Evaluation of sorghum flour as extender in plywood adhesives for sprayline coaters or foam extrusion

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A B S T R A C T

This study was conducted to evaluate sorghum flour as protein extender in phenol–formaldehyde-based plywood adhesive for sprayline coaters or foam extrusion. Defatted sorghum flour, containing 0.2% (db) residual oil and 12.0% (db) crude protein, was analyzed for solubility and foaming properties. Sorghum flour proteins were least soluble (≤12%) under acidic pH, most soluble (72%) at pH 10, and produced substantial and highly stable foam at pH 10. Sorghum flour was substituted (on protein content basis) for wheat flour in the standard glue mix. Mixing properties and bond strength of the sorghum-based glue were compared with those of the industry standard glue. The sorghum flour-based adhesive had mixing properties and appearance that were superior to those of the standard wheat flour-based plywood glue, but its viscosity and bond strength were markedly less. Doubling the amount of protein contributed by sorghum flour in the glue mix markedly improved both viscosity (1104 cP) and adhesion strength (1.37 MPa) of the sorghum-based plywood glue to acceptable levels. The modified sorghum flour-based plywood glue also produced foam that remained stable up to 3 h. These results demonstrated that sorghum flour is a viable extender in plywood glues for sprayline coater or foam extrusion.

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1. Introduction

The continuing global fuel energy situation that has been responsible for exorbitant gasoline prices and greater demand for biofuels like ethanol is spurring the re-evaluation of cereal grains other than maize as renewable energy raw materials. If the projected spike in the utilization of fuel ethanol occurs, then sorghum (Sorghum bicolor L. Moench), which has historical use in the production of industrial alcohol (Hahn, 1970; Rooney and Serna-Saldívar, 1991), could become a major substrate along with maize for the fuel ethanol industry. Sorghum ranks fifth among cereals in terms of total worldwide production, with the United States accounting for about a third of the quantity grown (Rooney and Serna-Saldívar, 1991). Sorghum production in the United States amounted to 350 million bushels (19.6 million tons) in 2010 (Feed Outlook, 2010). Livestock feed and export markets are the major users of sorghum grains (Rooney and Serna-Saldívar, 1991).

A likely consequence of an increased utilization of sorghum in ethanol fermentation is a corresponding boost in sorghum products available in the market, the implication of which would be a reduction in their value unless other or novel uses are developed for them. It is a recognized fact that sorghum products compete for the same outlets as other cereal products based on their lower cost and availability (Rooney, 1973). Among sorghum products, the flour has found the most use in non-food industrial applications. Gelatinized sorghum flours have been used in oil drilling as fluid loss-control agents and in the paper industry as coating and beater additives (Anderson, 1969; Hahn, 1970). The binding properties of starch and protein in sorghum flour have been exploited in the manufacture of building materials, such as insulation, gypsum board and ceiling tiles, foundry core binders, molding sand additives, low-grade tannic ore pellets, and charcoal briquettes (Anderson, 1969; Hahn, 1970). Sorghum hulls and flour were also used as filler or extender in urea–formaldehyde (U–F) plywood adhesives for roll-coaters (Alexander and Krueger, 1978; Edler, 1981; Ramos et al., 1984) and, more recently, in inexpensive adhesives for wallboard and packaging materials (Rooney and Waniska, 2000).

Our laboratory has been evaluating cereal and oilseed protein products as alternative extenders in phenol–formaldehyde (P–F)–based plywood glues for foam extrusion and sprayline coaters. We have developed a soybean flour–based foamed plywood glue that is now being used commercially (Hojilla-Evangelista, 2002) and a soybean meal–based sprayline adhesive (Hojilla-Evangelista, 2010). It was a logical step to test the performance of sorghum flour in these two types of plywood adhesives, especially given the flour’s past use in roll-coater plywood glue. The present study, then,
was conducted to evaluate the use of sorghum flour as extender in plywood glue intended for sprayerline coaters or foam extrusion. This report presents some functional properties of sorghum flour protein that were pertinent to wood adhesive preparation and the comparison of the mixing and adhesion properties of the sorghum-based adhesive with the industry’s wheat flour-based standard plywood adhesive.

