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ORIGINAL PAPER

Adhesion Properties of Plywood Glue Containing Soybean Meal as an Extender

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Abstract This study was conducted to evaluate the performance of soybean meal as a protein extender in plywood adhesive intended for sprayline coaters. Ground soybean meal, with 52.8% (dry basis, db) crude protein and 0.9% (db) residual oil, replaced the current industry extender, wheat flour, in the standard glue mix. Substitution was done on the protein content basis. Mixing and adhesion properties of the glue containing soybean meal were compared with those of the industry-standard glue. The soybean meal-based glue showed excellent mixing characteristics and its viscosity (1,275 cp) met the recommended value for this type of adhesive. Its wet tensile strength, however, was less than that of the standard glue and below the threshold value of 200 psi considered as an indicator of strong bonding. The soybean meal glue formulation was then modified further by increasing the amount of meal such that the amount of protein present was double that contributed by wheat flour in the original mix. This adjustment resulted in higher wet tensile strength (209 psi), which not only met the threshold value for strong glue bonds, but also equaled that of the standard glue. These results demonstrated that soybean meal is a viable extender in plywood glue for sprayline coaters.

The mention of trade names or commercial products in this paper is solely for the purpose of providing specific information and does not imply endorsement by the US Department of Agriculture.

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Introduction

Soybean meal is the predominant co-product from the processing of soybean, accounting for 80% of material output. Soybean meal production in the United States was 39.6 million tons in 2008, with 30.8 million tons consumed domestically and 8.9 million tons exported [1]. Soybean meal contains 48% crude protein (CP) and its primary use is as a protein supplier in livestock feed, which uses up to 98% of US soybean meal consumption. The remainder is used for human foods (e.g., baked goods, meat substitutes, infant formulas, and beverages). Soybean meal is the most inexpensive among the various soy protein products, selling for about $0.28/kg [1], compared with $0.48/kg for flour (55% CP), $2.05/kg for concentrate (65–70% CP), and $2.70/kg for isolate (at least 90% CP) (prices supplied by commercial manufacturers). Because of its abundant quantity, ready availability, and low cost, soybean meal has been, and continues to be, an attractive starting material for developing bio-based products, such as adhesives.

The technology of soybean-based adhesives is not a recent development. The wood products industry has had a successful history with glues containing proteins from soybean, other vegetables, and animals that dates back to the period before World War II [2]. Soybean meal was reported to have been used favorably in the wood veneer industry [3]. Alkali-dispersed soybean glues were used extensively during World War II for construction, packaging, and transportation [2]. The postwar years found soybean flour being blended with casein or blood in glues that were used to bond interior-grade plywood, doors, and millwork [4]. However, this was also the time that the petrochemical industry experienced its own expansion and generated low-cost, high-volume synthetic resin adhesives that eventually displaced the natural glues.

The availability and prices of the synthetic resins, however, were heavily dependent on the global petroleum situation. During the oil crisis of the early 1970s, when resin supply was limited and price was high, the wood industry made an immediate partial return to natural glues, but resorted to synthetic adhesive resins again as soon as oil prices stabilized [2]. Since that time, the wood products industry has had ongoing interest in alternative sources of adhesives to minimize the impact of disruption caused by another petrochemical crisis.

There has been renewed interest in soybean-based wood glues in recent years. Huang and Sun [5, 6] evaluated the adhesive properties of soy protein isolates modified by denaturants or reducing agents and found that their water resistance and bond strengths improved. Kreibich et al. [7] successfully developed an adhesive system that used soy protein isolate and phenol-resorcinol-formaldehyde for end-jointing green lumber. Yang et al. [8, 9] cross-linked soybean flour with phenol-formaldehyde (P-F) resins and found the glue to have satisfactory bonding performance in medium-density fiberboard and Southern pine plywood. Li et al. [10, 11] developed PF-free adhesive mixes containing soy protein isolate that gave high water resistance and strong bonds in wood composites and interior-grade plywood.

