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ORIGINAL ARTICLE

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Properties of wheat straw particleboards bonded with different types of resin

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Abstract Wheat straw particleboard bonded with a urea–formaldehyde (UF) resin, usually employed in the manufacture of wood-based particleboards, or with a resin based on epoxidised oil was manufactured using a compression molding machine. The effects of resin type on internal bond strength, flexural modulus, and thickness swelling were examined. The properties of boards using UF resins were poor. Internal bond strength and thickness swelling, linked to adhesion quality, were especially low. The high compatibility between straw particles and oil-based resin was explained in terms of straw surface free energy. In straw, this parameter exhibits a much lower polar component than wood species and leads to higher compatibility with resins based on oil than with water-soluble systems like UF.

Key words Wheat straw · Particleboard · Epoxidised oil · Urea formaldehyde resin

Introduction

During recent years, increasing attention has been paid to composite materials of vegetal origin. This interest is justified by the environmental advantages of these substances. They allow reduced consumption of raw materials from petroleum and/or forestry resources and, in the case of agricultural resources, are rapidly renewable. These materials might also constitute new outlets for agriculture and contribute toward sustainable growth.

Amongst agriculture production, wheat is the second most cultivated cereal plant worldwide. Wheat straw is the main by-product from cereal harvesting and is primarily used in animal husbandry. However, it can now be found in a wide range of industrial outlets[1] because straw demand and prices have decreased over the past 15 years. Straw could be available in very large amounts to new industrial applications.[2] Amongst these, the production of particleboard panels, that are at present almost exclusively produced from timber or timber by-products like saw dust, seems feasible. Low-density straw panels have already been suggested for applications in thermal[3] and acoustic[4] insulation. Panels having properties corresponding to furniture industry standards have also been described.[5–7] According to these studies, the use of straw fibers allows the production of panels over a larger range of densities, from 0.2 to 0.8 g/cm^3, than with wood panels in which case the production of panels having a density below 0.4 g/cm^3 is impossible. An important straw panel production unit using 4,4′-diphenylmethane diisocyanate (MDI)-based resin has recently been built,[8] although the use of such resins from petrochemical origin presents some health hazards. Moreover, the manufacture of such panels is often difficult due to problems in releasing the panels from the press after production.

In the study of renewable polymers, some work has been devoted to the development of thermosetting resins based on epoxidised vegetable oil.[9–10] This type of resin, which is of great interest from an environmental perspective, was chosen to produce straw panels. Our objective was to produce panels with acceptable properties for interior use, for example, in the furniture or flooring industries. Properties of such panels determined according to European standards are described in the French requirements CTB-S edited by the Centre Technique du Bois et de l'ameublement (CTBA). A urea–formaldehyde (UF) resin, a thermosetting polymer widely used for the manufacture of such CTB-S particleboard panels, was also used for comparison.

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Materials and methods

Materials

Raw materials were wheat (*Triticum aestivum L.*) straws. Straw was prepared using a hammer mill fitted with a 2- or 20-mm grid. All particles used were oven-dried at 80°C for 15 h to about 3% moisture content.

The thermosetting resin known as PTP (polymeric material from triglycerides and polycarbonic acid anhydrides) used in this study was elaborated at our facilities and is based on epoxidised linseed oil and anhydride as hardener. One percent of 2-methylimidazole based on the weight of the resin was added as a catalyst.

The UF resin (Pressamine 210) used in this study was supplied by Elf Atochem. This resin is water dispersed with a solid content of 65%. One percent of NH_4Cl based on the weight of the resin solid content was added as a catalyst.

Board manufacturing

The resins were sprayed onto the straw particles in a blender. The resin content varied from 5% to 17% based on the dry weight of straw particles. A hot press (Carver model M43196) was used to manufacture the boards. The platen temperature was fixed at 200°C, a temperature widely used for particleboard manufacture on an industrial scale. The manufactured boards were 200 mm side square and 6 mm thick. Such a thickness is usually used in flooring materials. For each manufacturing parameter, six boards were manufactured. The targeted board density was 0.7 g/cm^3. In the case of UF resin-based panels, a three-step pressing cycle, described in Table 1, was used to allow degassing and thereby avoid panel delamination. This cycle is similar to cycles used in industry to obtain stable and optimal properties when full resin cure is reached. In the case of oil-based resin, the cycle has to be modified taking into account its cure kinetics[10] and the absence of water when compared with 35% water content for UF resin.

