

PRODUCTION OF YELLOW POPLAR INTERIOR PLYWOOD WITH COTTONSEED-BASED PROTEIN ADHESIVES¹

Rubin Shmulsky[†]

Department Head and Professor
Department of Sustainable Bioproducts
Forest and Wildlife Research Center (FWRC)
Mississippi State University
Mississippi State, MS 39762
E-mail: rs26@msstate.edu

Michael K. Dowd

Lead Scientist and Acting Research Leader
Commodity Utilization Research and Cotton Structure and Quality Research Units
Southern Regional Research Center–USDA
New Orleans, LA 70124
E-mail: michael.dowd@usda.gov

Dercilio J. V. Lopes[‡]

Assistant Research Professor
E-mail: dvl23@msstate.edu

George D. Miller, Jr.[‡]

Research Associate II

Edward D. Entsminger^{*†}

Research Associate II
Department of Sustainable Bioproducts
Forest and Wildlife Research Center (FWRC)
Mississippi State University
Mississippi State, MS 39762
E-mail: edward.entsminger@msstate.edu

(Received April 2021)

Abstract. Defatted cottonseed (CS) and water-washed CS meals were prepared from glandless CS and were used in adhesive formulations to produce three-ply yellow poplar plywood panels. Adhesive resins were prepared from each protein meal with sodium bisulfite and one of two polyamido-amine-epichlorohydrin (PAE) wet strength agents, and the plywood panels were produced by hot pressing. Shear strength and water resistance were determined by American Society for Testing and Materials (ASTM) and American National Standards Institute for Hardwood and Decorative Plywood/Hardwood Plywood and Veneer Association (ANSI/HPVA) methods and were compared with the properties of plywood panels made with an adhesive formulated from a commercial soybean meal. Panels prepared from three protein meals had comparable shear strengths. The combinations of the two CS preparations and the two wet strength agents produced panels with acceptable wet resistant properties, whereas the soybean meal only produced acceptable panels with one of the wet strength agents. Because the panels prepared from the two CS meals had comparable properties, there appears to be no benefit to including a water-washing step to increase the meal's protein level. In contrast with a recent literature report suggesting the addition of alkali to elevate the formulation pH was necessary with CS meal, suitable panels were prepared herein without the addition of the base. This difference may have been due to the slightly higher pressing temperature and longer press times used in this work compared with earlier results. The CS meals showed promise as formaldehyde-free hardwood-plywood wood-based adhesives for interior applications.

* Corresponding author

† SWST member

‡ Deceased

¹ The copyright of this article is retained by the authors.

Keywords: Cottonseed meal, shear strength, soybean meal, water resistance, wood adhesives, yellow poplar hardwood plywood.

INTRODUCTION

The most commonly used adhesives in the wood composite industries are based on urea-formaldehyde, melamine-urea-formaldehyde, phenol-formaldehyde, or polyurethane formulations (Dunky 2003). Among them, urea-formaldehyde-based adhesive is the primary resin used for interior applications because of its bonding strength, rapid curing, and low cost compared with other adhesives (Conner 1996; Cheng et al 2016). However, due to concerns about formaldehyde and its reclassification as a carcinogen by the World Health Organization (WHO), there is an ongoing interest to eliminate the compound from the workplace (Liteplo et al 2002). As part of this effort, there is a need to develop applicable low-formaldehyde or formaldehyde-free, bio-based, environmentally safe, wood-based adhesives (Hodgson et al 2002).

One approach to this issue is to consider the reapplication of protein-based adhesives. Protein adhesives were commonly used in the wood products industry prior to the development of petroleum-based formulations (Lambuth 2003). Since the oil embargo crisis of the 1970s, there has been renewed interest in developing these natural materials as adhesives for a variety of reasons, namely eliminating formaldehyde to reduce employee exposure, consumer exposure, environmental release, reduced petroleum reliance, and improved process sustainability (Marra 1992).

