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Development of high-performance UF-bonded reed and wheat straw medium-density fiberboard

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Abstract Urea formaldehyde resin-bonded reed and wheat straw fiberboards were produced from the fibers made under different steam cooking conditions in refining processes at densities of 500 and 700 kg/m³. The effect of steam cooking conditions on the board properties was examined. The steam pressure and cooking time for reed and wheat straws were 0.4MPa/10min and 0.4MPa/5min, respectively, and 0.6 MPa/3 min and 0.6 MPa/10 min for both straws. The effect of steam cooking treatment before the fiber refining process on the wettability and weight losses of the straws was also investigated. The results indicated that the mechanical properties and linear expansion of the straw medium-density fiberboard (MDF) were improved with increasing steam cooking pressure and time during the refining process, whereas the thickness swelling (TS) did not vary much. The wettability of the straws was improved by cooking treatment. The steam cooking conditions had little effect on the wettability of the straw surfaces. For reed and wheat straws, the weight losses increased with increasing steam pressure and cooking time. In addition, it was found that the properties of MDF were significantly higher than those of particleboard, especially the internal bond (IB), where the IB values of MDF were more than 10 times higher than those of particleboard. All the properties of the straw MDF, except the TS of wheat board,

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can meet the requirement of JIS fiberboard standard. The high performances of MDF could be due to the improved wettability and the removal of extractives during the refining process.

Key words Urea formaldehyde resin \cdot Reed straw \cdot Wheat straw \cdot MDF

Introduction

The structure of straws from annual plants such as reed and wheat is less homogeneous than that of softwood and even hardwood. Morphologically, straw is more complicated than wood. Straw contains a relatively large number of elements, including the actual fibers, parenchymal cells, vessel elements, and epidermal cells, which contain a high amount of ash and silica. In a cross section of straw, the epidermal cells are the outermost surface cells, covered by a thin wax layer. This surface layer deteriorates the moisture absorbence of straw from water-based adhesives such as urea-formaldehyde (UF) resin. It therefore acts as a barrier to the gluing of straw with UF resin.¹⁻⁷ Removing this bonding barrier layer from straw materials has been a technical problem for performance enhancement of straw panels.

The previous studies reported that proper treatment such as ethanol-benzene (EB) extraction improved the wettability of reed and wheat straw surfaces. The upgraded properties of UF-bonded straw particleboards could be attributed to the improved wettability, which was caused by removal of the wax from straw surfaces.⁷⁻⁹ Based on these results, it was thought that the wax layer would be destroyed by a proper thermal-mechanical refining process. The object of this study was to examine the effect of refining conditions on the wettability and weight losses of straws and to develop high-performance UF-bonded medium-density fiberboard (MDF) using straw materials. This led us to explore an economical and feasible manufacturing process for utilizing the plant straw resources in the panel industry.

Materials and methods

Raw materials

Reed and wheat straw materials (including leaves), as for previous studies,⁷⁻⁹ were obtained from northeastern China. A commercial UF resin adhesive (TKB-1), with a solid content of 65% and a U/F molar ratio of 1.0:1.4, was supplied by Oshika Shinko (Japan). An NH₄Cl solution of 20% concentration was used as a hardener for the UF resin, with the addition level of 5% based on the resin solid weight.

Preliminary experiment

Oven-dried reed and wheat straws of 10cm length (about 5g) were cooked with steam using a thimble filter. The thimble filter was loosely tied with a wire before putting it into a cooking cell to prevent the straw blocks from bursting out of the filter. The steam pressures were 0.2, 0.4, 0.6, and 1.0MPa. The cooking times for each pressure level were 5 and 10min. The weights of samples after oven drying at 105° C for 24h were measured before and after cooking, and the weight losses were then calculated. The wettability of treated samples was evaluated. Wettability is expressed as the advancing contact angle of distilled water on the outer surface of the straw. Measurement of the contact angle was the same as in the previous study.⁷ Seven replicates were used for each condition.

