Combining Cellulosic Ethanol Fermentation Waste and Municipal Solid Waste-derived Fiber with a Kraft Black Liquor-derived Binder for Recycled Paper Making

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Rice straw cellulosic ethanol fermentation waste (CEFW) and municipal solid waste derived fiber (MSWF) were used as alternative fibers for recycled paper making. The fibers were mixed with old newspaper (ONP) fiber at different mass ratios to produce standard recycled papers and paperboards. A "green" adhesive binder derived from kraft black liquor (BLDB) was used to improve the physical properties of the waste-derived paper products. The values of these properties increased linearly with increasing average fiber lengths, regardless of the type of fiber used in the products. BLDB improved the physical properties of the products by 50% for papers and 85% for paperboards, and the performance of this binder was comparable to a commercial urea formaldehyde resin binder. Thermal pressing, however, did not improve the physical properties of the binder-enhanced paper products. With the addition of the adhesive binder, CEFW and MSWF showed reasonable substitution potential for ONP fiber by providing suitable tensile and bursting strength in the recycled paper products. The critical fiber length, which produced the minimum strength properties for the recycled paper products, was approximately 1020 μm.

Keywords: Municipal solid waste; Bioethanol fermentation waste; Recycled paper; Lignocellulosic biomass; Biorefinery

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INTRODUCTION

Developing biorefinery techniques to convert waste-derived lignocellulosic fiber into biofuels and value-added products is a sustainable approach for substituting fossil fuel products and mitigating global climate change (Limayem and Ricke 2012). Many pretreatment and bioconversion techniques have been developed to handle different types of lignocellulosic biomass through the well-known sugar platform (Hendriks and Zeeman 2009), *i.e.*, agriculture residues (Alvira *et al.* 2010) and forestry residues (Leu et al. 2013; Zhu et al. 2009). Some such techniques have been successfully utilized in pilot-scale applications(Zhou et al. 2013). However, because of the recalcitrance of the lignocellulosic biomass to hydrolysis, the economic barriers to the success of a

biorefinery still exist, preventing full-scale application of these processes (Sims *et al.* 2010). Therefore, a key concept for developing a biorefinery is to develop suitable outlets for all of the products to completely utilize the feedstock. Cellulosic ethanol fermentation waste (CEFW) recovered from the slurry after saccharification and fermentation processes, is an example of a lignocellulosic by-product that can be recovered from a biorefinery for beneficial uses.

Furthermore, diverting resources from municipal solid waste (MSW) is a new subject of consideration in supporting sustainable urban development. MSW often still contains a considerable amount of useful components before incineration or final disposal. The disposed MSW in landfill sites in the U.S., for example, contains approximately 14.8% paper and paperboard wastes and 17.2% wood and yard wastes. In 2011 the total amount of lignocellulosic biomass discarded was approximately 52.5 Mt (USEPA 2013). The fraction of the lignocellulosic fiber recovered may not increase through the existing recycling policy and technology because of difficulties in recycling some of the components. In Taiwan, MSW before incineration contained approximately 40% lignocellulosic product wastes even after mandatory recycling and sorting policies were conducted (Hung *et al*. 2012). Developing new processes to extract fiber from MSW could be useful in further recovering these resources and achieving "zero-waste" production to promote sustainable waste management. Extending the service life of the lignocellulosic fibers can also improve carbon sequestration (Heath *et al*. 2011).

Paper and paperboard are two possible products of the waste-derived fibers. Virgin papers contain different additives, *e.g.*, surface coating agents, fillers, and/or paper strength agents, which can enhance the physical strength of the products or improve the processability of paper products. Some papers and related products, *e.g.*, food containers, package papers, and paper towels, are specially treated and are not suitable for making recycled paper. These products are often disposed of directly in landfill sites or sent for incineration after simple uses. Pretreatment is often needed for separation of the lignocellulosic fibers and inorganic components before further use of these paper-related products.

Mechanical heat treatment (MHT), or autoclaving, is an applicable process for MSW pretreatment (Chang *et al.* 2011; Eley *et al.* 1995). MHT can be implemented with incinerators to reduce energy consumption and operation costs. The excessive steam generated by the incinerators can be used for autoclaving, and the non-recyclable residues can be directly incinerated for energy recovery after MHT. During MHT, plastics are softened or shrunk, while the physical condition and dimensions of most of the lignocellulosic fibers and metals or other inorganic matter are maintained. The compostable or fermentable organics can be separated from the MSW after MHT and fractionation. MHT was shown to be applicable in mixing with old corrugated containers fibers (Ashley and Hodgson 2003; Ashley and Hodgson 2004), improving the recyclability of wastepaper (Hung *et al*. 2012), wasted bamboo chopsticks (Hung *et al.* 2013), and poly(ethylene terephthalate) (PET) plastics (Holtman *et al*. 2012). Autoclave-pretreated MSW was also used to produce second-generation biofuels (Antizar-Ladislao and Turrion-Gomez 2008; Shi *et al*. 2009), and bioenergy (Antizar-Ladislao and Turrion-Gomez 2008), and it has been applied in the production of fermentable sugars after hydrolysis (Li *et al*. 2012) and the generation of biogases (García *et al*. 2012).

In this study, two waste-derived lignocellulosic fibers were used with old newspaper (ONP) fiber for recycled paper making. The first fiber was a CEFW fiber

derived from steam-exploded rice straw after simultaneous saccharification and fermentation (SSF) and solid-liquid separation. The second fiber was a municipal solid waste-derived fiber (MSWF). An adhesive binder derived from kraft black liquor (KBL) was developed and used in the paper making processes to enhance the physical properties of the paper products, and its performance was compared to a commercial urea-formaldehyde (UF) resin binder. Standard paper handsheet samples were produced, and their mechanical properties, *i.e.*, tensile, bursting, and tearing strengths, were evaluated. Fiber analysis was performed to investigate the relationship between the characteristics of the waste-derived fibers and the properties of the paper products. The results of this study can be used to evaluate the feasibility of adopting MHT processes as an operation in MSW treatment and management. New applications of the recycled KBL and additional benefits of the second-generation biofuel biorefineries can be also identified.

