COMPOSITE PANEL RESIDUES GROUP: Sawdust from the wood composite panel industry	
MDI panel	Panel sawing sawdust from an oriented strand board (OSB) mill using aspen wood (see above) and methylene diphenyl diisocyanate (MDI) adhesive.
PF panel	Panel sawing sawdust from a laminated veneer lumber (LVL) mill using about 15% aspen wood (see above), 85% birch wood (see above), and phenol formaldehyde (PF) adhesive.
SLUDGE GROUP: Residues from the pulp and paper industry	
Deinking sludge	Deinking sludge produced from recycling of mixed-grade office paper.
TMP sludge	Sludge from a pulp and paper mill using softwood chips and thermo-mechanical pulping process (TMP).
Kraft sludge	Sludge from a pulp mill using softwood chips and Kraft pulping process.

sieve, and sieved with a Ro-tap Laboratory Sieve Shaker. The 150-710 μm opening fraction (100 to 25 US mesh) was kept for WPC processing. Sludge was not milled because it would shorten fibers, nor was it sieved. Finally, fibers were oven-dried at 80°C to approximately 3% dry basis moisture content.

High-density polyethylene (HDPE) (DOW DMDA-8907 NT7, Dow Chemical) was used as polyolefin matrix. Maleated polyethylene (MAPE) (Fusabond E226, Dupon) was used as compatibility agent at 3% of the total composite dry mass.

Fiber Characterization

Cellulose and ash contents were measured for all residues. Cellulose content reflects the fiber reinforcing potential, and ash content reflects the degree of contamination. The dried material was placed in an airtight container to homogenize the moisture content. Cellulose content was determined by Kürschner and Hoffer's nitric acid method.[12] Ash content was determined by combustion in a muffle furnace at 600°C according to ASTM D 1102 standard. Fiber aspect ratio was measured using an OPTEST fiber quality analyzer. Each average aspect ratio was obtained from three samples of 5,000 fibers each. Based on image analysis, the FQA allows rapid determination of fiber length, width, and several other morphological properties. Thermogravimetric analysis was applied to determine the thermal sensibility of the different raw materials used. The information obtained was then used to select processing temperatures below the thermal degradation of the raw materials. Measurements were performed by heating 20 mg of fibers from 30°C to 650°C at 10°C/min heating rate in a N_2 atmosphere using a TGA Q50 analyzer (TA Instruments, New Castle, Delaware, USA).

WPC Processing and Characterization

All composites were prepared in two stages: compounding for pelletizing followed by injection molding. A counter-rotating, intermeshing, conical twin-screw extruder (Thermo Scientific HAAKE PolyLab OS Rheodrive 7 with Rheomex OS extruding module) was used to compound fibers, HDPE, and MAPE. The screws were 30 mm in diameter at the large end and 340 mm long, and a 3-mm-diameter die was used. Screw speed was 30 rpm and barrel and die temperature was 155°C. The extrudate was cooled in a water bath and ground into 3-mm-long pellets.

Samples were molded for tensile, bending, and impact specimens with an Arburg 370 A (600 kN) injection molding machine. Injection molding parameters were 30°C mold temperature, 160 MPa injection pressure, 1.6 s injection time, 70 MPa holding pressure, 9 s holding time, 180°C barrel and nozzle temperature, and 17 s cooling time. All specimens were 3.18 mm thick. Bending and impact type specimens were 12.7 mm wide. Bending and impact specimens were 127 mm and 63.5 mm long, respectively.

WPC specimens were characterized according to ASTM D 1037 standard for apparent density, water uptake, and thickness swell (TS) of water-soaked samples. Measurements were repeated three times on bending-type specimens. Three-point bending properties was measured according to ASTM D 790 standard with a span-to-depth ratio of 16:1 and at a speed of 1.4 mm/min. Tensile properties were measured according to ASTM D 638 standard using specimen Type I and at a speed of 5 mm/min. Unnotched impact resistance was measured according to ASTM D 4812. Bending, tensile, and impact tests were repeated 10 times.

Results and Discussion

Fiber Characteristics

Table 2 presents the cellulose and ash contents of the different fibers used in the WPC formulations. Cellulose is the structural component of wood and thus indicates the reinforcing potential for WPC. The cellulose contents found for the three wood species in the present study are in good agreement with previously reported values. Aspen wood has higher cellulose content than white birch wood, and hardwoods have higher cellulose content than softwoods.[13] Bark generally has lower cellulose content than wood. Aspen wood was sampled from composite panel mills. The cellulose content of the sawdust from these mills was therefore determined as the cellulose content of aspen wood minus the relative proportion of non-wood ingredients used in composite panel formulations. The sampled sludge materials showed high cellulose content despite their very high degree of contamination

Table 2
Cellulose and ash contents and aspect ratio of the fibers used in WPC formulations

Name	Cellulose (%)	Ash (%)	Aspect ratio
Aspen wood	50.7	0.7	5.8
Birch wood	47.3	0.3	2.5
Spruce wood[a]	43.0	0.3	5.8
Aspen bark	32.0	5.3	4.1
Spruce bark	33.5	1.6	4.6
MDI panel	48.2[b]	1.1	3.4
PF panel	47.7[b]	1.7	1.9
Deinking sludge	43.3	51.1	18.2
TMP sludge	29.5	19.4	47.4
Kraft sludge	32.8	37.8	83.2

[a]from [13]; [b]estimated with the non-wood content in panels.

(up to 50% ash). This result suggests that sludge has good reinforcing potential for WPC despite its high degree of contamination.

Fiber ash content, a measurement of the degree of fiber contamination, is also presented in Table 2. Fibers in the present study fell into three contamination degree categories. The first category, clean wood (wood group, Table 1), is considered uncontaminated (<1% ash content). The ash contents in Table 2 are typical for wood, and are due to the minerals present in wood cell walls and extractives.[13] The second category, low contaminated residues (bark and composite panel groups, Table 1), showed ash contents in the range 1% to 5%. The higher ash content in bark than in wood is due to bark's higher inorganic content. Bark is also contaminated with sand or dirt resulting from contact with the ground (e.g., after logging). Inorganics in the composite panel group residues are mainly due to non-wood ingredients used in panel formulations. The cured resin present in these residues is more desirable than other types of contaminants, such as sand. The third category is highly contaminated wood residues with 20% to 50% ash content, including all the pulp and paper sludge samples. Deinking sludge has the highest ash content. A large proportion of this ash comes from ink and clays recovered in the deinking process. Although the Kraft mill produces pulp only (no ink or other additives), this sludge showed higher ash content than the TMP sludge from an integrated pulp and paper mill. This result may be explained by the fact that the chemical Kraft pulping and bleaching processes generate more residual components.

