Industrial Crops and Products 54 (2014) 248–256

Contents lists available at ScienceDirect

Industrial Crops and Products

journal homepage: www.elsevier.com/locate/indcrop

The potential of paper mill sludge for wood–plastic composites

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ARTICLE INFO

Article history:
Received 7 October 2013
Received in revised form 6 January 2014
Accepted 8 January 2014
Available online 18 February 2014

Keywords:
Primary and secondary sludge
Sludge characterization
Pulping process
Wood–plastic composites
Mechanical properties
Water immersion

ABSTRACT

Recent studies have demonstrated the potential of primary sludge (PS) as reinforcing fibers and secondary sludge (SS) as binder or co-binder in wood–plastic composites (WPC). A comparative study was conducted using paper mill sludge produced by three different pulping processes at two SS to PS ratios. The objectives were to determine the impact of PS and SS on the development of high density polyethylene (HDPE) WPC properties. Sludge produced by thermomechanical pulping (TMP), chemithermomechanical pulping (CTMP), and Kraft pulping were used at three different proportions (20%, 30%, and 40%) for composite manufacturing. The use of mixed sludge containing 30% SS resulted in lower tensile, flexural, and impact performance of the WPC compared to mixed sludge containing only 10% SS for the three pulping processes. Sludge type had a significant impact on the WPC physical and mechanical properties. Kraft sludge produced the best WPC properties, followed by CTMP and TMP sludge. Increasing the sludge proportion produced increasingly negative impacts on water absorption and thickness swelling, but improved the flexural and tensile properties.

1. Introduction

The pulp and paper industry requires large quantities of water to operate. The wastewater must then be treated to reduce the total suspended solid content and the oxygen demand prior to disposal (Gilbride and Fulthorpe, 2004). The sludge is the solid residue generated by the water treatment plant. Each ton of paper produces about 45 kg of dry sludge (Son et al., 2004). Sludge disposal can account for up to 60% of the total water treatment cost (Mahmood and Elliott, 2006). Certain fine particles that are mechanically removed from the wastewater are called primary sludge (PS). These suspended solids contain mostly cellulose, hemicelluloses, and lignin, as well as other possible residues such as bark and additives used as filler in paper production (Mahmood and Elliott, 2006; Chen et al., 2002). The remaining suspended solids are sent to a secondary treatment plant for bacterial digestion. The mixture of biosolids and the remaining suspended solids is called the secondary sludge (SS). The large numbers of bacteria in the SS make it difficult to dewater and dry, because a large portion of the water is trapped inside the living cells, and therefore tends to jellify (Mahmood and Elliott, 2006; Tchobanoglous et al., 2003; Mabee, 2001). Bacteria are made up of approximately 80% water and 20% dry material, of which 90% is organic and 10% inorganic. An approximate formula for the organic portion is $C_5H_7O_2N$ (Cheremisinoff, 1996). The nitrogen content in the simplified molecular structure is a particular feature of SS. When the SS is digested in the presence of oxygen, the aerobic process generates mainly heat, water (H_2O), and carbon dioxide (CO_2).

When a secondary treatment plant operates in the absence of oxygen, the digestion produces methane (CH_4) and carbon dioxide (CO_2). This anaerobic process considerably reduces the biomass produced by the digestion of suspended solids (Baudez, 2001). The mixture of PS and SS is called mixed sludge. Paper mills usually dispose of mixed sludge by burning and landfilling (Beauchamp et al., 2002). However, burning nitrate-rich paper mill sludge in industrial boilers has been criticized because it generates powerful greenhouse gases such as nitrate oxide (NOx) and provides low calorific value. In Canada, the public has begun complaining about land filling because of the unpleasant smell of the active bacterial culture in the sludge and the potential to produce elements that are toxic to fauna and flora (Pearson, 2005). Studies have recommended composting, ethanol production, and other thermal treatments such as pyrolysis, vitrification, and gasification for sludge disposal, but these alternatives are either economically unviable or liable to generate other sub-products that could be protested by environmentalists (Beauchamp et al., 2006; Mahmood and Elliott, 2006). Due to the very high volume of sludge worldwide, there is growing interest in either increasing agricultural use of sludge or processing sludge for use in producing other products (Chen et al., 2002). For example, Ou-Yang and Wu (2002) observed a 50% higher plant

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http://dx.doi.org/10.1016/j.indcrop.2014.01.013

growth rate when composted paper mill sludge was added to peat as a container medium for seed germination. They also noted that the sludge application rate should be adapted according to the paper sludge composition, which varies across mills.

Recent studies (Geng et al., 2007; Migneault et al., 2010, 2011a,b) have assessed the potential of recycling paper mill sludge as filler and co-binder in medium density fiberboard manufacturing. Geng et al. (2007) reported that medium density fiberboards made with up to 20% primary and deinking sludge met the American National Standards Institute minimum requirements. Migneault et al. (2010) found that thermomechanical pulping (TMP) sludge was suitable for medium density fiberboards and met the minimum requirements using up to 25% sludge content with a secondary to primary sludge ratio up to 3:7. Medium density fiberboards made with sludge from chemico-thermomechanical pulping (CTMP) and Kraft pulping did not meet the American National Standards Institute minimum requirements. Soucy (2007) studied the mechanical properties of WPC made with pulp from three different processes and found that Kraft fibers made stiffer WPC compared to TMP fibers. Migneault et al. (2009) reported that increasing CTMP fiber length improved the mechanical properties of injected and extruded WPC. Injection molding and extrusion are the two most commonly used thermoplastic processing methods, and they dominate the plastic products market (Trotignon et al., 1996). Injection molding uses WPC pellets with a maximum of 40% wood content, whereas the extruded product can contain up to 60% wood content (Maine, 2007). In both processes temperatures in excess of temperatures are generally used 120 °C. At these temperatures all living cells are killed (Regnault, 1990). Since these processing methods kill all the microorganisms and pathogens in the sludge, they provide a promising alternative to sludge disposal.

