

Modified Formaldehyde-based Resin Adhesives for Rice Hull-Wood Particleboard

Chung-yun Hse¹ and Elvin T. Choong²

Abstract

A study was conducted to develop an effective and economical resin system to improve the physical and mechanical properties of rice hull-wood composites. Boards 0.5 inch (12.5 mm) thick were made from a mixture of rice hulls and pine particles with three main resin types: urea formaldehyde (UF), phenol formaldehyde (PF), and polyisocyanate (ISO), in various adhesive formulations. Results indicated that conventional UF and PF resins did not perform well with rice hull-wood composites. However, a modified resin system with ISO as minor component significantly improved the boards' strength properties and dimensional stability. Resin content had a significant effect on the quality of the boards, with the higher resin content resulting in stronger boards. The boards bonded with the 1%ISO / 6%UF resin system had higher internal bond strength, and attained the best dimensional stability and the highest bending strength (MOR) and stiffness (MOE).

As global population increases and developing countries increase their use of wood and paper products, there is new interest in producing and conducting research on composite board from agricultural fibres. Rice hull is quite fibrous by nature and requires little energy input to prepare, so its suitability for the manufacture of particleboards has been assessed in a number of studies (Vasishth 1971; Hancock and Chandramouli 1974; Mahanta et al. 1980; Viswanathan et al. 1987). However, particleboards made from rice hulls have not found commercial acceptance because substantially more adhesive is needed for rice hulls than for wood flakes to yield boards with acceptable properties (Vasishth 1971; Chen 1979).

The reason for the higher resin requirement for bonding rice hulls is not completely understood,

but a comparison between wood and rice hull showed that rice hull has less holocellulose and a much higher ash content (Houston 1972). The predominant component of its ash is silica (Luh 1991). The silica covers almost the entire outer layer of the rice hull surface which also contains the water repellent cuticle (Juliano 1985). This silica layer and the partially hydrophobic surface of rice hull are incompatible with aqueous urea formaldehyde (UF) or phenol formaldehyde (PF) resins and prevent the formation of a good bond between rice hull surfaces. Thus, a new and improved resin adhesive system is needed to produce high quality rice hull particleboards.

Urea formaldehyde resins are often fortified with melamine to increase the bond strength and water resistance of particleboard. More recently, highly reactive polyisocyanate (ISO) has been used to modify UF resins (Deppe and Ernst 1971; Deppe 1977; Pizzi 1981; Liu and Bingley 1992) and PF resins (Hse 1978, 1980) for board products. ISO modified adhesives improve the bond strength and performance of wood particleboards, so they might have similar beneficial effects on the bonding of rice hulls. Therefore,

¹Southern Research Station, USDA Forest Service, Pineville, Louisiana, 71360, USA.

²School of Forestry, Wildlife, and Fisheries, Louisiana State University Agricultural Center, Baton Rouge, Louisiana, 70803, USA. Email: chse@fs.fed.us

Table 1. Summary of resin type and resin application levels used in the study

Resin type	Resin application level	
	Low	High
ISO/UF	(5%): 1%ISO/4%UF	(7%): 1%ISO/6%UF
ISO/PF	(5%): 1%ISO/4%PF	(7%): 1%ISO/6%PF
ISO/PF/UF	(5%): 1%ISO/1%PF/3%UF	(7%): 1%ISO/1%PF/5%UF
UF	5%UF	7%UF
PF	5%PF	7%PF
ISO	2.5%ISO	5%ISO

the objective of this study was to investigate the feasibility of developing multi-polymer adhesive systems that could take advantage of the durability of phenol formaldehyde, the low cost of urea formaldehyde, and the reactivity of polyisocyanate for manufacturing rice hull-wood composite products. This paper reports preliminary findings and discusses further developments necessary to improve the properties of rice hull-wood particleboard bonded with modified ISO/UF adhesives.

Materials and Methods

Experimental variables

The resin types used in this study are polyisocyanate/urea formaldehyde (ISO/UF), polyisocyanate/phenol formaldehyde (ISO/PF), polyisocyanate/phenol formaldehyde/urea formaldehyde (ISO/PF/UF), UF, PF, and ISO resin adhesives. The ISO/UF, ISO/PF, and ISO/PF/UF adhesive systems were obtained by the alloying process described in a previous study (Hse 1978). The process involves applying minor amounts of polyisocyanate before adding major amounts of either UF or PF resin to the furnish, then letting the combined adhesive react in situ to obtain an improved thermosetting adhesive resin. The UF-, PF-, and ISO-bonded boards acted as controls. There were two resin application levels, based on percentage oven dry weight of rice hull furnish: high (7%) and low (5%), except for ISO which was applied at 2.5% and 5%. The various resin formulations are summarised in Table 1.

