### **Evaluation of Melamine-Modified Urea-Formaldehyde Resins as Particleboard Binders**

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**ABSTRACT:** Particleboards bonded with 6 and 12% melamine-modified urea-formaldehyde (UMF) resins were manufactured using two different press temperatures and press times and the mechanical properties, water resistance, and formaldehyde emission (FE) values of boards were measured in comparison to a typical urea-formaldehyde (UF) resin as control. The formaldehyde/(urea + melamine) (F/(U + M)) mole ratio of UMF resins and F/U mole ratio of UF resins were 1.05, 1.15, and 1.25 that encompass the current industrial values near 1.15. UMF

resins exhibited better physical properties, higher water resistance, and lower FE values of boards than UF resin control for all F/(U + M) mole ratios tested. Therefore, addition of melamine at these levels can provide lower FE and maintain the physical properties of boards. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 106: 4148–4156, 2007

**Key words:** urea-melamine-formaldehyde resins; urea-formaldehyde resins; particleboard; formaldehyde emission

#### **INTRODUCTION**

Urea-formaldehyde (UF) resins (Fig. 1),<sup>1–8</sup> the current binder resins of fiberglass mat and interior-grade wood composite board, such as particleboard (PB), medium density fiberboard, and hardwood plywood,<sup>9,10</sup> show drawbacks of low water resistance and formaldehyde emission (FE).<sup>11–13</sup> Formaldehyde is released into the environment during the hot pressing of boards and also from finished boards.<sup>14–19</sup> Formaldehyde is released from the degradation of methylol groups of UF resins during hot-pressing and the entrapped free formaldehyde in boards diffuses out slowly. Furthermore, unreacted methylol groups and methylene-ether groups present in cured resins can slowly break down to emit formaldehyde over the lifetime of boards. Methylene groups in cured resins can also slowly break down and emit formaldehyde when the humidity and temperature is high. Possible approaches to reducing the FE from boards include application of surface coating or barrier membrane, treatments of boards with ammonia, use of low F/U mole ratio resins, and addition of melamine to the UF resins.<sup>20-26</sup> Among these various methods, the approach of lowering the F/U

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mole ratio of resin had been the mainstay and the value currently is near 1.15 and various adverse effects on physical properties of boards are known to appear below this value.

Various methods of adding low levels of melamine to UF resins have been investigated to offset the adverse effects of lowering the F/U mole ratio of UF resins in the form of urea-melamine-formaldehyde (UMF) resins. These UMF resins differentiate themselves from the common melamine-urea-formaldehyde (MUF) resins currently being used widely in lumber lamination and other applications, in that the melamine contents are generally higher, 50% or more, in comparison to urea in the resins and with different curing parameters.<sup>27</sup> Melamine's higher functionality and more stable molecular structure in comparison to urea and its reaction with formaldehyde that is similar in formalism to that of UF resins led to variously synthesized UMF resins based on several typical UF resin synthesis methods.<sup>21,22</sup> Thus, depending on the method of resin synthesis, resins made with 4%-35% melamine levels often show varying strength and FE values that do not necessarily improve with increasing melamine lev-els.<sup>21,22,24,26,28</sup> One report on bonding of plywood with UMF resins indicated an improved durability ascribed to the higher basicity of melamine in comparison with urea.<sup>29</sup> In recent years, some UMF resins are used to bond PB and medium-density fiberboard in Europe and, to a less extent, in North America with improved physical properties and lower FE reported. 23,26,28,30-37

Since melamine costs significantly more than urea, we studied various UMF resin synthesis parameters

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Figure 1 Schematic representation of urea-formaldehyde (UF) and melamine-formaldehyde (MF) resin systems.

for maximizing the contribution of melamine toward the bonding performance<sup>36,37</sup> with incorporation of some new details reported on UF resin chemistry.<sup>38-44</sup> In our studies, the UF base resin was reacted to various different extents of polymerization at pH 4-5 and, after adjusting the pH to 6-9, melamine was added at various low levels, reacted to the target viscosity of resin, and the second urea added to the final mole ratio values of 1.05, 1.15, and 1.25. Adding melamine early with the first urea and formaldehyde and reacting at pH 6-9, a procedure commonly used by early researchers, resulted in a highly turbid resin because of the high extent of polymerization of MF resin components and only a little extent of polymerization for UF resin components. On the other hand, adding melamine after the UF base resin has been advanced to a high extent resulted in a very low

extent of polymerization for MF resin components with a low turbidity. It was found that the solubility of MF resin components during resin synthesis decreases rapidly forming solid particles at the dimer stage and, therefore, depending on the point and pH of melamine addition the resultant resin could contain various amounts of insoluble MF resin components as suspended particles to make the resin turbid. The MF component particles would vary in the thermosetting quality depending on their melting temperatures versus the curing temperature of board.

