Production of Particleboards from Steam-pretreated Rice Straw and Castor Oil-based Polyurethane Resin

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The high extractives content in rice straw inhibits the reactivity of castor oil-based polyurethane (CPUR) resin and hinders its bonding. This study employed steam treatment to pretreat rice straw to decrease the extractives content before using it for particleboard production. The effects of steam pressure and time on the chemical composition of the rice straw were investigated. Variable parameters tested were steam pretreatment time (5, 10 min) and steam temperature (110, 120, 130, and 140 °C). In addition, the mechanical properties and water resistance of particleboard manufactured from rice straw bonded with CPUR resin were evaluated. A three-phase hot press schedule at 110 °C was adopted. Other production parameters, such as resin content (20 wt%), hot press time (5 mm/s), and board density (800 kg/m^3) were held constant. The resulting particleboards were tested for their mechanical properties in terms of modulus of rupture (MOR), modulus of elasticity (MOE), and internal bond (IB). Further, the physical properties including water absorption (WA) and thickness swelling (TS) were analyzed. The results showed that steam pretreatment significantly decreased the hemicellulose and extractives contents of the rice straws. Furthermore, steam pretreatment improved the mechanical properties and water resistance of the rice straw particleboards.

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INTRODUCTION

There is a shortage of forest resources in China. With the development of the national economy and continuous improvement of people's living standards, the demand for wood and wood products has increased significantly. The limited wood supply cannot meet the rising demand. Traditional particleboard is made with wood as raw material. The reduction in wood raw material supply has put pressure on the particleboard industry. Therefore, it is necessary to seek alternatives to wood raw materials.

China is a large agricultural country. The annual production of agricultural residues (such as rice straw, wheat straw, cotton stalks, *etc*.) is enormous. If these fibrous agricultural residues are used as raw materials to produce particleboards, it can significantly reduce raw material costs and save wood resources.

At present, rice straw has not been fully and effectively utilized in China. Except for a small portion used for papermaking, livestock feed, and fuel, the vast majority of rice straw is buried or open burned. The main chemical components of rice straw are cellulose, hemicellulose, and lignin. Therefore, rice straw has potential to be used as raw material for particleboard production. Compared to wood, rice straw has lower cellulose and lignin content, but higher hemicellulose content. The outer surface of rice straw is waxy and hydrophobic, and it is rich in silica and extractives (Pan *et al*. 1999; Boquillon *et al*. 2004).

Urea formaldehyde (UF) resin is currently the most commonly used adhesive in the production of wood-based panels. However, UF is not suitable to produce rice straw particleboard. Rice straw particleboards bonded with UF exhibit weak strength and poor dimensional stability, even with high adhesive consumption (Li *et al*. 2010; Zhang and Hu 2014). The particleboards produced with UF resin release formaldehyde, causing environmental pollution and endangering human health (Boquillon *et al*. 2004; Luo *et al*. 2020a). At present, the production of rice straw particleboards mainly uses polymeric diphenylmethanediisocyanate (PMDI). Due to the high price of PMDI, the production cost of rice straw particleboard is very high, which limits the development and promotion of the particleboard (Zhang and Hu 2014; Luo *et al*. 2020a). Using low-cost renewable plant resources to produce particleboard not only does no harm to the environment, but it also avoids dependence on petroleum-derived resin, which has significant social and economic benefits. Castor oil-based polyurethane resin (CPUR) is formulated from castor beans (Gava *et al*. 2015). Its advantages include abundant renewable raw materials, low price, curing at both room temperature and elevated temperature, and low degree of toxicity (Garzón-Barrero *et al*. 2016). Extractives in lignocellulosic raw material inhibit the reactivity of CPUR resin and hinder the penetration and diffusion of CPUR adhesive (Gava *et al*. 2015; Martins *et al*. 2018; Faria *et al*. 2024). The extractives content of rice straw is high compared with wood (Li *et al*. 2013; Luo *et al*. 2020a). Pretreating straw raw materials with steam can reduce their extractives contents (Han *et al*. 2009). The modification of wheat straw was studied using a steam pretreatment by Han *et al*. (2009). Wheat straw was steamed under steam pressure of 0.2, 0.4, 0.6, and 1.0 MPa for 5 and 10 min, respectively. The results revealed that steam pretreatment significantly decreased the ash, extractives, and hemicellulose contents and improved mechanical properties of the wheat straw. The ash, extractive, and hemicellulose contents decreased constantly with increasing pretreatment pressure and time. Li *et al*. (2011) pretreated rice straw with 0.33% oxalic acid solution and steam for producing particleboard bonded with UF resin. The steam pretreatment was performed at four temperature levels (100, 120, 140, and 160 °C) and two time duration levels (5 min and 10 min). The resulting particleboards exhibited improved bending properties and internal bond strengths. The particleboards made with steam-pretreated rice straw had better mechanical properties in comparison to those made with oxalic acid-pretreated rice straw.

