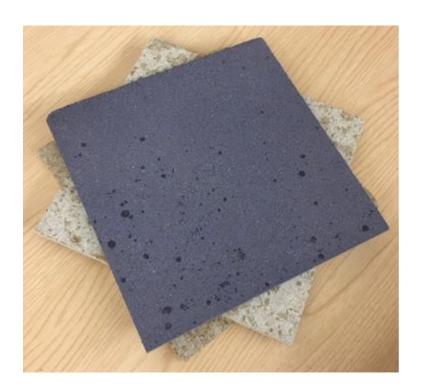






Report

$Composite\ panels\ produced\ from\ paper\ mill\ sludge$



Research title: Designing a circular business model from industrial by-products: A case study on

paper mills.

Researcher: Cynthia Adu

Academic Mentors: Professor Mark Jolly, Professor Stephen Eichhorn

Industrial Mentor: Tom Everitt

Date: 11/09/2019

Paper mill sludge is composed of fibres and minerals which can be used to produce composite panels. PMS has been used in various studies to substitute virgin fibres for medium density fibre (MDF) boards and wood plastic composites (WPC). In the literature, formaldehyde and Isocyanate based binding agents were used as they provide fast curing, improve internal bonding and are low cost. However, urea formaldehyde resins critically influence human health and indoor air quality, due to volatile organic compound emissions. To improve the value offered for PMS composite panels, a bio-based resin produced from the by-products of distillers dried grains (DDGs) was used to prepare boards from PMS by hot-pressing.

The composite panels were made from sludges obtained from mills UPH, JCR, LCR and MIX (mixture of sludges for 3 similar mills PDE, CTD and BRW). The panels tested for their modulus of elasticity, modulus of rupture, internal bonding and thickness swelling. The respective panels resulted in a MOR of 32.4 \pm 2.5, 23.5 \pm 3.3, 8.4 \pm 0.9 and19.5 \pm 2.4 N/mm². The DDGs based bio-adhesive was very compatible with the UPH fibre rich sludge which resulted in the highest internal bonding 1.8 \pm 0.2 N/mm². However, there were shortcoming for the panel due to water absorption of the fibres leading to increased thickness swelling. The DDGs resin wasn't suitable for deinking sludge LCR which had high inorganic minerals and fine particle size (200 mesh). The LCR panel resulted in brittle failure with low MOR at 8.4 \pm 0.9 N/mm². The study showed that composite panels prepared from UPH paper sludge could be used in load bearing and non-load bearing applications in dry conditions. JCR and MIX were limited by their internal bonding allowing them suitable to only underlay roof/wall board applications.

List of figures

Figure 1: Fibre board preparation process
Figure 2: TGA curve of paper mill sludge samples
Figure 3: Before (left) and after (right) disintegration of agglomerated PMS7
Figure 4: Aluminium mould with sliding frame and outer case (Left) and mould with PMS 8
Figure 5: Panel cutting template9
Figure 6: Specimen set up for 3-point bending test9
Figure 7: Internal bond test set up10
Figure 8: Appearance of composite panel from PMS11
Figure 9: Stress strain curve of PMS composite panels from 3-point bending test12
Figure 10: Structural properties of PMS panels showing MOR, MOE and IB (N/mm ²)12
List of tables
Table 1: Mechanical properties of paper mill sludge in composite panels
Table 2: Chemical composition of paper sludges used for composite panels6
Table 3: Mechanical properties of PMS panels compared with technical requirements of

1 Introduction

Paper mill sludge is a by-product of paper mill waste water treatment process produced at high volumes up to 1 million tonnes per annum in the UK. Efforts to utilise PMS have been shown in the literature ranging from land applications, energy recovery and integration in materials (Faubert *et al.*, 2016). One way to extend the life cycle of PMS is for manufacturing composite panels that can be used as building, furniture and construction materials. Composites panels range from medium density fibre (MDF) boards, particle boards, mineral boards, hardboard, plasterboard etc. Such products are embedded into buildings and can last up to 20 years. This could reduce the embodied energy or carbon footprint of paper mill process, if their by-product can be integrated in building materials.

PMS has been used in the literature to produce panels following similar methods used in production of medium density fibreboard or particle boards as shown in figure 1. PMS and wood chips are dried then milled and screened to obtain small particle sizes of 2-3 mm. Resin is added to the particle and formed into mats. The formed mats are pressed at temperatures between 150-210 °C and pressure of 0.5-5MPa, usually for ≤ 10 min to allow resin cure. The pressed panel are left to cool and finished by trimming of edges or sanding if required.