World production of high-density polyethylene (HDPE) was 29.8 million metric tons in 2007. About 10–15% of the HDPE is converted into construction products. HDPE is increasingly replacing traditional materials such as wood, glass, concrete, and paper in the construction industry, but its use is limited by its dependence on oil prices (Borruso, 2008). WPC have been proposed as an environmental solution for replacing a large proportion of plastic products, and the demand for it is expected to grow worldwide (Merle-Lamout and Pannetier, 2012; Ismail and Bakar, 2005a). The most commonly used thermoplastic polymer for manufacturing WPC is polyethylene, followed by polyvinyl chloride and polypropylene.

PS has proven to be a good reinforcing fiber source due to its high fiber content, whereas SS is used more as an adhesive or co-binder due to its high protein content (Zerhouni et al., 2012). Other paper mill additives found in PS and SS, such as clay and calcium carbonate, are currently used as filler in the plastic industry, and should be suitable for traditional plastic processing. For example, polypropylene is usually filled with mica, talc, calcium carbonate, and glass fibers to lower the price and strengthen the properties. More recently, several natural fibers such as wood, cellulose, jute, and bamboo have been investigated due to their low price, low density, high stiffness, and low abrasion during processing compared to traditional mineral fillers (Qiao et al., 2004).

Several studies have discussed the potential of SS, bacteria, and biofilm as binder and co-binder in material fabrication. Vu et al. (2009) described the molecular structure of some extracellular polysaccharides found in the biofilm of certain microorganism structures. They showed that alphaproteobacteria produce polymeric cellulose with a beta 1,4 bond, such as in wood cellulose. Cellulose is produced primarily by bacteria of the genera Acetobacter, but also by the genera Agrobacterium, Pseudomonas, and Rhizobium. Gerardi (2006) found that approximately 20% of the bacteria in an activated sludge process constituted gram-negative cocci and rods. This 20% also included species of the genera Acetobacteria and Pseudomonas. Haag et al. (2006) showed that glues derived from extracellular polysaccharides are VOC free and form sufficiently strong bonds at low and moderate relative humidity, achieving shear strength comparable to that of commercial polyvinyl acetate-based adhesives. Zerhouni et al. (2012) obtained higher internal bond strength from handsheets made with a higher SS:PS ratio and dried at 180 °C. The higher bond strength was attributed to a higher number of hydrogen bonds within the handsheet under these conditions. Recently, Xing et al. (2013) made particleboard panels using SS and urea-formaldehyde resins. All their tested formulations met most American National Standards Institute minimum requirements with significantly lower formaldehyde emissions.

Only a few studies to date have examined the use of paper mill sludge as filler, co-binder, or reinforcing fibers in WPC. Son et al. (2001) studied the effect of extrusion temperature and sludge particle size on the properties of sludge-filled thermoplastic. They observed that increased particle size resulted in improved mechanical properties, which could be explained by the high cellulosic content of the PS and the smaller particle size (fines) of the organic content in the SS. They also found improved dimensional stability with higher extrusion temperature and lower water absorption. Ismail and Bakar (2005a,b, 2006) studied the properties of a thermoplastic blend of polypropylene filled with paper mill sludge. Their results showed several similarities with conventional WCPs made with thermoplastics processes: (1) increased sludge content, water sorption, and Young modulus but reduced tensile strength and strain; (2) surface modifications such as sludge esterification and acetylation also improved most of the composites' physical and mechanical properties; and (3) the addition of sludge to a thermoplastic polymer reduced the cristallinity but did not affect the melting temperature.

The use of sludge for WPC application would benefit both the industry, by providing a new fiber source, and the environment, by reusing an industrial waste for high value products. However, the chemical composition of sludge varies depending on the pulping process, the sludge treatment processes and the chemical composition varies among SS to PS ratios. Consequently, using sludge in the manufacture of WPC is challenging and might lead to variable WPC properties. The sludge chemical composition is expected to play a major role in the WPC properties development. Thus, using sludge samples from different processes and at different SS to PS ratios is associated with important variations in both morphological and chemical properties of the sludge material used in WPC manufacturing. Thus, this study compares the physical and mechanical properties of HDPE WPC made with sludge from three different pulping processes and two SS to PS ratios. The objectives were to (1) investigate the role of PS and SS in the development of the WPC properties; and (2) to study the effect of the sludge's chemical composition on WPC properties.

2. Material and methods

2.1. Sludge sample preparation

The raw material was obtained from three paper mills, using softwood chips from different pulping processes. The TMP sludge is from White Birch Division of Stradacona Inc. in Quebec City, the CTMP sludge is from Lac-Saint-Jean Abitibi-Bowater in Dolbeau-Mistassini, and the Kraft sludge is from SFK Pulp in St-Félicien, all in Canada. All mills use chips from North American black spruce (*Picea Mariana*).

Mixed sludge samples were collected from the sludge presses at the dewatering facilities of the three mills at two different SS to PS ratios: SS:PS = 1:9 and 3:7. The SS:PS ratio vary across mills and within mills over time, but these are typical values. PS and SS were

Table 1
WPC formulations used in the experimental design.

Sludge source	Sludge ratio (SS:PS)	Sludge content (%)	HDPE content (%)	MAPE content (%)
TMP	1:9	20	77	3
		30	67	3
		40	57	3
	3:7	20	77	3
		30	67	3
		40	57	3
CTMP	1:9	20	77	3
		30	67	3
		40	57	3
	3:7	20	77	3
		30	67	3
		40	57	3
Kraft	1:9	20	77	3
		30	67	3
		40	57	3
	3:7	20	77	3
		30	67	3
		40	57	3

characterized separately by Migneault et al. (2011a). The two mixed sludges were preheated at 750 kPa for 90 s and then ground using an Andritz single disk refiner at 2000 rpm in the FPInnovations-pilot plant facility in Quebec City, Canada. The remaining moisture content was removed using a flash tube dryer to 5–15% moisture content and then dried to 1–3% moisture content in a rotary dryer.

2.2. Sludge properties characterization

A Fiber Quality Analyzer (Optest Equipment) was used to measure the sludge particle size distribution using three repetitions of 5000 fibers for each sludge sample. The PS and SS liquid sludge were air dried for two weeks to avoid protein degradation and the sludge samples were prepared as wood-based material in accordance with Tappi T264 prior to chemical analysis.