Manufacture of medium-density fiberboard

Fibers were made from reed and wheat straws of about 5 cm length using a pressurized single disk refiner with a refiner plate diameter of 305 mm (BRP45-300SS, Kumagai Riki Kogyo). The straw was steamed in a 6-1 pressure vessel and then passed to the refiner plate (plate gap 0.37 mm). A designated pressure was maintained in the system by a constant supply of steam. Based on the results of the preliminary experiment, three steam cooking conditions for each straw were used, as shown in Table 1. Refined fiber was vented from the refiner housing into a blowline connected to a continuous flash dryer. The moisture content (MC) of the obtained fiber was about 12%–15%. The dimensions of the fibers are shown in Table 2. The lengths and diameters of 200 randomly chosen fibers were

Table 1. Experimental variables of steam cooking in refining processes

Code	Straw types	Steam pressure (MPa)	Steaming time (min)
0.4/5	W	0.4 (140°C) ^a	5
0.4/10	R	0.4 (140°C)	10
0.6/3	R, W	0.6 (160°C)	3
0.6/10	R, W	0.6 (160°C)	10

R, reed; W, wheat

^a The figures in parentheses are the corresponding temperatures of the steam pressures

measured after photographs of the fibers were obtained at $25 \times$ magnification, and the length/diameter ratio of each fiber sample was calculated.

The fibers were dried again at 60°C to the target MC of 2%–3% before they were blended with adhesive. The UF resin was added to the fibers in an air-cyclic pipeline using a spray gun. The resin addition level was 15% based on the oven-dried weight of the fibers. Mats were formed by passing the blended fibers through another pipeline that ended in a forming box. A total of 12 mats were platen-pressed at 130°C for 5 min. A three-phase pressing schedule was used to avoid blowing. The board dimension was $370 \times 355 \times 6 \text{ mm}$ with target densities of 500 and 700 kg/m^3 for each condition.

Evaluation of panel properties

For conventional evaluation of mechanical properties and dimensional stability, the test boards were conditioned for 2 weeks under 20°C and 65% \pm 5% relative humidity (RH). The unsanded boards were then evaluated according to the Japanese Industrial Standard for fiberboard (JIS A 5905, 1994).

The static bending test in the dry condition was conducted on three specimens of $50 \times 200 \text{ mm}$ from each board, using a three-point bending test over an effective span of 150 mm at a loading speed of 10 mm/min. Five specimens with dimensions of $50 \times 50 \text{ mm}$ from each board were tested for internal bond (IB) and thickness swelling (TS).

In addition to the standard water soaking test, two specimens of $50 \times 200 \text{ mm}$ were prepared from each board to test linear expansion (LE), thickness change (TC), and equilibrium moisture content (EMC) under a relative humidity (RH) conditioning cycle of 33%, 66%, and 98%. The initial and final dimensions and weights were measured after oven drying at 50°C under vacuum for 36h followed by 105°C for 5h. The corresponding changes in length, thickness, and weight were examined after the test samples were conditioned to equilibrium at 33%, 66%, and 98% RH over saturated solutions of MgCl₂, NaNO₂, and CaSO₄, respectively, in airtight chambers at 20°C. The length was measured to the nearest 0.01 mm by means of a linear gauge

 Table 2. Dimensions of fibers under various cooking conditions during the refining process

Fiber types	Length (mm)	Diameter (µm)	L/D ^a
R 0.4/10	3.00 (1.32)	246.25 (127.99)	16.28 (12.49)
R 0.6/3	3.46 (1.75)	118.17 (72.26)	39.83 (31.22)
R 0.6/10	3.57 (1.68)	109.80 (56.56)	40.74 (26.30)
W 0.4/5	5.32 (2.63)	186.94 (117.56)	38.54 (26.71)
W 0.6/3	5.17 (2.60)	101.00 (57.79)	65.47 (48.83)
W 0.6/10	5.12 (3.19)	109.39 (76.02)	66.42 (50.01)

Refer to Table 1 for the explanation of fiber types

The results are given as averages and standard deviations (in parentheses) from the mean values of 200 randomly chosen fiber samples a L/D is the length/diameter ratio of each fiber sample sensor, which was fixed on a platform with the sensor parallel to the length direction of the specimen.

Results and discussion

Effect of steam cooking treatment on wettability and weight loss of straws

Figure 1 shows the contact angles and weight losses of reed and wheat straws under various steam cooking conditions. The contact angles of the straws were reduced after cooking treatment, and this reduction was greater for wheat straw. The wettability of the straws was therefore improved by cooking treatment. This improvement could be attributed to the removal of wax from the straws. The F-test statistical analysis revealed that the effect of cooking time on wheat straw and the co-effect of steam pressure and time on reed straw exist at the 95% significance level. This indicates that the steam cooking conditions in the studied range had little effect on the wettability of reed and wheat straw surfaces.