Mechanical tests

Mechanical properties were measured using an Instron 4201 testing machine. Each measurement presented is the average for eight samples cut from two different boards. Three-point flexural tests have been carried out following the NF-EN 310 standard to allow calculation of the modulus of elasticity (MOE) and the modulus of rupture (MOR). Sample size was 130 mm (length), 20 mm (width), and 6 mm (thickness). Internal bond strength (IB) was determined following the NF-EN 319 standard. Square samples had a 50-mm side length and a 6-mm thickness.

Thickness swelling measurements

Thickness swelling (TS) was measured after 24-h immersion in distilled water at 20°C, following the NF-EN 317 standard. Each measurement presented is the average of eight samples cut from two different boards. Square samples have a 50-mm side length and a 6-mm thickness.

Differential scanning calorimetry (DSC) measurements

Thermograms were obtained using a Setaram DSC92. Runs were carried out under a nitrogen atmosphere in aluminum pans. The sample (about 25 mg) was heated from ambient temperature to 280°C at a scanning rate of 10°C/min.

Contact angle measurements

Contact angles were measured using a goniometer Krüss G40 under standard conditions (50% relative humidity and 23°C). A 5-μl drop of the liquid to be tested was applied to the outer surface of the straw using a syringe. Contact angle change was observed over 100 s, and a measurement was made every 2 s. Eight measurements were made for each sample tested. Diiodomethane and distilled water were used for surface energy calculation. Table 2 gives the characteristics of these two liquids.

Results and discussion

Development of a pressing cycle for PTP-based panels

Figure 1 shows the change of temperature inside the fibers mat and the applied pressure. Step one and step two durations are linked to vapor release. During the first step, the pressure reached 4 MPa after 50 s and was maintained for 20 s. In the second step, the pressure was reduced to 0.75 MPa over 20 s to allow a release of vapor. For the platen temperature tested, we observed that the pressure relaxation occurred when fibers temperature (between 140° and 180°C) was high enough to allow vapor formation. The pressure was then increased to 4 MPa to the end of

Table 1. Different steps (time and pressure) of the pressing cycles used in this study for urea–formaldehyde (UF) resin-based panels

Resin type	Step 1		Step 2		Step 3	
	Time (s)	Pressure (MPa)	Time (s)	Pressure (MPa)	Time (s)	Pressure (MPa)
UF	40	4.00	40	0.75	40	4.00

Table 2. Liquid characteristics

Liquid	Interfacial tension γ_l (mJ/m^2)	Disperse component γ_l^d (mJ/m^2)	Polar component γ_l^p (mJ/m^2)
Distilled water	72.4	21.8	50.6
Diiodomethane	50.8	48.5	2.3

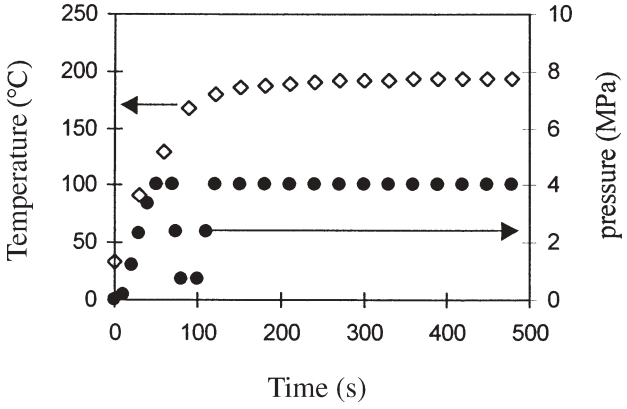

Fig. 1. Evolution of straw fiber mat temperature (*open diamonds*) and applied pressure (*open circles*) over time