Board manufacture

All boards were prepared in the laboratory with equal weight (50:50) mixtures of rice hulls and

wood particles. The rice hulls were brought to the laboratory from a local rice mill, passed through a Sears chipper to break the boat-shape hull particles, sieved to remove fines, and then dried at 105°C to an average moisture content of 4%. The dry southern pine (*Pinus* sp.) wood particles were obtained from a local particleboard plant and used without further treatment.

To prepare each board, a pre-weighed mixture of rice hulls and wood particles was placed in a rotating drum-type blender. The resin was weighed and applied to the mixture using air-atomising nozzles. After blending, the rice hull-wood furnishes were carefully felted into a 19 × 20 inch (475 mm × 500 mm) box to form a mat. The mat was transferred immediately to a 40 × 40 inch (1 m × 1 m) single-opening hot press with the platen temperature regulated at 370°F (188°C). Sufficient pressure (about 550 psi / 3790 kPa) was applied so that the platen, closed to 1/8-inch thickness, stopped in 45 s. The press time was 255 s.

Sampling and testing

All boards were conditioned in an environmental chamber at 50% RH and 80°F (40.6°C) ambient temperature to obtain an average equilibrium moisture content of 5.8%. After conditioning, each board was cut to yield ten 2 × 2 inch (50 mm × 50 mm) specimens for tensile strength perpendicular to the face (internal bond strength), five 2 × 14 inch (50 mm × 350 mm) static-bending specimens, and two 6 × 6 inch (150 mm × 150 mm) specimens for dimensional stability tests.

The mechanical tests were performed in accordance with ASTM standard D- 1037-99 (ASTM 1999) using an Instron universal testing

Table 2. Summary of resin type and resin application levels used in the study

Resin type	Resin content	Density (psi)	MOR (psi)	MOE (10 ² psi)	IB (psi)	TS (%)
PF	5.0	53.61	1735.5	387.2	29.2	33.7
PF	7.0	55.69	1663.1	408.2	43.0	23.7
UF	5.0	56.47	1434.5	399.2	22.2	57.0
UF	7.0	60.54	1769.7	470.9	44.3	46.6
ISO*	2.5	51.00	1645.6	321.0	25.5	28.3
ISO*	5.0	51.62	3022.1	464.3	59.5	12.6
ISO/PF	1/4	53.01	2285.8	410.9	60.5	21.1
ISO/PF	1/6	54.60	2521.6	461.5	78.0	18.5
ISO/UF	1/4	57.86	2252.8	400.4	85.9	23.7
ISO/UF	1/6	54.61	3162.4	494.1	97.6	12.3
I/P/U	1/1/3	54.20	2570.5	451.4	60.1	25.5
I/P/U	1/1/5	50.97	2412.2	447.2	73.8	13.5

*The average moisture content of particles was 11.2 and 9.25% for rice hull and pine, respectively.

machine. The dimensional stability test measured changes in thickness and weight after the specimens had been submerged in water at room temperature for 24 h.

Results

The average physical and mechanical properties of the particleboards are summarised in Table 2. The effects of resin types and resin contents were evaluated by analysis of variance at the 5% significance level ($P < 0.05$), and all differences discussed below were significant at that level.

Internal bond strength

The average internal bond strength ranged from 22.2 psi (153.1 kPa) for UF resin content at 5%, to 97.6 psi (672.9 kPa) for ISO/PF resin content at 1% / 6%. The analysis of variance showed that the internal bond strengths differed significantly among resin types and resin contents. Figure 1 shows that boards bonded with PF and UF resins had very low internal bond strength. The average values of 43.0 psi (296.5 kPa) (PF) and 44.3 psi (305.4 kPa) (UF) at high (7%) resin content levels are far lower than the 130 psi (896.3 kPa) required by the American National Standard of Mat-Formed Wood Particleboard. The results suggest that the inclusion of 50% rice hull in a wood particleboard substantially reduces internal bond strength. As expected, the internal bond strength of rice hull-wood particleboard increased as resin content increased. The internal

bond strength of 5% PF-bonded board was 29.2 psi (201.3 kPa), and that of 7% PF-bonded board was 43 psi (296.5 kPa), an increase of 47%. When UF was used, comparable internal bond strength figures at 5 and 7% were 22.2 psi (153.1 kPa) and 44.3 psi (305.4 kPa), respectively, an increase of 99%. These trends and values are similar to those reported by Chen (1980).