In 2002, our laboratory developed soybean flour-based foamed plywood glue that is now being used commercially [12]. In this earlier work, we included soybean meal among the commercial soybean protein products that we screened for foam extrusion glue [13]. We found that soybean meal was not a suitable extender because of its poor solubility and foaming properties. However, we observed that the major protein sub-units were still present in soybean meal and showed an SDS-PAGE pattern that was similar to those of undenatured water-extractable soybean proteins. This finding suggested that it may be possible to make the soybean meal protein more reactive by physical (e.g., grinding) and/or chemical means. The highly alkaline pH of plywood glues (as high as pH 12) could be an ideal environment for modifying the meal protein through unfolding of the structure and increasing the available reactive sites. This report describes the results of our present study to incorporate soybean meal in plywood glue as a protein extender and the comparison of the mixing and adhesion properties of a soybean meal-based adhesive with the industry's standard glue.

Materials and Methods

Materials

Soybean meal was provided by Archer Daniels Midland Inc. (Decatur, IL). GP RPPY-5779 P-F resin (43% non-volatiles) and Southern pine veneers were supplied by Georgia-Pacific Chemicals (Decatur, GA). Glu-X filler, a proprietary product milled from wheat intended expressly for the plywood adhesive industry, was provided by The Robertson Corp. (Brownstown, IN). Wheat flour from General Mills (Minneapolis, MN) was obtained from the Experimental Baking Laboratory of the Cereal Products and Food Science Research Unit, National Center for Agricultural Utilization Research (Peoria, IL).

Determination of Moisture, Oil, and Protein Contents

Soybean meal was ground into 40-mesh particle size by using a Cuisinart coffee grinder (Model DCG-12BC, East Windsor, NJ) for 2 min. No preparation was needed for the wheat flour. The drying of samples prior to analyses was not necessary. Moisture, crude protein (%N $\times$ 6.25; %N $\times$ 5.7 for wheat flour), and crude oil contents of the samples were determined by using AOCS standard methods Ba 2a-38, Ba 4e-93, and Ba 3-38, respectively [14].

Preparation and Selection of Plywood Glue Mixes

Soybean meal (10-mesh, as received) was ground further to 40-mesh particle size by using the Cuisinart coffee grinder for 2 min, a duration that was effective in attaining the desired particle size without heating up the grinding chamber. The 40-mesh particle size was selected based on our exploratory experiments on the relationship between particle size and extractable proteins (results not shown). The ground meal was then stored in screw-capped bottles until analysis or use. Two glue formulations were considered for testing the performance of soybean meal as an extender. One recipe was standard glue for Southern pine prepared by a paddle-type mixer [15], while the other was for glue applied by sprayline coaters (personal communication, Dan DiCarlo, Georgia-Pacific Resins, Inc.). Both glues do not require foaming like the adhesive for foam extrusion and would be more accommodating of the presence of minor quantities of other non-protein components, such as oil or starches. The compositions of the control adhesives and soybean meal-based glue are given in Table 1.

Ground soybean meal replaced the industry's current extender, wheat flour, in the formulation on a protein content basis, adapting this approach based on its success in our earlier work [12]. The amount of filler in the soybean meal glue was adjusted accordingly to meet viscosity

Table 1 Composition of standard and soybean meal-based plywood glues

Formulation and ingredients	Mixing time (min)	Quantity (g/100 g glue mix)	
		Standard glue (wheat flour, 13.8% CP, db[a])	Soybean meal-based glue (52.8% CP, db)
Glue for paddle-type mixer[b]			
Water		16.7	16.7
Protein extender	3	5.7	1.5
Filler (Glu-X)	2	5.7	9.8
P-F[c] resin	3	7.7	7.7
50% NaOH	10	1.8	1.8
P-F resin	5	61.3	61.3
50% NaOH	5	1.1	1.1
Glue for sprayline coaters[d]			
Water		13.6	13.6
Protein extender	3	6.1	1.6
Filler (Glu-X)	2	7.0	11.5
P-F[b] resin	2	18.2	18.2
50% NaOH	7	1.8	1.8
P-F resin	2	51.6	51.6
50% NaOH	2	1.2	1.2

[a] *CP* crude protein, *db* dry basis

[b] From Sellers [15]

[c] P-F, phenol-formaldehyde

[d] From Georgia-Pacific Resins Inc., personal communication to author

requirements of around 1,000–2,000 cp. Glues were prepared as in our previous study [12] and then allowed to stand overnight at room temperature prior to plywood processing. Overnight standing is a standard practice in the plywood glue industry that allows chemical interactions (e.g., breakage of internal bonds and crosslinking) among ingredients to occur. The viscosities of glue mixes were determined by a viscometer before and after overnight standing. The glue mix where soybean meal showed acceptable mixing behavior and viscosity was then selected for plywood processing and bond strength testing.

