

Application of Corn Straw Substitute Fiber in Gravure Printing

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Abstract

 Packaging has become an indispensable part of our daily lives, without which it would bring great inconvenience and insecurity to consumers. The current global trend is to reduce the amount of packaging waste by recycling the same materials or using renewable waste raw materials. After each year's corn harvest, a large amount of straw is discarded in fields, and these corn straws are becoming an alternative source for wood fibers in green packaging. This study evaluates the feasibility of using straw fibers for gravure printing packaging by observing the quality parameters of gravure printing and the ink penetration into the printing substrate. From the experimental results, it can be seen that the gravure printing ink penetrates too much into the printing substrate, resulting in uneven surface ink distribution. The viscosity of the ink and the printing process itself have a significant impact on this. Therefore, gravure printing needs to add additional coatings or more fillers in the paper structure to improve it.

Keywords: corn straw; wood fiber; gravure printing; alternative; eco-friendly packaging

Introduction

 Packaging materials refer to the items used for wrapping or protecting products, whose main function is to protect products from damage and provide identification and promotion [1]. In addition, packaging materials can also provide important information about the product itself, as well as regulatory and legislative requirements. Packaging materials play an indispensable role in the use of various products. According to statistics, almost 50% of printed products are used for packaging purposes [2].

 With the increasing awareness of environmental protection, consumers have with the increasing awareness of environmental protection, consumers have a growing demand for environmentally friendly and healthy packaging that is acceptable. This demand can reduce waste generation, increase reuse and recycling, and comply with the global three R principles (reduce, reuse, and recycle) [3]. However, with the development of urbanization and industrialization, the consumption of packaging materials continues to increase, bringing negative impacts on the environment such as soil, water, and air pollution. The amount of waste generated each year is also constantly increasing, with an estimated increase to 2.4 billion tons by 2026 and further increase to 28 billion tons per year by 2050. One-third of this waste comes from Asia, especially China. These wastes have become the biggest problem faced by contemporary people and a challenge for future generations [4]. To address this issue, the world is moving towards sustainability, bio-based, and circular economy. Due to regulatory measures prohibiting the use of disposable plastic products, the use of green packaging is expected to reach up to \$30 trillion by 2030. The increased global environmental awareness has also promoted the development of bio-based industries such as paper products, graphic packaging industry, bioplastics, biocomposites, biofuels, biochar, bioenergy (electricity and natural gas), biochemistry, and renewable lubricants [5]. At the same time, the global increase in demand for natural fiber materials has led to a faster rate of deforestation each year, which also means that non-wood fibers become one of the most important sources of fiber material alternatives in the 21st century [6].

 In addition to being used as packaging materials, nonIn addition to being used as packaging materials, non-wood biomass also adds value to crops by utilizing residues. The diverse characteristics and chemical composition of these residues provide potential for alternative wood raw material sources. Furthermore, the pulp and paper industry is a good starting point for developing lignocellulosic biorefineries, as they possess the necessary technology and infrastructure, as well as experience in converting lignocellulosic biomass. Lignocellulose is the main component of plants and the most abundant type of terrestrial biomass [7]. Lignocellulosic biomass is mainly composed of cellulose (40-60%), hemicellulose (10-40%), and lignin (15-30%), with small amounts of extractives, proteins, and inorganic compounds [8]. Lignocellulosic components can be found in various biomasses, including wood and non-wood biomass, the latter including plant residues and agricultural crop residues, among others [9].

 The content of cellulose in paper substrate has a positive impact on paper strength and makes the fiber bundles easy to bind with natural and synthetic inks. Meanwhile, hemicellulose is responsible for the water absorption of plant fibers and reduces the internal stress within the fibers [10]. Cellulose is a linear ordered polymer formed by the connection of D-dehydroglucose units through β-1,4-glucosidic bonds, with a degree of polymerization ranging from 15 to 10,000. Lignin is a three-dimensional phenolic acid polymer formed by the ortho-, meta-, and para-hydroxylation of corresponding p- hydroxyphenyl alcohol groups [9]. Plant fibers with a holocellulose content greater than 33% and lignin content less than 30% are considered promising candidates for papermaking [11].