Up to now, there have been no reports on the use of steam-pretreated rice straw to manufacture CPUR-bonded particleboard. Therefore, this study considered the steam treatment to pretreat rice straw, using the steamed rice straw as raw material and CPUR as adhesive to manufacture particleboard. The aim of this study was to evaluate the technical feasibility of steam pretreated rice straw in production of particleboard bonded with CPUR. Steam pretreatment effects are related to the conditions, such as pretreatment temperature, time, and moisture content, of raw material (Liu *et al*. 1999; Cullis *et al*. 2004; Zhu *et al*. 2005). The purpose of straw raw material pretreatment for particleboard production is to efficiently decrease the extractive content and minimize the degradation of cellulose, hemicellulose, and lignin (Luo *et al*. 2020b). Under harsh pretreatment conditions the degree of extractives removal increases, but hemicellulose content is decreased, and the fibers are damaged (Han *et al*. 2009; Luo *et al*. 2020b). To avoid excessive degradation of thermally unstable hemicellulose, mild pretreatment conditions were used in this study.

EXPERIMENTAL

Materials

The rice straw was obtained from Huiguan Village, Xiaozhan Township, Jinnan District, (Tianjin, China). The moisture content of the rice straw was approximately 8.8%. The leaves and remnant grains were manually removed. Then, the rice straw was broken to pieces using a fodder grinder (Model HLO; Zhengyuan Powder Engineering Equipment Corporation, Shandong, China). A mesh with 0.3-mm aperture size was used to remove small particles and dust, which contributed to decreased silica content and improved adhesive efficiency (Halvarsson *et al*. 2010; Kurokochi and Sato 2015).

The commercial CPUR adhesive used in this study was bi-component type in a ratio of 1:1 (50% polyol and 50% pre-polymer) purchased from Jinan Huakai Resin Corporation (Jinan, China). The CPUR was free of solvents, with a solid content of 100%.

Methods

Steam pretreatment was performed in a custom-built 15-L batch stainless steel cylindrical autoclave (Model ZQS; Tongda Light Industrial Equipment Corporation, Xianyang, China). The autoclave consists of an electrical heater, a motor actuator, a safety valve, a steam releasing valve, and the required instruments for measuring and controlling the pressure and temperature. The design pressure of the autoclave was 4 MPa (Luo *et al*. 2020b). About 600 g (dry matter) of the rice straw was loaded in the upper part of the autoclave, while 500 mL of tap water was added in the lower part of the autoclave. Then, the autoclave was tightly sealed and quickly heated to the predetermined temperature. The pretreatment temperatures were 110, 120, 130, and 140 ℃, and the residence times were 5 min and 10 min. At the end of each desired treatment time, the heating was turned off, and steam was released gradually. The autoclave was discharged when attaining ambient temperature. Then, the steamed rice straw was collected and dried at 100 ± 3 ℃ to attain 8% moisture content and stored in plastic bags for future use (Yano *et al.* 2020). The raw and steam-treated rice straws were analyzed for cellulose, hemicellulose, lignin, and ethanol/benzene extractives contents according to the procedures of Liu and Zhang (2020).

The CPUR resin at 20% level (based on oven-dried weight of the particles) was used. The particles were put in a blender and blended with the CPUR for 6 min to produce a mixture with a homogenous distribution of CPUR. The glued particles were then placed into an aluminum forming mold to manually form the mattress. The mattresses were prepressed at 1.5 MPa for 30 s and then hot-pressed at 110 °C in accordance with a three-phase pressing schedule. Initially the mattress was pressed under a pressure of 5 MPa applied for 180 s. Thereafter, successive pressures of 0.3 MPa for 60 s and 4 MPa for 120 s were used to release gases that can trigger blowing or blistering of the mattresses (Sugahara *et al.* 2019). The dimensions of the particleboard size were 500 × 500 × 6 mm^3. The target density was 800 kg/m^3. Three replicate panels were produced for each manufacturing condition. The particleboard made with raw rice straw was used as control. After fabrication, the particleboards were conditioned by storing them at 20 °C and 65% relative humidity for about 2 weeks prior to being cut into test specimens.

