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Effects of silane coupling agent level and extraction treatment on the properties of UF-bonded reed and wheat straw particleboards

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Abstract Urea formaldehyde resin bonded reed and wheat particleboards with a density of 0.7 g/cm³ were manufactured from two types of particle: fine and coarse particles. The effects of the silane coupling agent (SCA) level and ethanol-benzene (EB) treatment on the board properties were examined. For SCA, epoxide silane (SiEP) and amino silane (SiNH) were used for reed and wheat particles, respectively. The results are summarized as follows. (1) For both reed and wheat boards, the internal bond (IB) strength and thickness swelling (TS) were significantly improved at up to 5% SCA content, but the effectiveness of treatment kept constant at above 5%. (2) The level of SCA had little effect on the bending strength, especially for the boards composed of coarse particles. (3) EB treatment upgraded both the IB and TS of wheat board significantly. (4) SiEP incorporation improved the IB and TS of reed board significantly, whereas EB treatment was more effective for wheat board. (5) The dimensional stability of both reed and wheat boards under varying humidity could be improved by increasing the level of SCA and by EB treatment. EB treatment was more effective than SCA addition.

Key words Reed straw · Wheat straw · Urea formaldehyde resin · Particleboard · Chemical treatment

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Introduction

During the production of composite boards from the straws of herbaceous plants (e.g., wheat, reed, paddy) the inferior bondability between urea formaldehyde (UF) resin adhesive and these materials has been a major concern. Isocyanate is an alternative resin that can be used to improve the properties of these strawboards,1-4 but the application of isocyanate is hindered by its high cost; hence, it is not commonly utilized, especially in developing countries. The properties of wheat board could be improved significantly by using polyurethane-based binder (RUBINATE), even at a low resin content level.⁵ It has been reported that the properties of wheat and rape strawboards could be improved to a certain extent by incorporating wood particles into the straw materials.³ Chow concluded that the properties of boards made from steam pressure-refined corn crop residues were superior to those produced from hammer-milled furnish.⁶ Recently, pretreatment of these herbaceous materials to remove substances such as waxes and extractives has been identified as a possible means to improve their bondability with UF.³ However, only a limited research on this subject has been reported so far.

Our previous papers reported the upgrading of UF-bonded reed and wheat straw particleboards by using silane coupling agent (SCA) and the improvement mechanism of bondability after SCA and extraction treatments. As a continuation from the earlier work, this study investigates the effects of the SCA level and ethanol-benzene treatment on the properties of reed and wheat straw particleboards.

Materials and methods

Raw materials

The raw materials used in this study are the same as in the previous studies^{7,8} (i.e., reed and wheat straws, including

leaves, obtained from northeastern China). The resin used was a commercial UF resin (TKB-1) formulated by Oshika Shinko with a solid content of 65% and a U:F molar ratio of 1.0:1.4. NH₄Cl solution of 20% concentration was used as a hardener for the UF resin, with an addition level of 5% based on the resin solid content. Two types of SCA, namely, epoxide silane (SiEP) and amino silane (SiNH), were supplied by Shin-Etsu Chemical. The ethanol-benzene (EB) solution was prepared by mixing one volume of 95% ethanol with two volumes of benzene.

Board manufacture

The reed and wheat straws were first cut to about 8cm length using a hand-cutter, followed by disintegration in a hammer mill. The hammer-milled particles were then screened through an 8mm sieve, where particles retained on the screen and passing through the screen were classified into coarse and fine particles, respectively. The corresponding particle geometry is summarized in Table 1. A portion of the coarse particles was used for EB treatment, where the particles were first oven-dried at 60°C for 72h and then immersed in EB solution in a glass container placed in a 50°C waterbath for 24h. The control and treated particles were then oven-dried to 2%–3% moisture content (MC) at 60°C prior to board fabrication.

Three types of homogeneous particleboard were fabricated: control board, SCA-treated board, and EB-treated board. The boards were manufactured as $9 \times 350 \times 400 \,\mathrm{mm}$, at a targeted density of $0.7 \,\mathrm{g/cm^3}$.