EXPERIMENTAL

Materials

The CEFW was obtained from acidic, steam-exploded rice straw after enzymatic saccharification and solid-state fermentation as detailed elsewhere (Ko *et al*. 2012). Rice straw samples were soaked in 1.5% H_2SO_4 solution for 24 h before steam explosion. The pretreated substrates were heated to 190 °C for 10 min. The pretreated samples were washed with excess tap water until the samples were completely neutralized and then refrigerated before the experiments. SSF was conducted according to the NREL LAP 002 protocol (Dowe and McMillan 2001). The yeast was *S. cerevisiae* D5α, and the enzyme was a commercial cellulase cocktail (CTec2) kindly provided by Novozymes China Investment Co. Ltd. Enzymatic hydrolysis was carried out 3 hours before yeast was introduced in the reactors for fermentation, and the operation conditions was controlled at 50 °C, under a pH of 5.0, and with a shaking speed of 10.5 $rad \cdot s^{-1}$ (100 rpm). The enzyme load was 6 mL per 100 g dry solid. During the fermentation phase the buffer pH, temperature, and shaking speed of the SSF were modified to 4.8, 37 °C, and 10.5 $rad \cdot s^{-1}$ (100 rpm), respectively.

The MSW was obtained from Yilan County, Taiwan. The moisture content of the raw MSW was approximately 58.2% by mass. With voids, the wet volumetric density was approximately 282 $kg \cdot m^{-3}$. The composition of the oven-dried MSW was 41.8% lignocellulosic biomass (including paper, fiber, wood chips, bamboo, and yard wastes), 7% kitchen wastes, 37.4% plastics and rubber, 3.4% other organics, and 11.1% inorganic materials.

The MSW was pretreated by using a 530-L stainless steel rotating autoclave, as detailed in Chang *et al*. (2011). A bulk MSW sample of 50 kg (including moisture) was steam-pretreated at 155 °C for 30 min after the addition of 18 kg of water to the autoclave. The MSW was completely mixed in the autoclave at a constant rotating speed of 0.314 $rad \cdot s^{-1}$ (3 rpm). No chemicals were used in the autoclaving process. Excess water was removed during the cooling process by releasing the water vapor into a cooling tank. The wet mass of the MSW decreased to approximately 40 kg after pretreatment. The steamed MSW was fractionated to separate the particles of different sizes by using a Bauer-McNett type fiber fractionator with sufficiently large amount of water. Debris greater than 0.6 mm (28 mesh) was removed. The weight of the fractionated biomass, or the MSWF, was approximately 18.4 kg after pretreatment. After the dehydration process,

there were 17.6 kg of dry matter. Of this amount, 8.1 kg was MSW fiber on a dry basis, 9.5 kg was plastic on a dry basis, so there was a 70 % yield on total dry material for dehydration process.

KBL was obtained from a pulp mill operated by Chung-Hwa Pulp and Paper Co., and the feedstock of the paper mill was eucalyptus and acacia wood chips. The initial pH of the KBL was 10.5 to 11.0, and the solid content was approximately 15%. Neutralized kraft black liquor (NBL) was prepared by neutralizing the KBL using 37.5% (v/v) hydrochloric acid until the pH had decreased to 6.0. Formaldehyde-condensed black liquor (MBL) was prepared by mixing 14 g of formaldehyde and 6 g of water with 100 g of concentrated KBL (*i.e.*, 32.5%). The solution was heated to 105 to 115 °C for 3.5 h and then cooled for further uses. The solids content of the MBL was 37.3 to 37.8%, and the final pH was in the range 10.0 to 11.0. A small amount of HCl was added to the MBL to adjust the pH to 8.0. Urea-formaldehyde resin (UF) was a commercial product obtained from Wood Glue Industrial Co., Ltd., Taiwan, and ONP fiber was collected from United News Group, Taiwan.

Methods

Fiber analysis

Morphology analysis of the lignocellulosic fiber was performed using a MorFi fiber analyzer (Techpap). For each fiber sample, 30 mg of sample was mixed into 1 L of tap water for analysis (*i.e.*, 0.3% solids content). Fiber length, width, curl, kink angle, and distribution, as well as the percentage of broken ends were analyzed. The average fiber length (l) of the sample was calculated based upon the accumulation of the weight fraction (w_i) of each fiber length (l_i) divided by the total mass of the fiber as follows:

$$\text{Average fiber length } (l) = \frac{\sum w_i \cdot l_i}{\sum w_i} \quad (1)$$

The chemical composition of the fiber was evaluated using the standard methods of the Technical Association of the Pulp and Paper Industry (TAPPI) for analyzing the contents of holocellulose (TAPPI T 249 cm-00), α-cellulose (TAPPI T 203 cm-99), lignin (TAPPI T 222 om-02, UM 250), caliper (TAPPI T 411 om-05), and ash (TAPPI T 211 om-02) (TAPPI 2005).

Gel permeation chromatography (GPC)

The molecular weights of BL and MBL were measured by GPC analysis using a JASCO PU-2080 plus HPLC pump and a JASCO 875 UV Intelligent UV/VIS detector associated with a Shodex SB-802.5 column. The eluent used in the measurement was a 60:40 mixture of 50 mM LiCl and CH_3CN, respectively. The flow rate and pumping pressure were controlled at 0.4 mL/min and 1.5 to 1.6 MPa, respectively.