Because of the scale of the soybean oil industry and the large volumes of soybean meal that are available, research in this area has focused on soybean proteins (Kumar et al 2002; Pizzi 2006; Li 2012), and interior plywood adhesives based on defatted soybean meal are currently on the market (Malin 2005). In addition, other protein-based materials have been studied and reported to produce good wood bonding. Among these proteins are wheat gluten (Trischler et al 2018; Xi et al 2021), pea protein (Santoni and Pizzo 2013), peanut meal (Li et al 2015), and blood meal protein (Li et al 2018).

Considerable plywood manufacturing takes place in the southeastern region of the United States. Although the southeastern United States produces soybeans (*Glycine max* (L.) Merr.), the region has considerable agricultural acreage invested in cotton (*Gossypium hirsutum* L.) production (Dodds and Pieralisi 2021; Irby 2021). This production generates not only cotton fiber, but also an underutilized seed coproduct. By weight, more seed (1.4 kg) are produced per kilogram of fiber (Dowd et al 2018). Because of the presence of a polyphenolic compound gossypol in the seed that has antinutritional or even toxic properties toward several animal species, few uses have been developed for this protein. As ruminant livestock animals can tolerate gossypol, most of the seed is often used as feed for dairy cows. Alternatively, it is processed to recover a food grade vegetable oil and a protein-based meal, with the latter also being used for ruminant livestock animals. Because of the colocation of plywood manufacturers and the cotton processing plants in the southern United States, this protein could be used as a potential environmentally safe and friendly, bio-based, formaldehyde-free wood adhesive.

Initial work conducted at the laboratory bench scale has indicated that cotton protein, both in the form of an isolate (>90% protein) or a meal (40-50% protein), have potential to bind wood veneers together to make plywood (Cheng et al 2013, 2016, 2017; He et al 2014a, 2014b, 2019, 2020). This work has shown that significant adhesive strength is possible, and it has demonstrated that the water resistance of cottonseed (CS) proteins appears to be better than that of soybean proteins. The process of transferring this work to producing plywood panels, however, has only recently begun (Li et al 2019; Chen et al 2020).

As there are considerable differences in the formulations and pressing conditions used in the literature, our work was conducted to better understand the need for some formulation components, for example, sodium hydroxide, and to see whether pressing times could be reduced. In addition, this

work was designed to determine whether there was any benefit to increasing the protein level of the meal by water-washing and to start to prepare boards at conditions similar to those used in typical commercial operations.

Therefore, the objectives of this research is to use two different CS meal preparations and an in-house soybean meal (as a control) to make environmentally friendly bio-based plywood adhesives using three different hot press times of 7, 8.5, and 10 min and two different polyamido-amine-epichlorohydrin (PAE) solutions at a constant hot press temperature of 135°C (275°F) and pressure of 1.241 MPa (180 psi). In addition to prepare and test the properties of the plywood panels, another purpose in this work is to start trying to reduce press times, as this factor directly relates to productivity and the potential for any commercialization. To that end, the authors start to explore the development work needed for larger scale plywood production by focusing on CS meal preparation, press times, and the use of epichlorohydrin-based wet strength agents to improve performance of the adhesive formulations.

MATERIALS AND METHODS

Protein Preparation

The CS proteins used in this study were prepared in house. Glandless CS (ie a low gossypol genotype) was cracked with a 20.32-cm plate mill, and the bulk of the hulls were separated with a 45.72-cm vibratory shaker fitted with two stacked screens, a #4 mesh screen that retained the fuzzy hulls and a #12 screen that allowed fine pieces to pass. The intermediate fraction consisted of the bulk of the kernel pieces with some seed debris (ie hull fragments, fine sticks, etc). The kernel pieces were further cleaned by passing the material through a laboratory aspirator to remove the nonkernel material. The cleaned kernels were then milled with a 20.32-cm pin mill to produce full-flat flour.

