# Making Ultra-thin High Density Fiberboard Using Old Corrugated Container with Kraft Lignin

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Ultra-thin high-density fiberboards (HDFs), a newly developed variety of fiberboards, broaden and extend the applications of medium thick medium- and high-density fiberboards and are capable of replacing cardboards for most applications. Old corrugated container (OCC) is an important packaging solid waste. The mechanical strength of OCC deteriorates after repeated recycling processes. Application of OCC fibers for value-added ultra-thin HDFs can be of much interest. Because the OCC fibers have more surface area than the wood particles, the resin coverage per surface area of the OCC is much lower than wood particles during panel board formation. Therefore, the performance of the OCC fiber-based board is poor and the resin adhesive consumption is high. To overcome these problems, a novel method of using OCC to make ultra-thin HDFs was developed and investigated. In this work, the OCC was shredded and pulped before making the ultra-thin HDFs. To protect consumers from exposure to harmful formaldehyde, kraft lignin was used as a binder. The target density and thickness of the ultra-thin HDFs were 1.0 kg/m$^3$ and 2 mm respectively. The resulting ultra-thin HDFs were evaluated for their physical and mechanical properties. Comparisons with the Chinese Standards for Wet-Process Fiberboards are presented. The results indicate that OCC could be a potential sustainable resource for ultra-thin HDFs production**.**

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## INTRODUCTION

Corrugated container is the most popular paper-based packaging material used for the storage, shipping, and distribution of goods (Frank 2014). The global corrugated board production was about 3.0 million tons in 2017, and it increased at a pace of 5% annually (Miao *et al.* 2020). After use, old corrugated containers (OCC) are theoretically able to be recycled up to 6 times for papermaking (Chen *et al.* 2012). However, repeated pulping and papermaking processes lead to the breakdown of recycled fibers, which get too short and weak to make paper or paperboard products (Guo *et al.* 2011; Chen *et al.* 2012). The OCC is mainly composed of cellulose, with low content of hemicellulose, lignin, and other impurities, such as minerals, adhesives, coating polymers, inks, *etc.* (Wan *et al.* 2011; Sangtarashani *et al*. 2020), and therefore it has the potential to be utilized as alternative raw material for panel board manufacture.

Fiberboard is a fibrous-felted, homogeneous panel made from lignocellulosic fibers that are combined with a synthetic resin or other suitable bonding system and then bonded together under heat and pressure (Mancera *et al*. 2012). Urea-formaldehyde and phenolformaldehyde are primary adhesives used in fiberboard manufacturing because of their low cost, non–flammability, hardness, water solubility, and excellent adhesion to wood (Boran *et al*. 2011; Cavdar 2020). However, formaldehyde is released from fiberboard during manufacturing and during use, which is a serious issue to human health (Diop *et al*. 2017). Using formaldehyde-free adhesives from natural renewable resources is one of the best ways to address these issues (Prasittisopin and Li 2020). Lignin can be used as a binding agent for fiberboard production (Ang *et al*. 2019). Several studies have been conducted on the replacement of conventional formaldehyde-based resins by various kinds of lignin to fabricate fiberboards (Angles 2001; Mancera *et al*. 2012; Velasquez *et al*. 2003). The results showed that fiberboards bonded with kraft lignin yielded the best physical and mechanical properties (Mancera *et al*. 2012; Velasquez *et al*. 2003). Kraft lignin is a low-cost byproduct of the pulp and paper industry. The annual output of kraft lignin in the world can reach 130 million tons (Gellerstedt 2015; Zhang *et al*. 2021). Currently, kraft lignin is mainly burned as fuel for recovering chemicals in the pulp and paper industry (Hu *et al*. 2018; Bernhardt *et al*. 2021).