Fiber aspect ratios are presented in Table 2. Despite the fact that all particle types were prepared with the same procedure, their aspect ratios were slightly different. Within the clean wood group, the dense birch wood produces particles with lower aspect ratio than the soft spruce and aspen woods. Bark particles showed slightly lower aspect ratio compared to wood. Composite panel residues have the lowest aspect ratios. This is explained by the previous processing steps. The three sludge samples differ from the seven other fibers used in this study in that the proportion of wood in sludge is in the form of individual fiber cells (or tracheids) instead of milled stocky particles. Slender fibers with high aspect ratio (length-to-diameter ratio) have better reinforcing potential for WPC than stocky particles

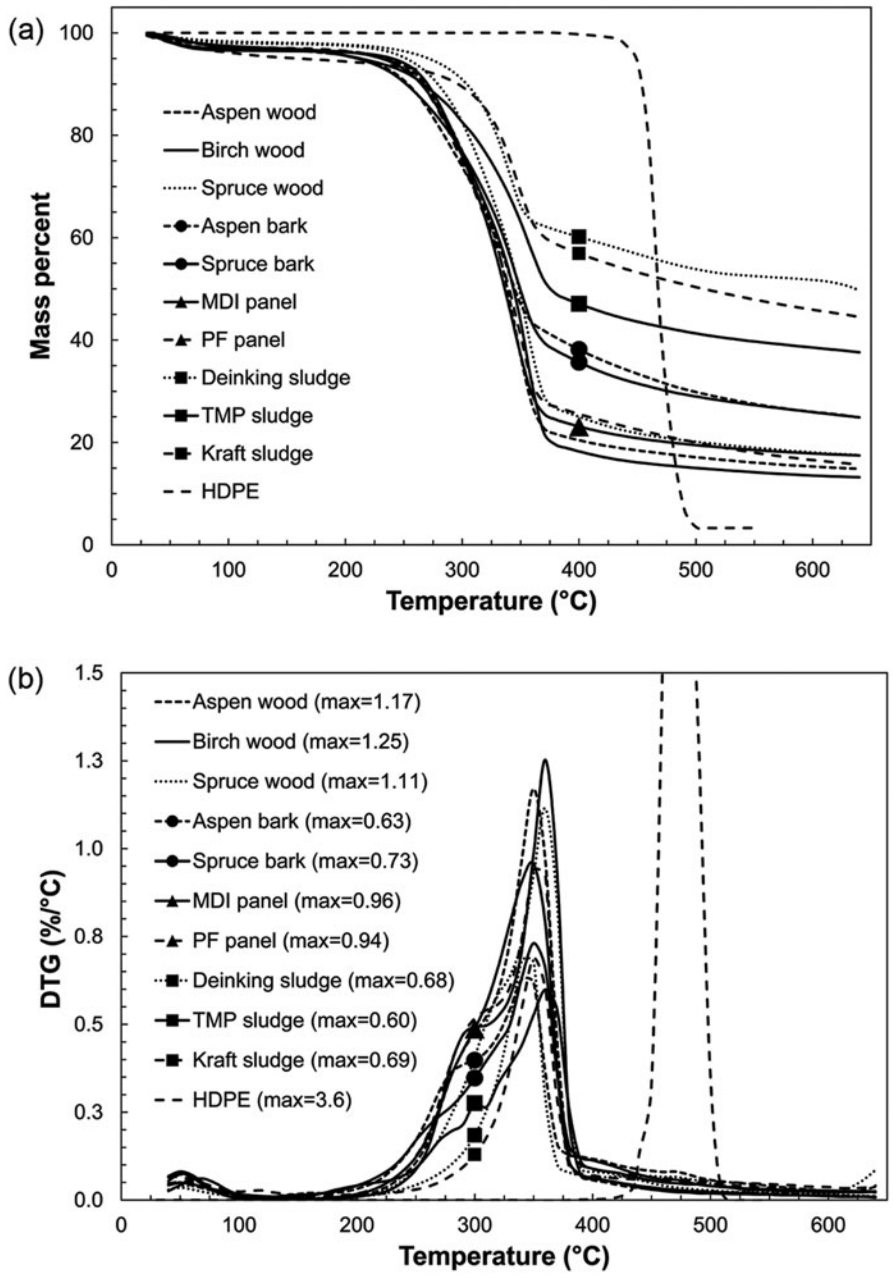

Figure 1. Mass losses (a) and temperature derivatives of the mass (DTG) (b) for the different fibers and HDPE in a nitrogen environment at 10°C/min heating rate.

with low aspect ratio.[14,15] Therefore sludge has greater reinforcing potential than the other materials in terms of fiber morphology.

Figure 1 presents the thermogravimetric curves of the different fibers and HDPE. Fibers are more thermally sensitive than HDPE, and begin to lose significant mass at around 250°C. In the present study, because samples were molded at 180°C, little thermal degradation occurred during processing. The small loss step at 100°C corresponds to moisture vaporization (about 3%). For some fibers, slightly more mass is lost before 250°C due to volatile compounds such as wood resin and extractives. All fiber curves are characterized by a dominant large pyrolysis step from 250°C to 400°C, where the wood polymers decompose.[13] This step begins about 50°C later (at around 300°C) for sludge because it has already undergone thermal or chemical changes in the pulping processes. As shown on the differential thermogravimetric (DTG) peaks (Figure 1b), the thermal decomposition of HDPE can be described by a single-step reaction. The DTG peaks of fibers are broader because wood is composed of different polymers with different degradation temperatures. Volatile compounds such as wood resin and extractives decompose from about 175 to 350°C, with a peak at about 250°C.[16] Hemicellulose polymers decompose from about 200 to 380°C, with a peak at about 295°C. [16] Cellulose decomposes from about 280 to 430°C, with a peak at about 360°C. [16] The derivative peaks shape and position vary with fiber type, showing that their thermal sensitivity is different. As reported,[16] the bark decomposition peak is slightly broader than the wood peak. All residues have a slightly broader decomposition peak than wood because of their more diversified chemical composition. The sludge peaks are remarkably different. These materials have already undergone thermal or chemical changes in the pulping processes. The TMP sludge decomposition peak is closer to those described for wood because these fibers have a similar chemical composition. However, the Kraft and deinking sludge decomposition peaks do not have the hemicellulose peak at 290°C because these polymers of lower molecular weight were removed in the pulping processes.

The percent material remaining at 650°C is higher for sludge materials due to their high inorganic content (Table 2). Bark has a roughly similar thermal resistance to clean wood, but more material remains at 650°C because of the higher inorganic content. The curves for the composite panel group are very similar to those for wood because the composite panels were processed at around 160°C, resulting in little thermal degradation.