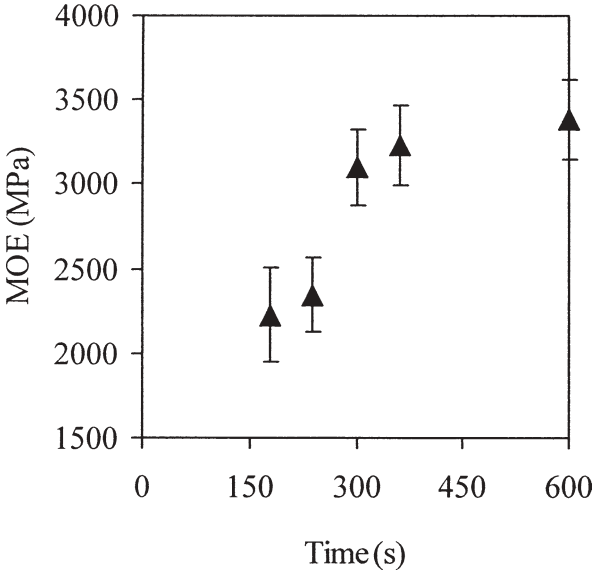

Fig. 2. Influence of pressing time on straw board modulus of elasticity (MOE)

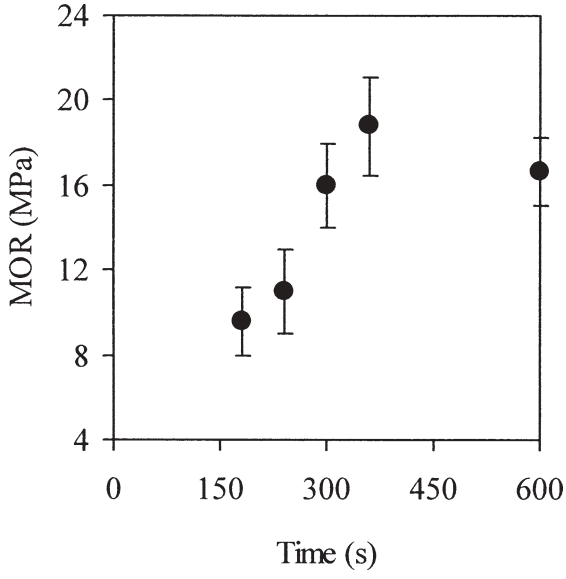

Fig. 3. Influence of pressing time on straw board modulus of rupture (MOR)

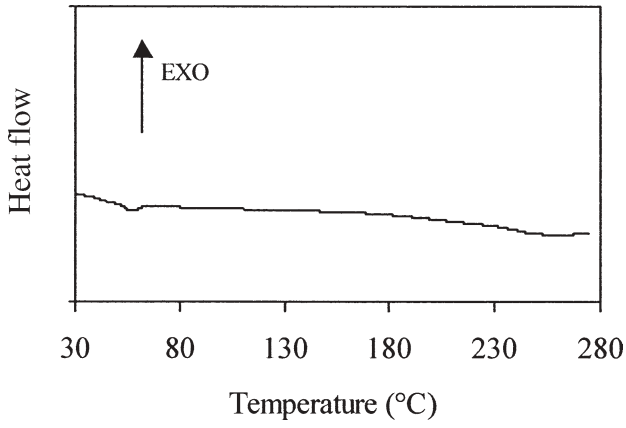

Fig. 4. Differential scanning calorimetry on pure PTP cured under the conditions allowing maximal PTP-based straw panel flexural properties. (PTP, polymeric material from triglycerides, and polycarbonic acid anhydrides)

the cycle. First and second step durations were shorter than with classical UF-based panels because of the lower water content of the mat when oil-based resin was employed.

Flexural properties as function of the pressing time were determined on panels having a density of 0.7 g/cm^3 and 17% PTP resin content. The results are presented in Figs. 2 and 3. MOE and MOR increased with increasing pressing time from 2200 and 10 MPa, respectively, at 180 s to 3200 and 18 MPa, respectively, at 360 s. For a pressing time as low as 180 s, the temperature inside the fiber mat just reached 180°C, close to the resin exothermic peak temperature determined elsewhere.[10] Consequently, full resin cure could not be reached under such pressing conditions. For pressing times from 360 to 600 s, flexural properties remained stable. An additional DSC experiment on a pure resin sample was carried out. For this experiment, the resin was cured under pressing conditions similar to that used in panel production over 360 s. The results (see Fig. 4) do not show any exothermic peak that could be linked to additional curing. Consequently, a 360-s pressing cycle (corresponding to 240 s for the third step) allows the production of panels with maximal properties linked to a full resin cure.