The particles were sprayed with UF resin in a rotating drum blender, at a resin content of 12% based on the ovendried weight of the particles. The resin was applied by means of an airless spray gun. Based on our previous conclusions about SCA-treated boards,7,8 SiEP and SiNH were used for reed and wheat particles, respectively, in this study. For coarse particles, the SCA was incorporated into the resin solutions at 2%, 5%, and 7% based on the resin solid content. For fine particles, only 2% SCA was added. Table 2 summarizes the processing parameters for reed and wheat particleboards. The resin-sprayed particles were then hand-formed in a forming box and hot-pressed at 130°C for 6min. A maximum pressure of about 3.0 MPa was applied during hot pressing. Because of the inferior permeability of straw mat, the breathing period at the end of the hot pressing cycle was monitored carefully to prevent delamination.

Table 1. Geometry of reed and wheat particles of different sizes

| Parameter | Reed | | Wheat | |
|---------------------------------------|-----------------------------------|---------------------------------|----------------------------------|---------------------------------|
| | Coarse | Fine | Coarse | Fine |
| Length (mm) Width (mm) Thickness (mm) | 12.7–28.8 0.8–1.6 0.17–0.39 | 2.5–6.0 0.4–1.0 0.08–0.22 | 9.8–23.1 0.8–2.3 0.16–0.42 | 2.8–7.5 0.5–1.4 0.08–0.28 |

Board evaluation

Prior to the evaluation of mechanical properties and dimensional stability, the particleboards were conditioned for 2 weeks at 20° C and $65\% \pm 5\%$ relative humidity (RH). The boards were evaluated in accordance with the Japanese Industrial Standard for Particleboards (JIS A 5908).

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

In addition, the linear expansion (LE), thickness changes (TC), and equilibrium moisture content (EMC) of two 50 \times 200 mm specimens from each board were examined after exposure to an RH cycle of 33%, 66%, 98%, 66%, and 33%. The initial and final dimensions of the specimens were measured after oven-drying until they reached a constant weight at 60°C; the specimens were then cooled in a desiccator at 20°C (8% RH). The RH in the desiccator was recorded through an RH recording meter. The corresponding changes in length, thickness, and weight were determined after the samples were conditioned to equilibrium at 33%, 66%, and 98% RH over saturated solutions of MgCl₂, NaNO₂, and CaSO₄, respectively, in air-tight moisture chambers at 20°C. The length was measured to the nearest 0.01 mm. After the RH cycle and measurements, the samples were subjected to drying again at 105°C until a constant weight was reacted and then weighed to determine the EMC.

Results and discussion

Bending properties

Figures 1 and 2 show the effects of the SCA level and EB treatment on the moduli of rupture (MOR) and elasticity (MOE) of reed and wheat particleboards. Irrespective of the addition level, SCA was found to have little effects on the MOR and MOE of either reed or wheat board. EB

Table 2. Processing variables for reed and wheat particleboards

| Code | Particle types ^a | SCA addition levels (%) ^b | EB treatment |
|--------------|-----------------------------|--------------------------------------|--------------|
| Control | С | _ | _ |
| SCA/2 | C | 2 | - |
| SCA/5 | C | 5 | |
| SCA/7 | C | 7 | |
| EB treatment | С | _ | 50°C, 24h |
| Control-F | F | | _ |
| SCA/2-F | F | 2 | |
| | | | |

SCA, silane coupling agent; EB, ethanol-benzene

^aC, coarse particles; F, fine particles

^bBased on the solid weight of resin

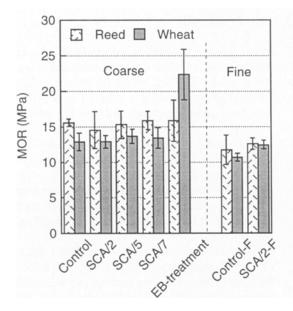


Fig. 1. Effect of silane coupling agent (SCA) level and ethanolbenzene (EB) treatment on the modulus of rupture (MOR) of reed and wheat particleboards. Refer to Table 2 for explanation of legend. Vertical lines through the bars represent the standard deviation from the mean value

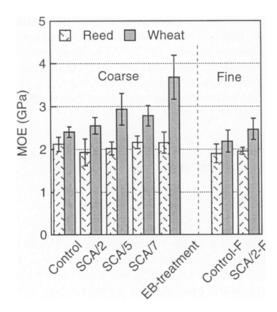


Fig. 2. Effect of SCA level and EB treatment on the modulus of elasticity (MOE) of reed and wheat particleboards. Refer to Fig. 1 for other explanations

treatment improved the MOR and MOE of wheat board by about 70% but had no significant effect on reed board. Similar to conventional particleboard, the bending properties of boards made from fine particles were relatively low compared to those composed of coarse particles. Based on the inherent characteristics of the raw materials, the high-strength reed straw contributed to the superior MOR of reed board, whereas the wheat board comprised of elastic wheat straw had higher MOEs.