Effect of Fiber Proportion on WPC Physical Properties

All WPC physical properties were significantly affected by fiber proportion in the matrix (Table 3). Density, TS, and water uptake increase mainly linearly with increasing fiber proportion, but show a somewhat significant higher order effect. HDPE density is 946 kg/m^3 and WPC density is 1100 kg/m^3 on average at 40% fiber proportion (Table 4). This result indicates that fiber lumens are filled with HDPE or collapsed, as the apparent basic density of solid wood varies from 350 to 550 kg/m^3 and the density of the fiber cell wall is around 1450 kg/m^3.[13]

In good agreement with the literature,[7] water mass uptake and TS increase with increasing fiber proportion (Figure 2 and Table 4). This result is due to the fact that wood material is the most hydrophilic of the composite components. Also, fiber networking increases as fiber proportion increases, and thus water diffusion increases. Figure 3 shows the variation in TS and water uptake with time. Water mass uptake increased up to the end of measurements at 84 days, whereas thickness swell reached a plateau at around 28 days. This behavior is similar to that of solid wood: dimensional changes reach a plateau while water mass absorption continues.

Effect of Fiber Proportion on WPC Mechanical Properties

WPC mechanical properties were significantly affected by fiber proportion (Table 3). As shown in Figure 2, the development of mechanical properties with increasing fiber proportion is mainly linear, but a significant second-order effect was found on all mechanical

Table 3
Analysis of variance (*F* values) in WPC properties

Source of variation	Density	Water uptake	Thickness swell	E[a]	Tensile strength	Tensile EB[b]	Impact energy	MOE[c]	MOR[d]
Fiber proportion	4 079**	356**	163**	3 312**	2 552**	1 688**	332**	8 013**	6 584**
Linear effect	8 151**	672**	301**	6 624**	5 066**	3 199**	619**	12 924**	11 306**
Nonlinear effect	5.9*	15**	8.8**	0.3^{NS}	37**	177**	46**	97**	14**
Fiber type	383**	33**	31**	670**	1 843**	264**	146**	1 319**	2 328**
Wood vs. All others	628**	19**	1.9^{NS}	3 327**	4 466**	145**	2.9^{NS}	812**	1 661**
Wood vs. Bark	21**	8.6**	18**	3 374**	11 421**	69**	101**	2 124**	3 251**
Wood vs. Panel residues	18**	10**	26**	389**	785**	1.6^{NS}	1.3^{NS}	7.8^{NS}	45**
Wood vs. Sludge	1 838**	80**	17**	2 836**	976**	277**	137**	707**	2 156**
Fiber type x Fiber proportion	20.9**	6.5**	4.1**	46**	124**	43**	10**	123**	122**

[a] tensile modulus of elasticity; [b] tensile elongation at break; [c] bending modulus of elasticity; [d] bending modulus of rupture; ** significant at 1% probability level; * significant at 5% probability level; NS not significant at 5% probability level.

Table 4

Mass uptake, thickness swell, and apparent density of WPC samples made with three proportions of the different fibers

Fiber proportion	Fiber type	Density (kg/m^3)	Water uptake (%)[1]	Thickness swell (%)[1]
20%	Aspen wood	1015^{cd}[2]	0.86^{abc}	0.25^{e}
	Birch wood	1012^{cde}	1.01^{a}	0.79^{abc}
	Spruce wood	1008^{d}	0.96^{a}	0.67^{bcd}
	Aspen bark	1012^{de}	0.60^{cd}	0.78^{abcd}
	Spruce bark	1010^{de}	0.60^{cd}	1.06^{a}
	MDI panel	1012^{de}	1.01^{a}	0.99^{a}
	PF panel	1011^{de}	0.87^{ab}	0.48^{cde}
	Deinking sludge	1055^{a}	0.69^{bcd}	0.29^{e}
	TMP sludge	1019^{c}	0.54^{d}	0.90^{ab}
	Kraft sludge	1038^{b}	0.46^{d}	0.46^{de}
30%	Aspen wood	1038^{g}	1.24^{bcd}	0.66^{cd}
	Birch wood	1045^{ef}	1.56^{ab}	1.51^{da}
	Spruce wood	1044^{f}	1.37^{bc}	0.93^{bc}
	Aspen bark	1050^{d}	1.10^{cd}	0.94^{bc}
	Spruce bark	1051^{d}	1.40^{bc}	1.18^{ab}
	MDI panel	1049^{de}	1.37^{bc}	1.44^{da}
	PF panel	1053^{cd}	1.82^{a}	1.17^{ab}
	Deinking sludge	1109^{a}	0.87^{de}	0.54^{d}
	TMP sludge	1057^{c}	1.10^{cd}	1.00^{bc}
	Kraft sludge	1084^{b}	0.66^{e}	0.54^{d}
40%	Aspen wood	1079^{g}	2.02^{d}	1.07^{de}
	Birch wood	1080^{fg}	2.31^{b}	2.17^{ab}
	Spruce wood	1085^{df}	2.07^{cd}	1.27^{d}
	Aspen bark	1088^{d}	1.92^{d}	1.63^{cd}
	Spruce bark	1089^{d}	2.28^{bc}	2.15^{ab}
	MDI panel	1084^{f}	2.10^{bcd}	1.72^{cd}
	PF panel	1090^{cd}	2.88^{a}	2.26^{a}
	Deinking sludge	1175^{a}	1.23^{e}	0.82^{e}
	TMP sludge	1094^{c}	2.67^{a}	1.92^{bc}
	Kraft sludge	1144^{b}	1.08^{e}	0.77^{e}
-	HDPE	946	—	—

[1] water uptake and thickness swell measured after 28 days of water immersion; [2] values followed with the same letter are not significantly different and values followed with different letters are significantly different at 0.05 probability level.

properties except for tensile modulus of elasticity (Table 3). As illustrated by the tensile stress-strain curves (Figure 4), the addition of fibers into HDPE and increasing fiber proportion yield material with higher stiffness, strength, and yield point, but lower ductility and deformation energy. Although this is a typical behavior for wood-filled polyolefin, a decrease in maximum strength with increasing fiber proportion has been observed in previous studies.[16] Compared to HDPE alone, the addition of 40% fibers into HDPE increases the