Preparation of paper handsheets

The paper handsheets were prepared according to the standard methods (TAPPI T 205 om-88) by using a TAPPI standard disintegrator similar to No.329, Yasuda, Japan. Two types of paper handsheets were produced, paper at 60 $g \cdot m^{-2}$ dry mass (with 0.32 mm averaged caliper) and paperboard at 200 $g \cdot m^{-2}$ dry mass (with 0.80 mm averaged caliper). Three manipulated variables were considered: (i) different combinations of the waste derived fibers (*i.e.,* MSWF, CEFW, and ONP), (ii) different mass contents of the fibers,

and (iii) treatment processes of the handsheets. The mass ratios tested were 100:0, 90:10, 80:20 and 50:50 between ONP and CEFW and ONP and MSWF. The dosages of the additives (*i.e.*, BL, NBL, MBL, and UF, if applied) were 2% (w/w on oven-dried paper). The hot press temperature, time, and pressure of the thermal treatment (TP) were 120 ^{o}C, 1 minute, and 25 kgf·cm^{-2}, respectively. The handsheets were cut into suitable sizes for the testing of physical properties.

Physical property tests of paper handsheets

The physical properties included tensile strength (ISO 1924-2: 1994), bursting strength (ISO 2758: 2001), and tearing strength (ISO 1974: 1985). Averages and standard deviations of 10 replicates were presented for comparison between each design condition.

RESULTS AND DISCUSSION

Fiber Analysis

The appearances of the CEFW, the MSWF, and a sample product are shown in Fig. 1(a) through Fig. 1(c), respectively, and the chemical compositions of the biorefinery feedstock and the two waste-derived fibers are shown in Table 1. The CEFW sample was more homogenous in particle size and was darker in color than the MSWF, which was expected due to the higher lignin content in CEFW. Compared to rice straw, the CEFW contained a significantly higher amount of lignin (*i.e.*, 53.6% *versus* 25.5%) and a lower amount of hemicelluloses (*i.e.*, 15.8% *versus* 45.7%), suggesting a significant conversion of the carbohydrates to monomers and bioethanol. The MSWF had a higher holocellulose content (67.8%) and lower lignin content (8.6%) because of the high fraction of waste paper in the MSW. It also contained 16.7% ash and 6.9% other components, which could be due to extractives or other volatile organic compounds (*i.e.*, resin polymers and other adhesives).

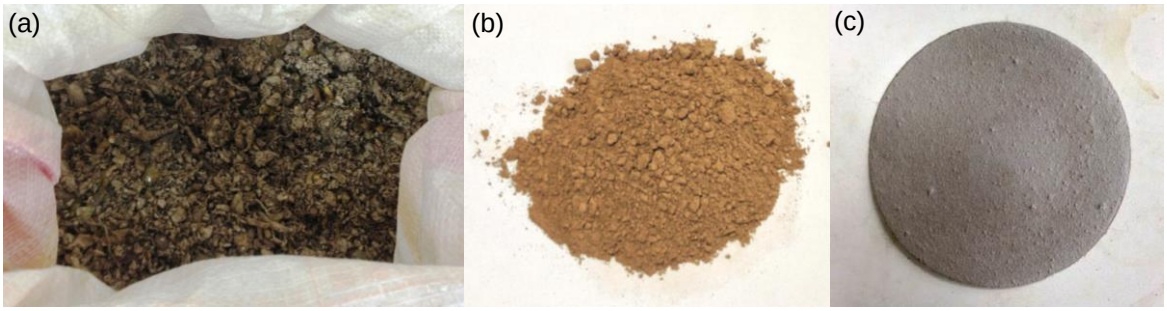


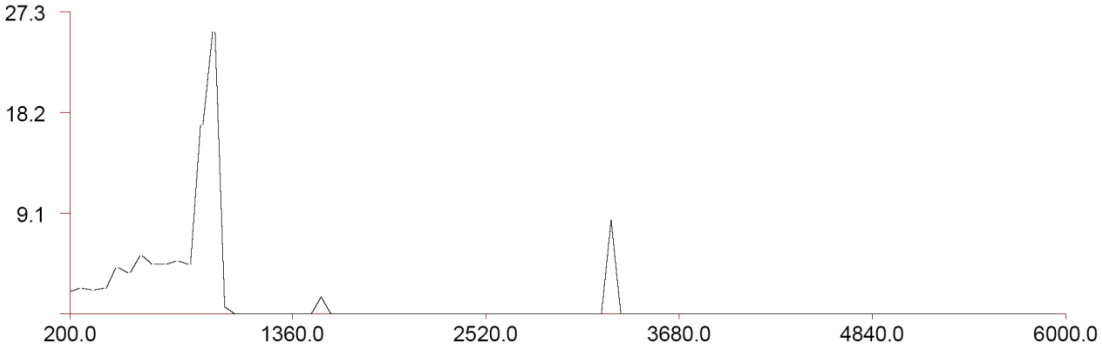
Fig. 1. Appearance of two waste-derived fibers: (a) CEFW, (b) MSWF, and (c) paper product.

Table 1. Chemical Compositions of the Two Waste-derived Fibers

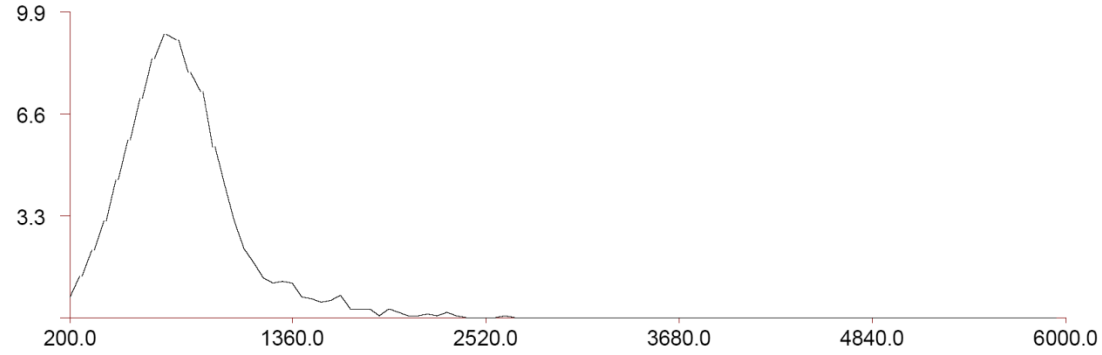
Substrate	Holocellulose		Lignin	Ash	Other
	α-cellulose	Other			
Rice Straw	21.3	45.7	25.5	10.8	~0
CEFW	23.4	15.8	53.6	11.2	~0
MSWF	37.4	30.4	8.6	16.7	6.9