Furthermore, the MF resin system reacts optimally at pH 6–9 and it becomes too fast to control at pH 4–5 and, on the other hand, the UF resin system polymerizes optimally at pH 4–5 and too slowly at pH 6–9, so that UF resin components cannot be effectively advanced once the melamine addition is done.



**Figure 2** Internal bond (IB) strength values of particleboards bonded with Resins UFA and UMF at an F/(U + M) mole ratio of 1.15.

The UF resin components need to be polymerized to a high extent in comparison to MF components for overall better performance of UMF resins as wood composite binders.45 The synthesized various UMF resins showed different handling and dynamic mechanical analysis (DMA) curing characteristics from the different molecular weight distributions attained as well as different melamine levels and it was generally found that the optimum melamine addition point is when the extent of polymerization of UF base resin reaches to  $D \sim K$  viscosity by Gardener-Holdt scale.36,37 UF resins are commonly cured using a latent catalyst, such as ammonium sulfate, which reacts with free formaldehyde present in resin to produce free acids. The UF resin-bonded wood composite boards are cured in industry at 150-170°C for about 3 min for 1.25-cm thick boards, but the hot pressing can often cause pre-cure/over-cure problems in surface layers and under-cure problems in core layers to cause deterioration of board properties. Since MF components cure slower than UF components, various higher hot pressing temperatures and longer pressing times were investigated as well as various melamine levels and final F/(U + M) mole ratios by manufacturing PBs and testing. The bond strength and FE values of the various UMF resin-bonded PBs are reported in this article.

#### **EXPERIMENTAL**

#### Syntheses of control UF resin and UMF resins

The synthesis of the UMF resins is briefly reiterated here (Fig. 1).<sup>36,37</sup> In the typical UF resin synthesis, urea is added in two parts, the first urea (U<sub>1</sub>) and

the second urea  $(U_2)$ . In the first step, the first urea and formaldehyde are reacted in a weak alkaline pH at an  $F/U_1$  mole ratio of about 2.1 to form various hydroxymethylureas and, in the second step, the polymerization reaction is carried out at pH 4-5 and 95°C to form mostly methylene groups. Some hydroxymethyl groups split off as formaldehyde because of reverse hydroxymethylation reactions and decreasing urea amide groups are available for reaction within the resin system. The second step is normally ended at viscosity "V  $\sim$  X" at a resin solids level of 60–65%, by increasing the pH to about 8.0, resulting in a mixture of polymeric methylene/methylene-ether hydroxymethylureas. In the third step, the reaction mixture is cooled to about 60-70°C and the second urea added to final  $F/(U_1 + U_2)$  of about 1.15 and the resin synthesis is completed by cooling to room temperature. Some of the second urea reacts with the free formaldehyde present in the reaction mixture to form monomeric hydroxymethylureas, normally leaving less than 0.5% free formaldehyde content in the resin. About 25% of the urea used in the resin synthesis remains as free urea and the rest as monomeric and polymeric UF polymers.