The rice straw particleboards were trimmed to a final dimension of 450 mm × 450 mm × 6 mm. Then, the particleboards were sawed into test specimens and measured according to the China national test standard GB/T 17657 (1999) for internal bonding strength (IB), modulus of rupture (MOR), modulus of elasticity (MOE), and 24 h thickness swelling (TS). The mechanical properties of particleboards were tested on a universal testing machine (Shimadzu AG-IC, Shimadzu Instrument (Suzhou) Co., Ltd, China).

Analysis of variance (ANOVA) and Duncan's mean separation tests were used to statistically analyze the data obtained with a SPSS (Statistical Package for Social Sciences, version 17, SPSS Inc., Chicago, IL, USA) software. The probability level for statistical significance was $P < 0.05$.

RESULTS AND DISCUSSION

Table 1 shows the chemical compositions of the steam-treated rice straw under different steam conditions. After steam pretreatment, cellulose and lignin contents increased, whereas hemicellulose, ash, and extractives contents were decreased. Cellulose and lignin are desired main components of rice straw particleboard. The increases in cellulose and lignin contents will contribute to the improvement of mechanical properties of the rice straw particleboards. Hemicellulose is thermally unstable (Han *et al*. 2009). Even at steam conditions as mild as 110 °C/5 min, degradation of hemicellulose was still observed. More severe steam pretreatment conditions resulted in a higher degree of hemicellulose degradation (Han *et al*. 2009; Li *et al*. 2011). Hemicellulose is more hydrophilic than cellulose and lignin. Partial removal of hemicellulose from rice straw helps to improve the dimensional stability of the rice straw particleboards (Li *et al*. 2011). The silica content in rice straw ash was more than 90% (Kurokochi and Sato 2015), which hinders uniform resin distribution (Hiziroglu *et al*. 2007; Kurokochi and Sato 2015). It can be predicted that the decrease of silica content will contribute to the improvement of bonding ability between rice straw particles and the CPUR resin. The presence of extractives on the surface of straw inhibits bonding among straw particles (Martins *et al*. 2018). As can be seen from Table 1, steam pretreatment effectively decreased extractives content of rice straw. As the steam pretreatment conditions intensified, the removal degree of ash and extractives also increased. Partial removal of extractives from rice straw surface is expected to enhance the compatibility between the rice straw particles and CPUR resin, and thus the performances of rice straw particleboards.

Table 1. Effect of Steam Pretreatment on Chemical Composition of Rice Straw

Pretreatment Condition		Cellulose (%)	Hemicellulose (%)	Lignin (%)	Ash (%)	Extractives Ethanol/Benzene (%)
Temp. (°C)	Time (min)					
110	5	38.8	21.2	15.9	14.8	6.5
	10	39.1	20.7	16.0	14.6	6.4
120	5	39.6	19.9	16.0	14.1	6.2
	10	40.3	19.4	16.1	13.7	6.1
130	5	41.1	18.6	16.1	13.4	6.0
	10	41.9	18.0	16.3	13.0	5.8
140	5	42.8	17.5	16.2	12.6	5.5
	10	43.7	16.9	16.4	12.3	5.3
Untreated		38.4	23.9	15.8	15.2	6.8

Table 2 shows the effect of steam pretreatment conditions on the physical and mechanical properties of rice straw particleboards. The physical and mechanical properties of the particleboards made from the steamed rice straw were significantly better than those made from untreated rice straw. The pretreatment conditions had a significant impact on the mechanical properties and dimensional stabilities of the rice straw particleboards. Higher pretreatment temperatures and longer residence times led to increased mechanical properties and dimensional stabilities.

Table 2. Properties of Rice Straw Particleboards at Various Steam Pretreatment Conditions

Pretreatment Condition		MOR (MPa)	MOE (MPa)	IB (MPa)	24 h TS (%)
Temp. (°C)	Time (min)				
110	5	10.14 ± 1.15 b	1476.6 ± 182.4 ab	0.24 ± 0.027 b	18.13 ± 3.08 d
	10	10.56 ± 1.37 b	1582.6 ± 176.1 bc	0.28 ± 0.029 bc	16.26 ± 2.91 cd
120	5	10.37 ± 1.23 b	1578.3 ± 184.3 bc	0.29 ± 0.028 bc	15.41 ± 2.82 cd
	10	10.75 ± 1.46 b	1637.7 ± 176.8 bc	0.30 ± 0.031 c	14.82 ± 1.92 bcd
130	5	10.78 ± 1.12 b	1662.2 ± 264.7 bc	0.30 ± 0.032 c	13.65 ± 2.43 abc
	10	11.36 ± 1.28 bc	1783.7 ± 277.4 bc	0.31 ± 0.034 c	12.87 ± 1.54 abc
140	5	11.12 ± 1.29 bc	1758.4 ± 282.5 bc	0.31 ± 0.031 c	11.74 ± 2.08 ab
	10	12.87 ± 1.33 c	1865.3 ± 297.6c	0.33 ± 0.035 c	11.23 ± 1.39 a
Untreated		7.58 ± 0.81 a	1223.5 ± 128.7 a	0.09 ± 0.011 a	33.27 ± 3.58 e

