VOLATILE ORGANIC COMPOUND HOT-PRESS EMISSIONS FROM SOUTHERN PINE FURNISH AS A FUNCTION OF ADHESIVE TYPE

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ABSTRACT

Three types of adhesives, urea-formaldehyde (UF) resin, phenol-formaldehyde (PF) resin, and polymeric methylene *bis*(phenyl isocyanate) (pMDI), were used for investigating the effect of pressing variables on volatile organic compound (VOC) emissions. The variables examined included press temperature and time, mat moisture content and resin content, and board density. The VOCs emitted during particleboard hot-pressing with Southern Pine furnish were collected with two scrubbers filled with water and methylene chloride, respectively. The water solution of emissions trapped in the first scrubber was analyzed for formaldehyde using a calorimetric method. The other VOCs contained in the scrubbers were extracted with methylene chloride and characterized and quantified by gas chromatography/mass spectrometry (GC/MS). The measurements showed that the effects of press temperature and press time on formaldehyde emissions were significant for all three adhesives. The effect of mat moisture content was also significant for both UF and PF resins, but mat resin content and board density did significantly influence formaldehyde emissions. There was good correlation between formaldehyde and amount of water vapor collected for both UF resin and pMDI. Formaldehyde press emissions with PF resin were lower than with pMDI, due to an additive in the PF resin. Other VOC pressing emissions included terpenes and their derivatives, lower molecular weight aldehydes, ketones, and some high boiling point linear alkanes. Total VOC (TVOC) emissions were significantly affected by press temperature for all three adhesive types, and by press time for the UF and PF resin. Moisture content also had a significant effect on TVOC emissions for pMDI. While UF and PF did not significantly affect TVOC emissions, application of pMDI caused a statistically significant decrease in TVOC emissions. Changes in pinene emissions with press conditions were similar to the changes seen in TVOC.

INTRODUCTION

Volatile organic compound (VOC) emissions from particleboard panel manufacture mainly arise from the drying of green wood particles, hot-pressing of particleboard panels, and post-treatment of pressed particleboard. Drying of green particles prior to pressing is the largest contributor to the VOC emissions during particleboard production, accounting for about 70 percent of the total VOC emissions. The second hugest source is hot-pressing, which contributes about 20 percent of the total VOC emissions [1]. Emissions from particleboard furnish drying have been investigated in previous research [2,3,4]. Past research on air emissions from wood products and hot-pressing focussed primarily on formaldehyde emissions from the adhesive resin. The 1990 Clean Air Act Amendments, and the reduction of the National Ambient Air Quality Standards for ground ozone level reduced from 0.12 to 0.08 ppm have increased the need to measure, understand, and control other types of VOCs that are emitted during hot pressing [5,6,7,8].

Emissions of VOCs during hot-pressing come primarily from the wood particles and the adhesive binder. Compounds emitted from the particles may be attributed to volatile and semivolatile extractive compounds, degradation products of the wood, and chemical reaction products of the wood extractives [2,9,10]. Compounds that have been identified in emissions from hot-pressing of Southern Pine particles include terpenes and their derivatives and low molecular weight aldehydes such as pentanal, hexanal, and octanal [4,11]. The VOC emissions that have been attributed to wood adhesives are formaldehyde, methanol, and phenol [7,9,10,12]. Terpenes have been shown to react with oxides of nitrogen (NOx) in the presence of ultraviolet light to form ground-level ozone [13,14]. Prominent among these ozone precursors are α-pinene and β-pinene, which are the main VOCs from Southern Pine wood extractives.

Press variables that affect VOC emissions during hot-pressing include press temperature, press time, mat moisture content, mat resin content, board density, and adhesive type. A preliminary study [11] was conducted in our laboratory to evaluate some of the effects of these press parameters on the VOC emissions during the hot-pressing of Southern Pine particleboard with urea-formaldehyde (UF) resin. In that study, emissions of both formaldehyde and total VOCs (TVOCs) were found to increase with press temperature, press time, resin application level, and panel density, with greatest effects for the first two variables and more modest effects for the latter two. Increasing mat moisture content showed mixed effects on TVOC emissions, with the maximum TVOC emissions occurring when the mat moisture content was between 10 and 14 percent. The design of our preliminary study [11] did not allow an evaluation of the interactions among press variables.