Fiber length distributions of the CEFW, MSWF, and ONP after the MorFi analysis are shown in Fig. 2(a) through Fig. 2(c), respectively. The MSWF was composed of a considerable amount of paper fiber and some particles. The majority of the inorganic compounds, metals, and plastics were removed after fractionation.

(a) CEFW (average length = 677 m)



(b) MSWF (average length = 802 m)



(c) ONP (average length = 1370 m)

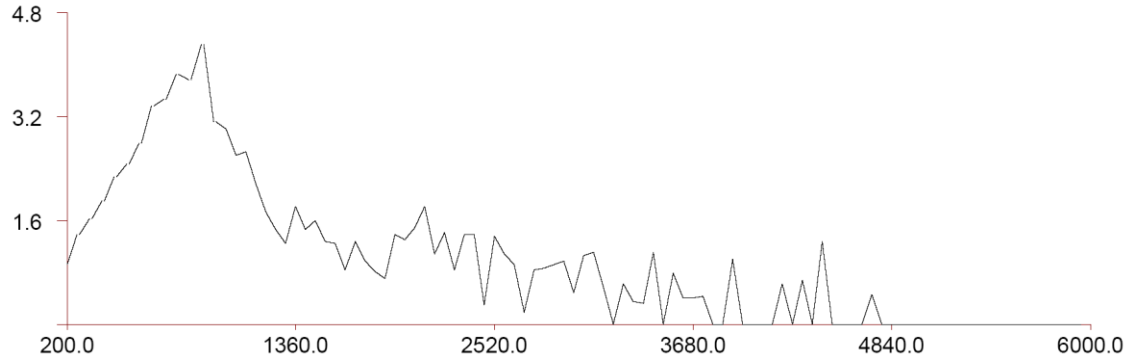


Fig. 2. Fiber length distribution of (a) CEFW; (b) MSWF; and (c) ONP fibers. Units of x-axis = mm and y-axis = % mass

The fiber length distribution of the MSWF covered a broader range of the spectrum (*i.e.*, from 200 μm to 1,360 μm) than the CEFW, which produced a sharp peak at approximately 928 μm. The ONP contained a large amount of longer fiber (*i.e.*, longer than 1360 μm), and the maximum fiber length of ONP fiber was approximately 4,800 μm. The mass-weighted average fiber lengths of MSWF, CEFW, and ONP, calculated based on Eq. (1), were 802, 677, and 1370 μm, respectively. The results of the chemical composition and fiber length analyses were compared with the changes in the physical properties of the paper and paperboard products.

Formaldehyde Condensation of Kraft Black Liquor

The results of the GPC analysis of the KBL lignin and the black liquor-derived binder (MBL) are shown in Fig. 3. The molecular weight of the lignin increased dramatically from 599 to 1,400 after formaldehyde condensation, which confirmed the effectiveness of the synthesis of the polymer binder. The binder was mixed with the recycled fiber for recycled paper making, and its performance was compared to the products of other manufacturing conditions, *i.e.*, no binder control, or BL, NBL, or commercial UF binder with or without thermal pressing.

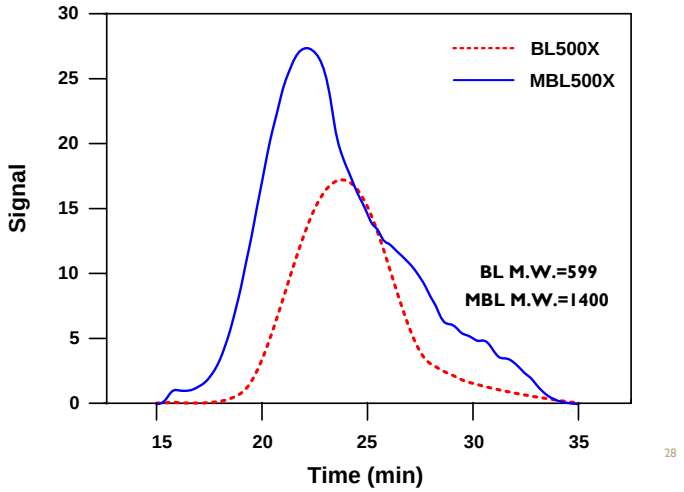


Fig. 3. Molecular weight increase for kraft black liquor by formaldehyde condensation

Properties of the Paper Handsheets

The experimental results of the physical properties of the recycled paper products are shown in Table S1, and the trends of these properties as a function of the mass ratio of the mixed fibers are summarized in Figs. 4(a) through 4(f) for paper products and Figs. 5(a) through 5(f) for paperboard products. The tensile, bursting, and tearing strengths of the paper/paperboard handsheets are presented from the top to the bottom of the three panels, respectively. The properties of the handsheets produced by the ONP/CEFW mixtures are shown in the three sub-figures in the left column (*i.e.*, (a) to (c)), with the products of the ONP/MSWF and MSWF/CEFW mixtures shown in the right column (*i.e.*, (d) to (f)). Control experiments for two out of three combinations (*i.e.* ONP + CEFW and ONP + MSWF) of raw materials were conducted. The control groups of MSWF and CEFW combination couldn't form handsheets with sufficient strength for physical properties measurement (results not shown). The parameter shown on the x-axis is the mass percent of the waste fibers, or the amount of shorter fiber mixed into the recycled

fiber mixtures. The results of different manufacturing conditions are distinguished by different symbols in Figs. 4 and 5, and the error bars display the standard deviations of the results.

