Effects of Wheat Protein as a Biological Binder in the Manufacture of Particleboards Using a Mixture of Canola, Hemp, Bagasse, and Commercial Wood

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Abstract

This article deals with the feasibility of the use of wheat protein glue to produce general purpose particleboards from bagasse, canola, and hemp chips and of decreasing the formaldehyde emission by using a bioproduct adhesive. Three series of panels were produced using wood chips in the surface layers and a mixture of annual plants with industrial wood in the middle layers. Particleboards were manufactured using various annual plants. Wheat protein was used in combination with urea-formaldehyde (UF) resin in the surface layers. Pure UF was only used in the middle layer. Panels were tested for some physical and mechanical properties. In addition, the formaldehyde emission according to the perforator method and the bottle method was determined. The data were compared with the respective properties specified by the Deutsches Institut für Normung EN 312-2 standard for commercial wood-based particleboard. The results showed that all mechanical properties greatly exceeded the standard requirements for wood particleboards. An increase of more than 50 percent canola particles in the core negatively affected the internal bond (IB) strength. All of the particleboards produced from hemp and bagasse had modulus of rupture and IB strength higher than required. With those containing up to 50 percent annual plant particles in the middle layer, thickness swelling values met the standard requirement. It was found that applying wheat protein as a bonding agent reduced the formaldehyde emission in comparison to when pure UF resin was applied. This study demonstrated that consistent, highperformance agricultural fiber composite panels with desirable environmental attributes can be successfully developed.

Particleboard is one kind of wood composite that is manufactured on the basis of mechanically chopped, milled, and ground wood particles and bonded by adhesives, usually by a procedure at high temperature and pressure (Maloney 1993, Youngquist et al. 1997, Kharazipour 2004). The particleboard was introduced in Germany about 70 years ago to use wood residues from secondary milling operations, but it succeeded so well that some producers are using wood from trees and primary milling operations in addition to other sources (Kloeser et al. 2007). More than 60 percent of panel board production in Europe and also in Germany is particleboards (European Panel Federation 2005, Marutzky 2006). The demand is high for wood composite industries. One of the ways the forest industry has responded to the global challenges in fiber use and processing efficiency was through the accelerated development of composites (Cooper et al. 1999). But wood supply for making wood composites

has become scarce and expensive in many countries for two reasons: first competition by the paper industry for wood fiber and, second, increasing energy prices and the use of wood as an energy resource (Europäischer Wirtschaftsdienst GmbH [EUWID] 2007).

Annual plants are the best substitute for wood in many aspects: sustainable forests, availability in huge quantities,

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environmentally reasonable, and low prices (Meadows et al. 1992). Various nonwood or annual plant properties have been studied for wood composite production in recent years; for example, waste grass clippings (Nemli et al. 2009), eggplant stalks (Guntekin and Karakus 2008), hazelnut husks (Çöpür et al. 2007), kenaf stalks (Gürü et al. 2006), almond shells (Kalaycıoglu and Nemli 2006), sunflower stalks (Alma et al. 2005), cotton carpel (Bektas et al. 2005), bamboo chips (Xu et al. 2004), kenaf core (Guler and Ozen 2004), cotton stalks (Papadopoulos et al. 2004), coconut and durian peels (Khedari et al. 2004), kiwi prunings (Nemli et al. 2004), and flax shaves (Papadopoulos and Hague 2003).

Nonwood and agricultural residues are excellent alternatives to virgin wood fiber for many reasons. Aside from their abundance and renewability, using agricultural residues will benefit farmers, industry, and human health, as well as the environment. Even in countries with huge forest resources, there is an increased demand and competition for agriculture residues (by-products of agriculture or industry that are usually thrown away or burned) and annual plants by the paper industry and the rapidly growing composites industry. This makes the use of alternative fiber such as agricultural residues more attractive and feasible. Finding new uses for by-products reduces pollution and wastes. Thus, producers of wood composites will be forced to seek nonwood plant fibers to supply the increasing raw material requirement in the future.