This study is a continuation of the preliminary study. Specifically, the goals of this study were

1. To develop an improved VOC collection system,
2. To employ a statistical design to evaluate the effects of press parameters and their interactions on VOC emissions from hot-pressing, and
3. To evaluate the effect of resin type on VOC emissions.

MATERIALS AND METHODS

Materials

The wood particles consisted of about 95 percent Southern Pine and 5 percent other species and were commercially prepared by Temple-Inland Corp. (Thomson, GA). The particles were different batches that those used for our preliminary study [11]. Commercial urea formaldehyde (UF) resin (Southeastern Adhesive Company), phenol formaldehyde (PF) resin (Neste Resin Corporation), and polymeric methylene diisocyanate (pMDI) resin (Bayer Corporation) were used to bond the panels.

Chemicals used for GC/MS calibration, 1-pentanal, hexanal, 2-heptanone, octanal, α-pinene, camphor, (-)-borneol, hexdecane, heptadecane, and octadecane, were Sigma research reagents. Methylene chloride (American Chemical Society certified) was purchased from Fisher Scientific.

VOC Collecting System

The system for collecting and trapping VOCs during hot-pressing consisted of a closed caul plate and a VOC emission-trapping system. The closed caul plate was designed to prevent the loss of VOCs during pressing and to construct 19-mm (3/4-in.) panels. Emission trapped by the enclosed caul plate were pulled through two 250-mL scrubbers. The first scrubber contained 100 mL of water chilled in an ice bath. The second contained 100 mL of methylene chloride and was chilled to between -5°C and -10°C in an ethylene glycol bath. Both scrubbers had fritted glass spargers on their inlets to assure dispersion of the vapors. Water vapor emitted from the panel was collected in the impingers and volume of water emitted was determined by subtracting the amount of water that was originally in the impingers.

Panel Manufacture and VOC Collection

The wood adhesive was sprayed onto the particle furnish in a drum blender by using a compressed air spray head. After blending, the furnish was hand felted into a 305- by 305-mm (12- by 12-in.) deckle box. The mat was then pressed in the hot press maintained at the desired temperature. The time and VOC emission collection system were started when the caul plate was closed. Ambient air was pulled through the caul plate and the collection system at a rate of 2.0 L per minute by a vacuum oil pump. The vacuum rate was set at the beginning of the collection and remained at 1.6 to 1.9 L per minute during the collection.

Analysis of VOC Emissions

A 10-mL sample of the water solution in the first scrubber was reserved and used for formaldehyde analysis. Formaldehyde was analyzed by using the chromotropic acid technique described by Carlson and others [7]. The remaining samples from the two scrubbers were combined. The two scrubbers and associated tubing were twice extracted with 20 mL of methylene chloride. The combined solution of the previous water solution with methylene chloride solution was put into a separatory funnel and extracted twice with 30 mL of methylene chloride. The methylene chloride extraction solution was analyzed by gas chromatography/mass spectrometry (GC/MS). The analysis was conducted on HP 5890II/HP 5971A GC/MS with autoinjector. The GC oven temperature was at 40°C for 4 min, then programmed to 280°C at 10°C per minute and held for 8 min. The carrier was helium at 0.7 mL per minute. The injector temperature was 270°C. A 30-m J&W Scientific DB-5MS GC-column was used to separate chemical compounds. The MS ionizer voltage was 70 ev, and ionizer temperature was 200°C. The mass scan range was 40 to 500 m/z at 1.6 scans per second. The VOCs were quantified using an hexadecane as an internal standard to minimize the effect of the mass detector response changes with time. The TVOC emissions in the methylene chloride extraction solution (excluding formaldehyde, methanol, and the other lower molecular compounds that elute earlier than methylene chloride) was calculated by combining the estimated amount of the 70 largest peaks.