2. Materials and methods

2.1. Starting materials

Sorghum flour was provided by the Grain Quality and Structure Research Unit, USDA, Agricultural Research Service (Manhattan, KS). The flour was prepared by grinding whole sorghum grains with a Udy mill (Udy Corp., Fort Collins, CO) equipped with a 0.5 mm screen and then stored in a screw-capped polypropylene container at room temperature until used. The GP RPPY-5779 phenol–formaldehyde (P–F) resin (43% non-volatiles) and Southern pine veneers were provided by Georgia-Pacific Chemicals (Decatur, GA). Glu-X filler, a proprietary wheat-based product created 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 Functional Foods Unit of our research center.

2.2. Moisture, crude protein, and crude oil contents

Moisture, crude protein (%N × 6.25; %N × 5.7 for wheat), and crude oil contents of the sorghum and wheat flour samples were determined using AOCs standard methods Ba 2a–38, Ba 4e–93, and Ba 3–38, respectively (AOCs, 1998).

2.3. Determination of protein solubility and foaming properties

Whole-grain sorghum flour was first defatted with hexane according to the method of Hojilla-Evangelista and Evangelista (2006), using multiple cycles of extraction until a residual oil content of less than 0.5% was obtained. The defatted flour was air-dried and then stored in screw-capped bottles until analyzed or used.

The protein solubility profile of sorghum flour was generated according to the method of Balmaceda et al. (1984). Aqueous dispersions containing 10 mg protein/mL were prepared and their pHs were adjusted to 4.0, 5.5, 7.0, 8.5, and 10.0. After centrifuging at 10,000 × g for 20 min, the amounts of soluble protein in the supernatants were determined spectrophotometrically using the Biuret method. Bovine serum albumin was the protein used to generate the standard curve.

Foam capacity and stability of sorghum flour protein (10 mg protein/mL) were determined at neutral pH and at pH 10, which is typical of plywood adhesives, by following exactly the procedure described by Myers et al. (1994). Foam capacity was the volume (mL) of foam produced in 1 min. Foam stability was expressed as the % foam remaining after standing for 15 min.

Sorghum flour-based plywood glue mix (see following section) was screened for foam extrusion using the whipping method (Hojilla-Evangelista and Dunn Jr., 2001). Two-hundred grams of glue mix was prepared for the whipping test and allowed to stand overnight at room temperature. The glue was poured into a mixing bowl and then whipped at maximum speed (setting no. 10) for 5 min using a KitchenAid mixer (model KSM 90, KitchenAid, St. Joseph, MI). The whipped glue mix was transferred into a tared, graduated beaker. Foam volume and mass were recorded to calculate foam density. Foam volumes after 1 and 3 h were recorded to determine foam stability. Duplicate determinations were done per sample.

2.4. Preparation of plywood glue mixes

Plywood glue intended for either sprayerline application or foam extrusion was selected as the media for testing the performance of sorghum flour as extender. These types of glues have similar compositions and viscosity requirements, but the protein extenders (wheat flour or spray-dried animal blood) and application techniques are different. The compositions of the industry standard for sprayerline coater and sorghum-based glue mixes are given in Table 1. Wheat flour is the current extender in the industry glue. Sorghum flour replaced wheat flour in the formulation on the basis of protein content, following the approach that we used successfully in our previous work on soy flour-based foamed plywood adhesive (Hojilla-Evangelista, 2002). Glues were prepared according to the manner described previously by Hojilla-Evangelista (2002). The ingredients were added in the mixing bowl of the KitchenAid mixer in the order presented, with the P–F resin and 50% NaOH being split into two portions and added alternately with each other in the last stages. Mixing time at each stage was usually 2 min, except for the first addition of caustic, which required 7 min. Glues were 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. Viscosities of glue mixes were determined by a viscometer before and after overnight standing.

2.5. Plywood processing and glue strength evaluation

Glues were tested on 3-ply wood panels using 30.5 cm × 30.5 cm (12 in × 12 in.) Southern pine veneers. The glue spread was 13 g for single-glue-line application and assembly time was 20 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 (2002). 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 (1984). The “vacuum/pressure soak” method was then applied, using a modified autoclave, to simulate accelerated aging before shearing (American Plywood Association, 1984). Specimens were immersed in water, placed under vacuum (~30 mm Hg) for 30 min, and then pressurized (30 psi) for another 30 min (U.S. Department of Commerce, 1983). After draining the water, speci-
mens were sheared immediately in a Globe Model Testing Machine and their wet tensile strengths were recorded. Wet tensile strength value of at least 1.38 MPa (200 psi) indicated a strong glue bond. The average strength per panel was calculated from 16 test specimens.