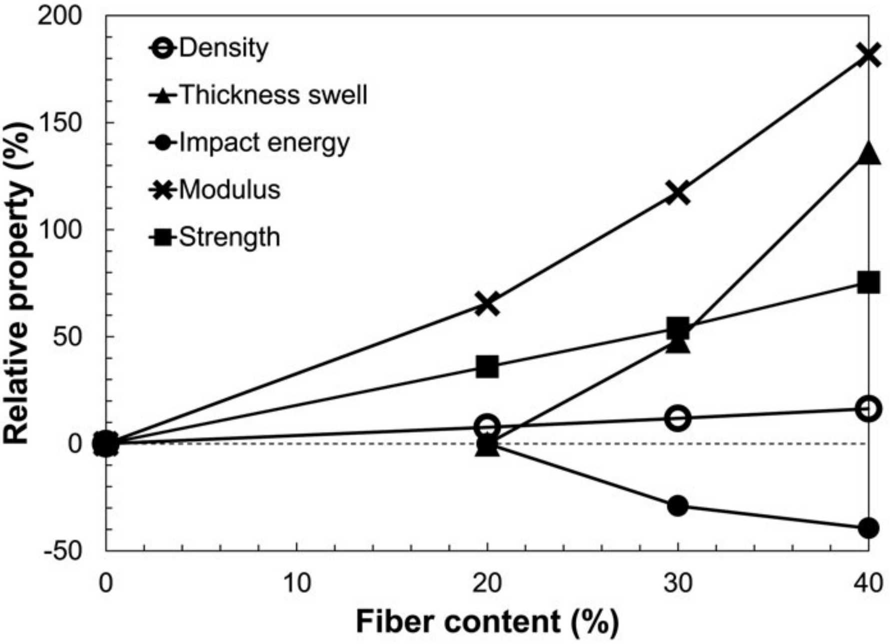

Figure 2. Variation in selected WPC properties with increasing fiber content.

modulus of elasticity by 182% and strength by 75% on average (for tensile and bending), and tensile deformation at break is 275 times lower (Table 5). These results indicate that WPC should be avoided for applications where toughness and ductility are crucial. Otherwise, the addition of fibers into the polymer improves mechanical performance. When comparing WPC formulations with 20% to 40% fiber proportion, the tensile modulus of elasticity increases by 68% on average (Figure 2), tensile strength increases by up to 28% on average, and toughness is reduced by 39% on average. The increase in modulus of elasticity was expected, because wood material is stiffer than HDPE, and this result is in good agreement with most previous works.[16] In the present study, the increased strength of HDPE observed for most formulations indicates strong fiber-matrix adhesion.

Effect of Fiber Type on WPC Physical Properties

The analysis of variance revealed significant variations in all WPC properties according to fiber type, but the differences are less significant than those for fiber proportions (Table 3). The graph in Figure 5 provides an overview of the WPC properties, showing the averaged relative properties for the three fiber proportions of the different residue groups. However, no direct conclusions can be drawn due to the significant interactions between fiber type and fiber proportion (Table 3). In other words, the effect of fiber type varies with fiber proportion. Specific differences are presented in Table 5.

WPC apparent density was significantly affected by fiber type (Table 3). Composites made with the clean wood group are the lightest, those made with the sludge group are the heaviest, and those made with the composite panel and bark groups are in between (Table 4). Composite density increases with increasing degree of fiber contamination, shown by the

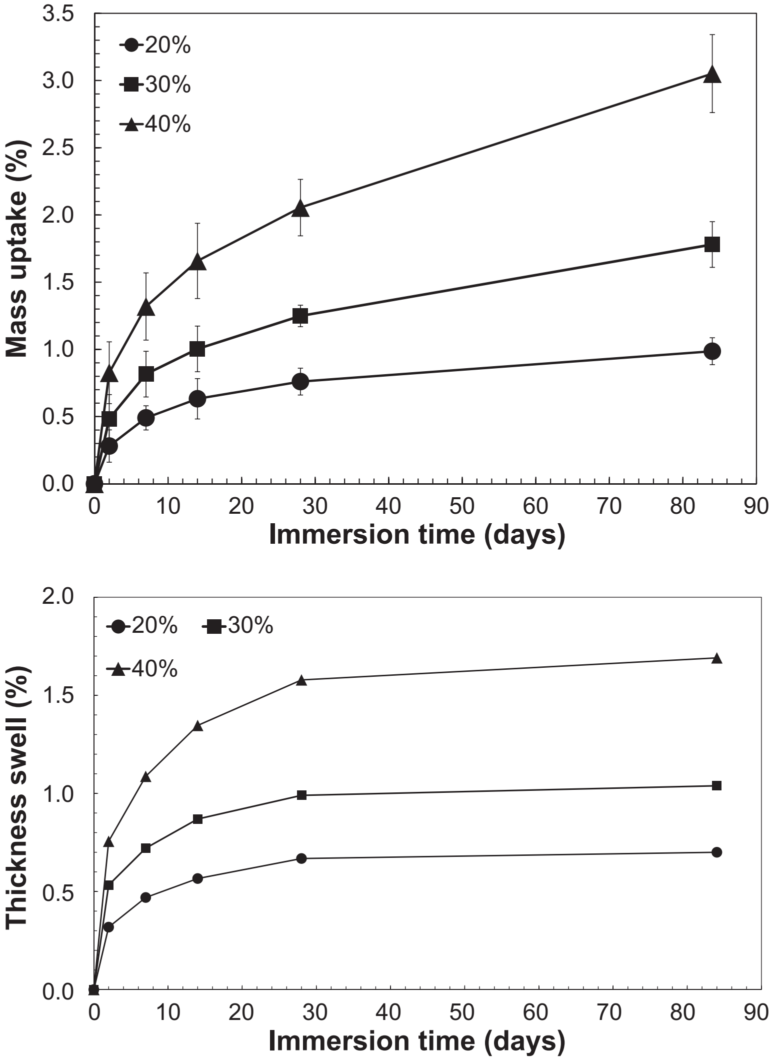

Figure 3. Mass uptake (a) and thickness swell (b) for the water-soaked WPC samples made from three proportions of the different fibers (averages for each proportion).

significant correlation between ash content and density (Table 6). This result is explained by the fact that inorganic materials such as metals are heavier and less bulky than wood. Clay in deinking sludge is another example of a dense inorganic material. Density also increases with increasing wood content and cellulose content (Table 6), because the wood fiber cell wall is denser than HDPE, as discussed above. Wood content was calculated using Equation (1). It should be noted that wood content is different from fiber proportion. Finally, fibers from the composite panel residues group were densified in previous processing steps and thus produced WPC with a slightly higher apparent density.

$$Wood\ content = \frac{Fiber\ proporation}{100} \times (100 - ash\ content) \tag{1}$$

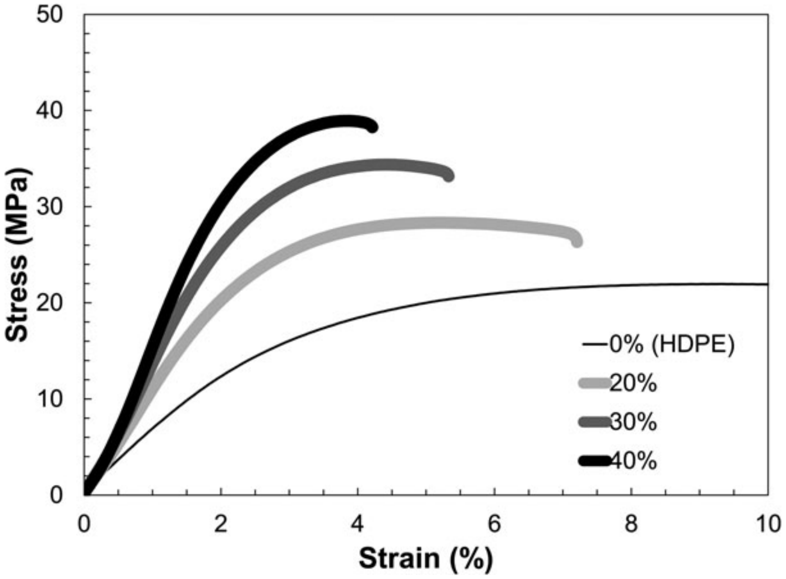

Figure 4. Tensile stress-strain curves for WPC samples with increasing fiber proportion (aspen wood given as example).