In addition, high formaldehyde emissions of derived timber products are no longer acceptable for customers. Although urea-formaldehyde (UF) and phenol-formaldehyde resin binders have contributed greatly to the progress made by the wood industry many years ago, they are still very controversial. Decreasing the emission levels of formaldehyde fumes from manufactured particleboard using UF resins has now become one of the major concerns of the timber and wood adhesives industry, particularly in the case of adhesively bonded wood products (Koontz and Hoag 1995). The source of formaldehyde emission is not only the phenol resin, but wood particles as well, which can emit formaldehyde (Schwarzman and Schedro 1987, Vasilgev et al. 1990). The formaldehyde release from wood-based products has been declining over the last five decades as a result of the development of new resins and new resin technologies (Roffael 1993).

New resins or adhesives from renewable resources (casein, soybean protein, blood, bones, and others) were known in ancient times and used up to the middle of the last century (Frihart 2005) and currently are receiving more attention because of prices for conventional crude-oil–based binders (Yang et al. 2003, EUWID 2007). Among the possible alternatives, wheat protein is an excellent renewable resource that can be used for replacing petroleumderived phenolic compounds. When cereals are used for starch or glucose syrup production, the storage proteins accumulate as a by-product in the watery milling solutions (Lawton 2002, Schöpper 2006). They are abundantly available at cheap prices (Schöpper 2006), and such protein concentrates are excellent raw materials for use as natural binders and can be delivered by starch companies (e.g., from Vilvoorde in Belgium and Cargill in Germany; Müller et al. 2007).

In a previous study (Nikvash et al. 2010) it was shown that bagasse, canola, and hemp are some lignocellulose materials that have acceptable mechanical and physical properties with pure UF resins in manufactured particleboards. Also, formaldehyde (methanal [HOCH]) values of the particleboards were determined by the perforator method but not published.

In this study, we investigated the feasibility of wheat protein as a binder in particleboard manufacturing using some annual plants and wood. Three types of panels were manufactured using bagasse, canola, and hemp. The experimental design used wheat protein mixed with UF adhesive in certain ratios only in the surface layers of the experimental particleboards. Some properties of the boards were investigated. In addition, formaldehyde emission of the wheat–UF boards were studied and compared with pure UF particleboards of the authors' previous study (Nikvash et al. 2010).

Materials and Methods

Experimental variables

Three annual plants species were chosen for this study: bagasse (Saccharum officinarum) from Iran, canola [Brassica rapa (syn. Brassica campestris)], and hemp (Cannabis sativa) from Germany.

Three-layer boards were manufactured using each species alone as well as in mixtures with wood in the middle layers. The species mix ratio varied from 0 to 100 percent and is listed in Table 1. The surface layers of all 40 boards were made of industrial wood.

Preparation of wheat protein binder

Wheat protein was delivered from Cargill, Krefeld (Germany). It is a by-product of glucose syrup production from wheat crops. The solid content of the protein suspension was 50 ± 1 percent and had a pH value of 3.5 to 4.7. This adhesive is in a slurry form and can be sprayed easily in a blending device. The wheat protein was added into the UF binder and stirred mechanically for 10 minutes to prepare a 50 percent mix wheat protein–UF binder.

Board manufacture

The raw material was chipped in a pulpwood chipper with knives set for 15-mm chip length and then flaked with a ring flaker. The particles were dried to about 3 to 4 percent moisture content. The target density was 0.7 g/cm³, and the particleboards were prepared in a random order according to the following procedure. The particles were loaded in a drum-type blending device that provided a uniform distribution of the resin on the chips (flakes). Resins including pure UF for the middle layers, 50 percent UF and 50 percent wheat protein for the surface layers, 1 percent wax, and ammonium sulfate $[(NH_4)_2SO_4]$ as a hardener were prepared, mixed together, and applied by air

Table 1.—The experimental design (in the middle layer).