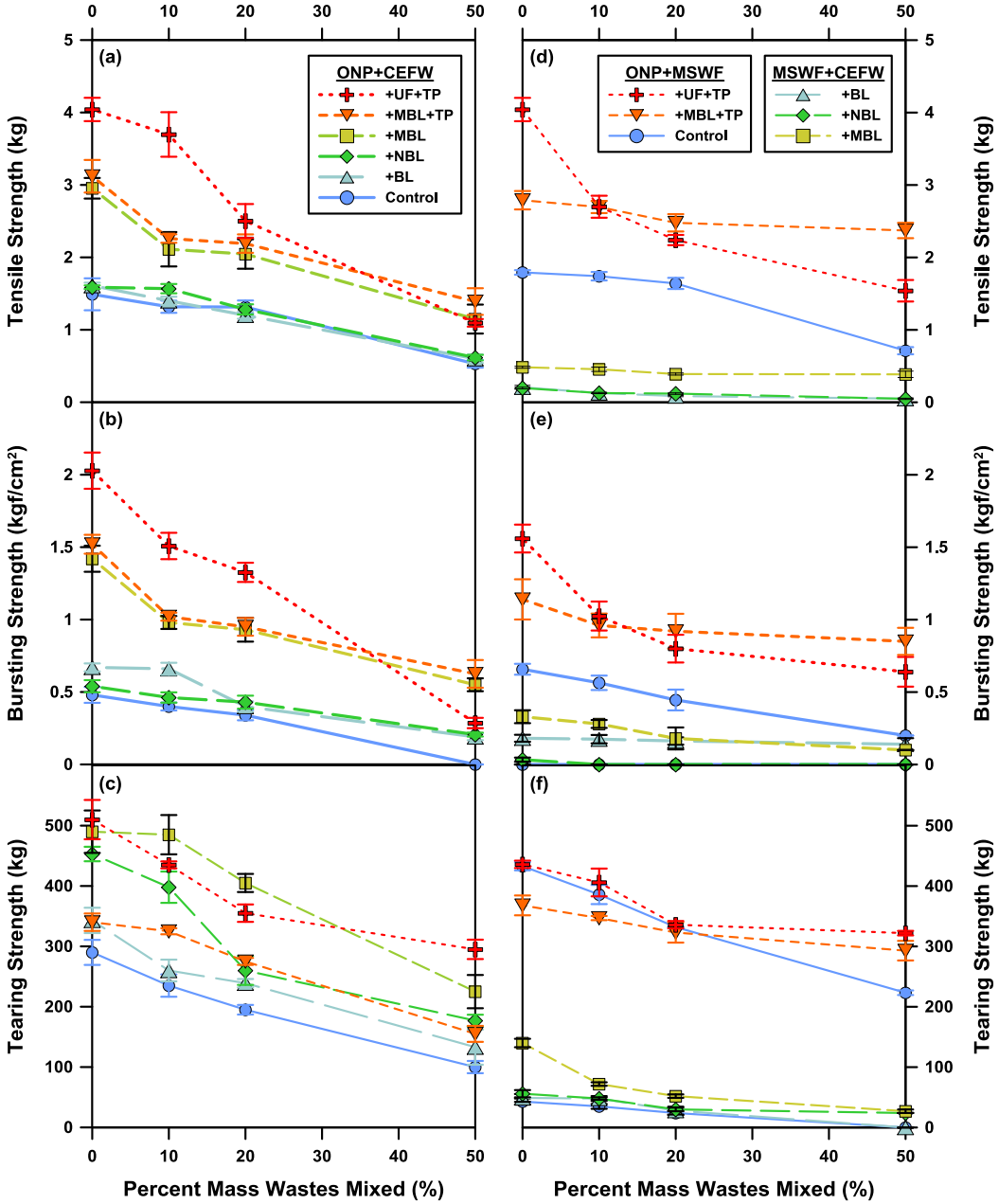


Fig. 4. Physical properties of the manufactured paper handsheets (mass = 60 g/m^2) under different manufacturing conditions

Tensile indexes of samples without adhesives in this manuscript were from 6.53 to 17.42 Nm/g for 60g/m^2 handsheets from 50% to 100% ONP, up to 43.55 Nm/g for ONP handsheets with adhesives. For the sake of comparison, the tensile index of reported pulp OCC is 30.65 Nm/g for 125 g/m^2 handsheets (Ashley and Hodgson 2003). Tensile index ONP in this manuscript is clearly inferior to those of US OCC reported

(Ashley and Hodgson 2003). Neither tensile indexes of CEFW nor those of MSWF demonstrated comparable strength to ones of MSW reported (Ashley and Hodgson 2003).