Fiber type also significantly affected WPC water absorption (Table 3). The WPC made with the sludge and bark groups are more hydrophobic on average than those made with clean wood (Figure 5 and Table 4), but with several exceptions. For example, the TMP

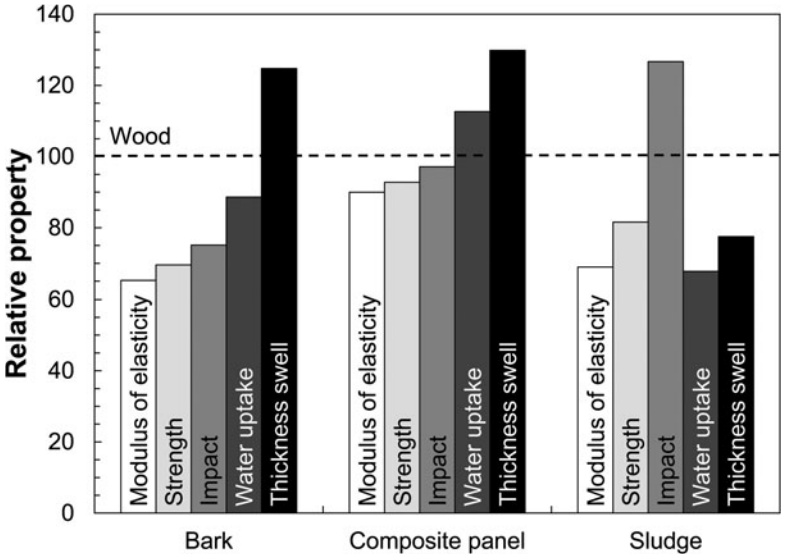

Figure 5. Relative values of selected WPC properties for the different fiber groups for the three fiber proportions (averages for each fiber proportion, modulus of elasticity and strength are averages for tensile and bending, groups described in Table 1).

sludge WPC shows very low water uptake at 20% fiber proportion, but very high water uptake at 40% fiber proportion. Significant correlations were observed between wood content (Equation (1)) and water absorption (Table 6 and Figure 6a), and between cellulose content and water absorption (Figure 6d). This result is explained by the fact that wood is the most hydrophilic component in composites. It is important to note that correlations with wood and cellulose contents presented in Table 6 implicitly includes the effect of fiber proportion.

For thickness swell, only the WPC made with the sludge group performed better than those made with clean wood (Figure 5 and Table 4). The two other residue groups, bark and composite panel, produced less dimensionally stable WPC compared to those made with clean wood, with a few exceptions. Composites made with deinking sludge, Kraft sludge, and aspen wood are the most dimensionally stable at all fiber proportions. Birch wood WPC are among the less stable at all fiber proportions, and the TMP sludge WPC also show low stability. Thus, large differences were observed within each fiber group. As expected, the significant correlations between wood content (Equation (1)) and thickness swell and between cellulose content and thickness swell suggest that the hydrophilic nature of the lignocellulosic material can partly explain these results (Table 6), with some exceptions. For example, aspen wood has the highest cellulose and wood contents but produced among the most dimensionally stable WPC. Obviously, fiber characteristics alone cannot explain all results. Other factors, such as fiber-matrix compatibility, could play important role and should be investigated.

Effect of Fiber Type on WPC Mechanical Properties

In most cases, WPC made with clean wood fibers have higher modulus of elasticity than those made with residues (Figure 5). For all fiber proportions and both bending and tensile modulus of elasticity, aspen wood produced the stiffest WPC (Table 5). The composite panel residues group generally follows the clean wood group (Table 5), and shows equivalent bending modulus of elasticity (Table 3 and Figure 5). This result is unsurprising, because the residues were made of aspen wood with some cured resin. The bark group ranks last for all proportions for both bending and tensile modulus of elasticity, coming slightly behind the sludge group. Modulus of elasticity increases with increasing wood content and cellulose content (Figure 6b, Figure 6e, and Table 6). For example, aspen wood has the highest cellulose content and is therefore the stiffest (Table 2 and Table 5). Another example is the sludge group with its low wood content and thus low modulus of elasticity. No significant correlation coefficient was found between fiber aspect ratio and modulus of elasticity except for the sludge group, where the correlation was highly significant (Table 6). Within the sludge group, the Kraft sludge fibers have the highest aspect ratio and formed the stiffest WPC. In terms of mechanical strength, the wood group performed the best on average of all the residue groups for both tensile and bending strength (Figure 5). Aspen wood produced the best WPC for all fiber proportions for both bending and tensile strength (Table 5). The only exception is the Kraft sludge, which performed equally well or better than clean aspen wood on tensile strength. The bark group ranks last for tensile and bending strength for all proportions. For the other groups (sludge and composite panel), the ranking varies with fiber proportion and test type (bending or tensile). Significant correlations were found between wood content and mechanical strength (tensile and bending) and between cellulose content and mechanical strength (Figure 6c, Figure 6f, and Table 6). This result is explained by the fact that cellulose is the structural component of the wood cell wall, and it contains many hydroxyl groups for the esterification reaction with MAPE.[8] Thus, the

Table 5
Mechanical properties of WPC samples made with three proportions of the different fibers