 Currently, only a small portion of paper (approximately 12%) is made from non- wood fibers, which are referred to as "tree-free" fibers. These fibers are mainly divided into two categories: agricultural residues and crops. In recent years, there has been an increase in research reports on the quality and usability of alternative fibers in paper production. Most non-wood resources include straw, bagasse, bamboo, hemp, sisal, banana stalks, etc. [12]. It is worth noting that in northern China, besides wheat straw, corn stalks are also an important non-wood fiber resource. The total planting area of corn in China is no less than that of wheat, and the annual output of corn stalks is also considerable. However, according to the consumption structure of papermaking fiber raw materials in China, as of 2022, corn stalks have rarely been involved in pulp and papermaking, at least not to the same extent as wheat straw in the Chinese paper industry. This has led to a serious waste of China's limited papermaking fiber raw materials resources.

 Corn stalks have a high content of cellulose and low lignin, making them well- suited for use in the pulp and paper industry. Under the same cooking conditions, corn stalk pulp obtained using the alkali-anthraquinone method has a higher yield compared to wheat straw pulp. Corn stalks are pure natural and high-quality raw materials for papermaking, composed of fiber components such as cellulose, hemicellulose, and lignin. Due to the high content of cellulose in corn stalks, it is beneficial for improving the yield of pulp and the strength of paper. Additionally, the moderate content of hemicellulose aids in water absorption by the pulp and facilitates refining. Furthermore, the lower lignin content in corn stalks is advantageous for the cooking and bleaching processes, thereby enhancing the quality of paper [13].

 In this study, we chose corn stalks as an alternative fiber source forIn this study, we chose corn stalks as an alternative fiber source for papermaking because a large amount of corn stalks are discarded in the field after harvesting every year [14]. The research mainly analyzed the printing substrate made from discarded corn stalk fiber and the green packaging printed on this substrate using the intaglio printing process. Intaglio printing was selected for this study because it is one of the main printing processes used for packaging printing on absorbent substrates.

Research Methods

 This study is divided into the following stages: (1) pulping of corn stalks using the alkali-anthraquinone method; (2) production of paper substrate from the corn stalk pulp; (3) intaglio printing on the paper substrate; and (4) evaluation of the printed quality.

Pulping of Straw Using the Alkali-Anthraquinone Method

 After corn harvest, agricultural residues are collected and the corn stalks are converted into woody fiber pulp using the process conditions summarized in Table 1.

Table 1: Conversion of Straw into Woody Fiber Pulp.

Chlorine or alkali dosage		Pulp Consistency	<i>pH</i> value	Temperature	Time
C	65%		1.8	24° C	30
stage					minutes
E	2%	.5	11	60° C	80
stage					minutes
H	35%	.5	9	40° C	90
stage					minutes

Table 2: Bleaching Process Parameters.

 In this study, sodium hydroxide was used as the pulping liquid in this study, sodium hydroxide was used as the pulping liquid, which is a major chemical for non-wood alkaline pulping [15]. After the pulping process, the slurry was washed with tap water in two stages to remove soluble substances from the slurry, and finally beaten to separate fibers from the aqueous suspension.

Production of Paper from Straw Pulp

The paper substrate produced had a weight of 52.5 ± 0.5 g/m² and was prepared by mixing recycled wood pulp with corn stalk pulp in a ratio of 6:4. A reference sample (X) made entirely of 100% recycled wood pulp was also prepared in an identical manner for comparison purposes in this study.

 Depending on the species of plant, fibers may differ in length, width, fineness or microscopic structure, as well as chemical composition. Longer fibers can provide greater strength to paper, while shorter fibers can increase the opacity and smoothness of the paper surface. Cork fibers (from coniferous trees) can be up to 5 millimeters long, while hardwood (from deciduous trees) has a less uniform anatomical structure and contains fewer fibers with an average length of about 1.5-2 millimeters [8]. The length of corn stalk fiber is roughly equivalent to the average length of hardwood fiber [16].

To better understand the structure of the paper substrate, the SEM images of the analyzed paper substrates are included in Figure 1.

Figure 1: SEM images of paper substrates: (left) X; (right) Y.

 Table 3 lists the characteristics of the paper substrates, wherein paper labeled as X is made from 100% recycled wood pulp and serves as a reference sample in this study. Paper labeled as Y is composed of 40% corn stalk pulp and 60% recycled wood pulp.