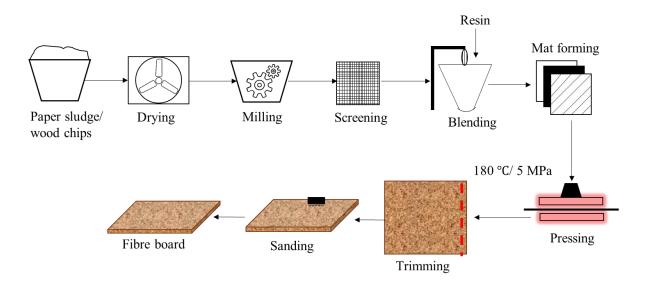


Figure 1: Fibre board preparation process.

Primary sludges (PS) containing low ash (< 30%) and deinking sludge (DS) with high ash content (> 30%) have been used to produced PMS panels. Taramian et al., (2007) was one of the earliest studies to produce PMS panels by binding low ash PMS (28%) and wood chips

with urea formaldehyde (UF). The research concluded that 15 % of wood chips could be substituted with PMS to meet ANSI A208.1-2009 for general use of particle board. UF resin is widely used in particle board, medium density fiber board, plywood manufacturing. However, panels bonded with UF resins emit formaldehyde which significantly affect indoor air quality and health (Pilidis *et al.*, 2009). Although PMS panels may not meet load bearing applications, the use of bio-based adhesive in their manufacture may offer a competitive indoor product. Thus, the research investigates the use of bio-based resin produced from distillers' dry grains and soluble as a binder in PMS boards.

1.1 PMS panels in literature

Geng, Zhang and Deng (2007) used DS and PS to substitute spruce pine fibres showing that 70 % of spruce pine fibres can be replace with PS and 70 % with DS to meet ANSI A208.1-2009 for interior applications. The relationship between panel properties and processing parameter showed significant effect caused by temperature, pressing time and panel density. Modulus of rupture is improved at higher pressing temperature and increased panel density. Similarly, thickness swelling of the panels reduced 50 % for pressing temperature of 210 °C compared to standard 180 °C. Other studies followed similar approach using 10-12% urea formaldehyde (UF) to prepare panels from PMS (Geng, Deng and Zhang, 2006; Geng, Zhang and Deng, 2007; Goroyias, Elias and Fan, 2004; Xing et al., 2013). Panels are tested for their bending strength or modulus of rupture (MOR), flexural strength or modulus of elasticity (MOE), internal bonding (IB) and thickness swelling (TS). Most studies found that an increase in the PMS content negatively affect the panel modulus of rupture (MOR) and modulus of elasticity (MOE). Moreover, the high mineral content of DS and short fibers present a challenge in substitution of materials in particle boards. Optimal parameters recommended for PMS panels were density of 1000 kg/m³, 210 °C and 8 mins curing time.

PMS have also been used to prepare panels without any binders, instead secondary sludge containing microbes and proteins were used as binders (Migneault et al., 2011). Secondary sludge (50%) with higher lignin content improved internal bonding of panels when used as a binder. At pressing temperature 210 °C the lignin softens and acts as a crosslinker. Apart from pressing, PMS panels have been produced by extrusion. Scott et al (2000) used

mixture of PMS and old newsprints to extrude panels of 3mm thickness for hardboards. A wet sheet forming process was employed by Tikhonova, Lecourt and Irle (2014) whereby webs/sheets of blended fibres were layered and hot pressed to produce panels. This study showed significantly improved panel properties for a 50:50 ratio of PMS and pulp. The MOR, MOE and TS of the wet formed panels were 45 MPa, 6179 MPa and 65 %. The table below summarises properties of panels made from PMS.