Fiber proportion	Fiber type	Tensile properties		Elongation at break (%)	Impact energy (kJ/m^2)	Bending properties	
		E[a] (GPa)	Strength (MPa)			MOE[a] (GPa)	MOR[b] (MPa)
20%	Aspen wood	1.11^{ac}	28.3^{b}	7.38^{e}	22.9^{c}	1.36^{A}	38.2^{A}
	Birch wood	0.96^{c}	25.2^{e}	8.75^{cd}	23.5^{c}	1.11^{D}	33.4^{C}
	Spruce wood	1.01^{b}	25.1^{e}	8.09^{de}	21.6^{cd}	1.10^{D}	33.2^{C}
	Aspen bark	0.76^{e}	20.3^{h}	10.5^{b}	18.4^{e}	0.83^{H}	26.8^{F}
	Spruce bark	0.73^{e}	19.8^{i}	10.2^{b}	19.4^{de}	0.87^{G}	25.8^{G}
	MDI panel	1.00^{bc}	25.8^{d}	8.03^{de}	23.2^{c}	1.22^{B}	34.2^{B}
	PF panel	0.89^{d}	23.8^{f}	9.12^{c}	23.3^{c}	1.00^{F}	32.2^{D}
	Deinking sludge	0.72^{e}	22.2^{g}	18.5^{a}	45.7^{a}	0.74^{I}	25.0^{H}
	TMP sludge	0.86^{d}	28.0^{c}	5.98^{f}	16.5^{e}	1.05^{E}	30.7^{E}
	Kraft sludge	1.08^{a}	29.9^{a}	7.46^{e}	28.3^{b}	1.15^{C}	32.9^{C}
30%	Aspen wood	1.43^{a}	34.3^{a}	5.38^{de}	19.5^{c}	1.93^{A}	45.6^{A}
	Birch wood	1.34^{b}	29.0^{c}	5.21^{e}	16.7^{d}	1.72^{C}	41.5^{C}
	Spruce wood	1.34^{b}	28.7^{d}	4.84^{f}	15.7^{d}	1.57^{D}	42.5^{B}
	Aspen bark	0.91^{f}	21.4^{i}	5.86^{c}	13.0^{e}	1.05^{G}	30.0^{H}
	Spruce bark	0.93^{e}	19.7^{j}	5.14^{ef}	11.7^{e}	1.08^{G}	28.5^{J}
	MDI panel	1.34^{b}	27.0^{f}	4.11^{g}	13.3^{e}	1.86^{B}	40.6^{D}
	PF panel	1.16^{c}	26.6^{g}	5.59^{cd}	16.8^{d}	1.38^{E}	38.4^{E}
	Deinking sludge	0.86^{g}	24.7^{h}	9.81^{a}	30.3^{a}	0.94^{H}	29.3^{I}
	TMP sludge	1.03^{d}	27.4^{e}	4.10^{g}	11.4^{e}	1.29^{F}	32.7^{G}
	Kraft sludge	1.15^{c}	31.6^{b}	6.24^{b}	23.7^{b}	1.31^{F}	34.2^{F}
40%	Aspen wood	1.83^{a}	38.9^{a}	4.14^{b}	18.1^{c}	2.55^{A}	52.9^{A}
	Birch wood	1.64^{b}	32.0^{c}	3.57^{de}	15.3^{d}	2.31^{C}	49.2^{B}
	Spruce wood	1.61^{bc}	31.8^{c}	3.44^{e}	11.7^{e}	2.45^{B}	46.9^{C}
	Aspen bark	1.09^{h}	22.5^{g}	3.88^{c}	10.7^{e}	1.38^{G}	32.9^{H}
	Spruce bark	1.20^{g}	23.0^{g}	3.83^{c}	9.4^{f}	1.42^{G}	33.7^{G}
	MDI panel	1.61^{c}	33.6^{b}	3.70^{cd}	15.2^{d}	2.25^{D}	48.8^{B}
	PF panel	1.35^{d}	31.0^{d}	4.17^{b}	15.0^{d}	1.95^{E}	45.0^{D}
	Deinking sludge	1.06^{i}	28.0^{e}	6.71^{a}	21.9^{a}	1.31^{H}	34.7^{F}
	TMP sludge	1.28^{e}	26.4^{f}	2.79^{f}	9.0^{f}	1.60^{F}	34.1^{G}
	Kraft sludge	1.24^{f}	33.3^{b}	4.17^{b}	20.7^{b}	1.95^{E}	42.8^{E}
0%	HDPE[d]	0.63	21.6	1100	—	0.56	19.9

[a]modulus of elasticity; [b]modulus of rupture; [c]for each property within the same fiber proportion, values followed with the same letter are not significantly different and values followed with different letters are significantly different at 0.05 probability level; [d]elongation at break taken from supplier technical sheet, no break on impact test.

Table 6

Pearson's coefficients (determination coefficients) between selected WPC properties and selected fiber characteristics

		Density	Water uptake	Thickness swell	Tensile E[a]	Tensile strength	Bending MOE[a]	Bending MOR[b]	Impact energy
Cellulose[c]	All formulations	0.591^{**}	0.773^{**}	0.566^{**}	0.838^{**}	0.640^{**}	0.847^{**}	0.841^{**}	-0.301^{NS}
	Without sludge	0.811^{**}	0.868^{**}	0.620^{**}	0.931^{**}	0.838^{**}	0.937^{**}	0.919^{**}	-0.386^{NS}
	Sludge only	0.978^{**}	0.437^{NS}	0.183^{NS}	0.328^{NS}	0.060^{NS}	0.392^{NS}	0.394^{NS}	-0.087^{NS}
Ash[c]	All formulations	0.685^{**}	-0.262^{NS}	-0.327^{NS}	-0.240^{NS}	0.067^{NS}	-0.210^{NS}	-0.255^{NS}	0.423^{*}
	Without sludge	0.298^{NS}	0.097^{NS}	0.197^{NS}	-0.302^{NS}	-0.432^{NS}	-0.273^{NS}	-0.377^{NS}	-0.479^{*}
	Sludge only	0.915^{**}	0.045^{NS}	-0.245^{NS}	0.124^{NS}	0.107^{NS}	0.196^{NS}	0.303^{NS}	0.273^{NS}
Wood content[d]	All formulations	0.330^{NS}	0.894^{**}	0.818^{**}	0.783^{**}	0.376^{*}	0.789^{**}	0.705^{**}	-0.743^{**}
	Without sludge	0.987^{**}	0.928^{**}	0.780^{**}	0.784^{**}	0.543^{*}	0.793^{**}	0.679^{**}	-0.754^{**}
	Sludge only	0.368^{NS}	0.843^{**}	0.883^{**}	0.792^{*}	0.321^{NS}	0.809^{**}	0.626^{NS}	-0.835^{**}
Aspect ratio	All formulations	0.287^{NS}	-0.319^{NS}	-0.286^{NS}	-0.099^{NS}	0.264^{NS}	-0.090^{NS}	-0.141^{NS}	0.188^{NS}
	Without sludge	-0.046^{NS}	-0.215^{NS}	-0.379^{NS}	0.136^{NS}	0.136^{NS}	0.105^{NS}	0.061^{NS}	-0.074^{NS}
	Sludge only	-0.182^{NS}	-0.154^{NS}	$-.007^{\mathrm{NS}}$	0.635^{NS}	0.854^{**}	0.565^{NS}	0.631^{NS}	-0.284^{NS}

[a]modulus of elasticity; [b]modulus of rupture; [c]chemical compositions obtained from mass weighted sums of fiber chemical compositions from Table 2; [d]as defined by Equation (1); **significant at 1% probability level; * significant at 5% probability level; $^{\mathrm{NS}}$not significant at 5% probability level.

surface chemistry of the Kraft fibers provides good compatibility with MAPE,[8] resulting in effective reinforcement. Similar to previous results,[9,11,17] the two barks produced the weakest WPC at all proportions (Table 5) due to low intrinsic mechanical resistance and poor compatibility with MAPE.

