Distillers' Grains Used as a Filler in Recycled Containerboard

Hongxia Gao,* Zhenyu Cai, Yao Sun, Zihao Wan, Chi Xu, Canwen Yang, and Yang Xu *

Distillers' grain (DG) was used as an ingredient in paperboard made in the laboratory from recycled containerboard fibers. The chemical composition and physico-chemical properties of DG fractions that had been isolated using varied screens were investigated. The effect of DG incorporation on the properties of the recycled paperboard was compared relative what was obtained with either precipitated calcium carbonate (PCC) or talc powder as fillers. The DG was found to mainly contain cellulose, hemicellulose, lignin, protein, and fat. At a filler addition level of 5%, the 100- to 140-mesh DG-filled handsheet exhibited the most satisfactory physical properties, with a tensile index of 25.4 $N \cdot m \cdot g^{-1}$ and a ring crush index of 7.95 $N \cdot m \cdot g^{-1}$. The strength values were generally higher than those of paper filled by PCC or talc powder at the same addition levels. The tensile index and ring crush index of hybrid-filled handsheets increased with increasing ratios of DG. The results suggest that DG can be used as a substitute for fiber content in some grades of paperboard, especially where a low-cost, bulky material could provide an advantage. Such usage of DG can resolve environmental challenges associated with storage and transportation of excess DG that is presently discarded.

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INTRODUCTION

Baijiu, a traditional distilled liquor unique to China, is typically made from grains such as sorghum, wheat, rice, and corn through a solid-state fermentation process (Tu *et al*. 2022). Efforts have been made to enhance the brewing efficiency to reduce the generation of solid-state by-products, *i.e.*, Baijiu distillers' grains (DG). However, over 30 million tons of DG are produced annually in China (Ma *et al.* 2023). Fresh DG mainly consists of organics and water, which are highly susceptible to mold and decay, potentially causing environmental pollution if not handled promptly (Liu *et al.* 2022).

Until now, DG has mainly been applied in producing animal feed (Gaillard *et al.* 2017), fertilizer manufacturing, and methane production *via* fermentation (Cheng *et al.* 2023b; Wang *et al.* 2022). Academia and industry are committed to recycling and utilizing DG and have proposed and implemented many related methods. For instance, non-starch polysaccharides can be extracted from DG using the extrusion method, which can then be further processed and utilized in multiple industries (Liu *et al.* 2023b). DG can also be used to produce biochar, which has broad applications in water treatment and environmental governance (Liu *et al.* 2023a). The antioxidant-active polysaccharides from DG can be

extracted *via* an enzyme- and microwave-assisted approach, which affords an option for the high-value-added application of DG (Li *et al.* 2025b). While these approaches have identified new pathways for the resource recovery of DG, the complex and laborious nature of these processes makes it challenging to utilize the substantial DG generated in industrial applications entirely. Therefore, it is imperative to develop a more straightforward, widely applicable, and recycling technology to consume substantial amounts of DG.

The DG produced during liquor production, which contains approximately 34 wt% cellulose, indicates its potential for application in pulping and papermaking. However, pulp from DG with low yield was obtained in practical experience (Gao *et al.* 2024). Furthermore, handsheet of DG pulp (bleached or unbleached) generally exhibits inferior physical properties when compared with handsheets comprised of herbaceous plant fibers, owing to the inherent morphological limitation of DG fibers.

Filling with mineral particles is comment in certain grades of commercially made paper products (Shen *et al.* 2011). Commonly used fillers, such as kaolin and precipitated calcium carbonate (PCC), primarily contribute to improving the formation, enhancing brightness, and optimizing the printability of paper (El Gendy *et al.* 2014). Additionally, the incorporation of fillers can reduce the consumption of cellulose fibers, thereby supporting cost-effective production of paper (Dong *et al*. 2008). However, the incorporation of inorganic filler negatively affected the physical properties of paper, resulting in issues such as dusting and linting (Shen *et al.* 2010). DG is rich in organics, such as cellulose, starch, and protein, which can promote the formation of hydrogen bonds between DG particles and cellulose fibers. Additionally, compared with conventional treatment approaches, utilizing DG directly as a filler represents a more straightforward and efficient strategy for recovering waste bioresources.

This study assessed the feasibility of utilizing DG as a filler in papermaking, providing a novel insight into the recovery of bioresources and sustainable waste management. The smashed and sieved DG was used as a filler in paper handsheets prepared from recycled containerboard (which is comprised of linerboard and corrugating medium). A simple approach was employed in this work, in which effects of different size fractions of DG were compared with those of handsheets to which either calcium carbonate or talc had been added at different levels. The microtopographies of sieved DG particles were observed, and their chemical compositions, sedimentation speeds, and chemical/crystalline structure were determined. Handsheets were prepared using old corrugated containers (OCC) as the fiber source. Fillers were added in the range of zero to 20% by mass, comparison different size fractions of sieved DG, PCC, talc powder, and their hybrid mixtures (including DG/PCC and DG/talc powder). The resultant handsheets were evaluated in terms of tensile index and ring crush index.

EXPERIMENTAL

Materials

DG was obtained from a distillery located in Yibin, Sichuan, China. It was dried at 40 °C to achieve a moisture content of approximately 10% prior to use. Recycled fiber was prepared from old corrugated containers (OCC) through a process of shredding, soaking, disintegrating, pulping, and dewatering to obtain recycled pulp with a beating degree of 41 °SR. Precipitated calcium carbonate (PCC) and talc powder were procured from Aladdin Biochemical Technology Co., Ltd. (Shanghai, China). All other chemical reagents

(analytical grade) were purchased from Aladdin Biochemical Technology Co., Ltd. and used without purification.