Resin UFA is the typical UF resin synthesized according to the earlier procedure using 50% aqueous formaldehyde solution  $(F_1)$  and first urea  $(U_1)$  at an  $F_1/U_1$  mole ratio of 2.1, reacted first at pH 7.0–8.0 and then at pH 4.6. The second urea  $(U_2)$  was added at pH 8.0 and at 60°C, attaining  $F_1/(U_1 + U_2)$  ratio of 1.15, and the resultant resin cooled to room temperature. Resins UMF6D, UMF6K, UMF12D, and UMF12K were synthesized, with numbers 6 and 12 indicating the melamine level based on liquid resin weight and letters D and K indicating the viscosity of UF base resin at the melamine addition point. At the melamine addition point, the second formaldehyde solution  $(F_2)$  was also added to maintain the  $(F_1 + F_2)/(U1 + M)$  ratio at 2.1 and the second urea  $(U_2)$  added after the reaction is complete and the mixture cooled to room temperature to obtain resins with an  $(F_1 + F_2)/(U_1 + U_2 + M)$  mole ratio of 1.15. Resins UFA<sub>1.05</sub> and UFA<sub>1.25</sub>, UMF12D<sub>1.05</sub> and UMF12D<sub>1.25</sub>, UMF12K<sub>1.05</sub> and UMF12K<sub>1.25</sub> were synthesized using the same procedures except the final mole ratio was adjusted to 1.05 and 1.25, respectively, by increasing or decreasing the amount of the second urea  $(U_2)$ .

#### Preparation of particleboard

Single-layer boards were made using dried southernyellow pine particles obtained from the Georgia-Pacific Corp. PB plant in Louisville, MS. A rotary drum blender, forming box, and automated Dieffenbacher hot press were used according to the standard laboratory procedure. Resins were catalyzed with 0.5% ammonium sulfate as a 25% water solution based on the liquid resin weight and resin loading levels were 8% resin solids based on oven-dry wood weight. No wax was added. The press closing rate was initially 1.27 cm/s to a mat thickness of 2.53 cm and then 0.08 cm/s to reach to the target thickness. One panel  $(860 \times 860 \times 12.65 \text{ mm})$  was made for each resin with a target density of 801  $kg/m^3$  (50 lb/ft<sup>3</sup>). Press temperatures were 325 and 365°F and press times were 4.0 and 5.0 min for boards bonded with Resins UFA and UMF at an F/ (U + M) mole ratio of 1.15. Press temperature of 340°F and press time of 4 min were used for boards made with Resins UFA and UMF at F/(U + M)ratios of 1.05, 1.15, and 1.25. The conditioned boards were trimmed to 66 cm  $\times$  66 cm in size, machined, and cut for internal bond (IB), modulus of rupture (MOR) and modulus of elasticity (MOE), thickness swelling (TS), water absorption (WA), and formaldehyde emission (FE) tests.

## Internal bond, bending strength, and dimensional stability tests of boards

All tests of boards were carried out according to the ASTM test method.<sup>45</sup> Cut test specimens were equilibrated for 3 weeks in a constant humidity chamber to about 7% moisture content. Wet IB samples were obtained by immersing samples in water at room temperature for 24, 48, 72 h and then oven-drying at 158°F for 24 h to a moisture content of about 8%. An Instron testing machine was used for both IB and static bending tests. TS and WA samples were immersed in water at room temperature for 2 and 24 h and then measured for TS and WA.

#### Formaldehyde emission tests

Among the methods of FE from boards<sup>46-55</sup> the small-chamber (dynamic micro-chamber) method<sup>51-53</sup> and the Perforator extraction (PE) method<sup>56-58</sup> were used. The good correlation between the smallscale chamber (SSC) and large-scale chamber methods established in industry has made the former method a lower cost, fast testing procedure providing information on FE levels encountered in the early periods of board use.59 The PE method commonly used in Europe determines the total formaldehyde content in board samples by extraction.<sup>55</sup> FE from boards continues for a year or longer although the emission level decreases with time<sup>60°</sup> and the perforator method is suited for measuring the long term FE potential of boards. Also, Sundin et al. showed that the FE of PB was largely due to the free formaldehyde trapped in boards and not much from the hydrolytic degradation of cured UF resins.<sup>61</sup>

Tests using the SSC method were carried out at Georgia-Pacific Resins laboratory (Decatur, GA).