Tensile elongation at break also showed significant variation with fiber type (Table 3). The between-fiber variation is low and without great practical interest, with the exception of deinking sludge fibers, which produced highly ductile WPC (Table 5). Deinking sludge is a unique fiber type because it contains clay recovered in the paper recycling process. Clay is a desirable filler material for thermoplastics because it has low impact on the mechanical performance (e.g., toughness, strength, elongation, etc.).[18]

In terms of impact energy, the sludge group produced higher toughness than the wood group, the composite panel group produced comparable toughness to the wood group, and bark produced lower toughness than the wood group (Figure 5 and Table 5). Much higher impact energy was found for deinking sludge WPC than for all other formulations. For example, at 20% fiber proportion, impact energy is approximately double that for the wood group. Kraft sludge WPC also performed well on impact tests. These results cannot be explained by either cellulose or ash contents (Table 6). Surprisingly, the fiber aspect ratio did not explain the results either (Table 6). This result is contradictory to previous reports that indicated that unnotched impact strength is inversely proportional to fiber aspect ratio.[19,20] The low values of aspect ratio (Table 2) for most of the analyzed materials (except TMP and Kraft fibers) is among the plausible explanations of the non-significant impact of the aspect ratio on the impact strength, except for the TMP and Kraft fibers (Table 2). The highest impact values were found for the deinked sludge composites (Table 5). This result could be explained by the high inorganic matters found in the deinked sludge (Table 2) compared to other materials. This result is in good agreement with previous findings[21] suggesting that small inorganic particles have a beneficial effect on toughness. Impact energy decreases as wood content increases, as shown by the highly significant correlation coefficients (Table 6). Thus, the wood material proportion of the residues has a more negative effect on impact resistance than the inorganic proportion of the residues. The very low impact resistance of the TMP sludge WPC is not explained. The uneven fiber dispersion in the TMP samples could be among the plausible explanations. Scanning electron micrographs did not show, however, evidence of bad fibers and small inorganic particles dispersion in the TMP composite (Figure 7). Other factors, such as surface roughness, fiber flexibility, and fiber-polymer interactions, are also among the plausible explanations, but further investigations are needed.

Opportunities for Recycling and Practical Implications

Although clean wood generally produces WPC with superior physical and mechanical properties compared to WPC made with wood residues, there are nevertheless opportunities for using some residues. Kraft sludge WPC ranked best or second-best for tensile strength and impact energy for all fiber proportions, and in many cases it performed better than birch and spruce fibers. Thus, according to the present results, Kraft sludge could be useful for WPC production. Deinking sludge could be used for applications where tenacity and water resistance are important. It could serve as a cost-effective plastic filler to replace more expensive raw fiber materials used in conventional WPC applications. It could also be used as an additive in WPC, in addition to conventional clean wood particles, to increase tenacity and water resistance. The composite panel industry residue group, containing around 95% wood, produced WPC with similar properties to those made with clean wood, with the

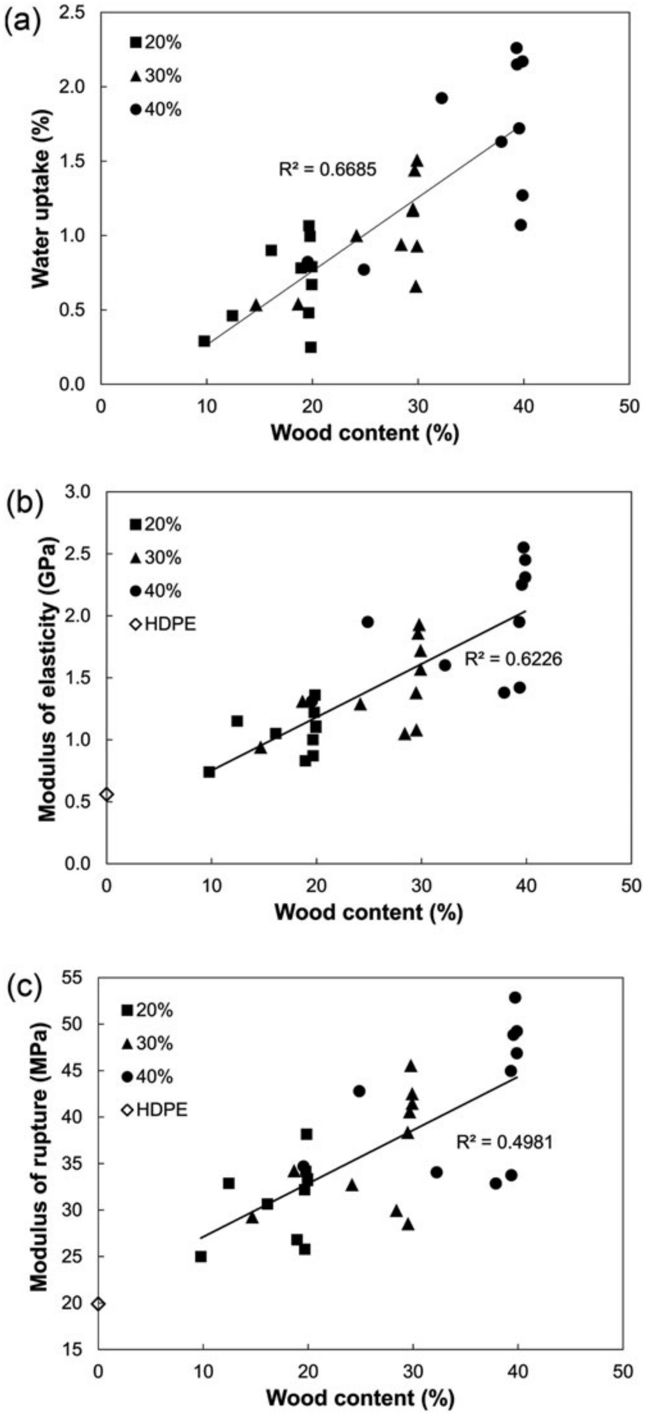

Figure 6. Correlation between wood content in WPC samples and (a) water uptake, (b) bending modulus of elasticity, (c) bending modulus of rupture. Correlation between cellulose content in WPC samples and (d) water uptake, (e) bending modulus of elasticity, (f) bending modulus of rupture. *(Continued)*