Methods

Preparation of fillers

Dried DG was pulverized and sieved using standard test sieves, which were used in sequence with 40, 60, 100, 140, and 200-mesh sieves. This process yielded a series of screened DG, which were hermetically stored in a refrigerator at 4 ℃. Commercial fillers, *i.e.*, PCC and talc powder, were used without further treatment.

Handsheet preparation using DG, PCC, or talc powder filling

The handsheets were produced according TAPPI T 205 sp-18 (2018). In detail, recycled pulp fiber and filler were weighed to achieve a target basis weight of 100 g/m², with various proportions of filler (0, 5, 10, 15, or 20 wt.%). The mixed pulp and filler were dispersed using a defibrizer (IMT-SJ00, China National Pulp and Paper Research Institute, China) until a complete dispersion was achieved. The slurry was transferred to a hand-sheet former (ASM-32N2F, China National Pulp and Paper Research Institute, China) to obtain handsheets. For simplicity, no retention aid chemical was used. Information reported later in this article about filler content refers to the added amount, not the retained amount, which is expected to be lower, especially for PCC and talc. The paper sheet was pressed (S005 laboratory sheet press, China National Pulp and Paper Research Institute, China), dried at 105 °C (RD-01 rotary drum dryer, China National Pulp and Paper Research Institute, China), then stored in a constant temperature-humidity chamber (23 ± 1 °C, 50 ± 2% RH) for at least 4 h prior to testing.

Handsheets using hybrid fillers

All processes used when preparing sheets with combinations of two fillers, *i.e.* hybrids, were consistent with the above-mentioned steps, except for the type of filler used. The used hybrid filler was a mixture of DG/PCC or DG/talc, with proportions of 0:20, 5:15, 10:10, 15:5, and 20:0.

Characterization of DG

The cellulose and hemicellulose content of DG was determined according to TAPPI T 203 cm-22 (2022). The acid-insoluble/soluble lignin content was determined according to TAPPI T 222 om-21 (2021) and GB/T 10337 (2008), respectively. The protein and fat contents of DG were determined according to GB/T 6432 (2018) and GB/T 5009.6 (2003), respectively.

The sedimentation volume of DG was determined according to GB/T 19281 (2014).

The micromorphology of DG was observed by scanning electron microscope (SEM; VEGA3 TESCAN, Libušinatřída, Czech Republic) at 15 kV. Before observation, the surface of specimens was coated with platinum under vacuum.

The particle size of various DG, PCC, and talc was analyzed by a Malvern 3000 mastersizer (Malvern, UK), using deionized water as dispersant after 1 h swelling at room temperature.

The crystalline structure of DG was analyzed using an X-ray diffraction (XRD) apparatus (D8-Advance instrument, Bruker, Germany) equipped with Cu Kα radiation (k = 1.54 A°) as X-ray source operating (40 kV, 30 mA) at a scan rate of 0.05 per second in a 2θ range of 10 to 60°.

The chemical structure of DG was analyzed using a Nexus 670 spectrometer (VERTEX-70, Bruker, Germany) *via* the KBr method at 25 °C, with a scan range of 4000 to 600 cm^{-1} for 64 scans.

The thermostability of DG was recorded using a thermogravimetric analyzer (STA 449F, Metzsch, Germany) from 50 ℃ to 700 ℃ under N_2 atmosphere with a N_2 flow rate of 50 $mL \cdot min^{-1}$, a heating rate of 10 $K \cdot min^{-1}$.

Determination of Physical Properties of Handsheets

The tensile index and ring crush index of handsheets were determined according to TAPPI T 494 om-22 (2022) and TAPPI T 818 cm-18 (2018), respectively.

Statistical Analysis

GraphPad Prism software was used for data analysis. All data were shown as mean ± SD, difference between groups was analyzed by one-way ANOVA. Statistical significant difference was defined as $P< 0.05$.

RESULTS AND DISCUSSION

Characterization of DG

Microtopography of DG with different screens

Figure 1 presents the SEM images of DG particles obtained under different sieve sizes. The surface of DG exhibited regular protruding structures. As the sieve size decreased, the particle size of DG also decreased. The size of the filler is the key factor in determining the physical properties of filled handsheets (Song *et al.* 2012; Hubbe and Gill 2016). For example, large-sized fillers tended to be inhomogeneously distributed, which will aggravate the two-sidedness in handsheet, *i.e*., the substantial discrepancies in physical properties on two sides of the handsheet and resulting in a declined usability of paper. In contrast, tiny fillers will reduce the filler retention efficiency, *i.e*., the decreased retention of filler following the formation of paper, and negatively affect the drainability of pulp.