The effect of cooking conditions on the weight losses of the straws shows that the weight losses of both reed and wheat straws increased with increasing steam pressure and cooking time. When the pressure was less than 0.6 MPa, there was a relatively slow escalating trend of the weight loss, especially for reed straw. The weight loss increased significantly at above 0.6 MPa pressure. The cooking time had a much greater effect on weight loss than did the steam pressure. At the same steam pressure, a longer cooking time resulted in more weight loss, especially when the steam pressure was above 0.6 MPa. In addition, the weight loss increased more in wheat straw than in reed straw. This means that much more of the extractives was removed from wheat straw. Lawther et al. reported that steam treatment removed some portion of pectic substances and hemicellulose from wheat straw.¹⁰ Because the pectic substances and high content of hemicellulose in nonwood lignocellulosic materials usually result in less adhesion between resin adhesive and these materials, extraction of these substances would certainly contribute to the enhancement of board properties.

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Properties of MDF under various steam cooking conditions during refining processes

Figure 2 shows the moduli of rupture and elasticity (MOR, MOE), IB, and TS of the straw MDF with the fibers under different steam cooking conditions during the refining process. The effect of densities on board properties is a major concern, so the densities among the boards and within the board were investigated. The results show that the range of densities among the boards are 490-570 and 690-720 kg/m³ for board target densities of 500 and 700 kg/ m³, respectively; the average and range of coefficients of variance for the densities in the board are 5.85% and 3.9%-7.5%, respectively. It was found that the deviations of the board densities among the boards and within the board were rather small. Considering that density usually has a significant effect on board properties, all the property values in this study were corrected to the board target densities of 500 and 700 kg/m³ based on the linear correlation between board densities and properties. Generally, the board properties MOR, MOE, and IB were improved with increasing steam pressure and cooking time during the refining process. For reed and wheat boards, the MOR and MOE were significantly increased when the steam pressure was up to 0.6 MPa. There was relatively little improvement in MOR but none in MOE as the cooking time increased from 3 to 10min under the steam pressure of 0.6MPa.

For reed board, greater upgrading of IB was observed when the steam pressure was increased to 0.6 MPa, and for wheat board this improvement occurred as the cooking time was extended from 3 min to 10 min under 0.6 MPa. Based on the results of the cooking treatment, the improved mechanical properties could be attributed to the removal of extractives from the straw materials, especially wheat board. A range of factors, such as wettability, extractives, and fiber dimensions have effects on the properties of MDF. Removal of extractives in wheat board play a more significant role in upgrading board performance under different cooking conditions. In contrast, the improved properties in reed board may be due to the greater increase of the defibration degree under higher steam pressure, as reflected in Table 2, where the length/diameter (L/D) ratio





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Fig. 2. Properties of reed and wheat straw medium-density fiberboard (MDF) with the fibers under different steam cooking conditions during the refining process. Refer to Table 1 for an explanation of the legend. *Vertical lines* through the *bars* represent the standard deviation from the mean value. *MOR*, modulus of rupture; *IB*, internal bond; *MOE*, modulus of elasticity; *TS*, thickness swelling



of the fibers under 0.6 MPa was about 2.5 times that of fibers under 0.4 MPa. The higher degree of defibration causes an increased surface area of fibers and the formation of fibrill, which makes the fibers blend together more readily and keeps them in more intimate contact during pressing.¹¹

Based on an F-test statistical analysis, the TS values of both reed and wheat boards were insignificant for the various cooking conditions during the refining process. This could be related to the fact that the water immersion time of 24h might be too short to allow complete springback of the heavily compacted fibers. In addition, even though there was little difference in the MOR and MOE values, the IB and TS of reed board were better than those of wheat board. In the previous studies, it was found that there is much more silica and wax in reed and wheat straw materials, respectively. The higher upgrading of reed particleboard by silane coupling agent (SCA) was thought to be due to the improved wettability, which might have resulted in part from some reactions between the SCA and silica on the straw surfaces. The greater improvement of wheat particleboard by extraction could be due to the removal of wax-like substances from wheat straw.⁷⁻⁹ Based on these conclusions, the superior IB of reed MDF in this study could be attributed mostly to the removal of silica in reed fibers during the defibration process, and this effect may be greater than the effect of the extractives removal on wheat MDF. The excellent TS of reed board is due to the inherent water resistance of reed straw,⁷ and its higher IB also contributes to the superior TS.

