J Wood Sci (1998) 44:282–286

ORIGINAL ARTICLE

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Upgrading of urea formaldehyde-bonded reed and wheat straw particleboards using silane coupling agents

Received: November 20, 1997 / Accepted: January 19, 1998

Abstract Reed and wheat straw particleboards bonded with urea formaldehyde (UF) resin were manufactured from two different material configurations (i.e., fine and coarse particles). The board densities were in the range of 0.55–0.90 g/cm^3. The effects of particle size and board density on the board properties were examined. The properties of particleboard produced from fine particles were better than those made from coarse particles. An increase in board density resulted in a corresponding improvement in the board properties. The properties of UF bonded reed and wheat straw particleboards were relatively lower than those of commercial particleboards. Three silane coupling agents were used to improve the bondability between the reed and wheat particles and UF resin. Results of this study indicate that all the board properties were improved by the addition of silane coupling agent. The degree of improvement achieved from each coupling agent was different; epoxide silane was found to be more effective for reed straw particleboard, and amino silane was better for wheat straw particleboard.

Key words Reed straw · Wheat straw · Particleboard · Urea formaldehyde resin · Silane coupling agent

Introduction

In view of the global decline in the forest resources and population growth, agro-based resources have been exploited in recent years to supplement the supply of wood materials from the existing forest resources. One of the main potential utilizations of these agro-based materials is conversion to composite products. Research studies have been conducted on these aspects for years, and the manufacturing technology of processing bamboo, bagasse, sorghum, and oil palm stems into composite products has been developed.[1–4] Some problems still exist with seasonality, storage, scattering sources, and bondability.[5] Among these factors, bondability remains a major unsolved technical problem, especially when urea-based resins are applied.[6–8]

Reed has traditionally been used to make paper, but in recent years other utilizations are being considered, especially in the case of growing areas remote from the paper production plants, where these resources are not being utilized. Among the agriculture plants in the world, wheat occupies the largest planting area: about 141 billion hectares (ha) with the highest annual production of 835 billion tons. At present, wheat straw is not being utilized on an industrial scale, and only a small amount is used for paper production.[9] It is therefore necessary to identify alternative applications so these materials can be fully utilized for the benefit of humankind and the environment.

Some research has been conducted on reed and wheat straw boards. The urea formaldehyde (UF)-bonded boards are reported to have inferior properties,[7,8] although high quality boards can be produced using isocyanate resin.[10,11] Silane coupling agents are generally applied for improving the adhesion between organic and inorganic materials. There are at least two functional groups in their molecules: One is the methoxyl or ethoxyl group, which decomposes in water or reacts with some groups of inorganic material; another is the amino or epoxy group, which can react with organic materials. In this regard, silane coupling agents are used to modify the characteristics of the inorganic surface by fixing some organic functional groups onto it.[12]

The objective of this study was to assess the fundamental properties of reed and wheat particleboards and to identify an effective method to improve the bondability of UF resin with these materials using silane coupling agents.

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Experimental

Materials

Raw materials used were reed (*Phragmites communis* Trin.) and wheat (*Triticum aestivum* L.) stems with air-dried densities of 0.57 and 0.31 g/cm^3, respectively. The stems were first cut into 150- to 200-mm lengths by a drum chipper and further refined using a ring flaker. The particles were then screened into two groups of fine and coarse particles. Table 1 summarizes the composition of fine and coarse particles based on mesh analysis. All particles used were oven-dried at 80°C to about 3% moisture content.

The UF resin used in this study was supplied by Zheng Yang River Wood Processing Company, China. The resin solid content and urea/formaldehyde molar ratio of the UF resin were 60%–65% and 1.0:1.4, respectively.

Three silane coupling agents (i.e., vinyl silane, amino silane, and epoxide silane) were used for improving the properties of particleboard produced from coarse particles in this study. The chemical structure of these compounds are as follows.

Vinyl silane (SiVN)
  Vinyltriethoxysilane (SiVN 1): $CH_2 = CH{-}Si(OC_2H_5)_3$
  Vinylytris (β-methoxyethoxy)silane (SiVN 2): $CH_2 = CH{-}Si(OC_2H_4OCH_3)_3$
Amino silane (SiNH)
  γ-Aminopropyltriethoxysilane: $NH_2{-}C_3H_6{-}Si(OC_2H_5)_3$
Epoxide silane (SiEP)
  γ-Glycidoxypropyltrimethoxy:

$$\overset{O}{\overbrace{H_2C{-}CH}}{-}CH_2{-}O{-}C_3H_6{-}Si\,(OCH_3)_3$$

Some properties of these silane coupling agents (Gai Xian Chemistry Company, China) are shown in Table 2.