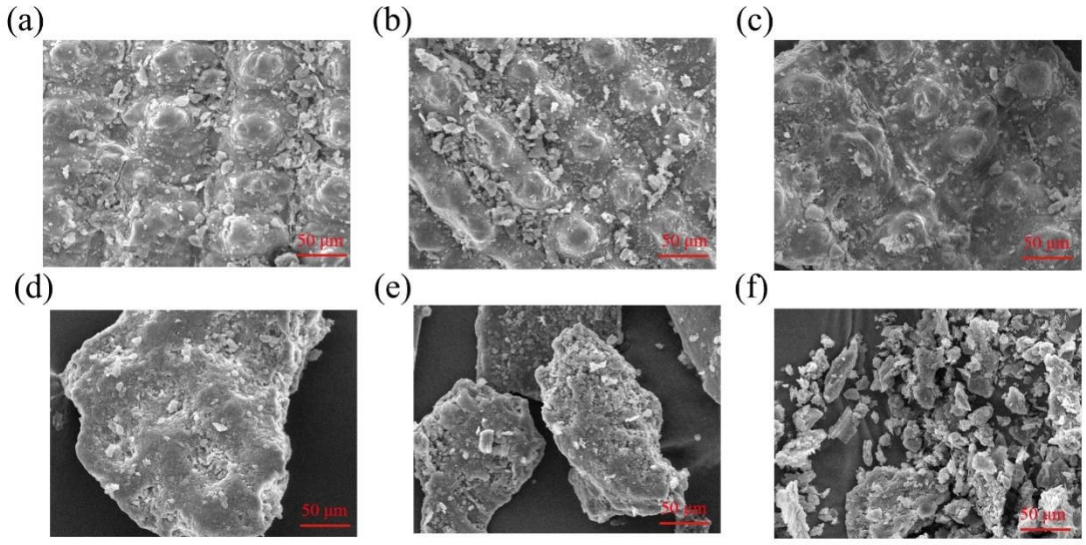

Fig. 1. Morphological characteristics of DG particles with various mesh sizes, (a) > 40 mesh; (b) 40-60 mesh; (c) 60-100 mesh; (d) 100-140 mesh; (e) 140-200mesh; (f) <200 mesh.

Table 1. Chemical Composition of Various Screened DG

Samples	Cellulose (%)	Lignin (%)	Hemicellulose (%)	Protein (%)	Fat (%)	Ash (%)
> 40-mesh	30.51±0.15	24.9±3.20	24.06±0.59	7.49±0.22	1.42±0.10	12.5±0.53
40-60 mesh	29.92±0.14	23.6±1.90	23.87±0.11	7.40±0.08	1.51±0.06	14.4±0.70
60-100 mesh	26.13±0.30	21.9±1.47	24.00±3.49	9.00±0.05	2.11±0.07	13.80±0.68
100-140 mesh	16.89±0.49	14.0±3.69	21.93±1.41	15.3±0.42	4.37±0.09	10.57±0.11
140-200 mesh	15.35±1.06	15.6±1.05	20.04±1.26	21.3±0.28	5.08±0.10	12.17±0.58
< 200 mesh	13.29±1.49	12.9±1.73	17.20±1.31	22.4±0.31	6.62±0.17	9.52±0.67

Table 2. TGA of DG with Different Screens

Samples	1st stage weight loss (%)	2nd stage weight loss (%)	3rd stage weight loss (%)	Peak temperature (°C)
> 40-mesh	4.54	44.26	21.23	343.95
40-60 mesh	3.69	38.67	27.62	352.92
60-100 mesh	4.62	40.82	27.56	352.43
100-140 mesh	2.99	45.07	23.15	348.90
140-200 mesh	4.37	45.72	24.25	346.91
< 200 mesh	3.43	44.85	22.45	342.91

Table 3. Sedimentation Volume of DG with Different Screens

Screened DG	> 40-mesh	40-60 mesh	60-100 mesh	100-140 mesh	140-200 mesh	< 200 mesh
Sedimentation volume /mL·(10g)$^{-1}$	29	27	28	28	29	36

Table 4. Particle Size of DG, PCC, and Talc

Specimens	> 40-mesh	40-60 mesh	60-100 mesh	100-140 mesh	140-200 mesh	< 200 mesh	PCC	Talc
D10 (μm）	7.51	6.7	6.59	6.13	5.14	5.03	3.48	3.52
D50 (μm）	541.39	301.79	156.67	42.94	26.32	24.39	12.49	12.39
D90 (μm）	862.82	722.74	416.4	141.37	96.62	93.14	37.15	37.8

Composition analysis of DG with different screens

The principal organic components of DG with various sieving specifications were determined, including cellulose, hemicellulose, lignin, protein, and fat (Table 1). Overall, the content of cellulose, lignin, and hemicellulose in DG decreased as the particle size decreased, whereas the content of fat and protein increased as the sieve mesh increased.

The variation in cellulose, protein and fat content might be attributed to the grinding process of DG, in which DG was broken up into particles of many different sizes. Cellulose serves as the primary structural component of the plant cell wall, exhibiting high mechanical strength and flexibility. Hence, cellulose tends to be present the larger particles due to its strong nature. Protein and fat, which are weaker and not tightly bound within the lignocellulose, will often be enriched in the smaller particles after grinding, resulting in an increased content with the decreased particle size of DG (Liu *et al.* 2008).

For lignin, it is typically embedded within the cellulose matrix in the plant, and can form strong associations with both cellulose and hemicellulose, resulting in the formation of lignin-carbohydrate complexes (LCCs) (Tarasov *et al.* 2018), leading to the variations in lignin content closely paralleled those of cellulose. Inorganics (ash content) exhibited irregular fluctuations, primarily due to the heterogeneous origin of mineral constituents in the dried DG (Liu *et al*. 2011), including silicates, calcium salts, phosphates, and residual fermentation additives.