Table 1: Mechanical properties of paper mill sludge in composite panels

Description and source	MOR	MOE	IB	TS	Density
	(N/mm²)	(N/mm²)	(N/mm²)	(%)	(kg/m³)
(Goroyias, Elias and Fan, 2004) WRAP					
MDF: 15 % UF, 45 % PMS, 40 % fibre	26.1	2892	0.5	11.8	995
Cement board: 30 % PMS, 70 % cement	11.9	10882	1	0.8	1618
Tile 80 % sludge, 20 % MDI Isocyanate	25	3000	-	4	1200
(Geng, Deng and Zhang, 2006)					
PS: SS 80:20 @ 210 °C 6 min	2.8	506	0.13	33.5	750
PS: SS 80:20 @ 210 °C 8 min	13.5	2165	1.25	24.3	1100
PS: SS 80:20 @ 180 °C 8 min	8.9	1770	0.29	51.7	1100
PS: SS 80:20 @ 180 °C 8 min	2.4	372	0.03	59.0	750
PS: SS 80:20 @ 210 °C 8 min	3.4	652	0.21	24.8	750
PS: SS 80:20 @ 210 °C 6 min	11.6	1998	1.00	27.4	1100
PS: SS 80:20 @ 180 °C 6 min	2.2	332	0.04	59.8	750
PS: SS 80:20 @ 180 °C 6 min	9.2	1554	0.25	75.2	1100
(Taramian et al., 2007)					
PMS: WC 15:85 10 % UF	12	-	-	15.5	-
PMS: WC 30:70 10 % UF	10	-	-	16.2	-
PMS: WC 45:55 10 % UF	7	-	-	16.6	-
PMS: WC 15:85 12 % UF	13.5	-	-	14.7	-
PMS: WC 30:70 12 % UF	9	-	-	14.5	-
PMS: WC 45:55 12 % UF	7	-	-	15.5	-
(Geng, Zhang and Deng, 2007)					
PS: SPF 3:7 12 % UF	25	2100	1	25	950
PS: SPF 7:3 12 % UF	18	1500	1.2	24	950
SS: SPF 3:7 12 % UF	10	1700	0.7	24	950
SS: SPF 7:3 12 % UF	23	1600	0.7	20	950
100 % PS 12 % UF	10	800	0.8	19	950

100 % SS 12 % UF	3	300	0.3	18	950
(Migneault et al., 2011) no binder					
SS:PS (1:9) TMP	11.4	2.03	0.96	50	1048
SS:PS (2:8) TMP	11.6	1.96	1.19	34	1105
SS:PS (3:7) TMP	14.4	2.58	1.29	26	1133
(Migneault et al., 2011) no binder					
SS:PS (1:9) CTMP	15.0	2.57	0.86	39	1085
SS:PS (2:8) CTMP	16.0	2.74	1.61	32	1126
SS:PS (3:7) CTMP	13.4	2.36	1.63	32	1106
(Migneault et al., 2011) no binder					
SS:PS (1:9) Kraft	6.7	1.10	0.06	245	925
SS:PS (2:8) Kraft	5.6	0.97	0.07	153	985
SS:PS (3:7) Kraft	4.0	0.60	0.07	187	951

^{*} PS (Primary sludge), WC (wood chips), SS (Secondary sludge), SPF (Spruce pine fibres), TMP (Thermochemical pulping), CTMP (Chemi-thermochemical pulping).

Some of the panels produced with mixtures with paper mill sludge and other materials met with BS EN 622-3:2004 requirements for medium boards. In humid conditions, panels \leq 10 mm in thickness, can be used for general purpose if they meet specification of \geq 18 N/mm² MOR, \geq 6 N/mm² MOR after boil test, 0.30 N/mm IB and \leq 12% TS (BSI, 2004). Meanwhile, general purpose boards \leq 10 mm for used in exterior conditions are required to meet the same parameters including MOE \geq 2200 N/mm². Load bearing boards \leq 10 mm for use in dry conditions are required to meet \geq 18 N/mm² MOR, 0.10 N/mm IB and \leq 15 % TS (BSI, 2004). Although the structural properties (MOR, MOE and IB) of some PMS boards are up to the specifications, they are disadvantage by the high thickness swelling. This is because high moisture absorbance of cellulose fibres and UF. UF is a cheap binder source however it is susceptible to hydrolytic degradation in the presence of moisture. Waxes have been used to counteract moisture absorption and addition of deinking sludge with high mineral content. Sludge from craft pulping process is found to be more prone to moisture absorption compared to CTMP and TMP pulping process.

1.2 Adhesives in composite panels

Adhesives are essential components in composite panel manufacturing; it determines structural properties, moisture absorption/resistance and processing time. Ureaformaldehyde was developed in the early 1930s as a cheap resin for large scale production of composite panels, required to meet increasing demand. 90% of MDF panels are produced with UF binders. UF is desirable for its water solubility, colourlessness, low flammability and fast curing. However, increasing legislation on formaldehyde due to emissions and environmental impact lead to the move to formaldehyde free resins such as MDI (4,4-diphenylmethane diisocyanate) which is more moisture tolerant compared to UF. MDI bonds with hydroxyl groups which also increases bond strength (Ormondroyd, 2015). Although MDI is non-toxic and does not emit any volatile organic compounds (VOCs) when cured, it has a very high toxicity in gaseous or droplet form (Vangronsveld, 2012). The monomeric substances in adhesives are crosslinking agents which promote intermolecular bonds between polymer chains. MDI is marketed as a formaldehyde-free adhesive because it is not a formaldehyde derivative. However, it can still release formaldehyde during decomposition.