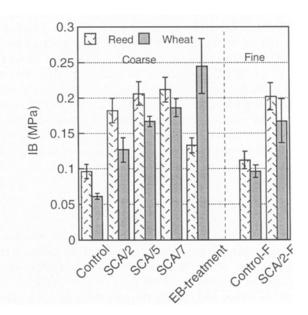


Fig. 3. Effects of SCA level and EB treatment on the internal bond strength (IB) of reed and wheat particleboards. Refer to Fig. 1 for other explanations

Internal bond strength

The IB strengths of reed and wheat boards with different SCA contents and EB treatment are shown in Fig. 3. The IBs of both reed and wheat boards were found to increase with higher SCA content. The IB improved significantly when up to 5% SCA was incorporated, but the effectiveness of treatment kept constant at above 5%. A significant improvement was observed in EB-treated wheat board, where the IB was four times that of control. Because the EB extractive content of wheat particles was about 7% and negligible in reed particles, the significant improvement of IB in wheat particleboard could be attributed to the removal of wax-like substances from the straw surface, facilitating the adherence of UF resin to the active hydroxyl sites of the cellulose.

Comparing the effectiveness of SCA and EB treatment, SCA was more effective for reed board and EB treatment was better for wheat board. Based on our previous conclusions that SCA was effective for improving the wettability of the reed straw surface and EB extraction was more effective for wheat straw, the improvement of the properties of reed and wheat boards using SCA and EB treatment reflects a good correlation with the trend of wettability improvement. In addition, the SCA added resincoated particles and EB-treated particles became less sticky and had better resin penetration than the untreated particles. This could be attributed to improved wettability of the straw surfaces. Similar to the earlier study, the IB values of both reed and wheat boards fabricated from fine particles were superior to those produced from coarse particles.

Thickness swelling

Figure 4 shows the effects of SCA level and EB treatment on the TS of reed and wheat boards produced from fine and

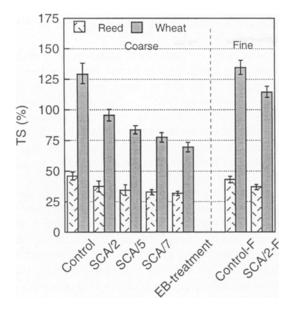
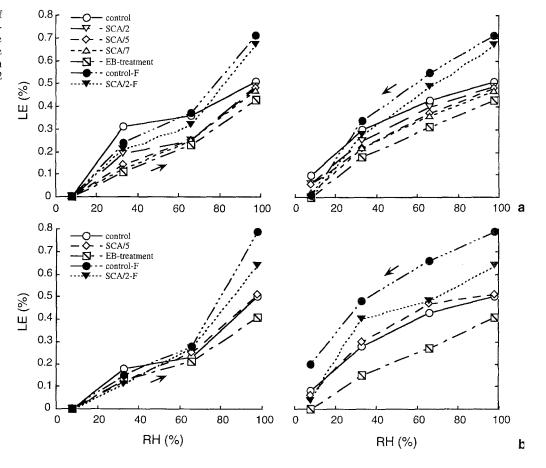


Fig. 4. Effect of SCA level and EB treatment on the thickness swelling (TS) of reed and wheat particleboards. Refer to Fig. 1 for other explanations

coarse particles. Generally, TS decreased with increasing the SCA content. This reduction in TS was significant when the SCA content was below 5%, but the significance of treatment almost kept constant when the SCA content was above 5%. The TS values of EB-treated reed and wheat boards were reduced by about 30% and 50%, respectively. EB treatment was more effective for reducing the TS of both reed and wheat boards than was SCA addition. This is attributed to improved interparticle bonding after EB treatment that resulted in lower water penetration into the board.