Crystalline structure, chemical structure, and thermogravimetric analysis of DG with different screens

X-ray diffraction (XRD) and Fourier transform infrared spectroscopy (FTIR) were employed to elucidate the crystalline and chemical structure of DG with various mesh sizes. DG particles with different mesh size exhibited distinct diffraction peaks at 2θ =16.2°, 22.3°, and 34.5°, corresponding to the (110), (200), and (004) lattice planes of cellulose I_β, respectively, confirming the present cellulose in DG with a crystal form of cellulose I_β (Parsai *et al*. 2024). Additionally, a diffraction peak at 2θ = 26.8° was detected, corresponding to the (101) lattice plane of SiO_2 (Nazopatul *et al.* 2018).

The FTIR spectra are shown in Fig. 2(b). All the characteristic absorbance peaks of organics in DG could be detected in the FTIR spectra. The broad absorbance peak near 3413 cm^{-1}, which corresponded to the O–H stretching vibration, was attributed to the presence of both intermolecular and intramolecular hydrogen bonding interactions among organics. The absorbance peaks at 2930 and 1375 cm^{-1} corresponded to the stretching vibrations of methyl, methylene, and dimethyl groups, respectively (Trache *et al.* 2016). The peak at 1650 cm^{-1} was attributed to the stretching vibrations of the C=O group from the protein amide I band (Lin *et al.* 2021). The absorbance peak at 1517 cm^{-1} was due to the stretching vibrations of the C=C in the aromatic ring of lignin. The absorbance peak at 1240 cm^{-1} was derived from the stretching of the C–O–C bond in lignin. The absorbance peak at 1080 cm^{-1} originated from the asymmetric stretching vibration of the Si-O-Si bond in SiO_2 (Nazopatul *et al*. 2018). The absorbance peak at 1050 cm^{-1} was assignable to C–O stretching vibrations in hemicellulose (Luo *et al.* 2021). The absorbance peak at 823 cm^{-1} was caused by the stretching of the ether bond in lignin and the deformation of the aromatic C-H (including the in-plane deformation of the aromatic C-H) (Cheng *et al*. 2023a). The absorbance peak due to the stretching vibration of C=O was present near 1737 cm^{-1}, which was attributed to the residual aliphatic esters or lactones produced during the process of liquor production.

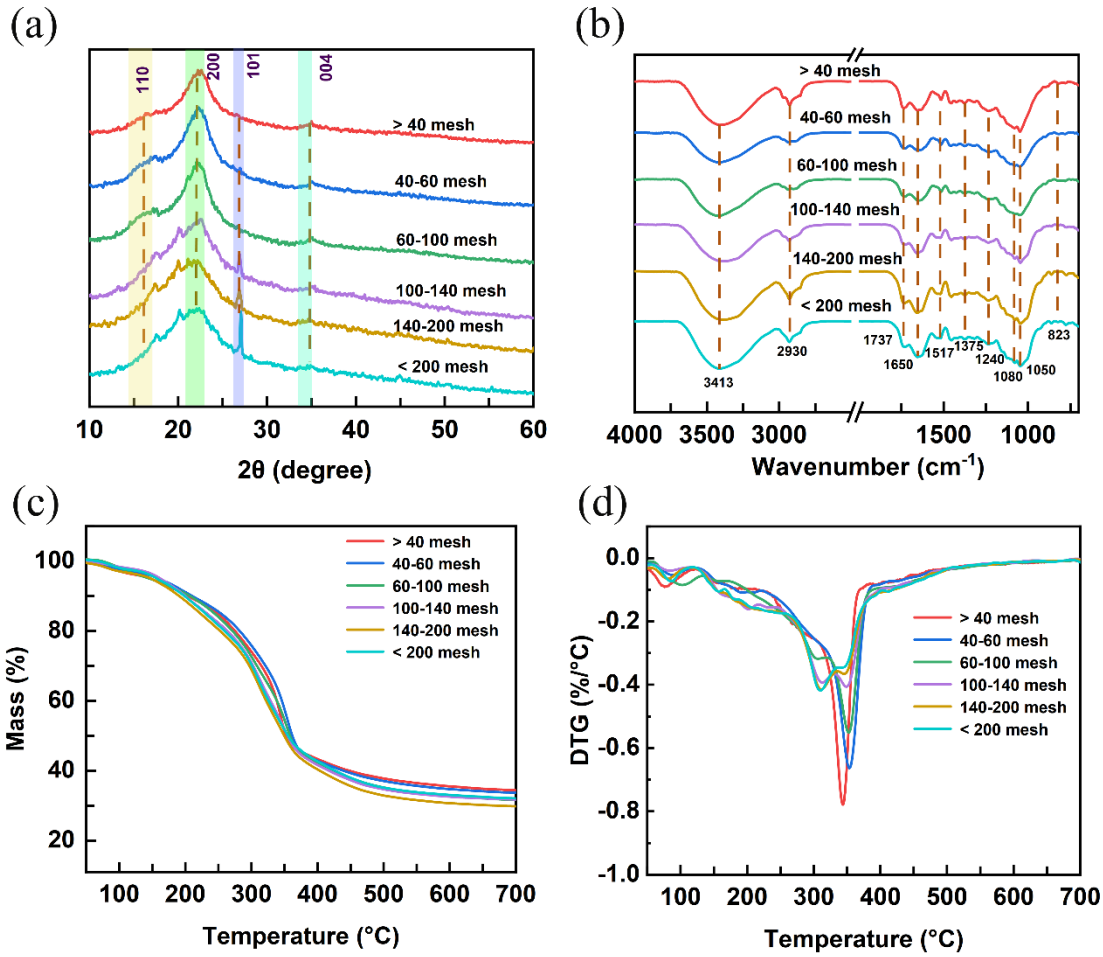

Fig. 2. Characterization of DG with different screens, (a) XRD pattern; (b) FTIR spectrum; (c) TG curves; (d) DTG curves