Crosslinkers such as polymeric methylene diphenyl diisocyanate (pMDI) has been used in particleboard as early as 1971, it is considered formaldehyde-free as the aldehyde cannot be released (Solt et al., 2019). There is still significant interest in developing bio-based adhesives from proteins. Traditional adhesives from the 20th century were made from proteins (Frihart, 2015). Protein adhesives are derived from plant proteins; soy, oilseeds, wheat gluten and, animal protein; collagen, casein, blood. Other sources of bio-based adhesives are plant oils such as castor oil, soybean oil and castor oil. Bio-based adhesive are non-toxic and can even be obtained from industrial by-products. For example, Fernandes et al., (2017) used waste vegetable oils from restaurants as epoxy resins and Liu et al., (2018) bonded wood composite panels with cotton seed meal, a by-product of cotton oil extraction.

Ethanol distillation process generates distillers dry grains and soluble (DDGs). DDGs is a by-product used as animal feed, it contains high plant protein. The DDGs was developed into an adhesive knowns as DIGLUE™, patented and sold by Cambond, UK (Zhao, 2015). Zhao et al. (2017) produced wood composite panels with DIGLUE using Low contents of pMDI 1-3% as a crosslinker. The pMDI forms a water-resistant polymeric network between the DDGs

to create a strong internal bond. Different DIGLUE formulations were used to produce wood composite panels for comparisons with urea formaldehyde. All panels with Diglue resulted in lower thickness swelling and water absorptions compared to UF panels. Some Diglue formulation also resulted in higher MOE and MOR than the UF panels.

1.3 Raw materials

Diglue was provided by CAMBOND, UK. It appears light brown in powder form with particle size ≤ 100 mesh (≤ 0.149 mm). Crosslinking agent pMDI Suprasec 5025 was supplied by Huntsman, UK . PMS used in this study were primary sludge from UPH mill and JCR mill, deinking sludge from BRW, PDE, CTD and LCR. 4 types of composites were made using UPH, JCR, LCR and equal portions of PDE, BRW and CTD referred to as MIX. The chemical compositions of the samples are given below.

Table 2: Chemical composition of paper sludges used for composite panels

Sample	Ash	Cellulose	Extractives	Total organics	TGA mass loss
	(%)	(%)	(%)	(%)	(%)
UPH	5 ± 2	61.2	16.5	66.2	68.5
JCR	27 ± 2	47.7	10.7	51.2	48.9
LCR	70 ± 5	13.2	12.3	16.3	21.1
MIX					
-PDE	65 ± 2	11.6	10.9	14.1	32.6
-BRW	64 ± 8	15.3	11.9	19.0	22.5
-CTD	69 ± 2	8.7	12.6	11.3	21.3

The panels were pressed separately based on their chemical composition. UPH sludge has very low ash content with predominantly long fibres and fine polyethylene JCR had low ash content compared to deinking sludges. Sludge from LCR was the highest ash content and had the least mass loss during TGA, it contained inorganic minerals compounds CaO, SiO_2 and Al_2O_3 in higher proportions. Thus, it was used for mineral composite panel. The mixture of

PDE, BRW and CTD was due to their similarity in chemical composition. The full chemical composition of the sludges can be found in submission 2 of the portfolio. The thermogravimetric analysis (TGA) curve is show below, the first degradation around 100 °C I moisture, 2nd degradation around 300 °C is the mass loss of organic compounds.

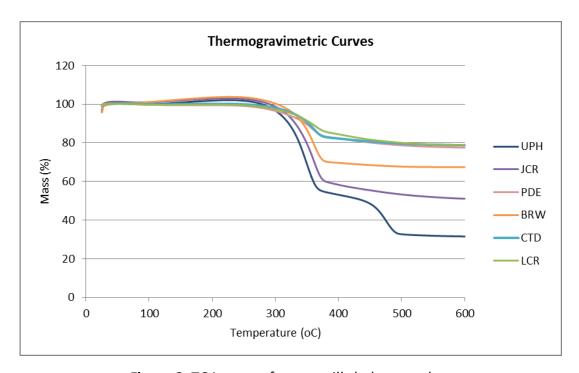


Figure 2: TGA curve of paper mill sludge samples.

This provides an indication of the suitable temperature during panel pressing to avoid thermal degradation. The PMS was dried which made it agglomerate into flakes (figure 3 left) thus an electric cereal grinder (CGoldenwall, China) was used to disintegrate the fibres at 28000 rpm and 20 second as shown if figure 3.



Figure 3: Before (left) and after (right) disintegration of agglomerated PMS.