Nine chemicals, 1-pentanal, hexanal, 2-heptanone, octanal, α-pinene, camphor, (-)-borneol, heptadecane, and octadecane, were used to estimate the relative response factors of VOCs to the internal standard, hexadecane. Six standard solutions, each containing all nine of the above compounds, were made up at concentrations ranging from 5 to 100 ppm in methylene chloride. These solutions were analyzed by GC/MS. The average relative response factor of each standard was used for VOC calculation. For VOCs other than the nine standard chemicals, relative response factors were estimated according to their carbon number and functional group by assuming the linear relationship between the carbon number and the relative response factor among the chemicals with the same functional group.

Statistical Experimental Design

A preliminary study showed that the possible press variables that affected VOC hot-pressing emissions from UF Southern Pine furnish were press temperature, press time, mat moisture content, mat resin content, and board density [11]. The VOC hot-pressing emissions are also affected by different types of wood adhesives, which have different physical and chemical properties. Because of the large number of runs that would need to be made to evaluate all variables at all levels, a statistical experimental design was used to evaluate the effects of the press variables and their interactions. Factorial experimental designs, such as $2^K$and 3^K, where K is the number of variables and 2 or 3 is the number of treatment levels of each variable. Due to the large number of runs needed for the factorial designs, one half of a factorial experimental design, denoted as 3^{K-1} and 2^{K-1}, are often used to evaluate the effects of any single variable and the simple interactions between any two variables, with the assumption of insignificant effects of interactions between three or more variables. The large number of press variables in this study made it impractical to employ a 3^{K-1} experimental design to investigate VOC hot-pressing emissions, so a 2^{K-1} design was chosen to evaluate the major effects of the variables while minimizing the number of experimental runs required. One center point with three replicates was added to the 2^{K-1} design to determine the nonlinearity of the response in relation to the variables. The experimental design with 2^{K-1} plus center point can evaluate any single variable effect and the interactions between any two variables among the ranges of all variables. Table 1 shows the experimental variables (press temperature, press time, etc.) and their levels (low, center, high) for each adhesive type. The ranges of the levels of the press variables were chosen to represent the press conditions across the industry. Because optimum press conditions are different for each type of adhesive, each type of adhesive was investigated separately. There were 19 runs for each type of adhesive. The detailed test schedule for each type of adhesive is summarized in the first five columns of Table 3. The run number was randomly chosen during the experiments to eliminate run order bias. In the analysis of variance (ANOVA) of the data, effects with a *P* value less than 0.05 were considered statistically significant.

For easy and visual comparison of VOC emissions as a function of press variables and adhesives, the relative effect of each variable was calculated. The relative effect, assuming no interactions between variables, is based on the mean effect of each variable and is calculated as follows:

$$Coef = \frac{\Delta \bar{Y} i}{\sum_{i=1}^{5} |\Delta \bar{Y} i|}$$

Where *Coef* is the relative effect of press variable i, ΔY_i is mean effect of variable i.

RESULTS AND DISCUSSIONS

Improved Collection System

In our preliminary study of VOC emissions during hot-pressing [11], we found that a dual scrubber collection system with water in both scrubbers did not provide optimum VOC collection efficiency. For this study, the second water scrubber was replaced with an impinger containing methylene chloride. Table 2 shows the collection efficiencies for both the original and modified collection systems. This change in collection system also allowed for higher efficiencies at longer press times, which had been a problem with the collection system with two water scrubbers.

Formaldehyde Hot-Pressing Emissions as a Function of Adhesive Type

Formaldehyde hot-pressing emissions from Southern Pine panels come from two main sources: free formaldehyde in the adhesives and emissions from the wood furnish itself. Two of the three predominant adhesives, UF and PF resins, contain free formaldehyde but the third, pMDI, does not. A portion of the free formaldehyde in UF and PF will be emitted from the mat during hot pressing, and the amount of formaldehyde emitted is affected by the press conditions. Appropriate press conditions are determined by the physical and chemical properties of the adhesive, such as curing rate and curing temperature.