The internal bond strengths of boards that were bonded with ISO were only slightly higher than the internal bond strengths of PF- or UF-bonded boards because the ISO resin content was substantially lower than the PF or UF controls. Note, however, that the internal bond strengths

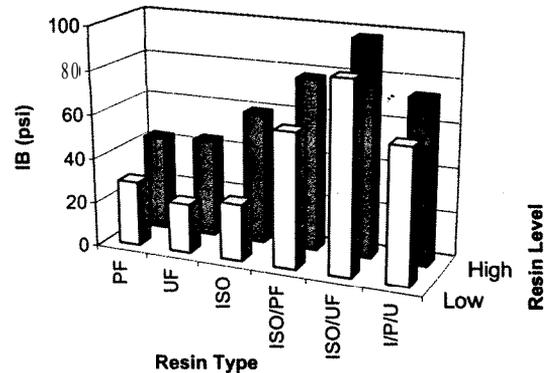


Figure 1. Effect of resin type and resin content on the internal bond strength of rice hull-wood composites

of **ISO-bonded** boards at 5% resin content level were 104% and 168% higher than those of **PF**- and **UF**-bonded boards at the same resin contents, respectively. The significant effect of **ISO** on bonding strength also resulted in significantly higher internal bond strength for all boards bonded with **ISO-modified** resin systems. For example, the internal bond strengths of 1/4 **ISO/PF** and 1/4 **ISO/UF** were 107% and 287% greater than those of 5% **PF** and **UF**, respectively.

Substantially higher bonding strength was found in boards bonded with the **ISO/UF** resin system than in those bonded with **ISO/PF**. The internal bond strengths of 1/4 **ISO/UF** and 1/6 **ISO/UF** were 42% and 25% higher than those boards bonded with the **ISO/PF** system. The low cost of **UF** resins suggests that there is merit in further investigation of the use of **ISO/UF** resin systems for the bonding of rice hull-wood composites.

Note, however, that the **ISO/PF/UF** resin system yielded the lowest internal bond strength of the three **ISO-modified** resin systems. This may be due to the basic incompatibility of the curing properties of **PF** and **UF** resins (i.e. **PF** resin cures at alkaline **pH**; **UF** resin cures at acidic **pH**).

Bending strength (MOR)

The relationships between modulus of rupture (MOR) and resin type for the two resin content levels are shown in Fig. 2. Boards bonded with polyisocyanate had the highest bending strength. The significant effect of **ISO** in increasing bending strength also occurred with the mixed **ISO** resin

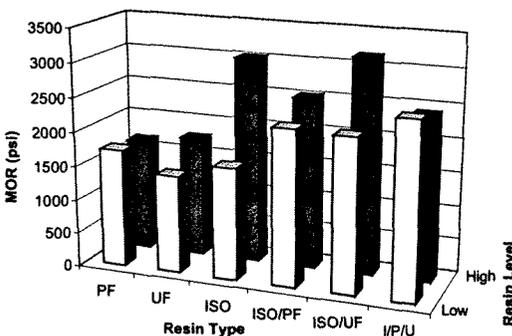


Figure 2. Effects of resin type and resin content on the MOR of rice hull-wood composites

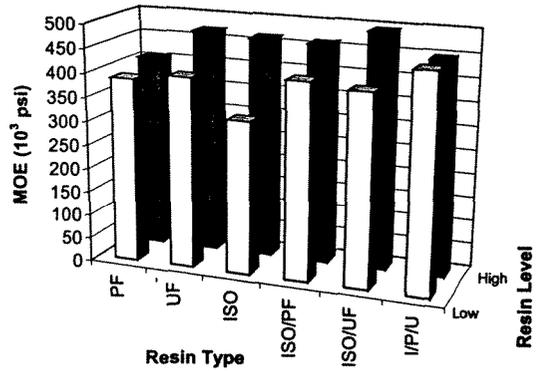


Figure 3. Effects of resin type and resin content on the MOE of rice hull-wood composites

adhesive system, i.e. **ISO/PF**, **ISO/UF** and **ISO/PF/UF**, even though the **ISO** content was only 1% in the resin systems. The **MORs** of boards bonded with 5% **ISO**, 1/6 **ISO/PF**, 1/6 **ISO/UF**, 1/1/3 **ISO/PFAJF** and 1/1/5 **ISO/PF/UF** met the American National Standard of Mat-Formed Particleboard standard of 2400 psi (16.5 MPa).