The panel target panel was 1.2 g/cm³. The panel was made from 380 g of PMS, diglue was 18 % of PMS (70 g) and pMDI 7 % (26 g) of PMS and Diglue. Water content used was 15 % (60 g) of dry mater. Samples were prepared by dissolving the 70 g of Diglue in 60 g of water stirred at 800 rpm for 30 mins, the 26 g of pMDI was added to the Diglue and stirred for another 30 mins to form the adhesive. The adhesive mixture was poured into the PMS fibres and mixed using a hand electric drill. The mixture was distributed evenly into the mould using a trowel. An aluminium mould with a cubic piston was used. The mould was available from another Cranfield student project producing MDF panels (Aluvihare gedara, 2019). The mould is made up of 3 parts, an outer case, sliding frame and piston to sit inside the frame. The volume of the frame is 210 x 210 x 40 mm (L x W x H). The sliding frame was inserted and aligned with the outer frame before the mixture is poured in as shown in figure 4. Chemlease release agent was applied on the mould (Chemtrend, UK) before the mixture is poured in. The release agent prevents adhesion of the PMS mixture to the mould, provides ease of removal and prevents wearing.

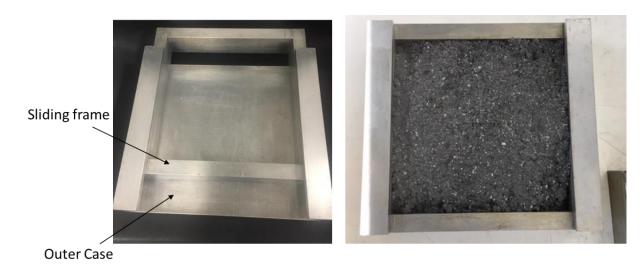


Figure 4: Aluminium mould with sliding frame and outer case (Left) and mould with PMS.

In industrial panel production, 12-15% of UF is used, a frame is used to form the mat after which it is removed for pressing. Mat-forming was not possible for PMS as the bio-resin had very low tack compared to when UF. UF provides tack to the material and keeps it in shape before pressing. Hence, the mould and piston was used to limit the material flow and provide a homogenous sample distribution during pressing. The sample was pressed at 180 °C at 4 MPa using a hydraulic press with heated plated.

2 Characterisation methods for PMS

Panels were cut for testing as shown in figure 5, in accordance with BS EN 326-1:1994 Wood based panels sampling, cutting and inspection (BSi, 1994).

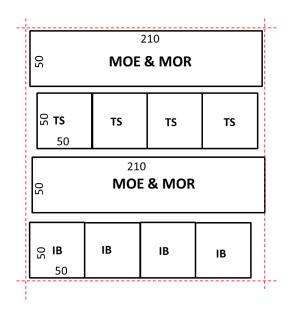


Figure 5: Panel cutting template.

The cut specimen was tested for their flexural strength, internal bond and thickness swelling. Where possible, the study followed technical British standards. The bending strength or modulus of rupture (MOR) was measured following BS EN 310-1993 Wood-based panels determination of modulus of elasticity in bending and of bending strength (BSi, 1993a). The standard uses a three-point bend test with 15 \pm 0.5 mm roller-bearing and 30 \pm 0.5 mm cylindrical loading head.



Figure 6: Specimen set up for 3-point bending test.

The dimensions of the cylindrical loading head in figure 6 was 15 \pm 0.5 mm as the appropriate loading head in the test standard was unavailable. 5 kN load cell was used at a constant loading rate of 2mm/min. MOR in N/mm² was calculated using equation 1.

MOR (N/mm²) =
$$\frac{3 F_{max} l_1}{2 h t^2}$$
 equation 1

 F_{max} is the maximum load in Newtons, I_1 is the distance between the centre of the rollers in mm (160mm), t is the thickness of the specimen and b is the width (50 mm). MOE was calculated using equation 2;

MOE (N/mm²) =
$$\frac{l_1^3 (F_2 - F_1)}{4 bt^3 (a_2 - a_1)}$$
 equation 2

 (F_2-F_1) is the difference between the maximum and minimum load from the straight line portion of the stress-strain curve. Whilst, (a_2-a_1) is the deflection of the sample. The load was applied at a constant rate of 2 mm/min.

Internal bond is used to measure the bonding strength of the specimen. The study followed the test standard BS EN 319:1993 Particleboards and fibreboards determination of tensile strength perpendicular to the plane of the board (BSi, 1993b). The method requires adhering test pieces onto metal blocks loaded in tension, perpendicular to the plane of the panel as shown in figure 7.

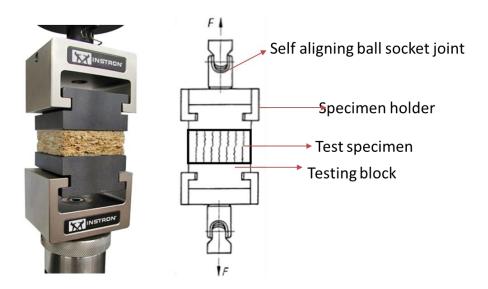


Figure 7: Internal bond test set up.