The cellulose content was determined by Kürschner and Hoffer's nitric acid method (Browning, 1967). Solvent extractives content and total Klason lignin content were measured according to Tappi Standard T204 and T222, respectively. The nitrogen content was determined using a Perkin-Elmer Series II CHNS/O 2400 analyzer according to Standard methods (1985). The ash content of the moisture content was determined after combustion at 525 °C according to Tappi Standard T211.

2.3. Composite manufacturing and testing

The WPC formulations (Table 1) were produced according to a factorial plan where factors are the mill pulping processes (TMP, CTMP, Kraft), sludge proportions (20%, 30%, 40%), and SS:PS ratios (1:9, 3:7), for a total of 18 different formulations.

Sludge composites were prepared in two stages: compounding for pelletizing followed by injection molding. A counter-rotating conical twin-screw extruder (Thermo Scientific HAAKE PolyLab OS Rheodrive 7 with Rheomex OS extruding module) was used to compound fibers, HDPE (SCLAIR®2907, Nova Chemical). 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. These processing parameters were optimized experimentally for good fiber dispersion without fiber thermal degradation. The extrudate was cooled in a water bath and ground into 3-mm long pellets. Pellets were mixed with a fixed proportion at 3% of maleated polyethylene (MAPE, Fusabond 226DE, DuPont). Samples were molded for tensile, bending, and impact specimens with an Arburg 370 A (600 kN) injection molding. 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. These processing parameters were selected for filling the molds with minimal thermal degradation. 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.

All samples were conditioned for at least 48 h at 20 °C and 65% relative humidity prior to testing. Three-point bending properties were 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. Un-notched impact resistance was measured according to ASTM D 4812. Bending, tensile, and impact tests were repeated ten times. Water uptake, and thickness swell (TS) of water-soaked samples were measured according to ASTM D1037. Composite tensile ruptured surfaces were analyzed using scanning electron microscopy (SEM) using an S-3500N variable pressure vacuum scanning electron microscope (Hitachi) combined with a Link ISIS Series 300 EDS analytical system (Oxford Instruments).

2.4. Statistical data analysis

Statistical analyses were performed by means of the SAS package version 9.2 (SAS Institute, 2007).

Data were subjected to variance analyses (ANOVA) using a GLM procedure. Least square Mean test was used to compare the properties means. Correlation analyses were also conducted using the CORR procedure to establish relationships between composites properties and the chemical properties.

3. Results and discussion

3.1. Sludge fiber distribution

The sludge fiber distribution was characterized by Migneault et al. (2011a) where similarities between the TMP and CTMP sludge fibers size and distribution were observed while the Kraft sludge showed longer fibers with fewer fines (fibers shorter than 0.2 mm) (Migneault et al., 2011a). Mechanical pulping tends to damage the fibers and produces high fines content, which could explain why the Kraft sludge ($l = 878$ μm, 5.6% fines) showed higher fiber length distribution and lower fines content compared to the CTMP ($l = 367$ μm, 19.6% fines) and TMP ($l = 317$ μm, 23.3% fines) sludge.

3.2. Chemical composition

The chemical composition of the sludges is presented in Table 2. PS and SS showed high ash content compared to pulps. Krigstin and Sain (2006) noticed 42–53% ash content in newsprint sludge depending on fiber size. Son et al. (2001) also observed 38–74% ash content in newsprint sludge with the highest concentration in the smallest fiber size class. Zerhouni et al. (2012) found a 30% ash in the TMP sludge. Inorganic material is heavier than water, and it is not recovered from any of the pulping processes. Some pulp and paper mills also use clay or bentonite in the water treatment step, thus increasing the ash concentration. The cellulose content of sludge was lower in the SS than in the PS for all three pulping processes. The Kraft pulp contained the highest cellulose content (88%), but its PS had slightly lower cellulose content (41%) than the CTMP PS (45%) and higher than the TMP PS (37%).

The lignin content was higher in the SS compared to the PS for all three pulping processes (Table 2). Zerhouni et al. (2012) observed lignin content in the range of 29–35% for the TMP, CTMP and Kraft

Table 2
Chemical composition of pulp, primary sludge (PS), and secondary sludge (SS) produced by the three pulping processes.

Material type	SPF	Pulping process								
	Wood	TMP			CTMP			KRAFT		
		Pulp	PS	SS	Pulp	PS	SS	Pulp	PS	SS
Ash test (%)	0.2–0.4	0.3 (0.0)	19.6 (0.2)	12.0 (0.1)	5.2 (0.1)	30.2 (0.1)	18.0 (0.2)	0.3 (0.0)	49.1 (0.4)	41.3 (1.4)
Cellulose (%)	43–46	49.7 (0.5)	36.5 (0.5)	19.7 (0.2)	52.7 (4.8)	44.6 (0.2)	26.6 (0.3)	87.9 (1.1)	41.4 (1.2)	18.9 (0.4)
Lignin (%)	27–30	25.3 (0.3)	23.6 (0.2)	50.2 (0.5)	29.9 (0.9)	22.6 (0.4)	50.0 (1.1)	0.0 (0.0)	20.3 (0.6)	36.4 (1.0)
Extractives (%)	5–8	7.3 (0.2)	15.5 (0.4)	21.5 (0.6)	4.6 (0.2)	2.9 (0.1)	1.7 (0.2)	3.4 (0.6)	0.4 (0.0)	7.9 (0.0)
Nitrogen (%)	–	0.1 (0.0)	0.5 (0.0)	7.7 (0.0)	0.1 (0.0)	0.2 (0.0)	5.4 (0.0)	0.0 (0.0)	0.1 (0.0)	1.3 (0.0)

All measured values for pulp, primary sludge, and secondary sludge from Migneault et al. (2011a). Standard deviations are in brackets. SPF, maximum and minimum values for spruce, pine, and fir wood species, from Rowell (2005). Tappi Standard Method T204 (extractives content) and T222 (Klason lignin content) were developed for wood and not for sludge. Results for secondary sludge may therefore be overestimated.