General properties of the straw panels

Influence of resin content

The influence of resin content was determined on panels prepared with PTP resin that had a density of 0.7 g/cm^3. These results are shown in Figs. 5–8. When the resin content increases from 5% to 17%, MOE increases from 2000 to 3200 MPa, MOR from 7 to 18 MPa, IB from 0.04 to

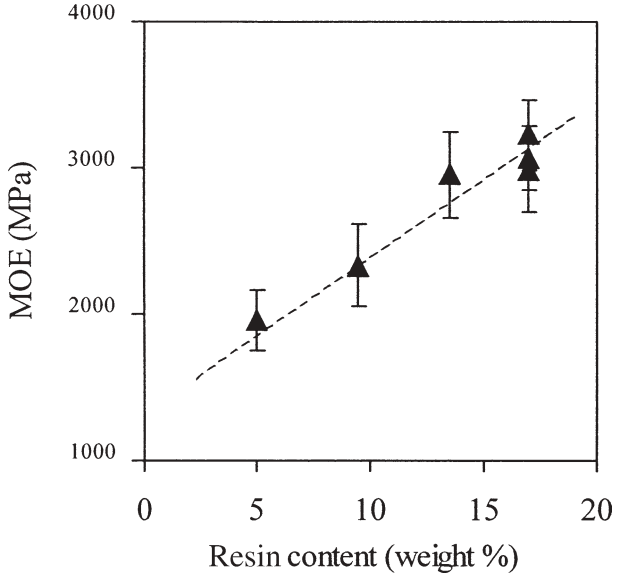

Fig. 5. Influence of resin content on straw board MOE

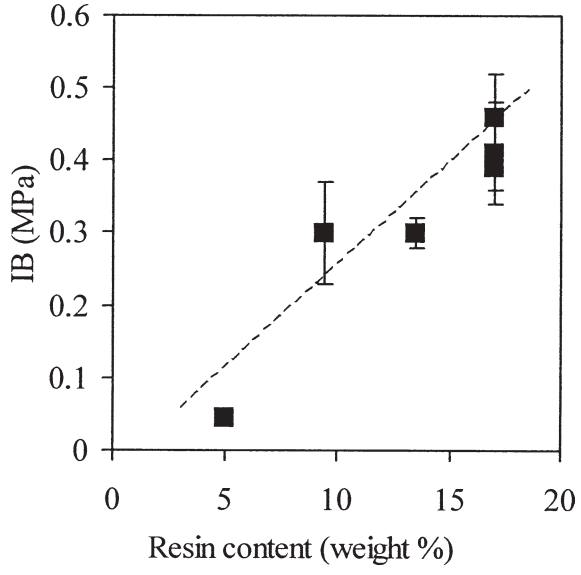

Fig. 7. Influence of resin content on straw board internal bond strength (IB)