The CS flour was defatted in 2 kg batches in a 20-L rotary evaporator operated with 8 L of hexane at 20 RPM and 50°C. The evaporator functioned as an extractor by using a fluted mixing flask and

redirecting the condenser condensate back to the extractor. Each batch extraction was conducted for 2 h. After extraction, the meal was separated from the miscella (mixture of solvent and extracted oil) by filtering on a pair of 250-mm i.d. Büchner flasks with medium-fast VWR Scientific (Radnor, PA) creped #417 filter paper and then was washed with additional fresh hexane (~2 L) to remove as much of the hold-up miscella volume as possible. The extraction, filtration, and washing were then repeated a second time to reduce the residual oil level to below 1%. After the second washing, the meal was allowed to off gas residual hexane under a laboratory fume hood. This CS meal sample was used as the first experimental protein. This material was analyzed for nitrogen with a Leco (Model 528, St. Joseph, MI) combustion nitrogen analyzer and found to have 9.4% nitrogen, corresponding to 56.5% protein ($N \times 6.0$).

Approximately half the above CS meal was water-washed to remove soluble carbohydrate (mostly raffinose). One and three-quarters ($1\frac{3}{4}$) kilogram of CS meal was mixed with 10 L of deionized filtered water in a 19-L (ie 5-gallon) bucket. The mixture was stirred at room temperature with a pail mixer for 30 min. The meal was then separated by centrifugation in 1-L bottles at $8000 \times g$ (speed $\times$ gravity) for 10 min, and the liquids were decanted. Each bottle was then refilled with fresh deionized water, the protein redispersed, and recentrifuged at the same conditions. The washed CS meal from the second centrifugation was recovered as previously stated and then freeze-dried. The process increased the protein concentration to levels slightly below those of a protein concentrate. This product formed the second experimental protein sample. After re-equilibrating at atmospheric conditions, nitrogen analysis yielded a total nitrogen level of 11.0% corresponding to 65.8% protein.

The commercial source soybean protein meal used was Prolia 200/70 from Cargill, Inc. (Wayzata, MN). It was specified as a 50.0% protein product that had been classified with particle sizes between 70 and 200 mesh and had a protein dispersion index of 70.0. Nitrogen analysis gave a nitrogen level of 8.18% corresponding to 49.10% protein.

Adhesive (Resin) Preparation

All adhesives were synthesized in the laboratory under ambient conditions. Component amounts are as described in Table 1 and varied as needed to yield adhesive with a suitable viscosity range. Each protein was mixed with deionized water at ambient temperature with moderate stirring until fully dispersed. While maintaining stirring, sodium metabisulfite ($\text{Na}_2\text{S}_2\text{O}_5$) was then added to modify viscosity and one of two PAE preparations was added to improved water resistance. The PAE preparations tested were Kymene Soyad CA1130 and CA1920A from Solenis, LLC (Wilmington, DE). The amounts added were based on the manufacturer's recommendation. The CA1130 had 20% solids, a higher molecular weight, and a medium cationic charge density was used at a 1:7 weight ratio, whereas the CA1920A had 30% solids, a reduced molecular weight, and higher cationic charge, was used at a 1:8 weight ratio. Hydrated lime (calcium hydroxide, $\text{Ca}(\text{OH})_2$) was added to the soybean preparations (10 mL of a 23% w/v [weight per volume] lime dispersion), as the addition of a base better simulated current and historical industry practice (Lambuth 2003). Hydrated lime was not added to the CS formulations, as preliminary experiments found it to be detrimental to the curing of the plywood panels and water resistance of the samples.

Panel Preparation

Three-ply plywood panels were pressed from 30.48 cm $\times$ 30.48 cm $\times$ 3.5 mm thick sheets of yellow poplar/tulip poplar (*Liriodendron tulipifera* L.) veneer. Adhesive was applied with a 10.16-cm wide roller to one side of a veneer. A second layer

of veneer was placed over the adhesive with its grain oriented perpendicular to the core layer's grain. The process was repeated with a second layer of adhesive and a third layer of veneer again oriented perpendicular to the middle layer. The amount of adhesive applied to each panel was 40 ± 2 g, which equates to approximately 220 g of resin per square meter of glue line (ie about 45 pounds per 1000 square feet). After preparation, the panels were cold pressed at 25°C and 1.03 MPa (150 psi) for 5 min, and then transferred immediately to a Clifton Hydraulic Hot Press (Model 0390; Clifton, NJ). The hot press conditions were 135°C and 1.241 MPa (180 psi). Hot press times were 7, 8.5, or 10 min, respectively. Four panels were prepared with each resin formulation and press time, for a total of 72 panels.