Board manufacturing

The UF resin was sprayed onto the particles in a blender at 13% resin content based on the oven-dried weight of particles. Two percent of silane coupling agent was mixed with the UF resin prior to blending, based on the weight of the resin solid. One percent of NH_4Cl based on the weight of the resin solid, was added as the curing catalyst.

Table 1. Composition of fine and coarse particles based on mesh analysis

Mesh size (mm)	Reed particles (%)		Wheat particles (%)	
	Fine	Coarse	Fine	Coarse
≥4.76	0	0.65	0	0.19
4.76–2.00	0.50	31.61	–	–
2.00–1.00	20.84	54.76	35.30	60.75
1.00–0.25	68.53	12.90	62.80	37.01
0.25–0.125	8.17	0.07	1.90	2.05
≤0.125	1.94	0.00	0	0

Components are expressed as percentage based on the total weight.

Table 2. Some properties of silane coupling agents

Coupling agent	Molecular weight (kDa)	Specific gravity at 25°C	Boiling point (°C)
Vinyltriethoxysilane (SiVN1)	190.3	0.90	161
Vinylytris (β-methoxyethoxy) silane (SiVN2)	280.4	1.04	285
γ-Aminopropyltriethoxysilane (SiNH)	221.4	0.94	217
γ-Glycidoxypropyltrimethoxy (SiEP)	236.3	1.07	290

The hand-formed mats were pressed into 8 mm thick boards using distance bars at 150°C for 7 min. A three-step pressing schedule was used to avoid blistering. During the first step the mat was pressed under a pressure of 3 MPa for 1 min, and during the second and third steps the mat was pressed at 2 MPa for 3 min and 1 MPa for 3 min, respectively. The board size was 450 × 430 × 8 mm with targeted densities ranging from 0.55 to 0.90 g/cm^3. Two boards were made in the same condition; altogether 52 boards were manufactured. The densities of the boards manufactured are shown in Table 3.

Board testing

Specimens were cut from the boards after conditioning and tested according to GB/T 4897-92 (Particleboard Standard of China). The specimen size for the bending test was 27 × 5 cm with an effective span of 15 cm. The sample sizes for internal bond (IB) and thickness swelling (TS) were 5 × 5 cm and 2.5 × 2.5 cm, respectively.

Thickness swelling was determined by measuring the changes of board thickness after immersing in 20°C water for 2 h. Four to eight replicates were used for each condition.

Results and discussion

Properties of reed and wheat boards

Figures 1 and 2 show the effects of particle sizes on the modulus of rupture (MOR) and the IB of reed and wheat boards at different densities. Similar to conventional wood-based particleboard, both MORs and IBs increased with increasing board density. The IB values of both the reed and wheat boards were much lower than those of conventional wood particleboard. This was true especially in the case of reed and wheat boards made from coarse particles, where the IB values were only 0.10 and 0.16 MPa, respectively, at a high density of 0.80 g/cm^3.

It is obvious from the figures that for both reed and wheat particleboards the MORs and IBs of boards produced from fine particles are better than those from coarse particles at the same board density levels. It is well known that the size and configuration of particles have

Table 3. Density of particleboards manufactured

Particle size	Without coupling agent (control) (g/cm³)							With coupling agent (g/cm³)			
								SiVN1	SiVN2	SiNH	SiEP
Reed board											
Fine	0.55	0.60	0.65	0.70	0.75	0.80	0.85				
Coarse	0.68	0.90						0.70	0.70	0.73	0.66
Wheat board											
Fine	0.55	0.60	0.65	0.70	0.75	0.80	0.85				
Coarse	0.66	0.75						0.72	0.72	0.76	0.68