The internal bond is by dividing the maximum force by the area of the test specimen, in N/mm².

Thickness swelling of specimen was measure in accordance with BS EN 317:1993 Particleboards and fibreboards- Determination of swelling in thickness after immersion in water (BSi, 1993c). The method requires full immersion of test piece in clean still water with pH 7 \pm 1 at temperature of 20 \pm 1 °C for 24 hours. The thickness of the specimen is measured using a micrometre gauge before and after immersion in water. The result is reported as a percentage of increase in thickness.

3 Results and discussion

The appearance of the composite panels is shown below in figure 8. The black/dark blue dots on the panel are due to poor dispersion and moisture from the resin. This similar effect was also found in panels from was found in MDF panels containing cottonseed meal bio-resin (Liu et al., 2018).

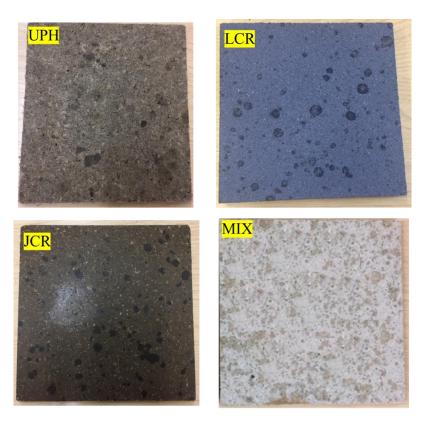


Figure 8: Appearance of composite panel from PMS.

Attempts to improve dispersion of the resin were made by using a spray bottle. However, the viscosity of the Diglue + pMDI mixture limited spraying. The panels appeared dimensionally stable and easy to cut. The stress strain curves of the boards from the three-point bend test is shown below in figure 9.

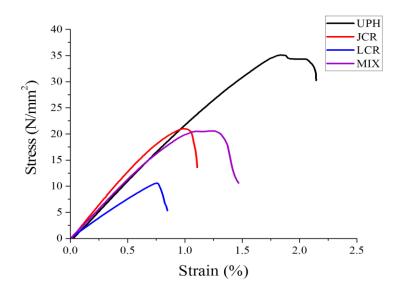


Figure 9: Stress strain curve of PMS composite panels from 3-point bending test.

The UPH, JCR and MIX panels showed similar stress-strain curve although UPH reached a higher stress and strain \sim 35 MPa and 2.2 %. UPH panel contained mostly organic materials which easily bind with the bio-resin. The polyethylene in the UPH panel also forms an interfacial bonding between the fibres, similar to wood plastic composites with paper mill sludge/wood fibres containing polyethylene (Soucy et al., 2014; Turku et al., 2017).

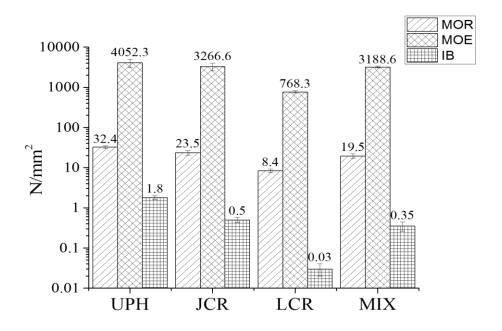


Figure 10: Structural properties of PMS panels showing MOR, MOE and IB (N/mm²).

The lignin content in the UPH sludge (< 2%) also plays a role in binding the cellulose fibrils to allow efficient stress transfer. The mechanical properties of JCR panel were lower

than UPH, although JCR is a low ash sludge it still contains higher inorganic minerals compared to UPH. This may have limited the internal bonding with the bio-resin. The LCR panel exhibited brittle failure at $\sim 10~\text{N/mm}^2$. The high mineral content in LCR and fine particle size of LCR limited the adhesives bonding. The DIGLUE binder was not suitable for LCR due to its high ash (70 \pm 5 %) content. The properties of the mix sludge were slightly lower than JCR. The thickness swelling of the composite panels after 24 h absorption in water were 17.5 \pm 2.8, 14.4 \pm 1.3, 6.5 \pm 0.3 and 3.4 \pm 0.5 for UPH, JCR, LCR and MIX. The thickness swelling is affected by the presence of lumens, fine pores and interfacial gaps and micro-cracks in the PMS-resin matrix. In humid conditions the panels have potential to take up water due to hydroxyl group interaction with water molecules. UPH had the highest thickness swelling as it contained a higher percentage of fibres. The mechanical properties of the panels are compared with requirements for various purposes based on technical standards in table 3.