The drying process, following the formation of handsheets, indicated that the used DG, as filler, required favorable thermal stability to avoid decomposition. Hence, the thermogravimetric (TG) and differential thermogravimetric (DTG) curves of DG were recorded by a thermal gravimetric analyzer, as shown in Fig. 2(c) and (d) and Table 2. The DG with a varied screen exhibited similar thermal decomposition behavior, which could be divided into three stages. The initial mass loss (3.9%) below 140 °C was primarily attributed to the evaporation of moisture. A significant mass loss (43.2%) within the temperature range of 140 to 350 °C resulted from the thermal decomposition of DG constituents. Further mass reduction (24.5%) within the temperature range of 350 to 700 °C was associated with the breakdown of carbonized residues. It is noteworthy that the maximum thermal degradation temperature (T_{max}) of DG generally decreased with the decrease in particle size, which might be attributed to the component differences of DG with different screens. The organics in DGs are different in thermostability. Among them, lignin and cellulose are more thermostable than hemicellulose, protein, and fat. Overall, DG particles of all screens can meet the required temperature for the drying process of handsheets.

Sedimentation volume of DG with different screens

During papermaking, the settling rate of filler can affect its distribution within the handsheet structure. Because the DG particles generally were much larger than typical mineral particles used in papermaking, such settling might affect the paper product. A fast or slow settling velocity of filler could result in an inhomogeneous distribution of filler, leading to two-sidedness of the handsheet. Hence, a sedimentation experiment of DG was carried out, as listed in Table 3, and results are shown in Fig. 3. In addition, the particle size of various DG was determined, as listed in Table 4. With the decrease in mesh size, the particle size (D10, D50, and D90) decreased accordingly. The result of sedimentation volume demonstrated the rapid settling behavior of large-sized DG (40 mesh and 40 to 60 mesh) in water. Medium-sized DG (60- to 100-mesh and 100- to 140-mesh) settled at a comparatively slower rate. Specifically, they achieved a sedimentation volume of 28 mL within 10 min and remained stable during 3 h of storage. Small-sized DG (140 to 200-mesh and <200 mesh) displayed significantly reduced settling velocities, with final sedimentation volumes of 29 mL and 36 mL, respectively, after approximately 50 min of storage (Table 3). The varied sedimentation volume was attributed to the reduced particle size of DG, which resulted in an increased specific surface area and promoted the dispersion and suspension of particles. Meanwhile, the increased settling resistance resulted in a slow settling velocity.

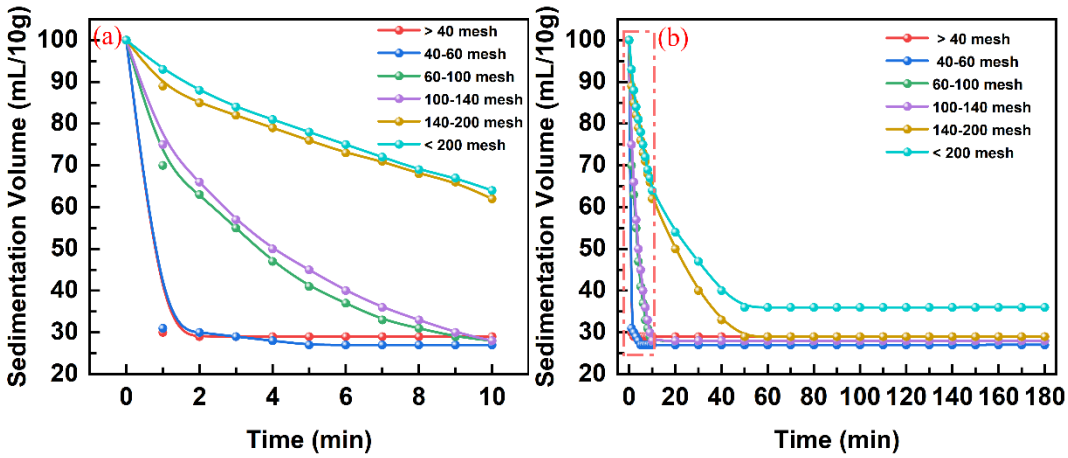

Fig. 3. The time-dependent sedimentation behavior of various DG

Performance Analysis of Handsheets

The screened DG with varied particle sizes affected its distribution of DG within handsheets and their properties (Hyll 2015). As shown in Fig. 4, the aggregated DG with an inhomogeneous distribution was easily observable, particularly with the incorporation of large-sized DGs (>40 mesh). With the decrease in particle size of DG (Table 4), handsheets with a fine and smooth surface were obtained, confirming the improved distribution of DG, despite optically discernible DG aggregation still being visible on the surface of handsheets filled with DG (40-60 mesh). Handsheets filled by DG with a sieving specification of 100 to 140, 140 to 200, or <200 mesh, exhibited a smooth surface with minimal two-sidedness, and negligible DG aggregation. Nevertheless, the decrease in drainability of the pulp, resulting from the decreased filler particle size, was apparent. Hence, DG with a sieving specification of 100 to 140 mesh may be a suitable choice for paper filling, considering the overall process and property of handsheets.