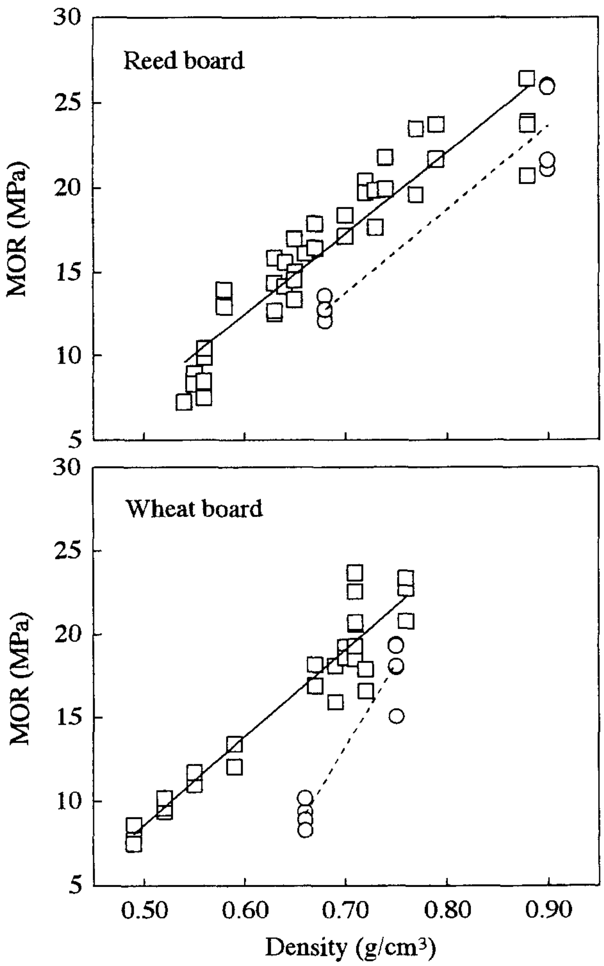

Fig. 1. Relation between density and modulus of rupture (*MOR*) of board made from reed and wheat straws. *Squares*, fine particles; *circles*, coarse particles

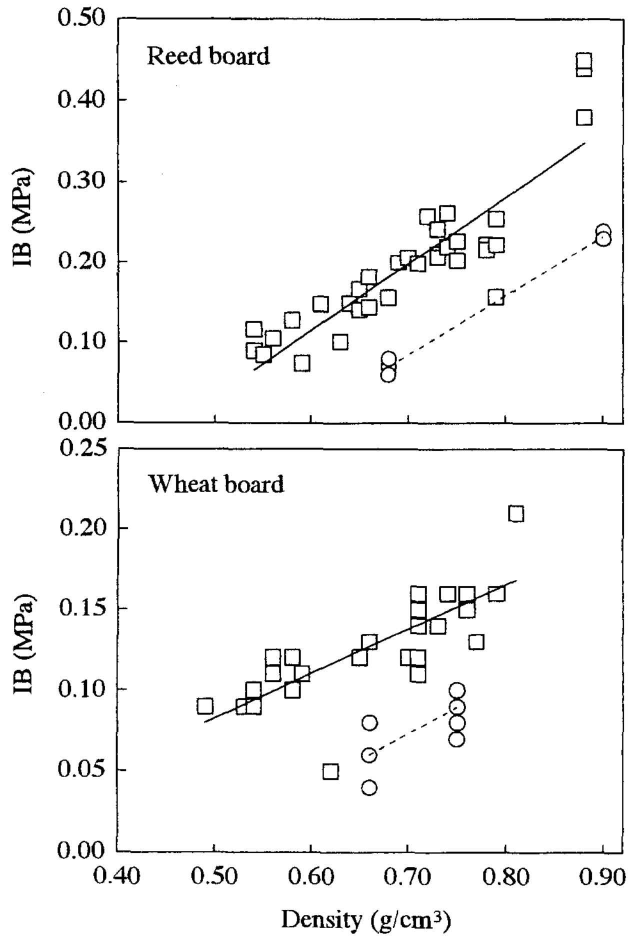

Fig. 2. Relation between density and internal bond (*IB*) of boards made from reed and wheat straws. *Squares*, fine particles; *circles* coarse particles

great effect on board properties.[13] Generally, small particles result in low MOR and high IB. The results of this experiment may be attributed to the inherent characteristics of the raw materials. Like other nonwood lignocelluloses, both reed and wheat straws have higher hemicellulose and ash contents than wood. The outer surface of these two straws are covered with much silica and wax.[7,8,11] The combination of the inherent nonpolar, hydrophobic characteristics of the surface of straws caused by silica and wax, and the polar, hydrophilic nature of urea-based resins result in difficult adhesion between these two components.[5] In the case of smaller particles, the specific surface of the particles is increased with a reduction in the surface containing silica and wax. This results in a reduced bonding inhibition effect caused by silica and wax, and so higher MORs and IBs are obtained.