Table 3 summarizes the results of the formaldehyde emissions by using the 2^{5-1} plus center point fractional factorial experimental design for each adhesive. Formaldehyde emissions during hot-pressing of panels with UF resin ranged from 80.05 to 445.98 mg per kg oven-dried board (mg/kg OD board), which was significantly higher than emissions from panels pressed with either PF or pMDI resin. Emissions from panels pressed with pMDI ranged from 0.10 to 60.11 mg/kg OD board and with PF ranged from 8.79 to 16.20 mg/kg OD board. The UF resins are known to contain higher levels of free formaldehyde and are subject to hydrolysis, releasing formaldehyde, so the higher levels of formaldehyde emissions for this resin were expected.

For panels bonded with PF and pMDI under similar conditions, the PF panels emitted significantly less formaldehyde than pMDI panels. Furnish pressed at the center level press variables with pMDI and with no resin exhibited emission levels of 33.07 and 41.10 mg/kg OD board, respectively. Thus, formaldehyde was emitted from the wood furnish alone and application of the pMDI resin reduced emissions by approximately 20 percent. However, panels bonded with PF resin showed even greater reductions relative to furnish pressed with no resin. At the center point of the press variables, formaldehyde emissions from particles with PF resin were 11.91 mg/kg OD board, which were also much lower than from the neat wood particles, 71.33 mg/kg OD board, a reduction by approximately 70 percent. While phenol is known to exhibit a negative interference in the chromotropic acid method, tests of spiking impinger solutions with known amounts of formaldehyde showed no reduction in response, eliminating the possibility of phenol interference. It was found that the commercial PF resin used in this study contained an additive with an amine functional group, which reacts rapidly with formaldehyde to reduce emissions during pressing. The source of formaldehyde in the particleboard furnish without applied resin was not determined. It is possible that a portion of the emissions from the furnish were due to particleboard trim recycled back into the commercial furnish used in this study.

Formaldehyde Hot-Pressing Emissions as a Function of Press Conditions

The ANOVA of the data in Table 3 showed that formaldehyde emissions from panels pressed with all three adhesive types were significantly affected by press temperature and press time. Moisture content also had a significant effect on formaldehyde emissions for UF and PF resins. Figure 1 shows the relative effects of press variables on formaldehyde hot-pressing emissions for the three adhesive types. In Figure 1, it is clear that the press variables causing the greatest relative changes in formaldehyde emissions for all three adhesive types were press temperature and press time. The combination of these two effects accounted for between 75 and 80 percent of the effect on formaldehyde emissions. This finding was also in accordance with the ANOVA analysis. The additive in the PF resin caused press temperature to exhibit a significantly negative effect on formaldehyde emissions from particles with PF resin. It is possible that the additive in the PF resin reacts more quickly with formaldehyde at high temperatures than at low temperatures. If the formaldehyde-additive reaction rate increases with temperature much faster than formaldehyde emission rate does, a decrease in formaldehyde emissions would be observed. Increases in mat moisture content were responsible for significant increases in formaldehyde emissions from panels pressed with UF and PF resins. High moisture content in the particle mat can decrease the curing rate of the resin and increase the hydrolysis speed of some wood components. Increased moisture content may also facilitate the transport of the formaldehyde out of the panel. Resin content and board density among all three adhesive types played insignificant roles in formaldehyde emissions in this study.

Relationship between Formaldehyde Emissions and Amount of Water Vapor Collected

Figures 2 and 3 show that formaldehyde emissions and amount of water vapor collected changed with press temperature and press time for panels pressed with the three adhesive types. Formaldehyde emissions increased with press temperature and press time for both UF- and pMDI-particle furnishes. However, it decreased with press temperature for the PF-particle furnish due to the additive in the PF resin. The values at the center point showed that the experimental data exhibited good linearity in the ranges of investigated press variables. Because of the longer press times and higher mat moisture contents, water vapor collected from the PF-particle furnish were higher than from UF- and pMDI-furnishes. As a result of the reaction of pMDI with water during the hot pressing, amount of water vapor collected from pMDI-particle furnish was much lower than from the UF-particle furnish at the same press conditions. There was a good correlation between formaldehyde and water vapor collected from UF- and pMDI-particle mats. The ratio of formaldehyde to water vapor collected is about 4.5 and 1.0 mg/g for panels pressed with UF resin and pMDI, respectively. Formaldehyde emissions did not correlate with water vapor collected in the PF furnish due to reactions between the additive in the PF resin and formaldehyde.