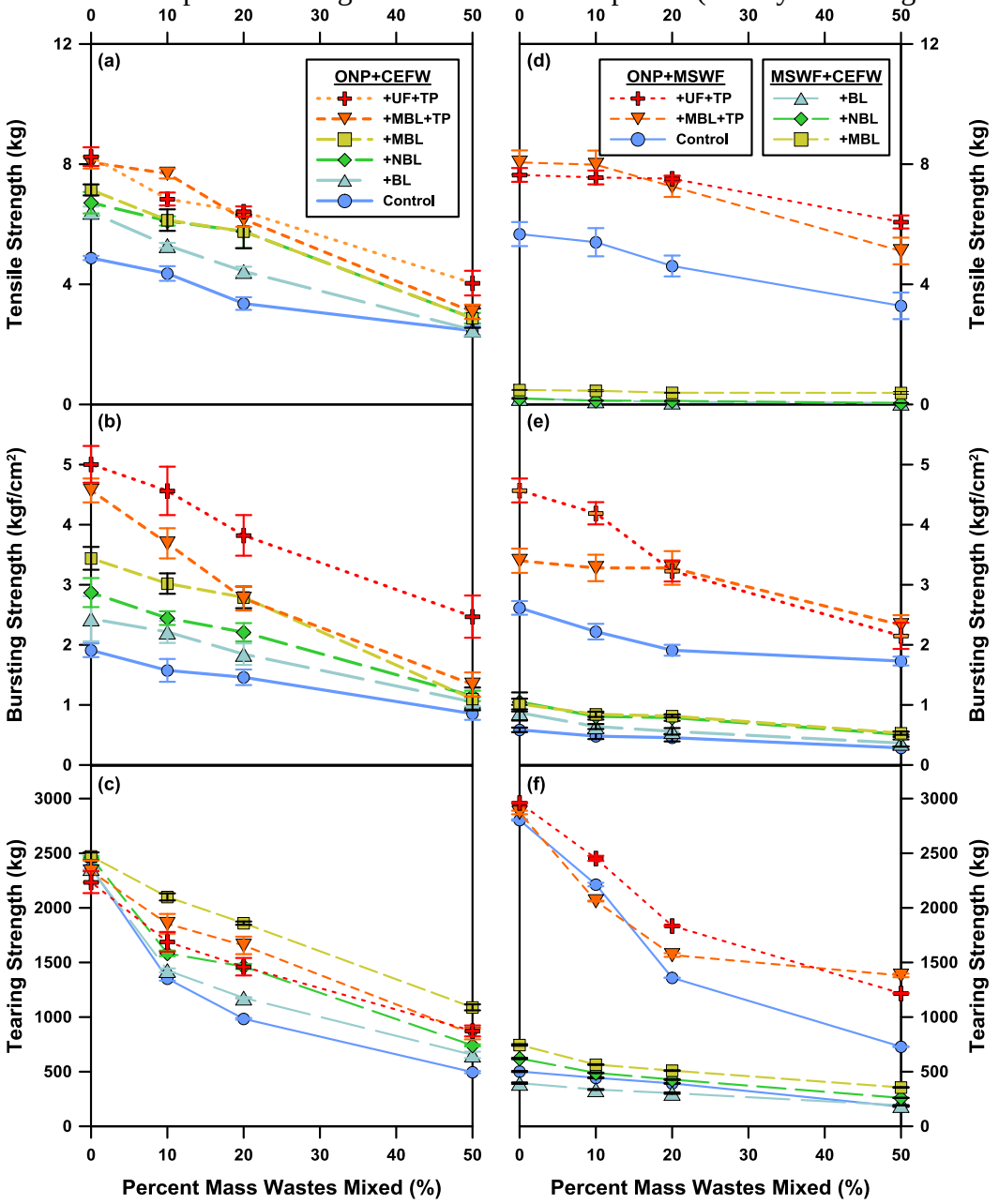


Fig. 5. Physical properties of the manufactured paperboard handsheets (mass = 200 g/m^2) under different manufacturing conditions

Higher lignin contents could contribute inferior physical properties of CEFW fiber. Inferior physical properties of MSW fiber might be attributable to higher separation temperature at 155 ^{o}C than one reported at 130 ^{o}C (Ashley and Hodgson 2003). In our previous pilot plant study better separation between fibrous and plastic wastes was achieved at 155 ^{o}C than at 130 ^{o}C (Chang et al. 2011). There is an extensive ongoing paper recycling program in Taiwan; hence the better fiber resources within municipal waste in Taiwan should be further deprived than those in the US Pacific Northwest, as

reported by (Ashley and Hodgson 2003; Leu et al. 2013). In addition, more adhesive and plastics remnants could be in MSW of this study. In addition, recycle rates of paper products Taiwan are usually exceeding 70%. Hence physical properties of MSW fiber in this study were shown to be inferior to those reported by Ashley and Hodgson in 2003 and 2004.

Effects of Different Manufacturing Processes on Handsheet Properties

Significant improvements in the physical properties of the waste fiber products were observed after the polymer binders (*i.e.*, UF and MBL) were introduced. For the ONP/CEFW fiber, applying UF resin with thermal pressing (TP) showed the most improvement in the tensile and bursting strengths of the fiber products. The tensile strength of the UF+TP samples increased from 1.49 kg to 4.04 kg, or more than 171% higher than the no-binder control (Fig. 4(a)); the bursting strength increased from 0.48 $kg_f \cdot cm^{-2}$ to 2.03 $kg_f \cdot cm^{-2}$, or 322% higher than the no-binder control (Fig. 4(b)).

Reasonable improvements were observed in the samples with the black liquor derived binder, MBL. The tensile and burst strengths of the MBL+TP samples were 3.12 kg and 1.52 $kg_f \cdot cm^{-2}$, or 109% and 216% higher than the controls, respectively. Thermal pressing, however, did not demonstrate significant beneficial effects on the physical properties of the paper handsheets. For example, no distinguishable improvements in tensile and bursting strengths were discovered for the MBL samples before and after thermal pressing. MBL may cure at room temperature without hot pressing. No improvement in tensile and bursting strengths was observed in the NBL- and BL-treated handsheets. Formaldehyde condensation is essential to create the binding capacity of the KBL lignin. Similar results were observed in the ONP+MSWF and MSWF+CEFW fiber mixtures, as shown in Fig. 4(d) and Fig. 4(e), respectively. The physical strengths of the MSWF/CEFW fibers were lower by at least one order of magnitude for all of the tested parameters compared to the ONP/CEFW and ONP/MSWF samples. This phenomenon could be due to the shorter fiber length or contamination of the MSWF.