The incorporation of DG had a significant impact on the properties of handsheets, including density, tensile index, and ring crush index, as listed in Table 5. The varied density of DG-filled handsheets was jointly affected by the size and content of DG. Larger-sized DG can be mostly retained in handsheets by filtration.

Two likely contributions to an apparent increase in bulk can be considered: On the one hand, the stiffness and irregular shapes of the larger DG particles might directly contribute to a looser structure of the paper with lower apparent density (*i.e.* increased bulk). On the other hand, relatively large particles can contribute to higher roughness, such that the paper appears to be thicker than its true average thickness when measured with caliper tests. Such effects can provide an erroneous lowering of apparent density, compared to its true value.

Conversely, the retention efficiency of smaller DG is usually lower than that of larger-sized (Maślana *et al.* 2023). Small-sized DG was better able to fill the tiny gaps among fibers, resulting in increased density due to its small volume, limited space occupation, and lesser impact on the fiber-to-fiber bonding of these DG. Additionally, the increased DG content resulted in a decrease in the density of the handsheets, which may be attributed to the fact that more particles were retained inside the handsheets, resulting in a bulkier structure of the fiber network. In principle, if a filler material tends to brace fibers apart from each other within the paper, this can weaken the potential bonding among fibers and reduce the density of handsheets, with the increase on DG content.

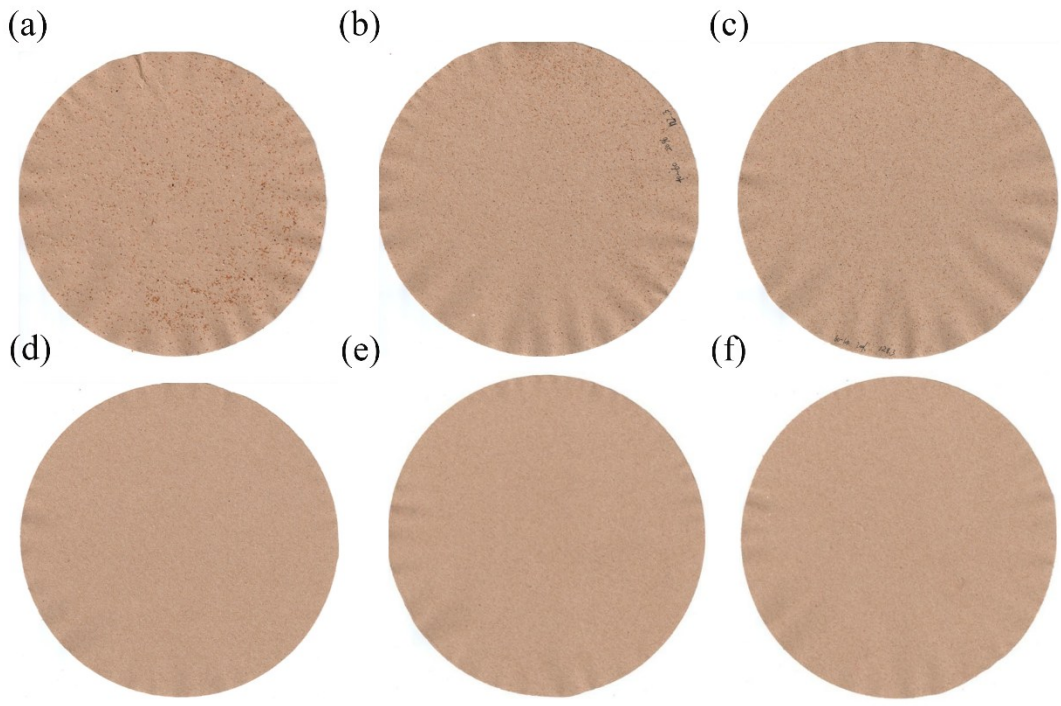

Fig. 4. Digital photograph of handsheets following DG filling with various screens, (a) > 40 mesh; (b) 40-60 mesh; (c) 60-100 mesh; (d) 100-140 mesh; (e) 140-200 mesh; (f) < 200 mesh

Table 5. Physical Properties of Handsheets with Different Screened DG Filling

Property	Filler addition (%)	> 40-mesh	40-60 mesh	60-100 mesh	100-140 mesh	140-200 mesh	< 200 mesh
Density ($g \cdot cm^{-3}$)	0	0.45	0.45	0.45	0.45	0.45	0.45
	5	0.25	0.40	0.42	0.46	0.46	0.46
	10	0.25	0.34	0.41	0.44	0.45	0.47
	15	0.21	0.34	0.41	0.43	0.45	0.46
	20	0.23	0.30	0.40	0.43	0.44	0.46
Tensile index ($N \cdot m \cdot g^{-1}$)	0	23.92	23.92	23.92	23.92	23.92	23.92
	5	19.38	23.75	25.15	25.45	23.45	23.07
	10	18.63	20.23	21.73	22.98	20.67	20.57
	15	17.91	18.46	18.85	21.71	20.16	20.23
	20	16.56	17.87	18.37	18.75	18.43	18.16
Ring crush index ($N \cdot m \cdot g^{-1}$)	0	8.31	8.31	8.31	8.31	8.31	8.31
	5	6.93	7.36	7.65	7.95	7.57	7.86
	10	6.24	6.83	7.51	7.72	7.54	7.35
	15	6.07	6.45	6.24	6.59	6.82	7.07
	20	5.25	6.13	6.15	6.23	6.21	6.59