2.6. Statistical analyses

Statistical analyses were performed by using the SAS® Systems for Windows software (SAS Institute Inc., Cary, NC). Analysis of variance and Bonferroni t-tests were performed on data to determine significant differences among the treatments (p < 0.05).

3. Results and discussion

3.1. Proximate composition

Sorghum flour, as received, contained 9.5% moisture, 3.1% crude oil (db) and 12.0% crude protein (db). The crude protein and oil contents were similar to those reported for roller-milled and dry-milled sorghum flour (Hahn, 1969). After defatting with hexane, the flour had 0.2% residual oil (db). Because oil is a defoamer, it was necessary to reduce the oil content to this extent to allow for the evaluation of foaming properties. Wheat flour had 10.5% moisture, 1.0% crude oil (dry basis, db), and 13.8% crude protein (db). The amount of protein we determined was slightly higher than that indicated on the product specifications sheet (10.5%), but still within the range of values reported for most commercial flours (Pratt, 1971).

Schober et al. (2005) reported that commercial grain sorghum flour contained 77.3% (db) starch, while flours from nine other sorghum hybrids had starch contents that ranged from 78.8 to 82.1% (db). These amounts are slightly greater than the 70.9% reported for wheat flour (Hoseney, 1994). However, the water-soluble carbohydrates, particularly the pentosans, have been reported to influence viscosity of the flour dispersion by their ability to absorb ca. ten times their weight in water (Kulp, 1968), as well as the water resistance of adhesive bonds (Huang and Li, 2008). Sorghum flour has 0.13–0.28% (db) water-soluble pentosans (Schober et al., 2005), which are markedly less than the 1.5% (db) found in wheat flour (Hoseney, 1994). These low amounts of water-soluble pentosans are actually advantageous for sorghum flour, because their presence at higher levels in plywood adhesives is considered detrimental. Frihart et al. (2010) reported that, generally, soluble carbohydrates raise dispersion viscosity, use up some of the cross-linker, and increase water absorption that consequently weakens the glue bond under high-moisture conditions. Ash contents of extenders for adhesives are recommended to be 0.5–3.0% to prevent increased abrasiveness of glue lines that would rapidly wear out saws, shaper knives, and spray tips (Sellers, 1985). The typical ash contents of sorghum flour (0.8%; Rooney, 1973) and wheat flour (0.4%; Hoseney, 1994) are well within the recommended range for plywood adhesive extenders.

3.2. Solubility profile and foaming properties

Proteins must be soluable first before they can express their functionality and solubility is an important factor that affects foaming, emulsification, and gelation (Kinsella, 1979). Sorghum flour proteins were least soluble under acidic conditions, but their solubility increased substantially at pH ≥ 7.0, with the maximum (72%) being obtained at pH 10 (Table 2). The presence of alkali generally improves solubility of proteins, especially if the pH is greater than 10.5, by causing dissociation and disaggregation of proteins (Kinsella, 1979). Our solubility profile followed the same trend that was observed by Elkhalifa and Bernhardt (2010) for ungerminated sorghum flour; but, the amounts we determined at pH 4 to pH 8.5 were less than the 35–55% they reported for the same pH range. The markedly higher solubility of sorghum flour proteins at pH 10 supported earlier findings that showed greater extractability of sorghum storage proteins under alkaline conditions (Belton et al., 2006) and also indicated that the proteins will likely be able to withstand the alkaline dispersion process during the preparation of plywood adhesive. Alkaline dispersion brings about the unfolding of the usually coiled protein structure, thus making the exposed reactive groups from amino acids available for adhesion to wood (Lambuth, 1994).