sludge. With around 50% lignin content in the SS for the TMP and CTMP mill, results in this study are significantly higher. Nadji et al. (2010) suggested that high insoluble lignin content in the SS may be overestimated due to interference by protein material in the sludge. The lignin dissolved in the Kraft cooking is typically consumed in the chemical recovery processes. Thus, we expected low lignin content for the Kraft sludge. However, lignin was present in high proportions in the Kraft PS and SS. This result suggests that lignin in the sludge comes from pre-cooking operations, such as debarking and chipping or from pulp washing steps. Lignin and extractives are less digested by the microorganisms in the SS, so their higher content in the SS than in the PS could be due to cellulose and sludge digestion. The CTMP sludge showed higher extractives content in the PS compared to that of TMP and Kraft sludge samples. The extractives content in the TMP sludge (PS and SS) was higher compared to the sludge from the two other pulping processes, because TMP does not require the use of chemicals that lixiviate the constituents. Cellulose conversion by biological reduction was also observed, as indicated by the increasing nitrogen content due to protein synthesis in the SS compared to the PS for all three pulping processes. Zerhouni et al. (2012) also observed higher nitrate content and lower cellulose content in the SS compared to the PS from three pulping processes.

Because the aerated lagoons used by the Kraft mill have long retention periods, they allow endogenous growth, whereas in the aerobic treatment used by the TMP and CTMP mills, the SS retains its slimy content, which facilitates dewatering (Cheremisinoff, 1996). The slimy content refers to biofilm or extracellular polymeric substances. The biofilm contains mainly polysaccharides, but also proteins, nucleic acids, lipids, and humic substances. Their composition depends on the type of microorganisms, age of the sludge, and the environmental conditions (Vu et al., 2009). Thus, the chemical composition of sludge produced according to paper manufacturing standards may be slightly affected by interference by the microorganisms found in the SS.

3.3. *Composite properties*

Table 3 presents the *F* values and level of significance for the physical and mechanical properties measured in this study. Pulp type, proportion of sludge in the WPC, and the SS ratio showed significant effects on all properties. The only exception was that the SS ratio had no significant effect on either water absorption or thickness swelling.

3.3.1. *Tensile properties*

Adding sludge at different proportions improved the tensile modulus of elasticity and strength for all three pulping processes (Table 4). However, the maximum strain and rupture energy decreased with increasing sludge proportion. These results were expected, and are in good agreement with previous reports (Ismail and Bakar, 2005a,b, 2006; Krigstin and Sain, 2006). Increasing the sludge proportion improved the mechanical properties of the material, whereas reducing the volumetric ratio of HDPE decreased the plasticity. The modulus of elasticity improved by 55 to 150% for the Kraft sludge composites compared to 55 to 135% for the CTMP and 35 to 125% for the TMP. Soucy (2007) also found that Kraft pulp fibers provided better tensile properties than CTMP and TMP fibers in WPC. These results could also be explained by the chemical treatment of cellulose fibers (pulp), which increases the numbers of hydroxyl groups at the fiber surface and reduces lignin and extractives contents. Fig. 1 illustrates SEM images of the rupture zones in tensile specimen of WPC composites made from TMP, CTMP and Kraft sludge. In the rupture zones, the TMP sludge fibers (Fig. 1a) and CTMP (Fig. 2b) seem to be totally separated of the polymer matrix and there is no evidence of fiber breakage due to tensile rupture. The Kraft sludge fibers do not show either evidence of fiber breakage due to tensile rupture but present a highly fibrillated surface and show some interlocking with the polymeric matrix (Fig. 1c).

Table 3
Analysis of variance (*F* values) for the physical and mechanical properties of WPC made with PS and SS.

Properties		Pulp	Sludge proportion	Secondary sludge ratio
Tensile	Modulus of elasticity (E)	21.98^{**}	403.12^{**}	162.71^{**}
	Maximal resistance (Rm)	582.43^{**}	49.98^{**}	208.43^{**}
	Work at break (W_b)	83.75^{**}	276.76^{**}	85.13^{**}
	Elongation at break (ε_b)	50.8^{**}	383.18^{**}	43.54^{**}
Flexural	Modulus of elasticity (MOE)	70.33^{**}	919.95^{**}	545.46^{**}
	Modulus of rupture (MOR)	122.38^{**}	218.01^{*}	515.10^{**}
Impact	Izod Impact energy (IE)	141.34^{**}	138.05^{**}	44.7^{**}
Immersion	Water absorption (WA)	11.51^{**}	152.37^{**}	0.92^{NS}
	Thickness swelling (TS)	12.07^{**}	122.71^{**}	0^{NS}

**Statistically significant at 0.01 probability level, ^{NS}not statistically significant at 0.05 probability level.

Table 4
Mechanical properties of the sludge-filled WPC for all tested formulations.