	% of raw materials					
Board types ^a	Annual plants (one species alone)	Industrial wood				
PB 0%		100				
PB 10%	10	90				
PB 30%	30	70				
PB 50%	50	50				
PB 100%	100	0				

 a PB = particleboard.

Table 2.—Effect of various percentages of annual plants and wheat protein binder on strength and stability properties.^a

	Ratio variability $(\%)$	Density	MOE (kN/mm)	MOR $(N/mm2)$	IB $(N/mm2)$	WA in water soak $(\%)$		TS in water soak $(\%)$	
Annual plants						2 _h	24 h	2 _h	24 h
Reference	PB ₀	0.73	3.92	19.27	0.79	14.6	46.26	3.9	16.63
Bagasse	PB 10	0.73	3.6	15.6	0.62	23.4	70.73	6.90	34.7
	PB 30	0.72	3.09	16.24	0.53	21.4	73.4	6.03	33.56
	PB 50	0.73	2.96	15.51	0.52	23.66	82.26	7.36	40.8
	PB 100	0.65	2.7	13.16	0.57	36.6	102.5	13.7	45.05
Hemp	PB 10	0.72	3.41	14.86	0.55	23.1	96.95	6.35	47.15
	PB 30	0.72	3.04	14.86	0.55	23.96	101.5	4.56	48.16
	PB 50	0.72	3.13	16.97	0.54	26.23	99.73	5.43	47.33
	PB 100	0.67	2.68	13.8	0.57	35.4	116.4	4.9	48.5
Canola	PB 10	0.73	3.01	13.02	0.2	29.6	124.8	16.9	62.5
	PB 30	0.72	3	13.42	0.2	33.4	132.7	23.33	76.56
	PB 50	0.73	2.76	11.36	0.1	71.7	137.5	26.95	76.45
	PB 100	0.68	2.42	10.71	0.09	78.8	156.9	76.45	84.7

^a MOE = modulus of elasticity; MOR = modulus of rupture; IB = internal bond strength; WA = water absorption; TS = thickness swelling; PB = particleboard.

spray to the tumbling particles. The target adhesive levels were 8 percent for the middle and 10 percent for the surface layers (based on the ovendry weight of particles). After removing particles from the blender, the moisture content and correct weight for face or core was determined. Then the mat was formed in a 70 by 46.5-cm forming box using a mat-laying device. The formed mat was loaded into a 60 by 90-cm single-opening hot press and pressed to 20-mm thickness. The pressure required for all particular boards was 220 bars. Press temperature and time were 200° C and 15 s/mm, respectively. After pressing, the finished board was acclimatized.

Preparation of specimens and testing

Perforator method.—The determination of the formaldehyde content was carried out according to the perforator method DIN EN 120 (Deutsches Institut für Normung [DIN] 1993a) as a quadruple analysis. For this purpose, approximately 110 g of the samples (cut into cubes with the size 25 by 25 by 20 mm) were extracted for 2 hours in a perforator apparatus using boiling toluene. The emitted formaldehyde was collected in a water recipient. The values of formaldehyde emissions were calculated using the acetylacetone method and were expressed as they relate to the dry weight of the material (milligrams of formaldehyde per 100 g of dry board).

Bottle method.—For this purpose, approximately 20 to 25 g of the solid wood cut into cubes (25 by 25 by 20 mm) was stored in polyethylene bottles (500 ml) containing 50-ml bidistillated water for 3 and for 24 hours in a cabinet conditioned to 40° C. The emitted formaldehyde was absorbed into the liquid solution and analyzed using the acetyl-acetone method. The mean value of 8 from four determinations was calculated and expressed in milligrams of formaldehyde per 100 g of dry board.