For the physical properties of handsheets, the effects of the DG screen and content are listed in Table 5. The tensile index of handsheets generally increased with the decrease in DG size. Furthermore, the tensile index of handsheets consistently peaked using DG particle sizes of 100 to 140 mesh. However, high filler proportion gradually weakened the bonding force between the fibers, thereby reducing the tensile strength of the handsheets. When extremely tiny DG (140- to 200-mesh and exceeding 200-mesh) was incorporated, the hydrogen bonding among cellulose fibers was scarcely disrupted, due to the small particle size of DG (Li *et al*. 2016; Przybysz *et al*. 2016). The result of physical properties confirmed the synergistic effect of DG particle size and proportion on the tensile index of handsheets (Song *et al.* 2018).

The ring crush index serves as a critical parameter for evaluating the strength characteristics of the corrugating medium. Table 4 listed the ring crush index of handsheets with varied DG particle sizes and proportions. The ring crush index of handsheets increased with the decrease in DG particle size and the increase in DG proportion, suggesting that smaller DG particles were more advantageous for enhancing the ring crush index of handsheets. The decline in the ring crush index with the increased DG proportion was attributed to the disruption or weakening of the fiber network. The adverse effect of DG incorporation on the ring crush index of handsheets was more serious when a large proportion of DG was adopted.

The effect of DG (100- to 140-mesh) on the physical properties of handsheets was further investigated and compared with conventional fillers (PCC and talc powder), as shown in Fig. 5. The tensile index and ring crush index of handsheets decreased with the increased proportion of filler. Handsheets filled by DG exhibited the highest tensile index and ring crush index compared to the other kinds of fillers at the same added amounts. In addition, DG-filled handsheets with a DG proportion of 5 wt% exhibited a higher tensile index when compared with unfilled handsheets. This was attributed to the organics in DG, which afforded additional positions for the formation of hydrogen bonding to enhance the

interfacial compatibility between cellulose fiber and filler (He *et al.* 2016; Przybysz *et al.* 2016). The 100- to 140-mesh was used in the following hybrid filler, considering its satisfactory effect on the physical properties of the handsheets.

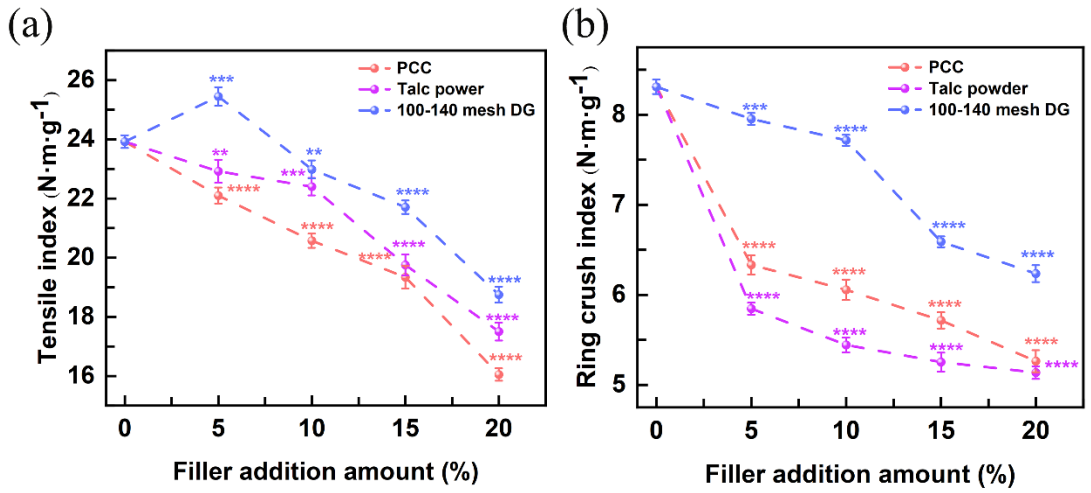

Fig. 5. Effect of filler on physical properties of handsheet, (a) tensile index; (b) ring crush index. Note: Statistical significance was analyzed using an unfilled handsheet as the control group. Data are presented as mean ± SD (n = 3).**P < 0.01, ***P < 0.001, ****P < 0.0001

The hybrid fillers, DG/PCC and DG/talc, with varying proportions and a total filling amount of 20%, were used to fill the handsheets for further investigation into the effect of DG on the physical properties of the handsheets. As shown in Fig. 6, with the increased DG proportion, the tensile index and ring crush index of the obtained paper increased consistently. The maximum tensile index (18.8 $N \cdot m \cdot g^{-1}$) and ring crush index (6.23 $N \cdot m \cdot g^{-1}$) of handsheets was achieved with a DG: PCC ratio of 20:0 (*i.e.*, the obtained handsheets was consisted with 20% of DG, 0% of PCC, and 80% of recycled fibers). The similar variation trend on the tensile index and ring crush index of handsheets, filled by DG/talc, can also be observed (Fig. 6b and d). In general, when DG was incorporated into paper either with PCC or talc powder, the increased DG particle ratio was beneficial for improving both the tensile index and ring crush index of handsheets, owing to the present organics in DG, which were capable of forming hydrogen bonds with the cellulose fibers (Song *et al.* 2022; Li *et al*. 2025a). Hence, DG can be considered as a potential organic filler in the paper industry.