Table 3: Mechanical properties of PMS panels compared with technical requirements of various products.

	MOR	MOE	IB	TS
	(N/mm ²)	(N/mm ²)	(N/mm²)	(%)
UPH	32.4 ± 2.5	4052 ± 908	1.8 ± 0.2	17.5 ± 2.8
JCR	23.5 ± 3.3	3266 ±713	0.5 ± 0.07	14.4 ± 1.3
LCR	8.4 ± 0.9	768 ± 52	0.03 ± 0.01	3.4 ± 0.3
MIX	19.5 ± 2.4	3189 ± 147	0.35 ± 0.09	7.6 ± 0.5
BS EN 622-5:2009	2	2	2	≤
-Non-load bearing dry	23	2700	0.65	17
-Non-Load bearing humid	27	2700	0.80	12
-Load bearing dry	29	3000	0.70	17
-Load bearing humid	34	3000	0.80	12
-Underlay roof/wall	14	1600	0.30	10

Although the UPH panels met the required MOR, MOE and IB for some panel applications, they were limited by thickness swelling. The UPH panels are suitable for in non-load bearing dry conditions, load bearing dry conditions and underlay roof/wall. JCR and MIX

were limited by their internal bonding allowing them suitable to only underlay roof/wall board applications. Maleic anhydride coupling agents have been used to strengthen reinforcement in the fibre matrix which can improve overall mechanical properties and act as a hydrophobic agent to reduce water absorption (Keener, Stuart and Brown, 2004; Valente et al., 2017). This may present an opportunity for some of the panels to be used in humid conditions. The LCR panel had very low thickness swelling due to inorganic minerals however the material does not meet any of the requirements for MDF according to BS EN 622-5:2009.

4 Conclusion

Paper mill sludge has been proposed as a substitute in composite panels such as medium density fibre boards and wood plastic composites. PMS panels can be limited by mechanical properties compared to products using virgin wood fibres. However, most products are bonded with urea formaldehyde adhesive, this is a growing concern due to interest in indoor air quality. To propose an environmentally competitive product, PMS panels were produced with a bio-adhesive from ethanol distiller by-product. The four PMS panels made were primary sludge in a virgin mill (UPH), primary sludge from coffee cup recycling mill (JCR), deinking sludge (LCR) and mixture of primary and deinking sludges from recycling mills (MIX). The UPH sludge showed the highest mechanical properties for MOR, MOE and IB however it was limited by its thickness swelling. Allowing it to be mainly used in dry conditions for load bearing, non-load bearing and underlay roof/wall applications. The JCR and MIX panel had low thickness swelling due to inorganic mineral content however their mechanical properties were lower than UPH sludge making them only suitable for underlay roof/wall. The LCR panel made from deinking sludge with high ash content showed very low thickness swelling (3.4 ± 0.3). However, the bio-adhesive is not a suitable binder for the sludge, this led to brittle failure and poor internal bonding. Nonetheless, the low thickness swelling of LCR panel may be suitable for flooring applications if a cementitious based binder is used. Other methods of producing PMS panels could be investigated such as extrusion which could lead to increased mechanical properties whilst thickness swelling can be improved by wax and surface treatment. There is also possibility of using some of the LCR deinking sludge in the UPH sludge with the potential of reducing it's thickness swelling.

Acknowledgements

The research was part-funded by the Engineering and Physics Research Science Council UK (Grant Number EP/L016389/1) and Ecoganix LTD. The The authors would also like to thank the following companies for their contribution; CAMBOND LTD for provision of bioresin and Huntsman, UK for provision of pMDI and the associated paper mill for providing sludge samples and data.

References

BSi (1994) 'Wood-based panels — Sampling, cutting and inspection —Part 1: Sampling and cutting of test pieces and expression of test results.'

BSi (1993a) 'BS EN 310:1993 Wood-based panels — Determination of modulus of elasticity in bending and of bending strength', , pp. 1–14.

BSi (1993b) 'BS EN 319:1993 Particleboards and fiberboards – Determination of tensile strength perpendicular to the plane of board.'

BSi (1993c) 'BS EN 317:1993 Particleboards and fibreboards- Determination of swelling in thickness after immersion in water', , pp. 1–10.

BSI (2004) BS EN 622-3:2004 Fibreboards Specifications Part 3: Requirements for medium boards.

Faubert, P., Barnabé, S., Bouchard, S., Côté, R. and Villeneuve, C. (2016) 'Pulp and paper mill sludge management practices: What are the challenges to assess the impacts on greenhouse gas emissions?', *Resources, Conservation and Recycling*, 108 Elsevier B.V., pp. 107–133.