Mechanical and physical testing

Mechanical and physical property tests were conducted on specimens cut from the experimental panels. Tests for 2 and 24-hour water absorption (WA) and thickness swelling (TS) were performed according to EN 317 (DIN 1993b). These measurements were made by immersing specimens in water in a horizontal position at ambient temperature. Three-point static bending modulus of rupture (MOR) and modulus of elasticity (MOE) values were evaluated according to EN 310 (DIN 1993c). Internal bond (IB) strength and screw-holding strength of the panels were determined according to EN 319 (DIN 1993d) standards (all specimens were conditioned to equilibrium at a temperature of 20° C and 65% relative humidity before any tests were carried out). Mechanical tests were performed using a universal-testing machine (Zwick/Roell). An average of 24 measurements was recorded. The mean values of all mechanical and physical properties of particleboards are shown in Table 2. Furthermore, all the data were statistically analyzed by using the analysis of variance (ANOVA).

Results

Mechanical properties

Table 3 shows the results of the ANOVA test for the mechanical and physical properties of particleboards produced in this study. It is well known that the MOR of the panels greatly affects the application area of the panels

Table 3.—Effect of species type and various percentages of particleboards (analysis of variance test).

^a Factor A = type species; Factor B = various percentages.
^b MOE = modulus of elasticity; MOR = modulus of rupture; IB = internal bond strength; WA = water absorption; TS = thickness swelling.
^c Value is not signifi

Figure 1.—Comparison of the bending strengths of particleboards containing bagasse, hemp, and canola, each in mixture with wood in the middle layer.

and is closely related to the density of the panel and the kind of resin being used. For general purpose particleboards, the 312-2 (DIN 2003) standard requires a minimum MOR of 13 N/mm². Table 2 indicates that the highest MOR value of 19.27 N/mm² was observed when only commercial wood was used in the manufacture of the particleboard (PB 0%). All various ratios of bagasse and hemp in the boards fulfill the minimum requirement of the EN standard. In the case of canola boards, which were manufactured with 10 to 30 percent canola (PB 10% and PB 30%), the minimum requirement was also achieved (Fig. 1). Based on the results, the MOR values of canola particleboards were reduced with the increase of canola percentage from 30 to 50 or 100 percent (Fig. 1). The types of raw material have an important influence on the bending properties of the panel produced, but the ratio of this material has a deeper influence on the MOR value. It can be easily seen in Figure 1 that there is a slight decrease in the MOR value when the percentage of annual plant material is increased. Figure 2 indicates the MOE of the particleboards. Based on the results, MOE values of all three series of boards are much higher than the minimum required values.

Figure 3 illustrates the IB strength of panels. According to EN 312-2 (DIN 2003), the minimum requirement of IB of the panels is 0.35 N/mm² for 20-mm-thick panels. As seen in the figure, all combination ratios of bagasse and hemp panels have met the required standard. The IB strength increases with the reduction of canola in the made boards (from PB 100% to PB 10%).

Figure 2.—Comparison of bending modulus of particleboards containing bagasse, hemp, and canola, each in mixture with wood in the middle layer.

Figure 3.—Comparison of internal bond strength of particleboards containing bagasse, hemp, and canola, each in mixture with wood in the middle layer.

Physical properties

Figure 4 shows the TS of the panels made from annual plants and commercial wood after 24 hours of soaking in water. As is shown in Figure 4, the TS rate of reference boards for 24 hours soaked in water is 16.63 percent. According to EN 317 (DIN 1993b), the minimum requirement of TS is 14 percent. As seen in Figure 4, after 24 hours none of the boards were able to fulfill this requirement. However, the lowest TS rate is obtained by bagasse boards and the highest value is present in canola boards. This pattern is also observed in the WA rates in Figure 5. Figures 4 and 5 show the relation between WA and TS and the different ratios of annual plants used in the particleboards. There are slight increases of the physical properties by adding increasing amounts of annual plants. The types of raw material and glue also influence the TS. The boards bonded with a combination of 50 percent UF and 50 percent wheat protein in surface layers obtained very high values in comparison with pure UF resin usage (Nikvash et al. 2010).

The results of ANOVA showed that the effect of species type and various percentages of them were significantly different ($P < 0.01$) except for the interaction MOE value (Table 3).