Formulations		Tensile properties				Flexural properties		Impact energy (J/m)
		E (GPa)	Rm (MPa)	ε_b (%)	W_b (kJ/m^2)	MOE (GPa)	MOR (MPa)	
SS:PS ratio 1:9								
TMP	20	$1.00^{J,K,L}$	28.12^{F}	8.96^{A}	2005^{A}	1.29^{G}	33.50^{J}	$257.0^{E,F}$
		(0.02)	(0.51)	(1.21)	(278)	(0.04)	(0.58)	(41.2)
	30	1.19^{F}	31.22^{D}	$3.90^{D,E}$	$817^{E,F}$	1.61^{F}	38.51^{G}	$177.9^{H,I}$
		(0.02)	(0.98)	(0.24)	(69)	(0.07)	(0.91)	(13.6)
	40	1.41^{C}	33.46^{C}	2.94^{F}	$595^{G,H,I}$	2.12^{C}	42.33^{E}	$134.8^{J,K}$
		(0.01)	(0.82)	(0.11)	(39)	(0.02)	(0.51)	(13.7)
CTMP	20	$1.04^{I,J}$	29.89^{E}	8.34^{A}	1960^{A}	1.27^{G}	35.12^{H}	315.5^{C}
		(0.02)	(0.49)	(0.91)	(224)	(0.04)	(0.73)	(31.3)
	30	1.18^{F}	35.73^{B}	4.41^{D}	$1030^{C,D}$	1.73^{E}	41.21^{F}	$240.6^{F,G}$
		(0.02)	(1.30)	(0.26)	(90)	(0.07)	(0.94)	(24.9)
	40	1.46^{B}	38.47^{A}	$3.21^{E,F}$	$731^{F,G}$	2.34^{B}	47.27^{C}	$182.8^{H,I}$
		(0.01)	(0.92)	(0.17)	(70)	(0.04)	(0.72)	(15.7)
Kraft	20	$0.99^{K,L}$	30.26^{E}	8.58^{A}	2030^{A}	1.71^{E}	45.07^{D}	410.2^{A}
		(0.02)	(0.59)	(3.07)	(869)	(0.08)	(0.51)	(50.8)
	30	1.36^{D}	35.44^{B}	7.19^{B}	1961^{A}	2.36^{B}	52.03^{B}	307.6^{C}
		(0.06)	(0.66)	(0.68)	(231)	(0.04)	(0.40)	(30.3)
	40	1.57^{A}	37.69^{A}	5.35^{C}	1465^{B}	2.62^{A}	57.33^{A}	$288.5^{C,D,E}$
		(0.06)	(0.88)	(0.76)	(251)	(0.20)	(2.56)	(51.5)
SS:PS ratio 3:7								
TMP	20	0.86^{M}	27.95^{F}	5.98^{C}	1202^{C}	1.05^{I}	$30.65^{M,N}$	$208.9^{G,H}$
		(0.03)	(0.23)	(0.59)	(156)	(0.02)	(0.21)	(35.0)
	30	$1.03^{K,J}$	27.44^{F}	4.10^{D}	$754^{F,G}$	1.29^{G}	32.74^{K}	144.5I,J
		(0.01)	(0.26)	(0.22)	(65)	(0.04)	(0.39)	(19.7)
	40	1.28^{E}	26.41^{G}	2.79^{F}	$494^{H,I}$	1.60^{F}	$34.06^{I,J}$	$114.1^{J,K}$
		(0.04)	(1.42)	(0.40)	(101)	(0.07)	(0.74)	(12.5)
CTMP	20	0.97^{L}	26.34^{G}	5.57^{C}	$1100^{C,D}$	1.01^{I}	30.14^{N}	199.1^{H}
		(0.03)	(0.59)	(0.53)	(120)	(0.03)	(0.23)	(33.5)
	30	0.96^{L}	24.30^{H}	$3.85^{D,E}$	$639^{F,G,H}$	1.30^{G}	30.99^{M}	$144.3^{I,J}$
		(0.09)	(1.32)	(0.13)	(44)	(0.04)	(0.37)	(18.2)
	40	$1.12^{G,H}$	23.95^{H}	2.55^{F}	392^{I}	1.64^{F}	31.60^{L}	103.0^{K}
		(0.02)	(0.87)	(0.21)	(48)	(0.03)	(0.37)	(8.7)
Kraft	20	$1.08^{I,H}$	29.91^{E}	7.46^{B}	1709^{B}	1.15^{H}	32.88^{K}	358.0^{B}
		(0.19)	(0.98)	(1.99)	(526)	(0.02)	(0.37)	(64.4)
	30	$1.15^{F,G}$	31.64^{D}	6.24^{C}	1514^{B}	1.31^{G}	34.24^{I}	$299.4^{C,D}$
		(0.03)	(1.05)	(1.12)	(320)	(0.03)	(0.50)	(63.4)
	40	1.24^{E}	33.31^{C}	4.17^{D}	$961^{D,E}$	1.95^{D}	42.78^{E}	$260.7^{D,E,F}$
		(0.07)	(1.67)	(0.68)	(210)	(0.05)	(0.69)	(30.4)

Standard deviations are in brackets. Results based on LSD procedure at $\alpha = 0.05$, means with the same letters in this table are not significantly different.

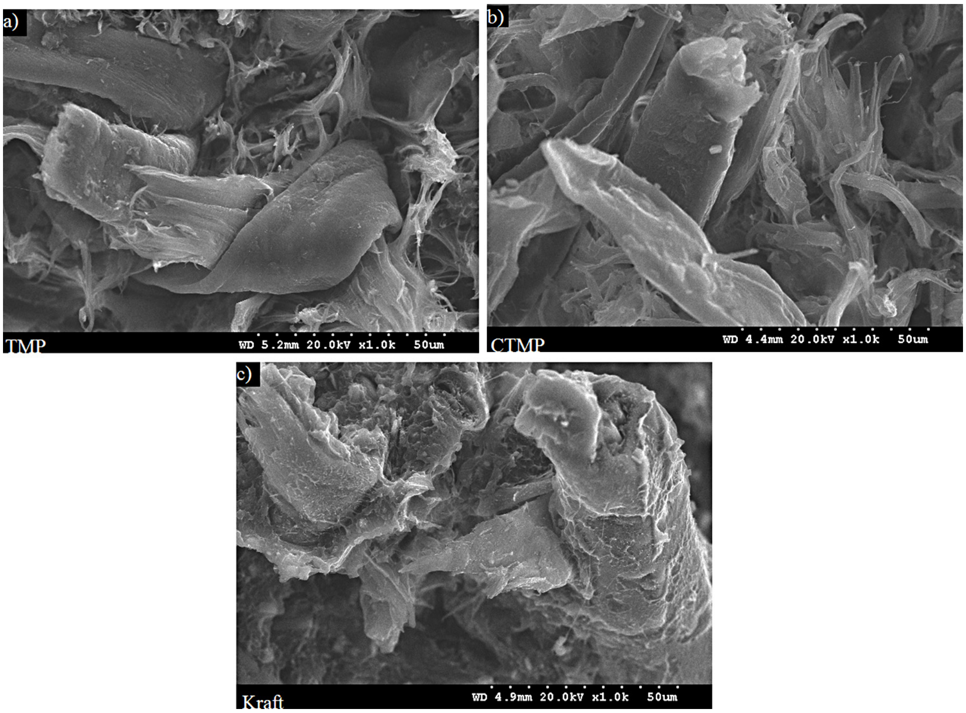

Fig. 1. SEM micrograph of TMP (a), CTMP (b) and Kraft (c) WPC at 1000×.