Sorghum flour solubel proteins produced foam volume at neutral pH (Table 2) that was substantially greater than the 136 mL of foam generated by soybean flour used in another study (M. Hijoilla-Evangelista, unpublished data). However, the foam bubbles disintegrated almost immediately, unlike soybean flour, which had 96% remaining foam after 15 min. At pH 10, less foam volume was produced by the sorghum flour proteins, but foam stability was significantly greater (Table 2). The large fraction of sorghum flour soluble proteins at pH 10 probably contributed to the creation of highly stable foam. German et al. (1985) explained that soluble proteins can rapidly reach the interface, lower the interfacial tension, and unfold to form surface films around air droplets, thus preventing the bubbles from combining and eventually collapsing. The foaming behavior of the sorghum soluble proteins in very alkaline media suggested that these proteins have strong potential as extender in foamed plywood glues.

3.3. Mixing and adhesion properties of sorghum-based plywood glue

Sorghum flour dispersed quickly and uniformly during the preparation of the glue mix. The resulting glue had no visible particulates and appeared smoother than the wheat flour-based standard glue: but, the viscosity was far below the minimum 1000 cP for this type of plywood adhesive (Table 3). Rooney and Serna-Saldívar (1991) noted that sorghum flour does not have proteins that produce viscoelastic properties like those of wheat gluten. Sorghum flour also contains five to ten times less water-soluble pentosans than wheat flour, which we pointed out in the discussion on proximate composition, and therefore, did not have much of these compounds to bind significant amounts of water (Kulp, 1968) that will consequently increase viscosity of the glue mix (Frihart et al., 2010). The amount of sorghum flour that we used was evidently not sufficient to effect thickening of the adhesive. Additionally, the low viscosity may indicate extensive hydrolysis or disintegration of the sorghum flour in the highly alkaline medium (Kinsella, 1979) such

<table>
<thead>
<tr>
<th>Table 2</th>
<th>Solubility profile and foaming properties of defatted sorghum flour or sorghum-based plywood adhesive. a</th>
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</thead>
<tbody>
<tr>
<td><strong>Functional properties</strong></td>
<td><strong>Soluble proteins (%) in sorghum flour</strong></td>
</tr>
<tr>
<td>At pH 4.0</td>
<td>7.8 ± 0.0</td>
</tr>
<tr>
<td>At pH 5.5</td>
<td>12.0 ± 0.1</td>
</tr>
<tr>
<td>At pH 7.0</td>
<td>28.8 ± 2.5</td>
</tr>
<tr>
<td>At pH 8.5</td>
<td>46.5 ± 2.1</td>
</tr>
<tr>
<td>At pH 10.0</td>
<td>72.0 ± 0.2</td>
</tr>
<tr>
<td><strong>Foaming capacity (mL) of sorghum flour soluble proteins</strong></td>
<td><strong>Foam volume (mL)</strong></td>
</tr>
<tr>
<td>At pH 7.0</td>
<td>174 ± 4</td>
</tr>
<tr>
<td>At pH 10.0</td>
<td>124 ± 1</td>
</tr>
<tr>
<td><strong>Foam stability (%) remaining foam of sorghum flour soluble proteins</strong></td>
<td><strong>Remaining foam after 1 h (%)</strong></td>
</tr>
<tr>
<td>At pH 7.0</td>
<td>9 ± 0</td>
</tr>
<tr>
<td>At pH 10.0</td>
<td>98 ± 1</td>
</tr>
<tr>
<td><strong>Remaining foam after 3 h (%)</strong></td>
<td><strong>Values are mean ± standard deviation of duplicate determinations.</strong></td>
</tr>
</tbody>
</table>

a
that functional properties were impaired. The weaker bond (lower mean tensile strength) of the sorghum-based adhesive compared with that of the standard glue (Table 3) appears to support this behavior.

To increase viscosity and improve bonding strength, we modified the sorghum flour-based adhesive by adding twice the amount of protein in the formulation. The presence of more protein can provide additional reactive groups for crosslinking and polymerization reactions with the resin and wood (Frihart and Wescott, 2004), which may then strengthen adhesion. Thus, 14.0 g sorghum flour was added per 100 g glue mix, while the filler amount was changed to 2.4 g/100 g glue mix. The viscosity of the modified sorghum flour adhesive was more than double that of the first formulation (Table 3) and within the required 1000–2000 cP for sprayline or foam extrusion plywood glues. The mean wet-tensile strength of the modified sorghum glue was also nearly equal that of the standard glue (Table 3) and met the threshold value of 1.38 MPa (200 psi) to be considered a strong adhesive. These results compared favorably with the findings of Ramos et al. (1984). They developed a U–F plywood glue extended with 20% sorghum flour that had bond strength and moisture resistance equal to that of the industry-grade U–F adhesive.