Hot-pressed boards were allowed to stand for 24 h in the laboratory and three test specimens (379.5 mm  $\times$  199.2 mm) were cut from each board and the edges were sealed with aluminum adhesive tape giving a total exposed board's face area of 0.4536 m<sup>2</sup> per board. The test samples were again aired for 7 days at 23.9°C at a relative humidity of 50% and then loaded in the test chamber, which was maintained with a makeup air flow of 8.93 L/min, providing a 1/2; air change per hour. The loading ratio was 0.13 ft<sup>2</sup> of the panel face area per cubic foot of chamber volume. The formaldehyde level in the exiting air was monitored over time and the steadystate values,  $C_{s}$ , attained in time were reported as the FE values of the test samples. The formaldehyde content (C, ppm) is calculated by the following formula [eq. (1)]:

$$C_s = (K \times A \times C_{eq})/(Q + K \times A)$$
(1)

where  $C_s$  is the steady-state formaldehyde concentration in ppm;  $C_{eq}$  is the equilibrium formaldehyde concentration in ppm; *K* is the mass transfer coefficient in m/h; *A* is the product surface area in m<sup>2</sup>; *Q* is the makeup air flow in m<sup>3</sup>/h.

The PE procedure was carried out with test samples (2.5 cm  $\times$  2.5 cm  $\times$  1.27 cm) cut from one-day aired boards, which were kept in a sealed plastic bag until the tests were done within 1 week using the setup described in DIN EN 120 (1992) European Standard Method. About 110 g of test samples were weighed to an accuracy of 0.1 g, put into the round bottom flask, and 600 mL of toluene were added. The flask was connected to the perforator and about 1000 mL of distilled water was poured into the absorption column, which left a space of 20-30 mm between the surface of water and the siphon outlet. Also, about 100 mL distilled water was placed in the exit gas absorption bulb connected to the apparatus. A controlled heating is started and maintained to cause toluene boil carrying the extracted formaldehyde to condense on the condenser and drop on the funnel and fall down to the perforator (glass filter) and come up in fine drops to the surface of the water layer. The formaldehyde is absorbed by the water layer during the rise of toluene bubbles and the collected toluene at the top of the water layer flows back to the flask through the siphon. The extraction was carried out for 2 h with a reflux rate of 70-90 drops of toluene per min maintained. Then the total extraction water in the column and exit trap was taken and diluted to 2000 mL with distilled water. Blank tests were done the same way without board samples. The formaldehyde content of the aqueous solution was determined photometrically using the acetylacetone method at the wavelength of 412 nm against distilled water. The blank value determined using distilled water was then subtracted. The perforator formaldehyde value (P) is expressed in mg formaldehyde per 100 g of oven-dry board (mg/100 g) calculated by the following formula [eq. (2)]:

$$P = \left[ (A_s - A_g) \times f \times (100 + H) \times V \right] / M_H$$
 (2)

where,  $A_s$  is the absorbance of the extraction solution;  $A_g$  is the absorbance of a blank control; F is the slope of the standard curve in mg/mL; H is the moisture content boards in %;  $M_H$  is the weight of board sample in g; V is the volume of total extraction water (2000 mL).

#### **RESULTS AND DISCUSSION**

## Particleboards bonded with Resins UFA and UMF at an F/(U + M) ratio of 1.15

#### Dry internal bond strengths of boards

Particleboard (PB) bonded with control Resin UFA and UMF resins at an F/(U + M) ratio of 1.15 and press times of 4 and 5 min at press temperatures of 325 and 365°F showed IB strength (IB) values shown in Figure 2. UMF resins exhibited higher IB values than Resin UFA throughout most conditions except that UMF resins made with 6% melamine level at the lower press temperature with the shorter press time gave strength values similar to that of Resin UFA (Fig. 1). Resin UFA showed very low IB values at the higher press temperatures probably because of the greater tendency of over-curing/degradation at high temperatures and long press times while UMF resins exhibited good bond strengths due to slow curing characteristics and better thermal stability as revealed from the DMA curing study.37 There was no significant differences in IB strength values of among UMF resins between "K" and "D" series indicating the optimum range of advancement of UF base resin components. UMF resins with 12% melamine level had higher IB values than those resins with 6% melamine level for all hot pressing conditions. It is believed that the higher melamine content enhanced the bond strength of boards because of the higher functionality and rigid ring structure of melamine. Higher press temperatures gave higher bond strength of boards bonded with UMF resins probably because of the higher temperature that compensated for the slower curing rates of UMF resins and also from the retarded degradation. Through all conditions, boards bonded with UMF resins exceeded the minimum strength requirements for IB required in the US ANSI H-3.<sup>62</sup> Statistical analyses using SAS program<sup>63</sup> showed that IB strength values were highly correlated with melamine contents. The Pearson correlation coefficients between IB strength values and melamine contents were 0.83 for #D and 0.79 for #K resin