Formaldehyde content

Formaldehyde (HCHO) contents in the panels were examined by using the perforator method (DIN EN 120; DIN 1993a). Figure 6 shows the perforator values of all manufactured boards with 50 percent wheat protein resin in the surface layers. Reference boards made of pure UF resin show the highest perforator value of about 8.16 mg/100 g (reference board PB 0%). Among the manufactured boards made of annual plants, bagasse and hemp boards fulfilled

Figure 4.—Comparison of thickness swelling of particleboards containing bagasse, hemp, and canola, each in mixture with wood after soaking for 24 hours.

FOREST PRODUCTS JOURNAL VOL. 62, No. 1 53

Figure 5.—Comparison of water absorption of particleboards containing bagasse, hemp, and canola, each in mixture with wood and references after soaking for 24 hours.

the requirement of lower than 7 mg/100 g perforator value. The particleboards in various percentages of canola remain under 8.17 mg/100 g.

Figure 7 shows the bottle method values. In comparison with the reference board, the bottle values of bagasse (PB 10%, PB 30%, and PB 50%) and hemp (PB 10% and PB 30%) are below the reference value. The hemp boards with PB 50 percent and PB 100 percent show higher values than the reference board (PB 0%).

Similar perforator values are shown in Figure 6. The particleboards with different percentages of canola were more or less within the reference values level.

Comparing Figures 6 and 7, it can be seen that the perforator values are proportional to those of the bottle values. The perforator method is more sensitive than the bottle method because the latter method only measures a certain area of the boards. With this method only very low

formaldehyde emissions are shown. Perhaps this method is not appropriate in determining the formaldehyde release from the panels since the panels contain a protein as a bonding agent.

Figure 8 shows the formaldehyde emission levels of particleboards bonded with pure UF adhesive in the surface layers. The particleboards with different amounts of annual plants reach higher perforator values than the reference board, whereas according to DIN EN 120 (DIN 1993a), particleboards should only emit ≤ 8 mg/100 g formaldehyde.

Discussion and Conclusions

Use of plant protein (wheat protein) in order to reduce petrochemical adhesives and their formaldehyde emissions has been studied by many researchers (Krug 2002, Krug

Figure 6.—Perforator values of the panels with wheat protein binder.

Figure 7.—Values of the panels in the bottle method with wheat protein binder.

and Heep 2006, Schöpper 2006). The effect of denaturing agents, pH, and temperature on wheat gluten and soy beans were investigated by Khosravi et al. (2010). They showed how these factors could improve viscosity and solubility of these plant proteins. El-Wakil et al. (2007) also indicated the effect of cylindrical reed fiber shape in enhancing adhesion between the wheat–UF binder and fibers. In their study, samples met the standard requirements using a modified wheat protein binder. These and other studies led us to use other annual plants and wheat protein for

investigating feasibility of wheat protein as a formaldehyde-scavenger binder in particleboard manufacturing.

In this article, the effects of wheat protein as an adhesive in annual plant particleboards were investigated. In addition, the influence of this binder was studied in terms of the quantity of formaldehyde emission. Because the World Health Organization has classified formaldehyde as carcinogenic to humans (IARC 2004) both in Europe and the United States, the formaldehyde emission levels of wood composites have been reduced (Global Insight 2005, 2007).

Figure 8.—Perforator values of the panels with pure urea-formaldehyde binder.

The European standard of formaldehyde emission limit is 6.5 mg/100 g for particleboard (tested by the perforator method). In this study, formaldehyde (HCHO) contents in the UF–wheat binder panels were examined by using both the perforator and bottle methods (DIN EN 120; DIN 1993a). Applying 50 percent wheat protein resin could achieve low emission of formaldehyde without deterioration of the mechanical properties of the particleboard. As in other studies, it is reported that applying resins based on biomass products or by-products (e.g., soy, tannin, and lignin) improved formaldehyde performance of particleboards (Marutzky 2008).