Paper Substrate	Thickness (µm)	Air Permeability (mL/min)	Ash Content (%)	Roughness (μm)	Surface Free Energy (mN/m)
	91.27 ± 2.47	234.18±0.80	4.94 ± 0.22	4.09 ± 0.34	40.87
	99.67±15.32	402.65 ± 2.45	3.82 ± 0.07	4.61 ± 0.51	42.64

Table 3: Characteristics of paper substrates.

 A video camera instrument was used to determine the water absorption and surface free energy of each paper substrate based on printing paper. To calculate the surface free energy, contact angle measurements were performed using 10 standard liquids (glycerol, water, formaldehyde, and dimethyl oxide) according to the OWRK method with a volume of 1 μ L and a dose rate of 5 μ L/s [17].

Gravure printing paper substrate

 Due to the high growth rate of packaging printing (over 6%) each year, gravure printing is mainly used for printing long-run luxury goods, publications, and packaging [18]. This study analyzes the practicality of waste corn stalk fiber paper in absorbent paper substrates, primarily for gravure printing on absorbent paper substrates.

 The printing on the produced paper substrate was carried out using a gravure printing equipment. Gravure printing uses a printing plate engraved with a laser, with a mechanical hardness (HS) of 83 Shore for the printing rollers. Printing was performed at a speed of 200 m/min, at a temperature of 123 °C and a relative humidity of 52% in full color tones. When printing on the produced paper substrate, Cyan (C), Magenta (M), Yellow (Y), and Black (K) inks were used for full-color printing.

Table 4 shows the viscosity values of gravure printing inks (C, M, Y, and K) measured at a temperature of 123 °C and a relative humidity of 52%. Viscosity is a property of liquids that describes the tension in liquid flow due to differences in liquid layer or its adhesion to surfaces. In other words, viscosity indicates the degree to which ink resists movement or flow. Liquids with high viscosity are also thicker and less fluid. The viscosity of a liquid can be described in two ways: dynamic viscosity and kinematic viscosity. Dynamic viscosity is determined by the coefficient η, which is determined by the force (F) required per unit area (A) to achieve a unit velocity difference between two parallel layers at a distance (x), while the kinematic viscosity (ν) of a Newtonian fluid is the ratio of dynamic viscosity (η) to fluid density (ρ) [19].

Kinematic viscosity can also be calculated based on flow time using the following equation.

$$
\nu = 4.57t - \frac{452}{t}
$$

Printing Ink	Kinematic Viscosity (mm²/s)
	147.54 ± 5.67
м	142.62 ± 7.83
	$136.98 + 4.47$
	284.27 + 8.75

Table 4: Viscosity of gravure printing inks (C, M, Y, and K).

Note: The above data were measured using a DIN 4 cup.

 Figure 2 shows the microscopic images of the printed product ink in cyan color on the X and Y printing substrates to observe the print quality obtained by gravure printing.

Print quality assessment

 In this study, the usability of waste corn stalk fiber paper for green packaging was analyzed by evaluating the print quality. The printed products on the paper produced by the new process were compared to those on paper made entirely from recycled wood pulp. The evaluation of print quality was based on several analytical parameters: mottling and graininess value, overall optical ink density, and ink penetration depth [20].

Graininess analysis

 Various models have been developed to define the uneven coverage of printing ink on a substrate, which are based on similar or identical factors. The main factors that lead to uneven coverage of printing ink on a substrate include the interaction between the printing machine and the ink, the interaction between the printing machine and the substrate, and the interaction between the printing ink and the printing substrate. The reasons for uneven coverage of printing ink on a substrate can vary and are often related to factors such as surface roughness, porosity, absorbency, surface energy of the substrate, viscosity of the ink, printing pressure, printing speed, and environmental conditions [21].

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 The graininess of the solid surface in the designated printing area refers to the irregular optical density variation with a spatial dimension smaller than the specified slice size. According to the International Protocol for Printing Quality (ISO 13660), this protocol uses a slice size of 42.3 μ m × 42.3 μ m to calculate the particle size. According to ISO 13660, the image area under inspection is divided into 100 equal areas (1.27 mm × 1.27 mm), wherein 900 reflectance measurements are performed in small non- overlapping square areas (42.3 µm × 42.3 µm) to obtain the amplitude of optical density variation. The graininess is defined by the following formula:

$$
G = \sqrt{\frac{\sum_{i=1}^{n} \sigma_i^2}{n}}
$$

Where σ_i is the standard deviation within the slice, *i* is the slice number, and *n* is the total number of slices.