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series. A regression analysis resulted in the following equations:

$$IB = 213.44 - 8.82 \times X_2 + 4.61 \times X_3(D resins)$$

$$IB = 382.61 + 0.33 \times X_1 - 11.68 \times X_2 + 4.64 \times X_3 - 5.48 \times X_4 (K resins)$$

where  $X_1$  = press temperature (°F);  $X_2$  = press time (min);  $X_3$  = melamine content (%) in resin;  $X_4$  = density (pcf, 1 pcf = 0.016 g/cm<sup>3</sup>) of board; IB: in psi (1 psi = 0.006895 MPa).

The board density ( $X_4$ , P value = 0.86) and the press temperature ( $X_1$ , P value = 0.72) were the least significant factors for IB in #D resins, so these factors were removed from the equation. The melamine content ( $X_3$ ) was the most significant factor affecting IB values of boards for both UMF resins.

#### Wet internal bond strengths of boards

IB strength values of boards bonded with Resins UFA and UMF using 4 min press time at 365°F after soaking in water for 0, 24, 48, and 72 h followed by oven-drying at 140°F for 24 h are shown in Figure 3. IB values decreased drastically in the first 24 h of soaking in water but not appreciably for the remaining immersion periods. UMF resins exhibited higher wet IB strengths than the control UF resin throughout all immersion periods. Among UMF resins, 12% melamine level resins gave higher wet IB strength values than 6% melamine level resins. UMF resins with "K" series exhibited better wet IB than those



**Figure 3** Internal bond (IB) strength values of particleboards bonded with resins UFA and UMF using a 4 min press time at  $365^{\circ}$ F after soaking in water at room temperature for 0, 24, 48, and 72 h followed by oven-drying at  $140^{\circ}$ F for 4 h.



**Figure 4** Modulus of rupture (MOR) and modulus of elasticity (MOE) values of particleboards bonded with resins UFA and UMF.

with "D" viscosity series on both 6 and 12% melamine levels. It seems that higher advancements of the UF resin component in UMF and higher levels of melamine resulted in better UMF resins for water resistance. Other press time and temperature data also show similar trends (data are not reported).

#### Bending strengths of boards

Most UMF resins exhibited higher MOR and MOE values than the control resin UFA (Fig. 4). Longer press times lowered both MOR and MOE values for Resins UFA and UMF at both high and low temperatures. The longer press time appeared to have overcured resins in the surface layers, resulting in degradation of strength values, but UMF resins were less affected than Resin UFA, which showed drastically lowered MOR and MOE values at the high press temperature. There were not much difference of bending strength values between "D" and "K" series on both 6 and 12% melamine levels.

#### Water absorption values of boards

Figure 5 shows that Resin UFA had lower water absorption (WA) values than the Resins UMF6 at a

press temperature of 325°F but higher WA values at a press temperature of 365°F. Resins UMF12 had lower WA values than Resin UFA under all conditions. Resins UMF6 and UMF12 showed lower TS values than Resin UFA under all condition. The effect of melamine levels on TS and WA in 24 h tests was not differentiated and it may require a more severe test, such as 2-h boil, three cycle test, etc. Resin UFA again exhibited higher WA and TS values when the higher press temperature and longer time probably because of the fact that the UMF resins cure more completely at the higher temperature and provide better water resistance while the control UF resin over-cure and lose its bonding strength.

# Effects of F/(U + M) mole ratios of resins on particleboards strengths and formaldehyde emission

#### Dry and wet internal bond strengths

In general, IB values of boards increased as the F/(U + M) ratio increased from 1.05 to 1.25. It has been known that higher F/U ratios give higher IB values because of the higher extent of cross-linking of resins in curing,<sup>26</sup> but the effects of melamine additions are



**Figure 5** Water absorption (WA) and thickness swelling (TS) values of particleboards bonded with resins UFA and UMF after soaking in water for 24 h.