TVOC and Pinene Hot-Pressing Emissions as a Function of Adhesive Type

The emissions from particleboard hot-pressing consist of a large number of volatile organic compounds. The GC/MS total ion chromatography showed that there were more than 100 chemical compounds in the collected VOC solutions. The limitations of the GC/MS instrumentation allowed quantification of no more than the largest 70 Peaks, which accounted for about 97 percent of the VOCs eluting from the capillary GC column. There were two main groups of peaks in the total ion chromatogram. One consisted of terpenes and their derivatives, eluting between 10 and 20 min. The other was a group of linear alkanes with carbon numbers larger than 17, eluting between 25 and 35 min. There were also C_5 to C_9 aldehydes and ketones in the collected VOC solutions. The predominant VOCs emitting from Southern Pine particles were α-and β-pinene. Although pMDI contains some lower molecular weight components, these were not detected in the VOC solutions collected from the pMDI-particle furnish. The pMDI molecules can react with water rapidly during hot-pressing to form amines, which can further react with other pMDI molecules to produce pMDI polymers [15]. It should be noted that the extraction method used for recovering the VOCs from the impinger solutions allowed only compounds with boiling points greater than approximately 40°C to be observed and quantified.

Application of UF and PF resins did not significantly change the TVOC emissions relative to the emissions from furnish that did not contain resin. For example, the TVOC hot-pressing emissions from the PF furnish and neat wood particles pressed at the center point conditions averaged 173.19 and 186.69 mg/kg OD board, which was not statistically significant. In contrast, application of pMDI resin can significantly reduce the TVOC emissions from wood particles, at certain application levels. For panels pressed at the pMDI center point conditions, the average TVOC emissions from wood particles were reduced from 128.62 to 93.77 mg/kg OD board when 4.5 percent pMDI was applied to wood particles. There was not enough evidence to explain the effect of pMDI resin on TVOC emissions from wood particles. One suggestion was that pMDI was cured faster on the particle surface to reduce VOC emissions from particles during hot-pressing.

TVOC and Pinene Hot-Pressing Emissions as a Function of Press Conditions

Figure 4 shows the normalized effects of press variables on TVOC hot-pressing emissions with all adhesive types. Statistical analysis of the TVOC hot-pressing emissios showed that increasing press temperature significantly increased TVOC emissions from panels pressed with all three adhesives. Increased press time caused significant increases in VOC emissions for UF and PF, but it did not affect TVOC emissions from panels pressed with pMDI. The combined effects of press time and press temperature accounted for more than two-thirds of the variance seen in the emissions from panels pressed with UF and PF. For pMDI, press temperature alone accounted for about 45 percent of the variance. Mat moisture content exhibited significant effects only for panels pressed with pMDI, increasing TVOC emissions with increased moisture content. While resin content and panel density did not exhibit significant effects individually, there was a significant interaction between UF resin content and board density. At 5 percent UF resin content, TVOC emissions increased with board density. But at 9 percent UF resin content, TVOC emissions decreased with board density.

Figure 5 shows the relative effects of press variables on the pinene emissions. These were similar to the effects on TVOC emissions for all three adhesive types, except that press temperature did not significantly affect pinene emissions for PF resin. Like TVOC emissions, pinene emissions from UF-particle furnish were significantly affected by the interaction between UF resin content and board density. At a UF resin content of 5 percent, pinene emissions increased from 266.30 to 293.37 mg/kg OD board when board density increased from 705 to 834 kg/m^3. But when UF resin content was 9 percent, pinene emissions decreased from 319.39 to 255.77 mg/kg OD board when the board density increased from 705 to 834 kg/m^3.