Improvements in tearing strengths after different manufacturing processes (trend shown in Fig. 4(c) and Fig. 4(f)) differ from the results from the tensile and bursting strength tests. Adding any polymer (*e.g.,* binders or even untreated black liquor) to the ONP/CEFW fibers improved the tearing strengths of the handsheets, and the enhancements imparted by MBL were close or even better than UF resins. This result could be due to the higher molecular weight of MBL than the UF resins. In addition, adding NBL and BL slightly improved the tearing strengths of the ONP/CEFW handsheets. For the ONP/MSWF samples, however, no improvements in tearing strength due to the addition of the adhesive binders were observed. The lowest tearing strength of the ONP/MSWF fibers with no binder was 223 kg, which was similar to the strength of the ONP/CEFW fiber after UF+TP enhancement (*i.e.*, 295 kg). The tearing strength of the control group of the ONP/CEFW fibers was much lower (*i.e.*, 100 kg).

The results of the paperboard products, produced by using the same fiber mixtures and manufacturing conditions (Fig. 5) at higher dry mass, were similar to the results observed from the paper handsheets. The overall physical properties were much higher for the paperboards than for the papers, except for the products using MSWF and CEFW fibers due to their short average fiber lengths.

Relationship between Fiber Length and Handsheet Strengths

Linear relationships were obtained between the tensile strengths of the paperboard handsheets and the average fiber lengths of the products, as presented in Fig. 6. The average fiber sizes evaluated by using the Techpap MorFi fiber analyzer and Eq. 1. Experimental results of two manufacturing conditions are presented in the figure for comparison: the physical properties of Group (1), the paperboard handsheets with no binder (symbol ☐) and the products with BL (symbol ◆), were plotted against the physical properties of Group (2), paperboards enhanced with MBL+TP (symbol △) and UF+TP (symbol●) binders. The dashed and solid lines display the linear fits of the relationships between the two parameters of the two sample groups, and the shaded areas show the 95% confidence intervals of the results. Similar to the tensile strength trends, positive linear relationships were observed for all of the other physical properties of the waste-derived paper and paperboard handsheets.

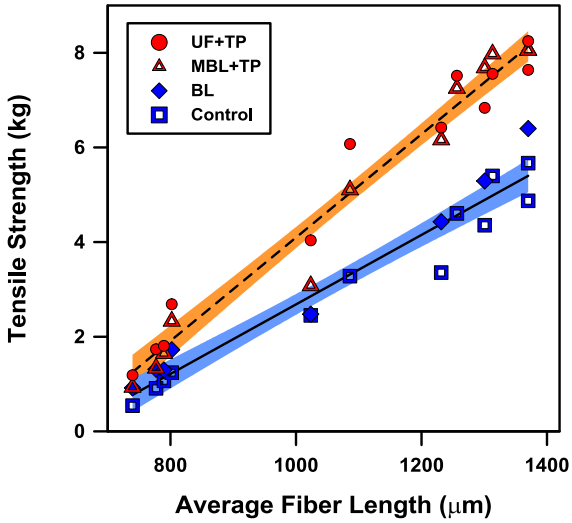


Fig. 6. Changes in tensile strength of the waste fiber paperboard products (mass = 200 g/m^2) over average fiber length. The dashed lines show the linear fits of the data after two groups of conditions (*i.e.*, UF+TP/MBL+TP *versus* BL/Control), and the shaded area shows the 95% confidence intervals of the data. Adding adhesive binders significantly improved the physical properties of the paper products.

In Fig. 6, the intercept between the linear fits and the x-axis represents the minimum fiber length required to provide the basic strength of the fiber mixture for papermaking. The minimum fiber length of the handsheets was approximately 670 μm, suggesting that fibers shorter than this length would not provide any support for tensile strength. In addition, the steeper slopes of the linear fit lines for MBL+TP and UF+TP than those of the two control groups may represent the factors of improvement provided by the adhesive binders.

The improvements in the fiber strength over the unit length of the fiber (*i.e.*, the slopes of the strength against fiber length) and the critical/minimum fiber length for different properties of the products under different manufacturing conditions are listed in Table 2. The results confirmed that adding binders (*i.e.*, both MBL and UF) can significantly improve the tensile strength and burst strength, but not the tearing strength. The effect of the binders was more significant for UF+TP samples than MBL+TP samples for paper products, but MBL+TP samples performed similarly to UF+TP

samples for paperboards. The improvements realized by adding MBL and UF binders to paperboard manufacture were 57.6% and 44.0% for tensile strength, and 84.5% and 98.6% for bursting strength, respectively. Finally, the minimum fiber lengths required to provide basic strength for the paper/paperboard products ranged between 670 μm and 1020 μm, although this length varied among the physical properties. The average fiber length of the waste fibers should be greater than 1020 μm for recycled paper making.

Table 2. Unit Improvement of Increased Fiber Length after Different Processes

Strength Property	Process Condition				Minimum fiber length (μm)
	Control	+BL	+MBL+TP	+UF+TP	
	Paper (dry mass = 60 g/m^2)				
Tensile (kg/mm)	2.76	2.46	3.98	8.17	950
Bursting (kgf/cm^2/mm)	1.54	1.47	1.62	3.86	1020
Tearing (kg/mm)	555	483	621	493	790
	Paperboard (dry mass = 200 g/m^2)				
Tensile (kg/mm)	7.24	7.87	11.41	10.43	670
Bursting (kgf/cm^2/mm)	2.91	4.06	5.37	5.78	770
Tearing (kg/mm)	3037	4317	3184	3110	970

CONCLUSIONS

1. The physical properties of the waste-derived papers made from rice straw cellulosic ethanol fermentation waste and MSW derived fiber were affected by the fiber lengths of the mixtures, regardless of the types and chemical contents of fibers used.
2. The positive linear relationships between paper strength and fiber length provide information to predict the physical properties of waste derived fibers for non-printing uses.
3. The "green binder" generated from kraft lignin can considerably improve the properties of recycled paper products. With the aid of MBL or UF binders, the waste-derived fibers CEFW and MSWF could substitute a portion of the ONP fiber used in the manufacture of recycled paper and paperboards.
4. The minimum average fiber length is 1020 μm, and therefore the mass fractions of the waste fibers in ONP fiber mixtures should be no more than 50.5% for CEFW and 61.6% for MSWF.