Mottling analysis

 According to the ISO 13660, the analysis of uneven coverage of printing ink on a substrate is divided into two levels: at the micron level, from 42 μm to 1270 μm, which is called graininess; at the macro level, greater than 1270 μm, which is called full-tone Mottling. Mottling is defined as the standard deviation of the average reflectance coefficient over 100 uniform slices, i.e., the variation of optical density from slice to slice.

$$
M = \sqrt{\frac{1}{n-1} \sum_{i=1}^{n} \left(m_i - \left(\frac{1}{n} \sum_{i=1}^{n} m_i \right) \right)^2}
$$

Where mi is the reflectance coefficient, *i* is the slice number and *n* is the total number of slices.

To perform particle mottling and graininess analysis, we used a digital microscope [22].

Overall optical ink density analysis

 The overall optical ink density (Di) parameter was also considered in this study to describe the thickness of the ink film on the paper substrate. Optical densitometer measurements were performed on all gravure prints through a 3-mm aperture using an optical densitometer. Higher values of optical ink density indicate higher ink coverage or higher pigment concentration and optical contrast compared to the substrate [23].

$$
D_i = \log \frac{I_0}{I}
$$

Ink penetration depth analysis

 The penetration of ink into the paper substrate is a complex issue, and therefore there are various methods to determine the extent of ink penetration into the paper interior. These methods can be differentiated based on sample preparation, image capture, sample size and resolution, as well as non-destructive or destructive methods. The most common destructive methods involve microscopic analysis using scanning electron microscopy (SEM), secondary ion mass spectrometry (SIMS), focused ion beam instrument (FIB), or confocal laser scanning microscopy (CLSM). However, due to other factors that affect the accuracy of results and limited observation range, non- destructive methods are more reliable.

 In this study, we used the spectral reflectance based on KubelkaIn this study, we used the spectral reflectance based on Kubelka-Munk theory to determine the ink penetration depth [24].

$$
H_p = \frac{\ln \frac{(1 - R_0 * R_\infty)(1 - R_p * R_\infty)(1 - R_q/R_\infty)}{(1 - R_0/R_\infty)(1 - R_p/R_\infty)(1 - R_q * R_\infty)}}{\ln \frac{(1 - R_0 * R_\infty)}{(1 - R_0/R_\infty)}} \times D
$$

This method involves measuring the reflectance values (R_{α}) of the unprinted paper against a white background, the reflectance values (R_o) of the unprinted paper against a standard black background, the reflectance values (R_p) of the printed paper on top of the unprinted paper, and the reflectance values ($\rm R_q$) of the reverse side of the printed paper on top of the unprinted paper. The ink penetration depth $(\mathtt{H}_{\mathtt{p}})$ is then calculated based on the average thickness (D) of the unprinted paper.

 To obtain the required reflectance values (R), we used a spectrophotometer and made measurements at a wavelength of 457 nm. We then derived the ink penetration depth $\left({\rm H}_{p} \right)$ from the average of 50 spectral measurements for each printed piece.

Research results

 In this study, the feasibility of green packaging paper printing using fiber paper made from waste straw was analyzed. We compared the ink penetration depth parameter with the characteristics of the printing substrate, as well as the properties of the ink such as its water absorption, surface tension, and viscosity, and equivalent parameters for gravure prints (Mottling and Graininess value) to evaluate the production quality of gravure prints [25]. Table 5 lists the attributes of gravure prints with overall optical density.

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 In addition, we also used an image analyzer to analyze the number of In addition, we also used an image analyzer to analyze the number of fibers in the printing substrate. Statistical methods were used to quantify the differences in form quality. The results of the analysis obtained on 10 samples of each printing substrate are shown in Figure 7.

Discussion of research results

 The composition and surface properties of the printing substrate have a significant impact on the ability of ink to penetrate the structure of the printing substrate. For example, coated paper has fewer pores and is more difficult to penetrate, making it harder to absorb ink compared to uncoated paper. This results in more diffusion of ink on the surface of the printing substrate, leading to a more uniform distribution of ink on the printed surface layer [26]. In addition, uncoated paper has higher porosity and a rougher surface. This explains why when using this type of paper, printing ink penetrates into the pores of the paper rather than staying on the surface, resulting in a less glossy print and lower density values for the printed pattern [27].

 This study also found that adding non-wood fibers to the pulp can significantly improve air permeability by 71.7%, roughness by 12.7%, and reduce water absorption by 53.2% (see Table 3 and Figure 4).