Fernandes, F.C., Kirwan, K., Lehane, D. and Coles, S.R. (2017) 'Epoxy resin blends and composites from waste vegetable oil', *European Polymer Journal*, 89(November 2016) Elsevier, pp. 449–460.

Frihart, C.R. (2015) 'Introduction to Special Issue: Wood Adhesives: Past, Present, and Future', Forest Products Journal, 65(1–2), pp. 4–8.

Geng, X., Deng, J. and Zhang, S. (2006) 'Effects of hot-pressing parameters and wax content on the properties of fiberboard made from paper mill sludge', *Wood and fiber science*

Geng, X., Zhang, S.Y. and Deng, J. (2007) 'Characteristics of Paper Mill Sludge and Its Utilization for the Manufacture of Medium Density Fiberboard', *Wood and Fiber Science*, 39(2), pp. 345–351.

Goroyias, G., Elias, R. and Fan, M. (2004) *Research into using recycled waste paper residues in construction products*.

Keener, T.J., Stuart, R.K. and Brown, T.K. (2004) 'Maleated coupling agents for natural fibre

composites', Composites Part A: Applied Science and Manufacturing, 35(3), pp. 357–362.

Liu, M., Wang, Y., Wu, Y., He, Z. and Wan, H. (2018) "Greener" adhesives composed of ureaformaldehyde resin and cottonseed meal for wood-based composites', *Journal of Cleaner Production*, 187 Elsevier Ltd, pp. 361–371.

Migneault, S., Koubaa, A., Riedl, B., Nadji, H., Deng, J. and Zhang, T. (2011) 'Binderless Fiberboard Made From Primary and Secondary Pulp and Paper Sludge', *Wood and Fiber Science*, 43(2), pp. 180–193.

Ormondroyd, G.A. (2015) Adhesives for wood composites. Elsevier Ltd.

Pilidis, G.A., Karakitsios, S.P., Kassomenos, P.A., Kazos, E.A. and Stalikas, C.D. (2009) 'Measurements of benzene and formaldehyde in a medium sized urban environment. Indoor/outdoor health risk implications on special population groups', *Environmental Monitoring and Assessment*, 150(1–4), pp. 285–294.

Scott, C.T., Simonsen, J., Klingenberg, D. and Zauscher, S. (2000) 'Pulp extrusions incorporating sludge from the pulp and paper industry', *TAPPI Proceedings - Environmental Conference and Exhibit*, 1, pp. 203–211.

Solt, P., Konnerth, J., Gindl-Altmutter, W., Kantner, W., Moser, J., Mitter, R. and van Herwijnen, H.W.G. (2019) 'Technological performance of formaldehyde-free adhesive alternatives for particleboard industry', *International Journal of Adhesion and Adhesives*, 94(May) Elsevier Ltd, pp. 99–131.

Soucy, J., Koubaa, A., Migneault, S. and Riedl, B. (2014) 'The potential of paper mill sludge for wood-plastic composites', *Industrial Crops and Products*, 54, pp. 248–256.

Taramian, A., Doosthoseini, K., Mirshokraii, S.A. and Faezipour, M. (2007) 'Particleboard manufacturing: an innovative way to recycle paper sludge.', *Waste management (New York, N.Y.)*, 27(12), pp. 1739–46.

Tikhonova, E., Lecourt, M. and Irle, M. (2014) 'The potential of partial substitution of the wood fibre in hardboards by reject fibres from the paper recycling industry', *European Journal of Wood and Wood Products*, 72(2), pp. 177–184.

Turku, I., Keskisaari, A., Kärki, T., Puurtinen, A. and Marttila, P. (2017) 'Characterization of wood plastic composites manufactured from recycled plastic blends', *Composite Structures*, 161 Elsevier Ltd, pp. 469–476.

Valente, M., Tirillò, J., Quitadamo, A. and Santulli, C. (2017) 'Paper fiber filled polymer. Mechanical evaluation and interfaces modification', *Composites Part B: Engineering*, 110, pp. 520–529.

Vangronsveld, E. (2012) 'Methylene diphenyl diisocyanate (MDI): EU classification; Impact of REACH; Worker and consumer exposure-perception versus facts', *International Wood Products Journal*, 3(1), pp. 58–63.

Xing, S., Riedl, B., Deng, J., Nadji, H. and Koubaa, A. (2013) 'Potential of pulp and paper secondary sludge as co-adhesive and formaldehyde scavenger for particleboard manufacturing', *European Journal of Wood and Wood Products*, 71(6), pp. 705–716.

Zhao, X. (2015) DDGS patent.pdf.

Zhao, X. (2017) 'Biomass-Based Formaldehyde-Free Bio-Resin for Wood Panel Process', Handbook of Composites from Renewable Materials, 2013, pp. 129–149.