Figure 3 indicates the effect of particle sizes on the TS of boards at different densities. In general, the TS after a long duration of water immersion would have a tendency to increase with increasing board density because of the greater springback of compacted particles in boards of higher density.[14] However, in this study, the TS values

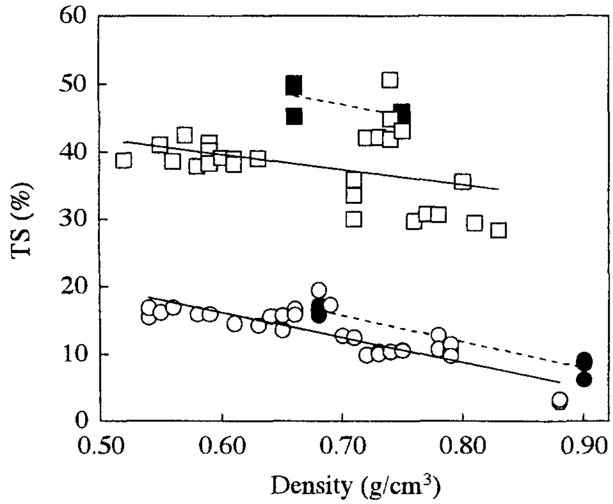

Fig. 3. Relation between density and thickness swelling (*TS*) of boards made from reed and wheat straws. *Open circles*, reed board from fine particles; *filled circles*, reed board from coarse particles; *open squares*, wheat board from fine particles; *filled squares*, wheat board from coarse particles

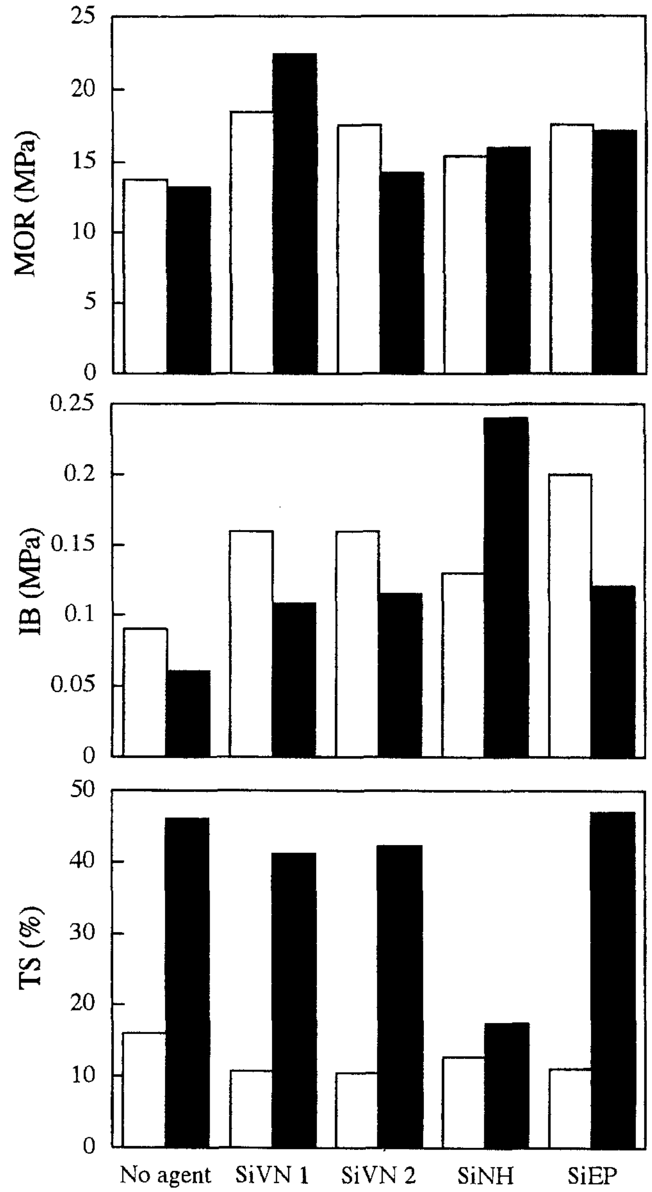

Fig. 4. Improvement of board properties using various silane coupling agents. All the values are corrected to the board density of 0.70 g/cm^3. *Open squares*, reed board; *filled squares*, wheat board

of both reed and wheat boards were found to decrease with an increase in board density, especially reed board. The reduction in TS in this case might be due to the following causes: The water immersion time was rather short. Although the sample size was small, a 2-h immersion may be too short to allow thorough penetration of water into the board; hence most of the compacted particles did not experience complete springback. There are more voids in the low-density boards than in the high-density boards. Consequently, more water was being absorbed, resulting in a greater springback. At high density, the higher bonding strength may play a more dominant role than the compaction ratio where water absorption is concerned during such short immersion.