**Figure 6** Wet and dry internal bond (IB) strength values of particleboards bonded with resin UFA, resins UMF6D, and resin UMF12 at three different formaldehyde/(urea + melamine) ratios. Wet IB tests were conducted after soaking in water for 24 h followed by oven-drying at  $140^{\circ}$ F for 4 h.

apparent. Both dry and wet IB strength values of boards were higher for UMF resins than Resin UFA for all F/(U + M) ratios used (Fig. 6). Resin UFA lost more IB strength than Resins UMF in 24 h water soak tests. Resins UMF6D and UMF12 gave better IB values than Resin UFA and, especially, Resins UMF at the F/(U + M) ratio of 1.05 exhibited higher IB values than Resin UFA at the F/(U + M) ratio of 1.25. These results indicated that UMF resins could be a good alternative to UF resins for lower FE at lower F/(U + M) mole ratios without lowering the mechanical properties of boards (Figs. 6 and 9). It is interesting to see that 6% melamine level provides better IB strength values than 12% melamine level under all conditions. The reason is unclear, but it is probably because Resin MUF12D did not cure completely at a press temperature of 340°F and a press time of 4 min, which is lower than the temperature in the first part of this study.

#### Bending strengths of boards

Resins UMF6D and UMF12D exhibited higher MOR and MOE values than Resin UFA regardless of the F/(U + M) mole ratios (Fig. 7). Resin UMF6D gave slightly higher MOR values than Resin UMF12D and there was not much difference in MOE values between Resin UMF6D and UMF12D.

#### Water-soak test results

Thickness swell values of boards decreased for Resin UFA with increasing F/(U + M) mole ratios whereas

such increases were not apparent for UMF resins (Fig. 8). On the other hand, UMF resins exhibited large decreases in WA values in increasing the F/ (U + M) mole ratio from 1.05 to 1.25. Both WA and TS values of boards bonded with UMF resins were lower than those of boards made with Resin UFA. In general, WA and TS graphs showed a trend opposite to IB strength graphs shown more obviously by the wet-IB data (Fig. 6). PB with higher IB values are known to show lower WA and TS values and the melamine addition appears to provide better water resistance as a result of better IB strength values.

#### Formaldehyde emission test results

The SSC and PE FE test results for boards bonded with Resin UFA and UMF resins made at F/ (U + M) mole ratios of 1.05, 1.15, and 1.25 are shown in Figure 9. UMF resins exhibited lower SSC values than Resin UFA under most conditions, especially at the F/(U + M) mole ratio of 1.25. Resin UMF12D showed lower SSC values than the Resin UMF6D under all conditions. PE test values showed a very similar trend with the SSC test values with a greater difference between Resin UFA and UMF res-



**Figure 7** Modulus of rupture (MOR) and modulus of elasticity (MOE) values of particleboards bonded with resin UFA, resin UMF6D, and resin UMF12D at three different formaldehyde/(urea + melamine) mole ratios.

ins. The PE test values showed a linear relationship with the SSC values ( $R^2 = 0.82$ ). The PE tests measure the total free formaldehyde in boards while the SSC tests measure only the level of formaldehyde emitted at the time of measurement.

SSC FE values of the PB were correlated to the F/ (U + M) mole ratios between 1.25 and 1.05, from which the Pearson correlation coefficients obtained for Resins UFA, UMF6D, and UMF12D were 0.97, 0.78, and 0.62, respectively. Thus, FE and F/U mole ratio values were highly correlated, especially for boards bonded with Resin UFA as shown below:

 $Y = 63.6X - 61.4 \ (R^2 = 0.9736)$  for Resin UFA.  $Y = 36.8X - 32.8 \ (R^2 = 0.9999)$  for Resin UMF6D.  $Y = 24.0X + 20.2 \ (R^2 = 0.9997)$  for Resin UMF12D.

where *Y*, FE (mg/100 g) using the PE test and *X*, formaldehyde/(urea + melamine) mole ratio.

The current F/U mole ratios of PB binder UF resins are at  $\sim 1.15$  (Ref. 23) and it has been known that lowering the mole ratio of resins further to lower the FE levels of boards would result in unacceptable board strength properties. Thus, the results of UMF



**Figure 8** Water absorption (WA) and thickness swelling (TS) values of particleboards bonded with resin UFA, resin UMF6D, and resin UMF12D at three different formalde-hyde/(urea + melamine) mole ratios.