Data shown are mean ± standard deviation; different letters in the same column indicate significant difference at P < 0.05

The particleboards made with steamed rice straw had IB values ranging from 0.24 to 0.33 MPa, showing a significant enhancement in comparison to the control, which was 0.09 MPa. Raising the steam temperature and/or prolonging residence time improved the IB. Steam condition of 140 °C/10 min resulted in the highest IB value of 0.33 MPa, around 3.67 times higher than that of the control. The increase in IB was attributed mainly to the elimination of the extractives from the surfaces of rice straws and the decreased silica content resulting from the steam pretreatment, which led to improved interfacial adhesion of rice straw particles with CPUR resin (Han *et al*. 2009; Li *et al*. 2011). The minimal IB requirement for general purpose particleboards is 0.31 MPa, as stipulated in the Chinese standards GB/T 4897.2 (2003). The IBs of the particleboards made with rice straws steamed at 130 °C/10 min, 140 °C/5 min, and 140 °C/10 min met the minimum requirement for general purpose end-use.

The MOR values of the particleboards made with steamed rice straw ranged from 10.14 to 12.87 MPa, which were significantly higher than that of particleboard made with untreated rice straw, 7.58 MPa. The highest MOR value was observed for particleboard made with rice straw steamed at 140 °C/10 min, which was about 70% higher than that of the control. The MOE exhibited similar trends as MOR. The particleboards made with steamed rice straw had significantly higher MOE than particleboards produced with untreated rice straw. Elevating steam temperature and/or prolonging residence time upgraded the MOE. The improvement in the static bending strength of particleboards made with steam pretreated rice straws reflected a good correlation with the trend of IB enhancement. Hence, the reason for these increases in bending properties was mainly attributed to improved interface compatibility between rice straw and CPUR resin resulted from the steam pretreatment. The minimal MOR requirement for general purpose particleboards is 14 MPa, as stipulated in the Chinese standards GB/T 4897.2 (2003). None of the produced particleboards met the MOR requirement. The MOE is not listed in the GB/T 4897.2 (2003) standard.

The 24 h TS values of the particleboards produced with steamed rice straw particles were significantly lower than those of particleboards produced with untreated rice straw particles. The main reasons for the lower 24 TS values are two-fold. Firstly, the partial removal of ash and decrease of extractives contents by steam pretreatment led to improved interface compatibility between rice straw particles and CPUR resin, resulting in a lower degree of voiding of the panel boards. Secondly, partial degradation of the relatively hydrophilic hemicellulose by steam pretreatment also helped to improve dimensional stability of the particleboards (Han *et al*. 2009). The 24 h TS values gained in this study were remarkably lower than those obtained by Li *et al*. (2011), who made particleboards using steamed rice straw bonded with UF resin. The results obtained in this study were consistent with the findings from the studies by Gava *et al*. (2015) and Sugahara *et al*. (2019) that the CPUR bonded particleboards were much more water resistant and dimensionally stable than UF bonded particleboards. The lower affinity of CPUR to water should be responsible for these phenomena (Pan *et al*. 2010; Gava *et al*. 2015).

CONCLUSIONS

1. Steam pretreatment significantly improved the physical and mechanical properties of rice straw particleboards bonded with castor oil-based polyurethane resin (CPUR). The physical and mechanical properties of particleboards made from steamed rice straw were significantly better than those made from untreated rice straw.
2. The strength properties and dimensional stability of the rice straw particleboards improved with the intensification of the steam pretreatment conditions.
3. The particleboards made with rice straw pretreated at 130 °C/10 min, 140 °C/5 min, and 140 °C/10 min, respectively, met the IB strength requirements by Chinese National Standard GB/T 4897.2 (2003). All the particleboards made with steamed rice straw satisfied the 2 h TS requirements by Chinese National Standard GB/T 4897.2 (2003).

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