Depending on the density, fiberboards can be classified into low-density fiberboards, with densities less than 400 $kg \cdot m^{-3}$, medium-density fiberboards, with densities ranging from 400 to 900 $kg \cdot m^{-3}$, and high-density fiberboards (HDFs), with densities ranging from 900 to 1100 $kg \cdot m^{-3}$ (Dominguez-Robles *et al*. 2018). HDFs are one of the most widely used wood-based products worldwide, with a range of thicknesses from 1.5 to 25 mm (Badin *et al*. 2018; Antov *et al*. 2021). Thin HDFs are less than 5 mm in thickness, while the thickness of ultra-thin HDFs is below 2.5 mm. Ultra-thin HDFs are a new variety developed in the past two decades. Ultra-thin HDFs widen and extend the applications of medium- and high-density fiberboards and are capable of replacing cardboards for most applications, such as decorated panels, electronic circuit boards, box boards, container boards, door skins, musical instruments, vehicles, ships, and medical devices (Hunt *et al*. 2015; Guan *et al*. 2016). Production of cardboards features very costly one-time investment and complex manufacture process. Particularly pulping process involves chemicals such as sulphite solution, sodium hydroxide solution, and lime solution, which cause severe water pollution (Smook 2016). The cost to treat sewage is very expensive. Production of ultra-thin HDFs dispenses with these issues. Ultra-thin HDFs are very promising and have vast market potential, with the profit much higher than medium thick medium- and high-density fiberboards (6 to 30 mm). Currently, medium- and high-density fiberboard production lines worldwide mainly produce conventional medium thick fiberboards, which are mainly used to make furniture, wood flooring and interior decoration. Up to now, only a few plants worldwide produce ultra-thin HDF. As ultra-thin HDFs have become very popular in the panel markets, these plants have experienced booming production and sales and gained remarkable economic benefits.

Up to now, studies dealing with OCC as raw material in the manufacture of panel board have commonly been carried out by cutting OCC into small pieces, then used alone or in mixture with wood particles blending with resin adhesives before being hot-pressed (Rassam 2008; Abdolzadeh and Doosthoseini 2009; Kordkheili *et al.* 2012; Lykidis *et al.* 2012; Mohammadkazemi and Doosthoseini 2015). The OCC is composed of two outside liners glued with a corrugated medium in between using starch adhesive. Compared with synthetic resin, the bonding force of starch adhesive is weak. Consequently, the produced panel boards exhibit poor internal bonding strength. Moreover, the OCC is prone to warp after mixing with water-based adhesive resin. The amassed and intertwined OCC pieces resist uniform mixing and forming. It is difficult to produce a homogeneous mat. As a result, the resulting boards exhibit various performances (Rassam 2008; Abdolzadeh and Doosthoseini 2009; Kordkheili *et al.* 2012; Lykidis *et al.* 2012). In addition, OCC tends to rebound after being compressed. As the amount of OCC increased in the OCC/wood mixture, the performances of the resulting boards deteriorated (Rassam 2008). The surface area of the recycled waste paper is much greater than that of wood particles. As a consequence, a lesser amount of adhesive resin was distributed onto the unit area of the waste paper. In addition, the inorganic components of OCC such as clay, calcium carbonate, *etc.*, impair bonding of the synthetic resin. A high amount of synthetic resin was required, and thus the cost of processing increased (Rassam 2008; Kordkheili *et al.* 2012). To address the above issues, the authors developed a novel process to produce ultra-thin HDFs using OCC. The OCC was first torn into small pieces and then disintegrated into separated fibers in water. The disintegrated fibers were mixed with kraft lignin and filtered to form a wet mat. Then, the wet mat was hot pressed to form fiberboard. No data have been published on this approach. Therefore, the objective of this study was to investigate the feasibility of using OCC as raw material for manufacture of ultra-thin HDFs bonded with kraft lignin.

## EXPERIMENTAL

### Raw Materials

The commercial kraft lignin M0010-25G (Mingcheng Chemical Products Co., Ltd., Nantong, China), in powdered form, was used in this study. The OCC, with moisture content of 8.3%, was collected from a local grocery store. The OCC was from standard corrugated boxes.

### Fiberboard Preparation

After removing all the impurities and non-fibrous materials (mainly tapes and staples), the OCC was torn into small pieces of about 25 × 25 $mm^2$ and weighed. The OCC was then soaked in water at ambient temperature for 24 h and subjected to slurrying by a slusher (Model TD 6-23; Xianyang Tongda Light Industrial Equipment Corporation, Shaanxi, China) at a 5% consistency for 30 min. Then, the pulps, along with desired amount of lignin, were disintegrated at 2,000 rpm for 10 min to ensure even fibrillation and lignin distribution. The mixture was then diluted to 1.2% consistency at ambient temperature. The amounts of OCC and kraft lignin used in the study are shown in Table 1.