**Figure 9** Formaldehyde emission values of particleboards bonded with resin UFA, resins UMF6D, and resins UMF12D at formaldehyde/(urea + melamine) mole ratio of 1.05, 1.15, and 1.25 using the small-scale chamber method and perforator extraction method.

resins provide some room for FE reduction while maintaining strength properties of boards.

#### CONCLUSION

UMF resins synthesized with 6 and 12% melamine additions were compared with a typical UF resin for binder and FE performances of PB at F/(U + M)mole ratios of 1.25, 1.15, and 1.05. Binder performances of resins tested by dry and wet IB and bending strengths and WA and entailing swelling extents of boards indicated significant improvements from the melamine addition at all F/(U + M) mole ratios examined. The binder performance improvements appear to be from increased stability of UMF resins over the course of the hot pressing period compared to UF resins. FE levels and emission potentials of resins tested by the SSC and PE methods for boards generally decreased with decreasing F/(U + M)mole ratios and UMF resins showed significantly lower values than the control UF resin at all mole ratios. These improvements in strength and watersoak test values and lower FE levels/potentials of boards led to conclude that addition of low levels of melamine to UF resins can be done to lower the FE of boards without suffering of physical performance properties of boards near the current F/U mole ratio of 1.15. However, it should be noted that even at 12% melamine addition levels, the SSC FE values of boards was about 0.10 ppm, a value that can seen as relatively high from the perspective of consumers.

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#### References

- 1. Crowe, G. A.; Lynch, C. L. J Am Chem Soc 1948, 70, 3795.
- 2. Crowe, G. A.; Lynch, C. L. J Am Chem Soc 1949, 71, 3731.
- 3. Smythe, L. E. J Am Chem Soc 1951, 73, 2735.
- 4. De Jong, J. I.; De Jonge, J. J Rec Trav Chem 1952, 71, 643.
- 5. De Jong, J. I.; De Jonge, J. J Rec Trav Chem 1952, 71, 661.
- 6. De Jong, J. I.; De Jonge, J. J Rec Trav Chem 1952, 71, 891.
- 7. De Jong, J. I.; De Jonge, J. J Rec Trav Chem 1953, 72, 139.
- De Jong, J. I.; De Jonge, J. J Rec Trav Chem 1953, 72, 1027.
  Wallace, P. D.; Kirksey, R. D. In TAPPI Nonwoven Symposium, Atlanta, GA, 1983.
- Maloney, T. M. Modern Particleboard and Dry-Process Fiberboard Manufacturing; Miller Freeman: San Francisco, 1997.
- 11. Myers, G. E. Wood Sci 1982, 15, 127.
- 12. Myers, G. E. Forest Prod J 1984, 34, 35.
- 13. Myers, G. E. Holzforschung 1990, 44, 117.
- Kelly, M. W. In Proceedings of the Fourth International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1970, p 137.
- 15. Christensen, R. L. Forest Prod J 1972, 22, 17.
- Johns, W. E.; Jahan-Latibari, A. In Proceedings of the 13th International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1979, p 355.
- Meyer, C. B. In Proceedings of the 13th International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1979, p 343.
- Markessini, A. C. In Proceedings of the 27th International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1993, p 207.
- 19. Risholm-Sundman, M. ; Wallin, N. Holz als Roh-und Werkstoff 1999, 57, 319.
- Roffael, E. In Proceedings of the 12th International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1978, p 233.
- 21. Shiau, D. W.; Smith, E.U.S. Pat. 4,536,245 (1985).
- 22. Druet, B.; Hopin, D.U.S. Pat. 4,997,905 (1988).
- Graves, G. In Proceedings of the 37th International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1993, p 221.
- 24. Breyer, R. A.; Hollis, S. G.; Jural, J. J.U.S. Pat. 5,681,917 (1997).
- 25. Andersen, B.; Brehmer, B.U.S. Pat. 5,681,917 (1999).
- Parker, R.; Crews, G. M. In Proceeding of the 33rd International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1999, p 57.
- 27. Wirpsza, Z.; Brezezinski, J. Aminoplasty; Khimia: Moscow, 1973; Chapters 2 and 3.
- 28. Oh, Y. S. Forest Prod J 1999, 49, 31.
- Dunwoodie, J. M. In Wood Adhesives and Technology, Vol. 1; Pizzi, A., Ed., Marcel Dekker: New York, 1983; Chapter 1.
- Hse, C. Y.; He, Z. In Proceedings Wood Adhesives; Conner, A. H.; Christiansen, A. W.; Myers, G. E.; River, B. H.; Vick, C. B.; Spelter, H. N., Eds.; Forest Products Society: Madison, WI, 1990; p 155.