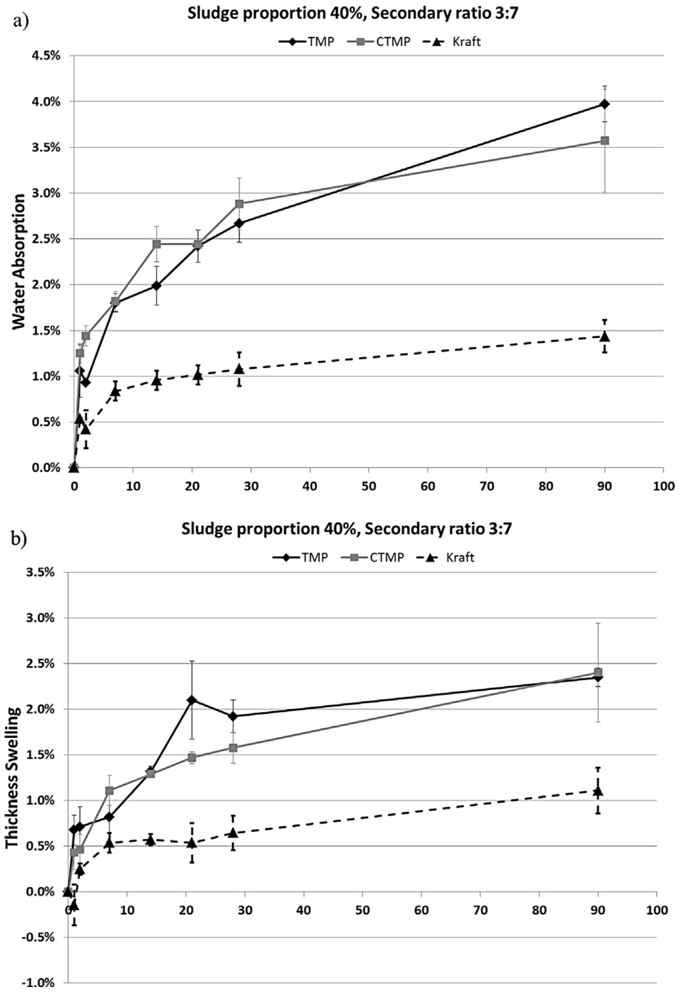

Fig. 2. Water absorption (a) and thickness swelling (b) as a function of the time in days for the WPC with an SS:PS ratio of 3:7.

Increasing the SS:PS ratio from 1:9 to 3:7 produced a significant decrease in tensile strength for all three sludge composites (Table 4), with a stronger decrease for CTMP sludge composite. A decrease in the Young modulus with higher SS ratio was observed for all tested formulations. For example, at 20% CTMP sludge proportion, the strain at break was reduced from 8.3% to 5.6%, and when the SS ratio was increased from 1:9 to 3:7, the mechanical energy was reduced from 1960 to 1100 kJ/m^2. Hence, the binding effect of SS on the CTMP and TMP composites cannot be confirmed. For the Kraft sludge, the higher SS ratio produced less variation in tensile properties compared to the CTMP and TMP WPC. The activated sludge process generates more biofilm containing short sugar chains, which may explain the lower strength produced by the inclusion of CTMP and TMP SS compared to the Kraft SS, which has a longer residence time (i.e., sludge age) because it is treated in an aerated lagoon.

The tenacity of the WPC decreased with increasing sludge proportion for all formulations (Table 4). For example, for the TMP sludge, when the sludge proportion was increased from 20 to 40% for the 1:9 SS ratio, the strain at break (ε_b) was reduced from 9 to 3% and the rupture energy (W_b) from 2000 to 600 kJ/m^2. At 20% sludge proportion, the WPC formulation showed similar behavior, but at 40% proportion, the Kraft WPC had significantly higher W_b and ε_b.

3.3.2. *Flexural properties*

The composites flexural modulus of elasticity and modulus of rupture increased with increasing sludge proportion for all formulations. When the sludge proportion was increased (from 20 to 40%), the flexural MOE and strength increased regardless of sludge type or SS ratio (Table 4). The addition of Kraft fibers to the WPC produced higher MOE and MOR values, thus making them a better candidate for WPC application compared to the TMP and the CTMP sludges. The TMP and CTMP WPC bending properties were similar for several formulations, but the CTMP WPC tended to have slightly better flexural properties (Table 4). The better flexural performance of the Kraft sludge composites compared to that of CTMP and TMP composites can be explained by the interlocking between the fibrillated fiber surface and the polymer as shown in Fig. 1c.

The SS content reduced the flexural properties for all tested formulations. When the SS:PS ratio was increased from 1:9 to 3:7, the MOE decreased by 19 to 44% and the flexural strength by 9 to 34%, regardless of sludge type or proportion. For example, for the 40% Kraft sludge composite, when the SS ratio was increased from 1:9 to 3:7 the flexural MOE decreased from 2.62 GPa to 1.95 GPa. For the WPC with 30% CTMP, when the SS ratio was increased from 1:9 to 3:7, the flexural strength dropped from 41 to 31 MPa. Thus, the adhesive properties of the SS did not appear to contribute to the WPC mechanical strength. Further surface chemistry analysis are needed to identify the adhesion mechanisms of the PS and SS in the WPC.

For the TMP and CTMP WPC, the negative impact of increasing SS content on the flexural properties was amplified with increasing fiber proportion. For the TMP composites, when the sludge proportion was increased from 20% to 30 and 40%, the flexural MOE decreased by 19, 20 and 25%, respectively. When the SS ratio was increased for the 20, 30 and 40% proportions, flexural strength was reduced by 14, 25, and 33%, respectively. For the Kraft WPC, increased SS content had a greater effect on the bending strength and stiffness compared to the CTMP and TMP WPC.

Maine (2007) found that the flexural modulus and strength in a first-generation of commercial WPC was in the range of 2 to 4 GPa and 20 to 25 MPa, respectively. Our results show that all formulations met the minimum requirements for strength, but only four formulations met the minimum requirements for rigidity. Increasing the sludge proportion from 40% to 60% in an extrusion process could help meet the minimum requirement for the flexural MOE, but the loss of tenacity should be monitored. Under these conditions, WPC made with HDPE and 40% paper mill sludge from any process would provide a competitive fiber source as long as the SS content is less than 10%. The Kraft sludge therefore appears to have higher potential for commercial applications.