CONCLUSIONS

The VOC hot-pressing emissions from Southern Pine particles with three adhesive types were investigated using a 2^{5-1} fractional factorial design, which allowed evaluation of the effect of individual press variables and the interaction between any two press variables. The modified collecting system in which the second scrubber is filled with methylene chloride was more suitable to collect VOC emissions at longer press times. Press temperature and press time significantly affected formaldehyde emissions for all three adhesive types, increasing emissions from panels bonded with UF and pMDI and decreasing emissions from panels bonded with PF. Formaldehyde emissions from UF-particle furnish were the highest among the three adhesive types. Despite the free formaldehyde in the PF resin, formaldehyde emissions from PF-particle furnish were even less than those from pMDI-particle furnish in which formaldehyde emissions only arose from the wood particles. Further study showed that an additive in the PF resin greatly reduced the formaldehyde emissions from the PF-particle furnish, and it may have been responsible for the observed decrease in formaldehyde emissions with increased press temperature. Mat moisture content also significantly increased formaldehyde emissions from UF and pMDI resins. There was good correlation between formaldehyde and water vapor collected for UF resin and pMDI. Nonformaldehyde VOCs emitted from during hot-pressing consisted of terpenes and their derivatives, C_5 to C_9 aldehydes and ketones, and high boiling point linear alkanes. The TVOC emissions were significantly affected by press temperature for all three adhesive types, and by press time for UF and PF resins. Increasing mat moisture content caused a significant increase TVOC emissions for pMDI. Application of UF and PF resin did not significantly affect TVOC emissions, but pMDI caused a decrease.

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Table 1. Experimental variables and their treatment levels during pressing of panels.

Treatment level and adhesive	Press temperature (°C)	Press time (min)	Mat moisture content (%)	Resin content (%)	Board density (kg/m^3)
Low level					
UF	165	4	10	5	705
PF	165	6	11	4	705
pMDI	165	4	10	3	705
Center point					
UF	182	6	12	7	769
PF	182	8	13	6	769
pMDI	182	6	12	4.5	769
High level					
UF	199	8	14	9	834
PF	199	10	15	8	834
pMDI	199	8	14	6	834

Table 2. Comparison of VOC Collecting Efficiency between the Original and Modified VOC Collection System

Name	Injected (mg)	Mean collected (mg)		Mean recovery (%)		Standard deviation (%)	
		Original	Modified	Original	Modified	Original	Modified
Hexanal	10.80	8.48	10.00	78.51	92.62	2.16	3.14
α-pinene	180.70	123,38	157.41	68.28	87.11	1.8	9.10
Heptadecane	7.17	5.94	6.52	82.88	90.96	7.27	8.67

Table 3. Experimental design and summary of the results'

Press conditions[b] (Levels are given in Table 1)					Formaldehyde (mg/kg OD board)			TVOC (mg/kg OD board)			Pinenes (mg/kg OD board)		
Press temp	Press time	Resin level	MC	Panel density	UF	PF	pMDI	UF	PF	pMDI	UF	PF	pMDI
-	-	-	-	+	96	13	0.10	267	151	26	190	52	13
+	-	-	-	-	189	9	21	389	217	109	237	61	42
-	+	-	-	-	221	18	29	448	209	98	279	59	37
+	+	-	-	+	321	9	40	629	304	136	349	77	46
-	-	+	-	-	80	11	0.78	322	85	14	235	32	8
+	-	+	-	+	163	9	17	377	163	71	249	44	30
-	+	+	-	+	240	13	26	454	189	80	290	57	32
+	+	+	-	-	362	11	47	671	274	145	401	73	52
-	-	-	+	-	91	13	14	309	147	84	199	38	41
+	-	-	+	+	178	11	23	466	226	185	284	61	65
-	+	-	+	+	262	14	46	606	191	94	351	58	37
+	+	-	+	-	446	11	60	673	202	113	350	41	44
-	-	+	+	+	136	15	6	247	147	73	165	45	24
+	-	+	+	-	232	11	23	504	167	261	325	45	100
-	+	+	+	-	347	16	42	520	229	79	316	59	30
+	+	+	+	+	553	15	60	536	218	149	320	51	51
0	0	0	0	0	244	11	35	455	196	95	270	46	36
0	0	0	0	0	257	13	32	583	203	95	332	48	37
0	0	0	0	0	249	12	32	529	161	91	300	38	35

[a]There was a 2^{5-1} statistical experimental design for each adhesive type. The order listed was not the run order.