**Table 1.** Kraft Lignin Dosage and the Amounts of OCC and Kraft Lignin Used in the Study

| Kraft Lignin Dosage (%) | 10 | 15 | 20 |
|---|---|---|---|
| Kraft Lignin Amount (kg) | 0.0045 | 0.0068 | 0.0090 |
| OCC Amount (kg) | 0.0442 | 0.0417 | 0.0393 |

The pulp was added into a laboratory paper sheet former. Excess water was removed *via* a filter and vacuum suction to retain the fibers and lignin. The wet webs were transferred into a laboratory mold of 150 mm × 150 mm and hot-pressed in accordance with a three-phase hot press schedule at target temperature. During the first phase, the mat was pressed at 15 MPa for 90 s, and during the second and third phases, the mat was pressed under 3 MPa for 90 s and 1.0 MPa for 60 s, respectively. Then, the fiberboard was gradually decompressed. The lignin levels were kept at 10%, 15%, and 20% levels based on the oven-dried weight of the OCC fibers. The hot press temperatures were 210 and 230 ℃. The board thickness achieved was 2 mm and the target density was 1000 kg/m$^3$.

## Fiberboard Evaluation

After manufacture, the fiberboards were conditioned at 20 ℃ and 65% relative humidity for 2 weeks. Then, the boards were sawn into test specimens and tested according to the Chinese national test standard GB/T 17657 (1999) for density, internal bonding strength (IB), modulus of rupture (MOR), modulus of elasticity (MOE), 24 h water absorption (WA), and thickness swelling (TS). Analysis of variance (ANOVA) and Duncan's mean separation tests were used to statistically analyze the data obtained with a SPSS software (SPSS Inc., Version 19, Chicago, IL, USA).

# RESULTS AND DISCUSSION

The mechanical properties and water resistances of the produced ultra-thin HDFs are presented in Table 2. Clearly, the addition of kraft lignin remarkably enhanced the mechanical properties and water resistances of the panels.

**Table 2.** Properties of OCC Ultra-thin HDFs with Various Amount of Kraft Lignin under Different Hot Press Temperatures

| Hot Press Temp. (°C) | Kraft Lignin Content (%) | Actual density (kg/m$^3$) | MOR (MPa) | MOE (MPa) | IB (MPa) | 24 h TS (%) |
|---|---|---|---|---|---|---|
| 210 | 0 | 973 | 16.28±1.5a | 1707.9±160ab | 0.53±0.05a | 32.5±3.1f |
| | 10 | 966 | 19.11±1.7b | 2018.3±200b | 0.76±0.06b | 29.3±2.8e |
| | 15 | 958 | 22.76±2.2cd | 2433.6±230c | 0.97±0.09c | 26.4±2.5cd |
| | 20 | 977 | 25.52±2.4e | 2818.3±260d | 1.25±0.10d | 23.1±2.3ab |
| 230 | 0 | 965 | 15.67±1.3a | 1683.2±150a | 0.51±0.04a | 28.5±2.8de |
| | 10 | 956 | 18.83±1.8b | 1967.3±180ab | 0.72±0.06b | 24.3±2.3bc |
| | 15 | 969 | 22.55±2.1c | 2390.8±230c | 0.93±0.08c | 21.6±1.9ab |
| | 20 | 982 | 24.76±2.3de | 2775.2±250d | 1.22±0.11d | 20.8±2.0a |
| Data shown are mean ± standard deviation; different letters in the same column indicate significant difference at P < 0.05 | | | | | | |

The IB values of the ultra-thin HDFs made with kraft lignin (10 wt% to 20 wt%) were between 0.72 and 1.25 MPa, which represented a noticeable increase compared with the control (Table 2). Increasing kraft lignin content significantly increased the IB value. Kraft lignin additions of 10% into the ultra-thin HDFs hot pressed at 210 and 230 ℃ increased the IB values by 43% and 41%, respectively. For kraft lignin additions of 15% into the ultra-thin HDFs at 210 and 230 ℃, it increased the IB values by 83% and 82%, respectively. As the kraft lignin content increased to 20%, the IBs were further increased, by 1.36 and 1.39 times, respectively. This clearly showed that the IBs of the ultra-thin HDFs with kraft lignin addition were much higher than for the controls. As the amount of lignin was increased, the IBs increased significantly accordingly. The poor IBs of the controls were attributed to the limited amounts of lignin, and the shorter (Velasquez *et al.*

2003; Gulsoy *et al.* 2013) and hornified OCC fibers (Hubbe *et al.* 2007; Gulsoy *et al.* 2013). The ultra-thin HDFs produced at 210 °C displayed better IBs than that formed at 230 °C. This is a good finding, which means that stronger panels can be made at lower temperature with less energy consumption. All panels satisfied the IB requirement (0.35 MPa) for general purpose used in dry conditions as stipulated by Chinese National Standard GB/T 12626.4-2015 (Table 3).