Plywood adhesive that uses this rate of replacement by sorghum flour (i.e., double the amount of wheat flour) will be cost-competitive as long as the price of sorghum flour remains about half of that of wheat flour. Should the price of sorghum flour rise beyond this level, sorghum flour may be blended with wheat flour wherein its proportion can be maximized until the point where the final cost of the resulting adhesive will still be equal to that of the standard wheat flour-extended plywood glue. The decreased amount of Glu-X filler in our sorghum-based adhesive (from 6.1 to 2.4 g/100 g glue mix) will also help reduce the final cost of the adhesive.

We then subjected the modified sorghum flour adhesive to the whipping test to evaluate its potential use in foam extrusion application. Our earlier study (Hojilla-Evangelista and Dunn, 2001) showed that the laboratory whipping method correlated well with the results of bench-scale foam extrusion and may be used to screen various protein products as possible substitute for blood in foamed glues. The modified sorghum-based adhesive produced foam volume (Table 2) that was about 50–60% of those generated by several soybean flours (425–550 mL) we tested previously, and was markedly less than the nearly 600 mL foam produced by the blood-based glue (Hojilla-Evangelista and Dunn Jr., 2001). Although the amount of foam decreased sharply within 1 h, the remaining foam was fairly stable even after standing for 3 h (Table 2), but still not as stable as those generated by soybean flour- and blood-based glues (>95% and >80% remaining foam after 1 h and 3 h, respectively). It should be pointed out that the sorghum flour-based adhesive we tested had a much lower protein content compared with the standard blood-based or soybean-based foamed glues. It would not be practical to attempt a protein content-based replacement of animal blood extender with sorghum flour; given the great disparity in the protein contents of these materials (99% versus 12%, respectively), a significant quantity of sorghum flour (ca. 45 g) will be required to match the amount of protein contributed by animal blood in the glue mix and problems with mixing and viscosity will likely arise. Despite its lower protein content, the modified sorghum flour-based plywood glue showed promise as an alternative extender in adhesives intended for foam extrusion. It may be possible to improve the foaming properties of the sorghum-based adhesive by combining sorghum flour with soybean flour.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Protein extender</th>
<th>Sorghum flour (12.0% CP, db)</th>
<th>Sorghum flour (modified glue mix, 2× protein content)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glue viscosity after mixing, cP</td>
<td>2190 ± 90 a</td>
<td>410 ± 50 c</td>
<td>985 ± 106 b</td>
</tr>
<tr>
<td>Glue viscosity after overnight standing, cP</td>
<td>1950 ± 120 a</td>
<td>480 ± 30 c</td>
<td>1104 ± 136 b</td>
</tr>
<tr>
<td>Wet tensile strength, MPa</td>
<td>1.45 ± 0.34 a (211 ± 50 psi)</td>
<td>1.27 ± 0.26 b (184 ± 38 psi)</td>
<td>(199 ± 31 psi)</td>
</tr>
</tbody>
</table>

Values across columns followed by different letters are significantly different (p < 0.05). CP, crude protein; db, dry basis.

* Values are mean ± standard deviation of duplicate preparations of glue mixes for viscosity and six boards (16 test specimens per board) total from duplicate glue mixes for tensile strength.

4. Conclusions

The soluble proteins in sorghum flour showed solubility and foaming properties that were desirable for the highly alkaline conditions in plywood glues for sprayline coaters. The adhesive containing sorghum flour as protein extender had mixing properties and appearance that were superior to those of the standard wheat flour-based plywood glue, but its viscosity and bond strength were markedly less. Both viscosity and adhesion strength of the sorghum-based plywood glue were substantially improved to acceptable levels by increasing the amount of sorghum flour in the glue mix. The modified sorghum flour–based plywood glue also produced notable volume of stable foam. These results demonstrated that, technically, sorghum flour is a viable extender in plywood glues for sprayline coater or foam extrusion. Cost-competitiveness of the sorghum flour–extended plywood adhesives will undoubtedly be dependent on the differential between the prices of wheat flour and sorghum flour.

Acknowledgments

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