3.3.3. *Izod resilience*

The impact energy (Table 4) is strongly dependent on the sludge type and proportion and the SS ratio. The WPC with Kraft sludge showed the highest impact resistance, followed by WPC made with CTMP and TMP sludge. For example, the 20% sludge-filled composite with an SS ratio of 1:9 achieved 410 J/m for the Kraft WPC compared to 316 J/m and 257 J/m for the CTMP and TMP WPC, respectively. However, the Kraft sludge WPC showed higher standard deviations at up to 35% of the average value. Some plausible explanations for the better performance of the Kraft sludge include longer fibers and higher apparent density which increases fiber-matrix contact and reduces voids where crack initiation occurs. Some Kraft fibers collapsed in the injection molding process (Fig. 1c). Collapsed fibers take fewer space and may explains the higher density and lower volume of the Kraft WPC compared to the TMP and the CTMP.

Increasing the sludge content from 20 to 40% reduced the rupture energy for all formulations. For the 1:9 ratio, this reduction

Table 5
Water immersion test results after 90 days for all tested conditions.

		WA_{90days}		TS_{90days}	
		Ratio 1:9	Ratio 3:7	Ratio 1:9	Ratio 3:7
TMP	20	$0.68\%^{F,G}$	$0.60\%^{F,G}$	$0.86\%^{C,E,F,D}$	$1.00\%^{E,B,C,D}$
	30	$1.29\%^{D,E,F}$	$1.69\%^{D,E}$	$1.11\%^{B}$	$1.36\%^{B}$
	40	$3.28\%^{B}$	$3.97^{A}\%$	$2.29\%^{A}$	$2.35\%^{A}$
CTMP	20	$0.88\%^{E,F,G}$	$0.88\%^{D,E}$	$0.75\%^{D,E,F,G}$	$0.54\%^{F,G}$
	30	$1.42\%^{C,D}$	$1.69\%^{C}$	$1.18\%^{B,C}$	$1.00\%^{B,C,D,E}$
	40	$3.94\%^{A}$	$3.57\%^{A}$	$2.43\%^{A}$	$2.40\%^{A}$
Kraft	20	N/A	$0.66\%^{G}$	N/A	$0.47\%^{G}$
	30	N/A	$1.00\%^{E,F,G}$	N/A	$0.72\%^{E,F,G}$
	40	N/A	$1.44\%^{C,D,E}$	N/A	$1.11\%^{B,C,D}$

Results based on LSD procedure at $\alpha = 0.05$, means with the same letters in this table are not significantly different.

Table 6
Correlation coefficients between WPC and sludge chemical composition.

	Extractives	Lignin	Cellulose	Nitrate	Ash
E	0.09^{NS}	0.48^{**}	0.62^{**}	0.1^{NS}	0.52^{**}
Rm	-0.20^{**}	-0.1^{NS}	0.44^{**}	0.15^{*}	0.48^{**}
ε_b	-0.36^{**}	-0.70^{**}	-0.50^{**}	-0.1^{NS}	-0.20^{*}
W_b	-0.41^{**}	-0.63^{**}	-0.38^{**}	-0.1^{NS}	-0.1^{NS}
MOE	0.0^{NS}	0.47^{**}	0.80^{**}	-0.1^{NS}	0.62^{**}
MOR	-0.1^{NS}	0.12^{NS}	0.55^{**}	-0.38^{**}	0.59^{**}
IE	-0.54^{**}	-0.56^{**}	-0.32^{**}	-0.73^{**}	0.29^{**}
WA_{90days}	0.38^{**}	0.84^{**}	-0.60^{**}	0.59^{**}	0.0^{NS}
TS_{90days}	0.2^{NS}	-0.62^{**}	-0.59^{**}	0.2^{NS}	-0.28^{*}

**Statistically significant at 0.01 probability level, *Statistically significant at 0.05 probability level, ^{NS}not statistically significant at 0.05 probability level.

reduced the rupture energy by 42, 72, and 90% for the TMP, CTMP, and Kraft WPC, respectively. The greater energy drop in the case of the Kraft sludge is not understood, but the 20% Kraft WPC formulation had a much higher toughness compared to the TMP and the CTMP formulations. Increasing the SS ratio further reduced the rupture energy. For the CTMP formulations, when the SS ratio was increased from 1:9 to 3:7, the reduction in the impact energy ranged from 60 to 80%. The TMP and Kraft WPC were less affected by the SS content, with 20 and 10% decrease in rupture energy, respectively.

3.3.4. Thickness swelling (TS) and water absorption (WA)

Fig. 2 shows the change in thickness swell (TS) and water absorption (WA) with immersion time for the WPC with 40% sludge content and SS:PS ratio of 3:7. The TMP and CTMP WPC swelled more and absorbed more water compared to the Kraft WPC. For all tested formulations, both TS and WA tended to increase steadily from zero up to 30 days, followed by a plateau or a slow increase up to 90 days. Nevertheless, the total increase in thickness swell and water uptake for all formulations was well below 5% and 3%, respectively (Table 5).

Table 4 presents the TS and WA after three months (90 days) of immersion for the TMP, CTMP, and Kraft WPC. As the sludge proportion increased, the weight and thickness gain increased for all formulations. These results were expected, due to the hydrophilic character of the fibers in the sludge. The difference in the TS and WA between the WPC made with 1:9 and 3:7 SS:PS ratios was not statistically significant (Tables 3 and 5). A comparison of the three sludge types shows that the TMP and CTMP WPC had similar TS and WA for both conditions (Table 5). The Kraft WPC has significantly lower TS and WS. Soucy (2007) also observed lower WS and TS for Kraft pulp fibers compared to CTMP and TMP fibers, even though the particle size distribution for the Kraft pulp was larger compared to the two other fiber sources. The higher compatibility of the MAPE coupling agent and the Kraft pulp due to its higher oxygen content at the surface, described by Bouafif et al. (2008), could explain the lower water absorption of the Kraft WPC.