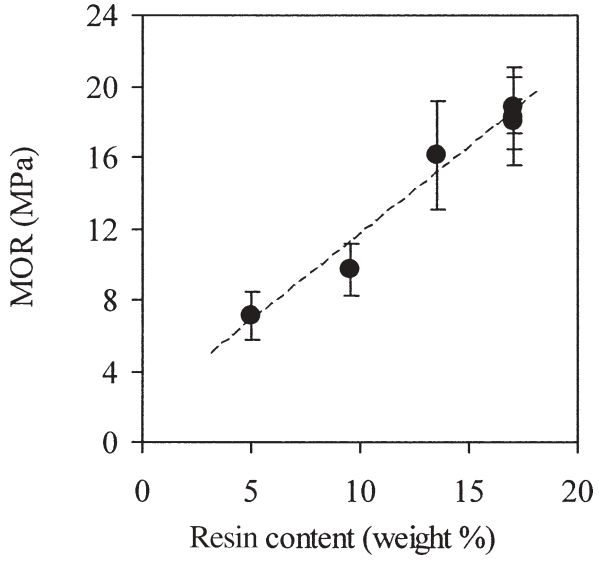

Fig. 6. Influence of resin content on straw board MOR

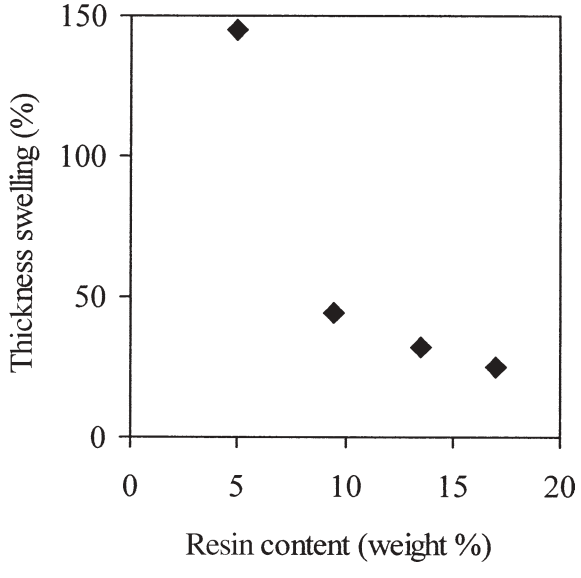

Fig. 8. Influence of resin content on straw board thickness swelling (TS)

0.42 MPa, and thickness swelling decreases from 145% to 25%. These changing properties are caused by an increase of surface contact between the resin and straw fibers, leading to improved bonding quality.

Influence of particle size

The influence of wheat straw particle size was determined on panels that had a density of 0.7 g/cm^3 and a 17% PTP resin content. The results, presented in Table 3, show that MOE decreases from 3200 to 2600 MPa, TS from 25% to 15%, and IB increases from 0.42 to 0.81 MPa when the hammer grid size changes from 20 to 2 mm. MOR remains unchanged. The MOE decrease is probably due to the decrease of the fiber form factor (length/diameter ratio) when the hammer grid size changes from 20 to 2 mm. Other studies of natural fiber-reinforced composites have shown that the MOE is widely influenced by the fiber form factor.[11] A decrease of the particle size leads to an increase of the contact area between the resin and the straw particles. As a consequence, IB and TS, which evaluate the panel adhesion, are improved. This was also observed for wood flakeboards.[12] In the case of straw particles, an earlier study[13] stated that the crushing of straw leads to a cracked epidermis and allows greater resin penetration inside the stem. This phenomenon could explain the influence of particle size on IB. Table 3 describes the CTB-S requirements for particleboard panels having a thickness between 4 and

Table 3. Properties of straw boards bonded with PTP resin

Hammer grid size (mm)	MOE (MPa)	MOR (MPa)	TS (%)	IB (MPa)
2	2600 (200)	17.6 (2.6)	15	0.81 (0.09)
20	3100 (200)	18.0 (2.5)	25	0.42 (0.05)
CTB-S standard	≥2200	≥17	≤19	≥0.45

Values in parentheses are standard deviations
PTP, Polymeric material from triglycerides and polycarbonic acid anhydrides; MOE, modulus of elasticity; MOR, modulus of rupture; TS, thickness swelling; IB, internal bond strength; CTB-S, french standard for inside use particleboard panels

Table 4. Properties of straw panels bonded with UF or PTP resin

Resin type	MOE (MPa)	MOR (MPa)	TS (%)	IB (MPa)
UF	660 (80)	3.0 (0.4)	240	0.02 (0.002)
PTP	2300 (250)	9.7 (0.8)	44	0.30 (0.07)

6 mm. We note that the use of smaller straw particles allows these requirements to be met. In the case of bigger particles, IB and TS values are close to these requirements.

Influence of resin type

Panels were manufactured from straw particles prepared using a 20-mm hammer grid size. The manufacture of particleboards based on UF resin and straw crushed using a 2-mm hammer grid size led to delamination of the panels, probably due to hindered vapor release. The resin content was 9% and the board density was 0.7 g/cm^3. Higher resin contents also led to panel delamination because of excessive water content. The results are presented in Table 4. Straw panels bonded using UF resin exhibit poorer mechanical properties than when using PTP resin. MOE values were 660 and 2300 MPa, respectively, and MOR values were 3 and 10 MPa, respectively. IB was close to zero for boards bonded with UF resin and was 0.30 MPa when PTP resin was employed. The situation is similar for TS with values of 145% and 44% for UF and PTP resins, respectively. These results clearly indicate poor compatibility between straw particles and UF resins.

The same propensity was observed in studies[14] that substituted wood with wheat straw in UF-bonded panels. An important IB reduction was observed for increased amounts of straw. MOE, more linked to the particle shape, was less affected.