Stiffness (MOE)

The relationships between modulus of elasticity (MOE) and resin type for the two resin content levels are shown in Fig. 3. The **MOEs** of most boards exceeded the 350 000 psi (2.4 GPa) minimum requirement of the American National Standard. The exceptions were boards that were bonded with 2.5% **ISO**. The significant effect of **ISO** on MOE was again evident. Boards bonded with 5% **ISO** had the highest MOE when compared to boards with the same resin content levels, and exceeded even the MOE of boards bonded with **PF** and **ISO/PF/UF** resins at the higher resin content. As with **MOR**, boards bonded with **ISO/UF** had the highest MOE of all boards.

Thickness swelling

Figure 4 shows the relationships between thickness swelling and resin type for the two resin contents. Thickness swelling was greatly affected by resin content. Increasing the resin content from 5% to 7% reduced thickness swelling by 18.0% and 23.7% for boards bonded with **UF** and **PF**, respectively. All **ISO-based** resin formulations exhibited excellent dimensional stability. Boards

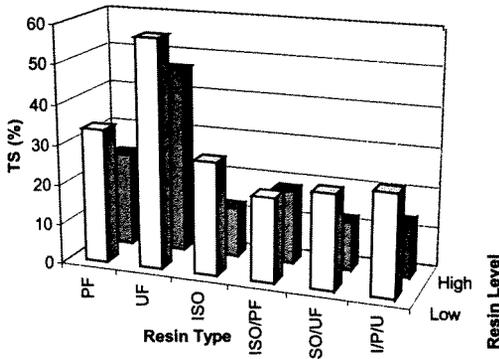


Figure 4. Effects of resin type and resin content on the thickness swelling of rice hull-wood composites

bonded with 1/6 ISOAJF had the lowest thickness swelling. The improvement in thickness swelling was more noticeable with increased resin contents. The reduction in thickness swelling was 55%, 48%, and 47% for ISO, ISO/UF and ISO/PF/UF, respectively, as resin content increased from low to high levels. These values show that an ISO-modified resin system can effectively enhance moisture resistance of rice hull-wood particleboards.

Discussion and Conclusion

Modified resin systems with ISO as a minor component significantly improved the strength properties and dimensional stability of rice hull-wood particleboards. As expected, resin content had an appreciable effect on board performance. In most cases, the higher the resin content, the better the performance of the boards, in terms of both strength properties and dimensional stability. The superiority of the resin system containing ISO is apparent for all resin levels in the test. Conventional UF and PF resins by comparison did not perform well with rice hull-wood composites.

The most interesting result in the study was the overall improved performance of the ISO/UF resin system. The boards bonded with 1% ISO/6% UF resin had the highest internal bond strength, the lowest thickness swelling and the highest MOE and MOR. Since the cost of UF (US\$0.17 lb⁻¹) is much lower than the cost of ISO (US\$0.85 lb⁻¹), there could be substantial

economic gains by using UF resin as the major component in an ISOAJF system.

The internal bond strength was the only board property evaluated in the study that did not meet the requirements of the American Standard of Mat-Formed Particleboard. The study showed that the internal bond strength could be increased most appreciably by raising the resin content level, and the ISOAJF resin system would be expected to provide the greatest potential for further improvement on an internal bond strength per unit cost basis. Note, however, that to maintain optimum internal bond strength : cost ratio, the increase in resin content would have to be obtained from an increase in UF resin level and not from the ISO content. Without the increase in the ISO content, the application efficiency of ISO in the resin could be critically important in further improving the performance of the ISO/UF resin system. The recent development of emulsifiable PMDI (polymeric methyl diisocyanate) for the medium-density fibreboard industry (Moriarty 1999) provides an additional avenue for efficiency improvements. A study is in progress, based on emulsifiable isocyanate adhesives in conjunction with a solvent-extended isocyanate resin system, to improve the efficiency of ISO application. It is anticipated that satisfactory internal bond strength will be achieved by improving the ISO efficiency and by increasing the UF content in the resin system.

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