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REFERENCES CITED

Alvira, P., Tomás-Pejó, E., Ballesteros, M., and Negro, M. (2010). "Pretreatment technologies for an efficient bioethanol production process based on enzymatic hydrolysis: a review." *Bioresource Technology*, 101(13), 4851-4861.

Antizar-Ladislao, B., and Turrion-Gomez, J. L. (2008). "Second-generation biofuels and local bioenergy systems." *Biofuels, Bioproducts and Biorefining*, 2(5), 455-469.

Ashley, C. R., and Hodgson, K. T. (2003). "Papermaking properties and morphology of cellulose fiber recovered from municipal solid waste." *Tappi journal*, 2(10), 19-22.

Ashley, C. R., and Hodgson, K. T. (2004). "Morphological properties of cellulose fibre recovered from municipal solid waste." *Appita journal*, 57(3), 210.

Chang, C.-C., Wang, Y.-C., Li, Y.-S., Shie, J.-L., Chen, Y.-H., Ho, C., Hung, Z.-S., Chiang, S.-W., and Chang, C.-Y. (2011). "Investigating the autoclaving technologies for municipal solid waste treatment." *Proceeding of the 7th Cross Straits Climate Change and Sustainable Energy Development Forum*.

Eley, M. H., Guinn, G. R., and Bagchi, J. (1995). "Cellulosic materials recovered from steam classified municipal solid wastes as feedstocks for conversion to fuels and chemicals." *Applied Biochemistry and Biotechnology*, 51(1), 387-397.

García, A., Maulini, C., Torrente, J. M., Sánchez, A., Barrena, R., and Font, X. (2012). "Biological treatment of the organic fibre from the autoclaving of municipal solid wastes; preliminary results." *Biosystems Engineering*, 112(4), 335-343.

Heath, L. S., Smith, J. E., Skog, K. E., Nowak, D. J., and Woodall, C. W. (2011). "Managed forest carbon estimates for the US greenhouse gas inventory, 1990-2008." *Journal of Forestry*, 109(3), 167-173.

Hendriks, A., and Zeeman, G. (2009). "Pretreatments to enhance the digestibility of lignocellulosic biomass." *Bioresource Technology*, 100(1), 10-18.

Holtman, K. M., Kodama, A., Klamczynski, A. P., Flynn, A., Bozzi, D. V., Torres, L., Franqui-Villanueva, D., Mao, J., Glenn, G. M., and Orts, W. J. (2012). "Thermal properties of poly (ethylene terephthalate) recovered from municipal solid waste by steam autoclaving." *Journal of Applied Polymer Science*, 126(5), 1698-1708.

Hung, Z.-S., Chang, C.-C., Chang, C.-F. H., Lin, Y.-S., Ji, D.-R., Chang, C.-Y., Tseng, J.-Y., Chiang, S.-W., Shie, J.-L., and Chen, Y.-H. (2013). "Autoclaving treatment of wasted disposable bamboo chopsticks." *Journal of the Taiwan Institute of Chemical Engineers*, 44(6), 1010-1015.

Hung, Z.-S., Chang, C.-C., Chang, C. H., Wang, Y.-C., Chang, C.-Y., Ji, D.-R., Tseng, J.-Y., Ko, C.-H., Chen, Y.-H., and Shie, J.-L. (2012). "Autoclaving of waste plastic-free papers for resource recovery and reutilization." *FRESENIUS ENVIRONMENTAL BULLETIN*, 21(8 C), 2486-2493.

Leu, S.-Y., Zhu, J., Gleisner, R., Sessions, J., and Marrs, G. (2013). "Robust enzymatic saccharification of a Douglas-fir forest harvest residue by SPORL," *Biomass and Bioenergy*, 59, 393-401.

Li, S., Zhang, X., and Andresen, J. M. (2012). "Production of fermentable sugars from enzymatic hydrolysis of pretreated municipal solid waste after autoclave process." *Fuel*, 92(1), 84-88.

Limayem, A., and Ricke, S. C. (2012). "Lignocellulosic biomass for bioethanol production: current perspectives, potential issues and future prospects." *Progress in Energy and Combustion Science*, 38(4), 449-467.

Shi, A. Z., Koh, L. P., and Tan, H. T. (2009). "The biofuel potential of municipal solid waste." *Gcb Bioenergy*, 1(5), 317-320.

Sims, R. E. H., Mabee, W., Saddler, J. N., and Taylor, M. (2010). "An overview of second generation biofuel technologies." *Bioresource Technology*, 101(6), 1570-1580.

TAPPI, T. (2005). "TAPPI Test Methods." *Tappi Committee, Atlanta, GA, USA*.

USEPA. (2013). "Municipal Solid Waste Generation, Recycling, and Disposal in the United States: Facts and Figures for 2011."

Zhou, H., Zhu, J., Luo, X., Leu, S.-Y., Wu, X., Gleisner, R., Dien, B. S., Hector, R. E., Yang, D., and Qiu, X. (2013). "Bioconversion of beetle-killed lodgepole pine using SPORL: process scale-up design, lignin coproduct, and high solids fermentation without detoxification," *Industrial & Engineering Chemistry Research*, 52(45), 16057-16065.

Zhu, J., Pan, X., Wang, G., and Gleisner, R. (2009). "Sulfite pretreatment (SPORL) for robust enzymatic saccharification of spruce and red pine." *Bioresource Technology*, 100(8), 2411-2418.

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