[b]Refer to Table 1 for the values of press time, press temperature, resin content, moisture content (MC) and panel density. The symbols -, +, and 0 represent low level, high level, and center point, respectively. For example, in run #1 for the UF resin, ----+ means press temperature, 165°C, press time, 4 minutes, mat resin content, 5%, mat moisture content, 10%, and board density, 834 kg/m^3.

FIGURE CAPTIONS

Fig. 1. Relative effects of press variables on formaldehyde emissions during hot-pressing of panels bonded with UF, PF, and pMDI.

Fig. 2. Formaldehyde emissions and amount of water vapor collected during hot-pressing as a function of press temperature for panels pressed with UF, PF, and pMDI.

Fig. 3. Formaldehyde emissions and amount of water vapor collected during hot-pressing as a function of press time for panels pressed with UF, PF, and pMDI.

Fig. 4. Relative effects of press variables on TVOC emissions during hot-pressing of panels bonded with UF, PF, and pMDI.

Fig. 5. Relative effects of press variables on pinene emissions during hot-pressing of panels bonded with UF, PF, and pMDI.

Figure 1

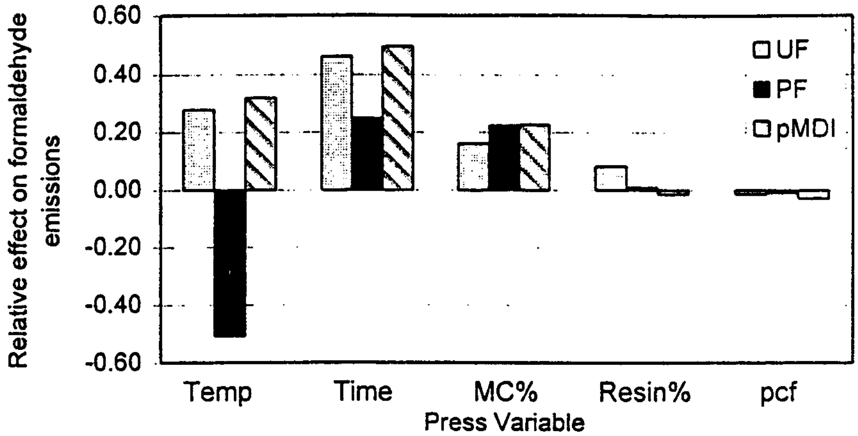


Figure 2

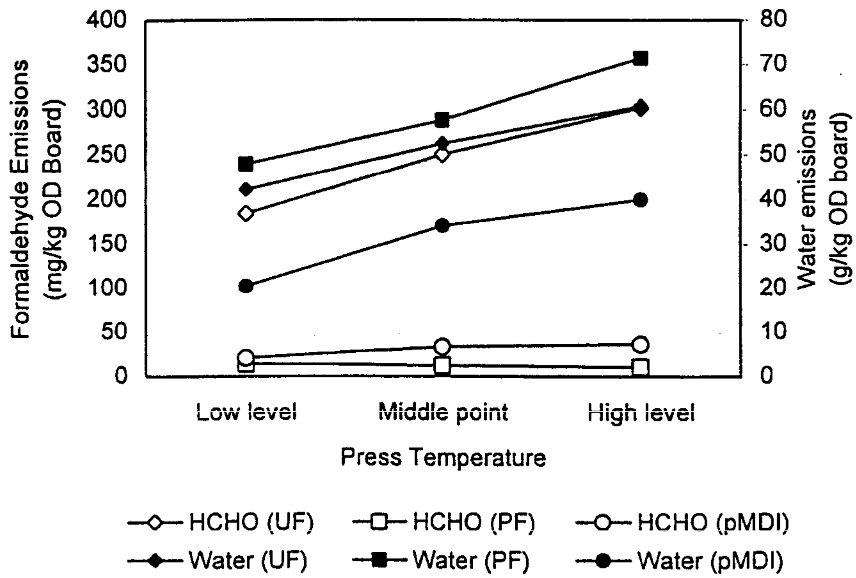


Figure 3

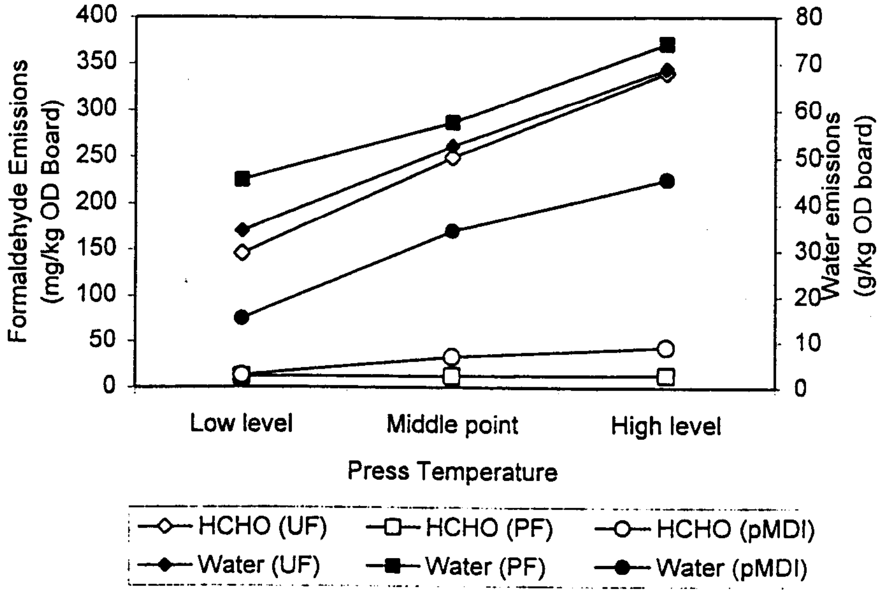


Figure 4

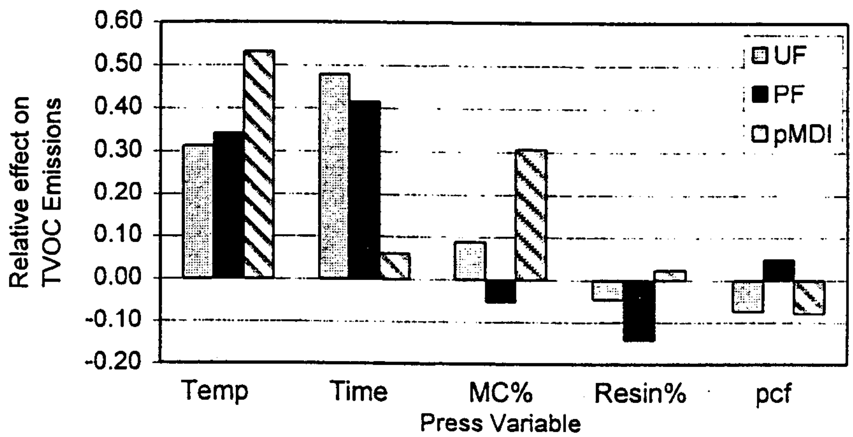
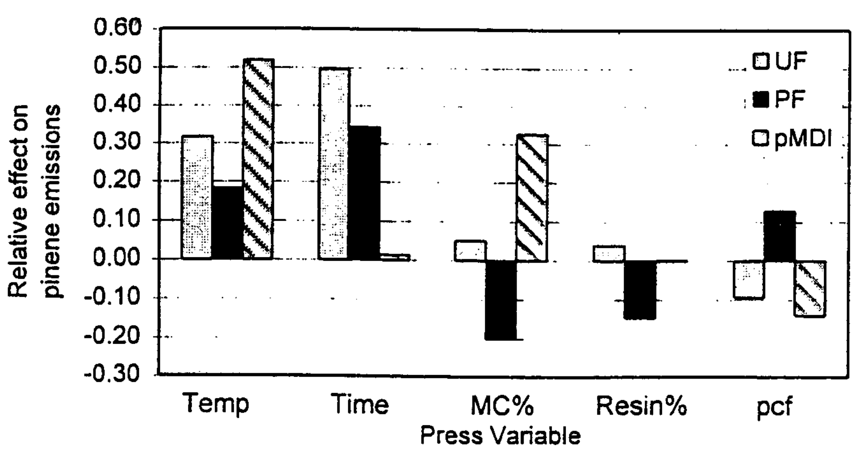


Figure 5



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