- Pizzi, A. In Advanced Wood Adhesives Technology; Pizzi, A., Ed.; Marcel Dekker: New York, 1994, p 67.
- Rammon, R. M. In Proceeding of the 31st International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1997, p 177.
- 33. Tomita, B.; Hse, C. Y. Mokuzai Gakkaishi 1995, 41, 349.
- 34. Tomita, B.; Hse, C. Y. Mokuzai Gakkaishi 1995, 41, 490.
- 35. Tomita, B.; Ono, H. J Polym Sci 1979, 17, 3205.
- 36. No, B. Y.; Kim, M. G. J Appl Polym Sci 2004, 93, 2559.
- 37. No, B. Y.; Kim, M. G. J Appl Polym Sci 2005, 97, 377.
- 38. Kim, M. G. J Polym Sci Part A: Polym Chem 1999, 37, 995.
- 39. Kim, M. G. J Appl Polym Sci 2000, 75, 1243.
- 40. Kim, M. G.; Watt, C.; Davis, C. R. J Wood Chem Technol 1996, 16, 21.
- 41. Kim, M. G. J Appl Polym Sci 2001, 80, 2800.
- 42. Kim, M. G.; Wan, H.; No, B. Y.; Nieh, W. L.-S. J Appl Polym Sci 2001, 82, 1155.
- 43. Kim, M. G.; Amos, L. W. Ind Eng Chem Res 1990, 29, 208.
- 44. Kim, M. G.; No, B. Y.; Lee, S. M.; Nieh, W. L.-S. J Appl Polym Sci 2003, 89, 1896.
- 45. American Society for Testing and Materials. ASTM D 1037-93; ASTM: West Conshohocken, PA, 1997.
- American Society for Testing and Materials. ASTM E 1333-96; ASTM: West Conshohocken, PA, 1996.
- Christensen, R. L.; Anderson, W. H. In Proceedings of the 23rd International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1989, p 55.
- 48. Groah, W. J.; Gramp, G. D.; Heroux, G. L.; Haavik, D. W. Forest Prod J 1998, 48, 75.
- Lehmann, W. F. In Proceedings of the 16th International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1982, p 35.
- Zinn, T. W. In Proceedings of the 18th International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1984, p 71.
- 51. American Society for Testing and Materials. ASTM D 6007-96; ASTM: West Conshohocken, PA, 1996.
- Lehmann, W. F.; Roffael, E. In Proceedings of the 26th International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1992, p 124.
- Liles, W. T. In Proceedings of the 27th International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1993, p 233.
- American Society for Testing and Materials. ASTM D 5582-94; ASTM: West Conshohocken, PA, 1994.
- 55. DIN EN 120, DIN EN 120-92; Deutsches Institut für Normung: Berlin, Germany, 1992.
- Sundin, E. B. In Proceedings of the 12th International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1978, p 251.
- Sundin, E. B. In Proceedings of the 16th International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1982, p 3.
- Sundin, E. B.; Risholm-Sundman, M.; Edenholm, K. In Proceedings of the 26th International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1992, p 151.
- Myers, G. E.; Nagaoka, M. In Wood Adhesives; Myers, G. E., Ed.; Forest Product Society: Madison, WI, 1980, p 93.
- Zinn, T. W.; Cline, D.; Lehmann, W. F. In Proceedings of 24th International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1990, p 95.
- Sundin, E. B.; Mansson, B.; Endrody, E. In Proceedings of the 21st International Particleboard/Composite Materials Symposium, Washington State University, Pullman, WA, 1987, p 139.
- Composite Panel Association. ANSI A208.1-1999; American National Standards Institute: New York, 1999.
- SAS Institute. Release 7.00 Software; SAS Institute: Cary, NC, 1998.