Straw has been identified as a material for which bonding presents important difficulties. In fact, as described earlier, straw stems have an outer layer with very low porosity[13] which disrupts resin penetration. Moreover, the rate of resin penetration into straw was recently observed to be several orders of magnitude slower than that into wood.[15] This phenomenon may explain the low adhesion between straw and UF resins. Another important straw property is its outer layer of wax,[16] that might help explain the changing properties between UF- and PTP-based panels. This aspect has not yet been studied in detail. A compatibility study using contact angle measurements is necessary to explain the observed differences.

Table 5. Straw surface free energy determination according to the Wu method

Contact angle		Surface free energy	Disperse component	Polar component
Distilled water (°)	Diiodomethane (°)	γ_s (mJ/m^2)	γ_s^d (mJ/m^2)	γ_s^p (mJ/m^2)
90 (4)	62 (3)	34.5 (1)	26.0 (0.9)	8.5 (1)

Compatibility study

Straw surface energy determination

The determination of the polar (γ^p) and disperse components (γ^d) of the surface free energy γ_s of the outer surface of straw stalks used the method described by Wu.[17] This method is based on the combination of Eq. 1, described by Young, and Eq. 2.

$$\gamma_s = \gamma_{sl} + \gamma_l \cdot \cos\theta \quad (1)$$

where γ_s is the straw surface free energy, γ_{sl} is the interfacial tension between the straw and the liquid (distilled water or diiodomethane) tested, and γ_l is the liquid surface free energy.

$$\gamma_{sl} = \gamma_s + \gamma_l - \frac{4\cdot\gamma_s^d\gamma_l^d}{\gamma_s^d + \gamma_l^d} - \frac{4\cdot\gamma_s^p\gamma_l^p}{\gamma_s^p + \gamma_l^p} \quad (2)$$

where γ_s^d is the disperse component of the straw surface free energy, γ_s^p is the polar component of the straw surface free energy, γ_l^d is the disperse component of the liquid interfacial tension, and γ_l^p is the polar component of the liquid interfacial tension.

The results presented in Table 5 show that the straw surface free energy is 34.5 mJ/m^2. The polar and disperse components are 8.5 mJ/m^2 and 26.0 mJ/m^2, respectively. The straw surface free energy is much lower than that of Douglas pine wood or redwood[18] calculated using the same method. For these wood species, surface free energies are approximately 50 mJ/m^2. The major difference is in the polar component, which is much lower for straw than in the former wood species where the polar component is in the range of 20–30 mJ/m^2. The low polar component of the straw surface free energy is probably due to the waxy layer and explains the low compatibility between straw and UF resin which, like other major components such as water, formaldehyde, and urea, has a high polarity. On the other hand, the major component of PTP is oil which is nonpolar. The difference in the polarity of the resins explains why PTP is more suited to bonding with a low polarity surface like wheat straw.

Table 6. Contact angle between straw outer surface and UF or PTP resins

Resin type	Initial contact angle (°)	Contact angle after 100 s (°)
UF	84	82
PTP	57	56

Contact angle measurements between the straw outer surface and the resins employed, presented in Table 6, confirm this result. The initial contact angle of UF resin was 84° compared with 57° for the PTP resin. This result shows that the wettability of the straw is much better when using PTP resin. Assuming that without good wetting, good bonding cannot be expected, the poor straw wetting by UF resin explains the very poor panel properties attained. The contact angle between straw and PTP is similar to the contact angle between straw and MDI resin measured elsewhere.[15] For both of these resins, the contact angle reduction is less than 2° after 100 s, indicating a very low resin diffusion inside the straw. This phenomenon, due to the low porosity of the straw outer layer, probably reduces the adhesion quality.

Conclusions

The use of PTP resin for the bonding of straw particles is efficient. The resultant panels have a high organic content and exhibit properties reaching industrial standards. This application may provide new industrial uses for wheat straw. Studies have also shown the straw surface can be modified to make it more suitable for use with traditional binders. These treatments are efficient and may widen the range of potential applications if combined with PTP resin.

The main drawback of PTP is the cure kinetic, which is rather low compared with UF resin. Additional work on resin formulation and chemistry is now under way. Our laboratory studies will soon be performed at pilot scale in order to optimize the processing parameters and to evaluate the process economics.

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