Plywood Processing and Bond Strength Evaluation

Glues were tested on 3-ply wood panels using 30.5 cm × 30.5 cm (12 in. × 12 in.) Southern pine veneers. For this area, the glue spread was 13 g for single glue-line application. The assembly time was 30 min. Two glue samples were prepared for each protein extender being tested and triplicate 3-ply boards were prepared per glue treatment.

Laboratory-scale plywood processing and evaluation of glue bonding strength were done by following exactly the conditions and methods described by Hojilla-Evangelista [12]. Test panels were cut into 8.3 cm × 2.5 cm (3.25 in. × 1.0 in.) shear specimens according to the specifications of the American Plywood Association [16]. The "vacuum/pressure soak" method was then applied, using a modified autoclave, to simulate accelerated aging before shearing [17]. Specimens were immersed in water, placed under vacuum (−30 mm Hg) for 30 min, and then pressurized (30 psi) for another 30 min [18]. After draining the water, specimens were sheared immediately in a Globe Model Testing Machine and their wet tensile strengths were recorded. A wet tensile strength value of at least 200 psi indicated a strong glue bond. The average strength per panel was calculated from 20 test specimens.

Statistical Analyses

Statistical analyses were performed by using the SAS® Systems for Windows software program (SAS Institute Inc., Cary, NC). Analysis of variance and Bonferroni *t*-tests or Duncan's multiple-range test were performed on data to determine significant differences among the treatments ($p < 0.05$).

Results and Discussion

Moisture, Oil, and Protein Contents

Soybean meal, as received, contained very little oil (0.9% db) and 52.8% (db) crude protein. The amount of oil we determined was close to the typical 0.6% (db) in commercial soybean meal and the protein content was, likewise, within the 49–54% (db) reported for the meal [19]. In our previous research [12, 13], we demonstrated that soy protein products containing no more than 1.0% crude oil could be used in a plywood glue mix without adverse effects on mixing or bonding properties. Because of the low residual oil content of the meal used for this study, further defatting was not necessary before incorporation in the glue mix. Wheat flour had 13.8% crude protein (db) and 1.0% crude oil (db). These values were also within the range reported for most commercial flours [20].

Selection of Plywood Glue Mix

The wheat-based standard formulation for paddle-type mixers (Table 1) had a viscosity of 7,053 ± 1,276 cp immediately after mixing, which increased only slightly to 7,480 ± 1,209 cp after overnight standing. When ground soybean meal was used in the formulation (Table 1), the resulting freshly prepared glue had an average viscosity of 18,680 ± 760, but after overnight standing, the glue thickened notably (31,500 ± 2,300 cp). The final viscosity of the soybean meal-containing glue was more than four times greater than that of the standard glue, which may be attributed to the higher amount of Glu-X filler and also the intrinsic traits of protein and carbohydrates in the soybean meal that may have fostered more expansive networking reactions with the resin [4]. We also noted that the mixture of water, soybean meal, filler, and first resin portion transformed into a very thick, dough-like mass after the first addition of 50% NaOH, even when the caustic was mixed in very slowly. Moderate quantities of lumps covered in gelatinized material were also detected. The viscosity of the soybean meal glue decreased substantially to 13,600 ± 3,650 cp when adjustments were made in the mixing method, such as increasing the resin amount in the first addition to facilitate dispersion of solids, decreasing the second resin portion correspondingly, and slow additions of the resins and caustic. However, the glue's thickness was still considerably greater than that of the standard glue.

In the sprayline coaters formulation (Table 1), the standard wheat flour-based glue had a viscosity of 23,340 ± 2,060 cp. We believe that this unexpectedly high viscosity was probably caused by an older batch of resin that we used. We had observed that the resin tended to thicken over time, despite being stored in the frozen state. However, this same batch of resin was used to make the soybean meal-based glue, which had a viscosity of 21,740 ± 805 cp. This value compared favorably with that of the standard glue. We also detected the presence of some particulates in the soybean meal glue, but, generally, the final consistency and appearance of the soybean meal glue was very similar to that of the standard glue. We found it easier to work with the sprayline glue formulation and had better success in meeting the viscosity limits (once we started using the newer batch of resin), so this was the media that we selected for testing further the adhesion properties of the soybean meal glue.