 As can be seen from Figure 3, the penetration depth values As can be seen from Figure 3, the penetration depth values of gravure ink determined using a non-destructive method based on Kubelka-Munk theory are different in different printing substrates (Y substrate with 40% straw fiber addition and X substrate without straw fiber). When observing the prints obtained using gravure printing, it is noteworthy that the ink penetrates deeper into the Y printing substrate (average penetration value of 59.05%). For gravure prints using UV curable ink, a larger penetration of ink into the Y printing substrate is also observed when using ink with higher viscosity and drying through UV radiation, so it can be concluded that the Y substrate with waste straw fiber leads to greater ink penetration into the substrate.

 The viscosity values of the gravure inks for cyan, magenta, and yellow are very similar (ranging from 147.54 to 136.61 mm2/s), while the high viscosity value of black ink (284.23 mm2/s) does not significantly affect the penetration of ink into the printing substrate (Figure 5). Penetration of ink deep into the substrate was observed in all prints, indicating that paper containing waste straw fiber is permeable to liquid gravure ink. However, when low-viscosity printing ink is applied to uncoated porous paper substrates, strong ink penetration occurs [28].

 For gravure prints, the integral optical density values are very similar, ranging within a variation of 0.08, with the highest values obtained for prints on printing substrates made solely from recycled pulp (0.9 for yellow prints to 1.05 for cyan prints) (Table 5), which indicates that the high permeability of low-viscosity gravure ink does not have a significant impact on the layer of ink retained on the surface of the printing substrate.

 The images printed using magenta and cyan inks were found to have higher values for mottling and graininess compared to the images printed using other inks. Specifically, the graininess ranges for magenta prints were 0.14 to 0.16 and for cyan prints were 0.12 to 0.17; the mottling value ranges for magenta prints were 0.12 to 0.14 and for cyan prints were 0.11 to 0.15. In contrast, the graininess ranges for yellow prints were from 0.05 to 0.08 and for black prints were from 0.06 to 0.07; the mottling value ranges for yellow prints were from 0.06 to 0.08 and for black prints were from 0.06 to 0.07. According to Karlovits et al. [29], different surface free energies and different ink penetration depths do not directly affect the uneven ink coverage on the substrate when printing with low-viscosity inks.

 Since the chemical composition of the printing substrate plays a key role in the penetration of ink into an uncoated printing substrate, the amount of fillers and cellulose fibers determines the permeability of the substrate itself. As can be seen from Figure 7, the printing substrate with 40% straw fiber addition contains more fibers larger than 0.12 mm compared to the printing substrate made solely from recycled wood fibers.

 Considering the number and length of fibers analyzed in the paper, the ink will first move along the cellulose fibers before penetrating the pores between the filling fibers [30]. Therefore, for substrates containing straw fibers, the penetration force of ink is stronger.

 According to the research conducted, printing products that use According to the research conducted, printing products that use straw fibers added to paper substrates cannot be directly used for gravure printing of green packaging because the ink penetrates deeply. However, by adding modified straw stalks to the pulp, as the amount of modified straw stalks increases, the gaps between pulp fibers are gradually filled. The straw stalks penetrate into the interior of the gaps, eliminating the original air and anchoring the fiber bundles together. Therefore, by providing these substrates with an additional coating or increasing the filler content in the pulp, it is possible to adapt to the requirements of gravure printing for green packaging.

Conclusions

Based on the research results, we can draw the following conclusions:

- By adding straw fibers to the pulp of printing substrates, it is possible to increase the surface free energy of the substrate. The addition of straw fibers can increase the surface free energy of the printing substrate by up to 2.33%, and compared to printing substrates without straw fibers, the average penetration value of ink can be increased by up to 23.92%.
- The addition of straw fibers leads to an increase in the depth of ink penetration into the printing substrate. This is due to a reduction in ash content and an increase in the number of hydrophilic fibers in the printing substrate.
- In gravure printing, the higher the ink concentration, the better the printing results.
- In gravure printing, the high penetration depth of ink into the substrate results in uneven coverage of the surface by the ink.

 Based on the results of this study, it can be concluded that fibersBased on the results of this study, it can be concluded that fibers from waste corn stalks have the potential to become a raw material for green packaging printing paper. However, when printing these substrates made of printed fibers using gravure printing, additional coating or more fillers are required.

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