The TS values of boards with fine particles tended to be lower than those made from coarse particles. This may be caused by the closer structure of the board, where the contact among the fine particles is better. Higher IB strength may also contribute to the reduction in water penetration. Figure 3 also shows that the TS of wheat boards is much greater than that of reed boards, which may reflect the higher IB values of reed boards with the same densities as shown in Fig. 2.

Improvement of board properties using silane coupling agents

The results above indicate that the board properties are not satisfactory and must be improved. Figure 4 and Table 4 show the effects of silane coupling agents on the properties of board manufactured from coarse particles. All the values were corrected to a board density of 0.70 g/cm^3, based on the linear equations obtained from the correlation between board density and properties. The board properties were generally improved by the addition of silane coupling agents. For reed board, even though there was no substantial increase in MOR, the IB improved significantly. After adding SiEP the IB value was twice as high as that of the control. It was also found that the TS decreased by about 5% compared to that of the control. For wheat board, incorporation of SiNH resulted in an IB of 0.24 MPa, which is four times that of the control. SiNH also reduced the TS of wheat board to about one-third of that of the control, resulting in a final TS of 17%.

The improvement caused by each coupling agent was different. Addition of SiNH results in great improvement in IB in wheat board, but SiVN1 and SiVN2 were not as effective. This difference may be related to the difference in chemical structure and the optimal adhesive type in which they can function. SiVN1 and SiVN2 are compatible with

Table 4. Improvement of board properties using various silane coupling agents

Property	Wheat board				Reed board			
	SiVN1	SiVN2	SiNH	SiEP	SiVN1	SiVN2	SiNH	SiEP
MOR	1.70	1.08	1.21	1.30	1.35	1.28	1.12	1.28
IB	1.80	1.92	4.00	2.00	1.78	1.78	1.44	2.22
TS	0.89	0.92	0.38	1.02	0.67	0.66	0.80	0.69

Improvement is expressed as the ratio of the properties of board with silane coupling agent to that without silane coupling agent (control).
MOR, Modulus of rupture; *IB*, internal bond; *TS*, thickness swelling.

polyethylene and polyester resins, respectively; and SiNH and SiEP are compatible with formaldehyde and epoxy resins

Epoxy silane (SiEP) is considered to be more effective for reed board, whereas the properties of wheat board were greatly improved by adding SiNH. This improvement might be due to reactions among the coupling agent, UF resin, and the particles. There are two functional groups in the SiEP molecule: the methoxysilane group and the epoxy group. The methoxysilane is readily hydrolyzed, and the silanols formed may react with silica on the material surface to form strong siloxane bonds. It is speculated that reactions between the epoxy and amide groups in the resin molecules could also have taken place, and the amido formed is capable of reacting with the hydrate of formaldehyde. Ethoxysilane and amino are the two main functional groups of the SiNH molecule. Ethoxysilane may experience a reaction similar to that of methoxysilane, and the reaction between amino and hydroxyl groups in UF resin may occur. The curing reactions of UF resin after adding silane coupling agents could be more complicated, and the above reactions must be further investigated.

Conclusions

The properties of UF-bonded reed and wheat particleboards manufactured from two types of particle at different densities were determined. Particle size was found to have a profound effect on the board properties. The properties of both reed and wheat boards produced from fine particles were better than those made from coarse particles because of their higher bondability.

The board properties are closely related to the board density. An increase in board density resulted in higher mechanical properties and dimensional stability. However, the IBs of the samples at 0.50–0.80 g/cm^3 density were lower than the minimum requirement of GB/T 4897-92 (0.35 MPa). The TS values for both reed and wheat particleboards exceeded the standard requirement of 8%.

Silane coupling agents can be used to improve the board properties of reed and wheat particleboards. The improvement was more obvious for IB than for MOR or TS. It seems that SiEP was more effective for reed board, whereas SiNH was better for wheat board. The mechanism of bondability improvement caused by the silane coupling agent is not fully understood at this stage, and more detailed investigation is necessary for clarification.

Acknowledgments The authors thank Professor Zhenguo Lui, Forest Products Institute, Heilongjiang, China, for his invaluable support and assistance throughout this study, and Ms. Ee Ding Wong for her critical reading.

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