Glue Bond Strength Evaluation

Soybean meal dispersed uniformly in the resin and the resulting glue had a flow behavior that was similar to that of the standard glue, even though its consistency appeared to be thinner. The viscosity values support this, as they were only about half of that observed for the standard glue, both immediately after mixing and after overnight standing (Table 2). This was probably the effect of reduced quantity of soybean meal extender. Even though the amount of filler was increased, this lignocellulosic material does not have sufficient quantities of proteins and starches that can provide rheological properties similar to those of wheat flour or soybean meal.

The mean wet tensile strength of the soybean meal glue was less than that of the standard glue (Table 2) and lower than the threshold value of 200 psi, indicating that the glue bond was slightly weaker. To improve bonding strength, we modified the soybean meal-based glue by doubling the amount of protein in the mix. The presence of more protein can provide additional reactive groups that can participate in crosslinking and polymerization reactions with the resin and wood [21], which may then strengthen adhesion. The amount of soybean meal was, thus, increased to 3.2 g/100 g glue mix, while the filler amount was changed to 9.6 g/100 g glue mix. The wet tensile strength of the modified soybean

Table 2 Viscosities and bonding strengths of plywood glues with wheat flour (standard) or soybean meal as a protein extender

Parameters	Protein extender		
	Wheat flour (13.8% CP, db)	Soybean meal (52.8% CP, db)	Soybean meal (modified glue mix, 2× protein content)
Glue viscosity after mixing, cp	2,435 ± 205a	1,010 ± 85b	1,145 ± 265b
Glue viscosity after overnight standing, cp	2,330 ± 170a	1,275 ± 95b	1,080 ± 305c
Wet tensile strength, psi	211 ± 42a	186 ± 36b	209 ± 44a

Values are mean ± standard deviation of duplicate preparations of glue mixes for viscosity and six boards (20 test specimens per board) in total from duplicate glue mixes for tensile strength. Values across columns followed by different letters are significantly different ($p < 0.05$)

CP crude protein, *db* dry basis

meal adhesive increased to nearly equal that of the standard glue (Table 2), and it also exceeded the threshold value of 200 psi.

In plywood adhesives, finely ground soybean protein products will wet and swell in water, but remain undispersed until treatment with strong alkali [4]. The alkaline dispersion process releases useful adhesive properties of the meal by breaking the internal hydrogen bonds of the usually coiled protein molecule, unfolding the structure, and making the exposed reactive groups from amino acids available for adhesion to wood [4]. Lambuth [4] reported that, for soybean meal to perform well as a protein glue, the adhesive grade (44–52% CP) meal must be ground to a fine flour, milling until 60–80% will pass through a 46-μm (325-mesh) screen. The particle size we used (40-mesh) was considerably larger, but it was adequate to allow for uniform dispersion in the mix and provide satisfactory flow properties, as well as tensile strength. However, these mixing and bonding properties may still be substantially improved by using a much smaller particle size for the meal.

Babcock and Smith [22] used soybean meal in their formulations of phenolic resin plywood glues that were tested on both hard and soft wood veneers in the laboratory. The soybean meal glues passed the strength specifications and also showed favorable resistance to mold action and exterior weathering conditions. They also mentioned that the typical solvent-extracted soybean meal, which contains 25–30% water-soluble components (sugars and non-protein nitrogen), was not suitable as an extender for their purpose, but the soybean meal from which such water-soluble fractions have been removed would be satisfactory in their formulation [22]. Despite the different formulation and age of the study, the findings showed that an effective plywood glue was possible with soybean meal as an extender.

Conclusions

The glue containing soybean meal as a protein extender had mixing and viscosity properties that were comparable to that of the standard wheat flour-based sprayline plywood glue. The bond strength of the soybean meal-based plywood glue was initially weaker, but was substantially improved to acceptable levels by increasing the amount of soybean meal in the glue mix. These results demonstrated that soybean meal is a viable extender in plywood glues for sprayline coaters.

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