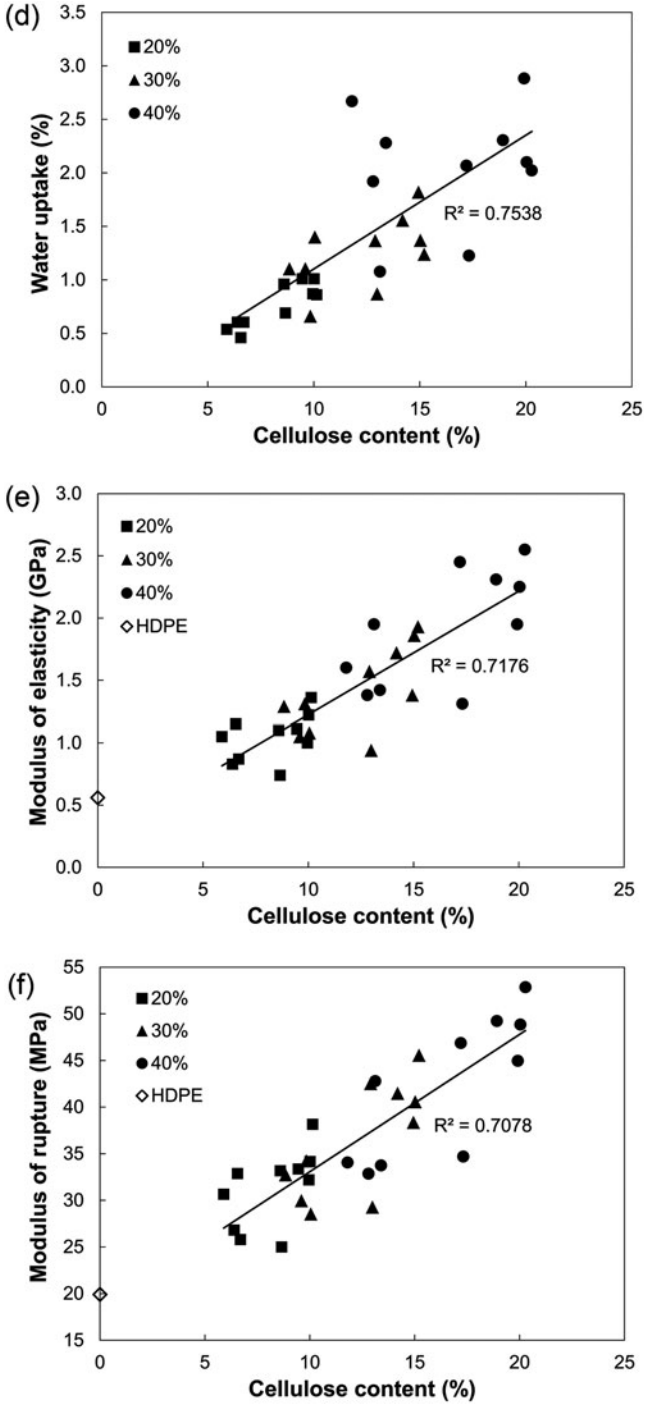

Figure 6. (Continued)

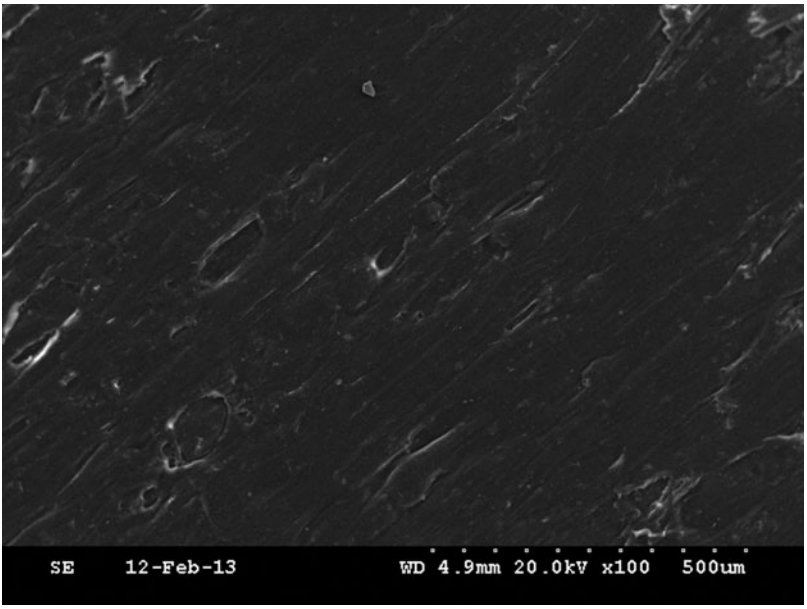

Figure 7. Scanning electron micrograph showing the fibers and inorganic material dispersion in TMP sludge WPC at 300X enlargement.

exception of moisture properties. It could therefore have a wide range of applications. Of the residues examined here, only bark and TMP sludge should be avoided for WPC production. Finally, the underused poplar fibers produced the best WPC in terms of mechanical strength and stiffness.

One interesting result is that the degree of contamination (ash content) did not explain any of the variations in WPC properties (Table 6), indicating that fiber contamination is not a limiting factor for WPC application. However, even low fiber contamination, as low as 5%, is undesirable for traditional wood-based composites such as particleboards and medium density fiberboards.[22] For example, Migneault et al.[23] observed a decrease in mechanical and physical properties with increasing ash content for medium-density fiberboards made with pulp and paper sludge.

Aside from the properties of the produced WPC, the low cost, availability, and environmental benefits of using residues are notable advantages. Moreover, the small particle size of these residues, which may be an obstacle for many applications, is adequate for WPC. On the other hand, there are many drawbacks to handling residues or waste. For example, sludge has very high moisture content and is typically difficult to dry. The high proportion of inorganic particles may increase wear of processing equipment. Social acceptability and health concerns would also need to be addressed prior to industry-scale production.

Conclusions

Results from this study indicate that thickness swell, water uptake, mechanical strength, and stiffness increased and elongation at break and impact energy decreased with increasing fiber proportion. In general, clean wood produced WPC with superior physical and mechanical properties compared to those made with wood residues, except for the Kraft mill sludge. The latter produced WPC with higher tensile strength and impact energy compared to those made with virgin birch and spruce fibers. Deinking sludge produced WPC with the highest tenacity and ductility.

The variation between the mechanical and physical properties of the WPC from different sources was mainly explained by the cellulose content and the wood content, while the fiber aspect ratio was not an important factor. The significant correlations between the wood content and WPC properties suggest that the wood content of fibers partially explains variations in WPC properties. However, the insignificant correlation between the ash content and WPC properties suggests that the presence of ash is not a limiting factor for WPC application.

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