Future Work

This work investigated DG using for the manufacture of paperboard, which can provide an example for the organic waste with graininess. These organic particles end up in the paper rather than remaining in the water. Sieving for size classification can also give a typical example for these organic waste particles using to produce paper/paperboard. For instance, large-sized DG is not suitable to produce paper products using in the surface of materials, but can be used in the eye-invisible part of paperboard. One such application would be in the middle ply of low-grade folding boxes, as are commonly used for breakfast cereals. Such plies need to be bulky and cheap. They don't have to be smooth, since they are the middle ply. Fortunately, the future work does not need to consider PCC or talc, due to the enough of a comparison with those fillers in this work. The contrast was so obvious that it was not even important to know how much of the different fillers were retained.

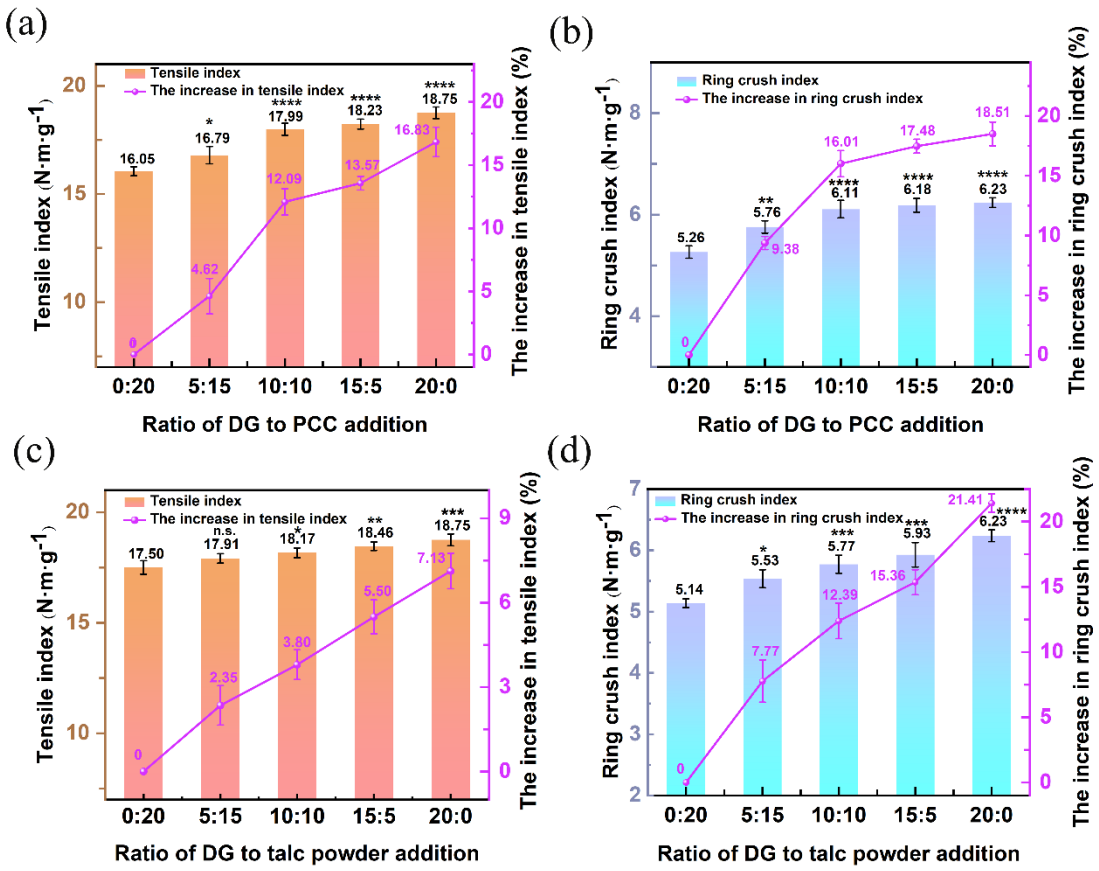

Fig. 6. The influence of DG ratio in the mixed filler (DG/PCC) on the (a) tensile index and (b) ring crush index of handsheet. The influence of the DG ratio in the mixed filler (DG/talc powder) on the (c) tensile index and (d) ring crush index of handsheet. Note: For significance analysis, handsheet without DG filler was used as the control. Data are presented as mean ± SD (n = 3). *P < 0.05, **P < 0.01, ***P < 0.001, ****P < 0.0001, while "n.s." indicated that significant difference was not observed (P >0.05T)

CONCLUSIONS

1. The primary components of distiller's grains (DG) were found to include lignocellulose. The structural characteristics were consistent with cellulose Iβ, proteins, and fats. With the decrease in particle size of DG, the lignocellulose content (cellulose, hemicellulose, and lignin) of DG was decreased, while the fat and protein content of DG was increased. In addition, SiO_2 was detected in DG, according to the results of X-ray diffraction (XRD) and Fourier transform infrared (FTIR) spectrometry.
2. The particle size of DG (varied screen) was correlated negatively with the density of the handsheets, and positively with the tensile index and ring crush index of the handsheets. Among them, DG with a screen of 100 to 140 mesh was a satisfactory option for filler.
3. For paperboard products requiring relatively high strength, DG was found to be superior to traditional fillers (PCC or talc) due to the better tensile index and ring crush index of the DG-filled handsheets, compared to PCC- or talc-filled handsheets.

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