The TS values of wheat board were generally higher than those of reed board. This is related to the inherent characteristics of the raw materials, where reed straw is more water-resistant than wheat straw. This superior water-repellent property of reed straw is due to its higher silica content. Figure 4 shows that the TS of wheat board produced from fine particles is higher than that for those made from coarse particles, whereas the TS of reed board was not affected by particle size. This may be attributed to the superior water resistance of reed straw, which prevented thorough penetration of water into the board despite immersion in water for 24h. Consequently, most of the compacted particles might not have experienced complete spring-back, making it impossible to determine the true effect of particle size on the board TS.

Fig. 5. Linear expansion (LE) of reed (a) and wheat (b) particle-boards under different relative humidity (*RH*) during moisture absorption (left) and desorption (right) processes. Refer to Table 2 for other explanations



Dimensional stability under varying relative humidity

Figure 5 shows the LE of reed and wheat boards during moisture absorption and desorption processes under different RHs. The LE of both the reed and wheat boards improved with increasing SCA content. The LE of the board with EB treatment was superior to that added with SCA. Generally, the LE increased gently when the RH was increased up to 66% but recorded exceedingly high values after attaining equilibrium at 98% RH. The LE of the EB-treated board was completely reversible upon over-drying at the end of the RH cycle. This superior LE could be attributed to the stronger interelement bonding in EB-treated board.

The corresponding TC of the reed and wheat boards under the above RH cycle is shown in Fig. 6. Similar to LE, the TC increased gradually as the RH was increased up to 60% but rose rapidly when subjected to 98% RH. For reed and wheat control boards, the thickness increased about 2.0 mm and 2.5 mm, respectively, after redrying at the end of

RH cycle, resulting in the residual TC to be 22% and 28%, respectively.

The LE of both reed and wheat boards produced from fine particles registered higher values than those prepared from coarse particles, but a reversed trend was observed in the TC. This may be due to a higher proportion of vertically oriented elements in boards composed of fine particles compared to those made from coarse particles; hence, the dimensional stability in the thickness direction improved, but there was reduced longitudinal stability.

Similar to conventional particleboard, the degree of spring-back in both reed and wheat boards was highly dependent on the EMC, as shown in Figs. 6 and 7, where wheat board recorded a higher TC and EMC than did reed board when subjected to 98% RH. The higher TC of wheat board may be caused by a greater expansion due to higher moisture absorption, in addition to the recovery of a greater compressive set resulted from a higher compaction ratio (about 2.26).⁷

Fig. 6. Thickness changes (TC) of reed (**left**) and wheat (**right**) particleboards under different RHs during moisture absorption and desorption processes. Refer to Table 2 for other explanations

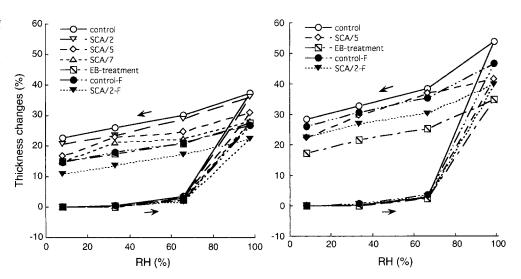
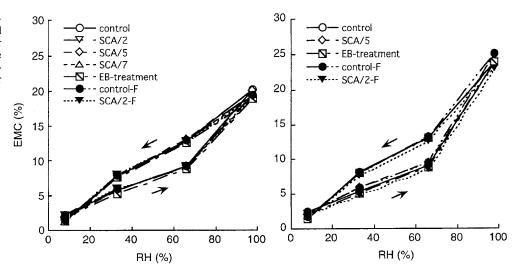


Fig. 7. Equilibrium moisture content (EMC) of reed (left) and wheat (right) particleboards under different RHs during moisture absorption and desorption processes. Refer to Table 2 for other explanations



Conclusions

The IB and TS of both reed and wheat particleboards increased significantly when up to 5% SCA was incorporated, but the effectiveness of treatment kept constant above 5% SCA content. EB treatment was found to improve the IB and TS of wheat board significantly. Whereas SCA incorporation was more effective for reed board, EB treatment was better for wheat board. Improvement of the IB and TS of reed and wheat boards by SCA and EB treatment showed good correlation with the improved wettability of straw surfaces reported in the previous study.8 The dimensional stability of both reed and wheat boards under various RHs also improved with increasing SCA content. EB treatment resulted in greater improvement in board dimensional stability than with SCA. The significant improvement of board properties by EB treatment may be attributed to improved interparticle bonding, which is related to the exposure of reactive components on the straw surfaces.

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