**Table 3.** Property Values Specified in GB/T Standard for Wet-Process Fiberboards with Thickness Less Than 3.5 mm

| Standard | Use | MOR (MPa) | MOE (MPa) | IB (MPa) | 24 h TS (%) |
|---|---|---|---|---|---|
| GB/T 12626.4-2015 | General purpose used in dry conditions | 25 | N/A | 0.35 | 40 |

The MOR and MOE values for the control were between 15.67 and 16.28 MPa, and between 1683.2 and 1707.9 MPa, respectively (Table 2). After the addition of kraft lignin, both MOR and MOE were remarkably increased. When the load of kraft lignin was at the 10% level, the MOR and MOE values increased 17.38% and 20.16%, respectively. With the increased kraft lignin contents, the MOR and MOE also tended to rise. The values of flexural properties depend on the bonding strength among fibers, and individual fiber length and strength (Mancera *et al.* 2012). Stronger inter-fiber bonds contribute to an improvement in flexural strength (Velasquez *et al.* 2003; Mancera *et al.* 2012). These findings agree with those of Velasquez *et al.* (2003), who reported that the addition of kraft lignin markedly increased the flexural strength of the particleboard. Further, the addition of kraft lignin significantly improved the mechanical performances of the ultra-thin HDFs because the adhesive properties of lignin increased the links between the cellulosic fibers (Angles *et al*. 2001). It is interesting to note that the ultra-thin HDFs hot pressed at 210 °C exhibited better mechanical properties than those hot-pressed at 230 °C. Since less energy is consumed, there could be substantial economic gains by hot pressing at 210 °C. Panels hot-pressed at 210 °C with kraft lignin content of 20% satisfied the MOR requirement for general purpose used in dry conditions as stipulated by Chinese National Standard GB/T 12626.4-2015. MOE is not specified in the GB/T 12626.4-2015 Standard for Wet-Process Fiberboards with Thickness Less Than 3.5 mm (Table 3).

The 24-h TS values of the ultra-thin HDFs are shown in Table 2. Compared to the control, the TS of the ultra-thin HDFs made with kraft lignin was decreased remarkably. This was mainly due to better interfiber adhesion that resulted from the presence of lignin at the surface of the fiber (Flandez *et al.* 2012; Mancera *et al.* 2012). With increasing kraft lignin content, the TS decreased significantly. The results obtained in this study are in good agreement with those reported previously (Angles *et al.* 2001; Velasquez *et al.* 2003). Kraft lignin was capable of providing a resistance to water penetration (Angles *et al.* 2001). Increasing hot press temperature from 210 to 230 °C led to reduced TS. Similar observations have been reported by previous researchers (Karr *et al.* 2000; Velasquez *et al.* 2003; Boon *et al.* 2013). The 24 h TS of all ultra-thin HDFs produced met the GB/T 12626.4-2015 Standard requirement (40%).

## CONCLUSIONS

1. This study investigated for the first time an upgrading approach to produce old corrugated container (OCC) ultra-thin high-density fiberboard (HDF) bonded with kraft lignin. Ultra-thin HDFs based on OCC were prepared and characterized. OCC ultra-thin HDFs bonded with kraft lignin exhibited clearly better physical and mechanical properties than those made without kraft lignin addition. The kraft lignin content had a significant effect on the physical and mechanical properties of OCC ultra-thin HDFs. As the kraft lignin content increased, the physical and mechanical properties of OCC ultra-thin HDFs increased significantly.
2. The OCC ultra-thin HDFs hot-pressed at 210 ℃ outperformed those that has been hot-pressed at 230 ℃.
3. The OCC ultra-thin HDFs manufactured with 20% kraft lignin at 210 ℃ satisfied the requirement specified by the Chinese National Standard GB/T 12626.4-2015 for Wet-Process fiberboards for general use under dry conditions. Based on the findings of this study, it can be concluded that OCC is feasible as raw material, in combination with kraft lignin, for ultra-thin HDFs production.

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