3.4. Effect of chemical composition on physical and mechanical properties

Table 6 presents the correlation coefficients between the WPC physical and mechanical properties and the chemical composition of the sludge fibers. Most of the correlations were highly significant. Significant correlations were found between the cellulose content and all physical and mechanical properties, except for impact energy. This result is due to the variation in total cellulose content across WPC due to variations in sludge proportion and to differences in chemical composition between the different sludge types used in this study. Cellulose content is known to have a positive impact on the mechanical and physical properties of WPC. At constant fiber content, all mechanical properties improved significantly with increasing cellulose content in the WPC, with the exception of a few conditions where the effect of cellulose content did not appear to be significant. However, the cellulose content showed mainly positive correlations with tensile strength and flexural strength and elasticity, thus supporting the potential role of PS as reinforcing fibers in WPC application.

The correlation between lignin content and WPC mechanical properties was moderate, negative, or non-significant (Table 6). The non-significant and negative correlations could be explained by the fact that lignin at the fiber surface inhibits the esterification reaction, as reported by Bouafif et al. (2008). The moderate positive correlations found between lignin content and some mechanical properties were not due to a beneficial impact of lignin on the mechanical properties of the WPC, but instead to the reinforcing effect of increasing fiber content. At constant fiber proportion, the lignin content appeared to have significant negative impact on all the mechanical properties. The only two exceptions, where the amount of lignin in the WPC did not appear to be significant, were for the W_b and ε_b at the 40% proportion. One possible explanation for these exceptions is the interference between the lignin and nitrate contents in the analysis, as reported by Nadji et al. (2010). In the present study, the nitrate content in the SS has a clearly a negative effect on all investigated WPC properties (Tables 2–4).

Surprisingly, the ash content was positively correlated with tensile and flexural strength and elasticity (Table 6). This result is contradictory to what has been reported for MDF panels, where ash content showed a negative impact on strength properties (Migneault et al., 2011a). Thus, the sludge proportion in WPC was not limited by the ash content. At constant fiber proportion, the ash content had a significant positive impact on all mechanical properties. The ash found in sludge generally contains inorganic matter, which may play a similar role to that of mineral fillers used in the plastic industry.

The correlation between nitrogen content and physical properties was negative and highly significant (Table 6). The highest significant correlation (−0.73) was found for impact energy. Because nitrogen is present in the SS, this result confirms that SS did not act as a binder, contrary to expectation. The extractives content correlation coefficient was moderate to high, with a positive effect on the water absorption test. The fiber proportion in the WPC might explain this correlation, because extractives may be volatile, soluble, leachable, and present in low concentration in chemical pulp and sludge (Table 2). However, we observed a negative correlation coefficient of 0.36 and 0.41 for ε_b and W_b, respectively. This concurs with Bouafif et al. (2009), who obtained lower elongation and energy at break in WPC made with bark and white cedar fibers due to high concentrations of extractives at the surface compared to black spruce and jack pine fibers. At constant fiber proportion, the extractives content appeared to either negatively affect all mechanical properties or to be statistically non-significant.

The chemical pulping process results in cellulose fibers with better integrity and thus a higher reinforcement. In addition, the high cellulose content on the surface of chemical pulp resulted in a better affinity with MAPE. Those two explanations also applied in the present study, because sludge is composed of fibers rejected in the papermaking process.

The reinforcing effect of adding MAPE to increase the affinity in WPC is well described by Bouafif et al. (2008). Because PS has higher fiber content compared to SS, better mechanical properties is expected in WPC having higher PS content.

The negative impact of SS content on WPC properties has been observed for all experiments. A reduction of surface reaction between sludge and HDPE with increasing SS content is suspected. The hydrophilic nature of microorganisms may interfere between sludge fibers and MAPE, and 2) some by-products such as low molecular weight sugars found in the SS may inhibit the adhesion between the fibers and the polymeric matrix.

3.5. *Practical implications*

We observed increased stiffness, water swelling, and water absorption with increasing sludge content, but reduced impact energy and elongation at break, similar to the behavior of traditional wood flour filled HDPE WPC in the same range of loading (20–40% fiber proportion). Paper mill sludge WPC allows retaining much higher ash content compared to other traditional wood products such as MDF panels, which appear to have an ash content threshold at from 5 to 8% before the mechanical properties begin decreasing considerably. Therefore, it may be possible to use not only dewatered sludge but also deinking sludge and incinerated sludge as fiber sources or fillers for WPC. Incinerated sludge may redress the problem of the high energy consumption required to dry the sludge.

Recycling WPC made from paper mill sludge may become difficult, because the HDPE cannot be well separated after processing. Sludge is compostable, and an emerging generation of new compostable polymers on the market, such as polylactic acid polymer, polyhydroxybutyrate-valerate, and some aliphatic/aromatic copolyesters, could potentially replace HDPE. Adding sludge could be a way to address market entry problems due to the high cost of these new polymers compared to cheaper polyolefins such as HDPE. The compatibility between the new polymers and paper mill sludge needs to be determined, along with their processing ability.

4. Conclusion

Paper mill sludge has been proposed as a raw material alternative to wood flour in WPC manufacturing. Our results revealed that Kraft sludge contained higher cellulosic content and longer fibers, and would be the most suitable candidate for producing better composite properties compared to sludge produced by chemithermomechanical pulping and thermomechanical pulping. Increasing the sludge proportion improved the composite strength, but also increased the water sorption and thickness swelling for the three tested processes. The results confirmed the role of primary sludge as reinforcing fibers for WPC application. The secondary sludge content had a negative impact on the physical and mechanical properties of the composites.

Acknowledgements

The authors are grateful for financial support by the Canada Research Chairs Program, the Natural Sciences and Engineering Research Council of Canada (NSERC), Le Fonds de Recherche sur la Nature et les Technologies du Québec (FQRNT), and FPInnovations. The authors would also like to thank the three pulp mills for providing the materials (White Birch Division of Stradacona in Québec City, Lac-Saint-Jean Abitibi-Bowater in Dolbeau-Mistassini, and SFK Pulp in St-Félicien).

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