The dimensional stabilities at three relative humidities for the boards made from different fibers are shown in Figs.



Fig. 3. Linear expansion (LE) of reed (a) and wheat (b) straw MDF under various relative humidities (RH) during moisture absorption and desorption processes. Refer to Table 1 for an explanation of the legend

3, 4, and 5. The LE of both reed and wheat boards produced from fibers registered higher values under 0.4 MPa than under 0.6 MPa, but a reversed tendency was present in the TC, especially at 98% RH. This may be due to a larger proportion of vertically oriented elements in boards composed of the fibers made under 0.4 MPa, which is related to their lower L/D (Table 2). The TC of the boards increased steadily when the RH was below 66% but recorded rather high values at 98% RH. In addition, wheat board represents a higher LE and TC than reed board. The residual TC of wheat board was about twice that of reed



Fig. 4. Thickness changes (TC) of reed (a) and wheat (b) straw MDF under various RH levels during moisture absorption and desorption processes. Refer to Table 1 for an explanation of the legend



Fig. 5. Equilibrium moisture content (EMC) of reed (**a**) and wheat (**b**) straw MDF under various RH levels during moisture absorption and desorption processes. Refer to Table 1 for an explanation of the legend

board. This greater irreversible swelling is caused by the release of higher compressive stresses imparted to the wheat board during the pressing process. It was also found that the degree of springback in both reed and wheat boards was dependent on the equilibrium moisture content (EMC), where wheat board recorded a higher TC and EMC than did reed board when subjected to 98% RH.

Comparison of MDF and particleboard

Figure 6 shows the properties of the straw MDF and particleboard. The MDF is 700kg/m³ board with the fibers prepared under the steam cooking condition of 0.6MPa/ 10min. The particleboard is the board without SCA and EB treatment made from coarse particles at 700kg/m³ density level using the same UF resin with 12% content reported in our previous paper.⁹ Though the resin addition content in MDF was 3% higher than in particleboard, the resin content per unit surface area of the elements in MDF



Fig. 6. Comparison of particleboard (PB) and MDF

is supposed to be much lower than that in particleboard. The results in Fig. 6 show that all the properties of both reed and wheat MDF were better than those of particleboard, especially the IB. The IB values of reed and wheat MDF were about 13 and 16 times that of particleboard, respectively.

The superior performances of MDF are thought to be attributed to the defibration process. Based on previous conclusions^{7,9} and the result of steam cooking treatment, the high performances of wheat MDF could be attributed to the improved wettability due to wax removal and the removal of extractives from the straw material through the defibration process. The excellent properties of reed MDF may be partly due to the improved wettability and the extractives removal, and mostly because silica was removed from reed straw by the defibration process.

All the properties of straw MDF, except for the TS of wheat board, can meet the JIS fiberboard standard. Dimensional instability has been a problem with nonwood lignocellulosic composites and is a major reason for their restricted use. Recently, it has been reported that the dimensional stability of agro-based fiber composites was greatly improved by chemical modification of agricultural fibers.¹²

Conclusions

The wettability of the straws was improved by steam cooking treatment. The steam cooking conditions in the range of this study had little effect on the wettability of the straw surfaces. The weight losses of the straws increased with increasing steam pressure and cooking time. Wheat straw shows a higher increasing extent of weight loss than reed straw, which means that more extractives were removed from wheat straw.

The mechanical properties and LE of the straw MDF were improved by increasing the steam cooking pressure and time during the refining process, although the TS varied little with the various cooking conditions. The improved performances could be due to the removal of extractives and increased defibration degree. In addition, even though there was no major difference in MOR and MOE, the IB and TS of reed board were better than those of wheat board.

The most interesting finding is that all the properties of both reed and wheat MDF were significantly higher than those of the particleboard. The much higher performance of wheat MDF could be attributed to the improved wettability and removal of extractives from the straw material. The excellent properties of reed board may be partly due to the improved wettability and the removal of extractives, but mostly because silica was removed from reed straw during the refining process. It is thought that MDF is one of the most feasible products for utilizing such straw materials.

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