The results indicate that by using wheat protein as a binder, it is possible to produce particleboards with different percentages of hemp, canola, bagasse, and wood chips. An increase in the percentage of hemp, bagasse, and canola chips in the composites matrix results in physical and mechanical properties of particleboards that almost fulfill the required standard values. The particleboards containing hemp or bagasse meet the required MOR, MOE, and IB strength values with the exception of the results after soaking for 24 hours. Owing to the hydrophilic character of the proteins, the boards show a high water absorption behavior. The advantage of the particleboards bonded with high amounts of wheat proteins is the low formaldehyde emissions. The biological binder appears to act as a formaldehyde scavenger.

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CONTENTS

420 Gaining Competitive Advantage in the New Normal by Rajat Panwar, Richard Vlosky, and Eric Hansen

REVIEWED ARTICLES

- 429 Trends in Lumber Processing in the Western United States. Part III: Residue Recovered versus Lumber Produced by Keith A. Blatner, Charles E. Keegan III, Jean M. Daniels, and Todd A. Morgan
- 434 Crack Detection in Computer Tomographic Scans of Softwood Tree Discs by Martin Wehrhausen, Norvin Laudon, Franka Brüchert, and Udo Hans Sauter
- 443 Modeling Air Drying of Sitka Spruce (Picea sitchensis) Biomass in Off-Forest Storage Yards in Ireland by Glen Murphy, Tom Kent, and Pieter D. Kofman
- 450 Effects of Compressive Deformation on Wettability and Water Uptake Behavior of Poplar by Yang Zhang, Siqun Wang, Omid Hosseinaei, and Zhaobing Zhou
- 456 Estimation and Analysis of Sanding Length in Brush Sanding Specially Shaped Wood Products by Wang Baogang, Zhang Zhankuan, and Peng Xiaorui
- 462 Thermal Conversion of Pine Wood Char to Carbon Nanomaterials in the Presence of Iron Nanoparticles by Sung Phil Mun, Zhiyong Cai, Fumiya Watanabe, Umesh P. Agarwal, and Jilei Zhang
- 467 Efficacy of Pentachlorophenol in Biodiesel versus Diesel by Saeed Keshani Langroodi, Hamid Borazjani, Darrel D. Nicholas, Lynn M. Prewitt, and Susan V. Diehl
- 474 [Technical Note] Demonstration Results from Greenhouse Heating with Bio-Oil by Philip Steele, Don Parish, and Jerome Cooper
- 480 Substantial Bark Use as Insulation Material by Günther Kain, Marius-Catalin Barbu, Alfred Teischinger, Maurizio Musso, and Alexander Petutschnigg

ERRATUM

The article ''Effects of Wheat Protein as a Biological Binder in the Manufacture of Particleboards Using a Mixture of Canola, Hemp, Bagasse, and Commercial Wood'' by Neda Nikvash, Alireza Kharazipour, and Markus Euring, Forest Products Journal 62(1):49–57, contained instances of missing or incorrect information.

The first full sentence in the second column on p. 50 should read ''Also, formaldehyde (methanal [HCHO]) values of the particleboards were determined by the perforator method but not published.''

The fifth sentence under ''Board manufacture'' on p. 50 was missing information and should instead read ''Resins including pure UF for the middle layers, 50 percent UF and 50 percent wheat protein for the surface layers, 1 percent wax, and ammonium sulfate $[(NH_4), SO_4]$ (1% in the middle layer and 2% in the surface layer) as a hardener were prepared, mixed together, and applied by air spray to the tumbling particles.''

The unit of measure for MOE in Table 2 on p. 51 should be "kN/mm²."

The y axis labels in Figures 1 through 3 and 7 on pp. 52, 53, and 55 are incorrect. The correct labels are "MOR [N/mm²]" (Fig. 1), "MOE [kN/mm²]" (Fig. 2), "IB [N/mm²]" (Fig. 3), and "HCHO Emission [mg/1,000 g]" (Fig. 7).

The legend for Figure 7 on p. 55 is incorrect. It should read ''Bottle method values of the panels with wheat protein binder after 24 hours.''