Shear Strength Test

The shear strength of the plywood was determined using a universal testing machine in accordance with American Society of Testing Materials D906-17 Standard Test Method for Strength Properties of Adhesives in Plywood Type Construction in Shear by Tension Loading (ASTM 2017). Six test specimens (25.4 mm $\times$ 82.6 mm) were cut from each prepared panel. The outer veneer layers of each specimen were cut to yield a 25.4 mm $\times$ 25.4 mm bonded area for testing. The specimens were then conditioned at approximately 25°C and 65% RH. The testing machine was operated with a crosshead speed of 23 mm/min, and shear strength values were calculated as follows:

$$\text{Shear Strength (MPa)} = \frac{\text{Maximum Tension Force at Break (N)}}{\text{Gluing Area (mm)}^2} \quad (1)$$

Table 1. Formulation of cottonseed and soybean protein meal wood adhesives.

Protein type ^a	PAE type ^b	PAE weight (g)	Water weight (g)	Protein weight (g)	$\text{Na}_2\text{S}_2\text{O}_5$ weight (g)	Viscosity (cP)	pH
CS meal	CA1130	100.0	500	200	2.8	230×10^3	5.6
CS meal	CA1920A	87.5	500	200	2.8	466×10^3	5.5
WW CS meal	CA1130	100.0	500	200	3.3	794×10^3	5.4
WW CS meal	CA1920A	87.5	500	200	3.3	288×10^4	5.6
Soybean meal	CA1130	85.7	400	200	2.5	212×10^3	10.3
Soybean meal	CA1920A	75.0	400	200	2.5	380×10^3	10.5

^a CS, cottonseed meal; WW, water-washed cottonseed meal to remove soluble carbohydrates.

^b PAE, polyamido-amine-epichlorohydrin wet strength agents.

Water Resistance Test

The water resistance of the plywood panels was determined with a three-cycle soak test in accordance with the American National Standards Institute for Hardwood and Decorative Plywood/Hardwood Plywood and Veneer Association (ANSI/HPVA HP-1 2016). Six specimens (5.08 cm × 12.70 cm) were cut from each panel and were soaked in tap water at $24 \pm 3^\circ\text{C}$ for 4 h, and then dried between 49°C and 52°C for 19 h with sufficient air circulation to lower the MC of specimens to 12% or below of the oven-dry weight (ANSI/HPVA HP-1 2016). This soaking and drying process was repeated three times. The degree of delamination of each specimen was measured after every cycle according to the standard as any continuous opening between two veneer layers longer than 5.08 cm, deeper than 0.635 cm, and wider than 0.008 cm.

The test requires that five of the six specimens pass the first soak cycle and four of the six specimens pass the third soak cycle for a given plywood parent panel to pass the test. It also requires that for any treatment to have passed the test, 95% of the specimens must pass the first soak cycle and 85% of the specimens tested must pass the third soak cycle.

Statistical Analysis

The shear strength data were analyzed in a full factorial arrangement of treatments (protein meal types, PAE types, and pressing times) in a completely randomized with subsampling design, based on the replicated panels. The data were reported as mean value and standard deviation. The treatment factors were evaluated using analysis of variance (ANOVA) (F-test: $p \leq 0.05$). Additionally, a Tukey multiple mean comparison test was used to assess whether any differences were apparent among the tested proteins. All analyses were conducted using SAS software version 9.4 (SAS Institute 2013; Cary, NC).

RESULTS

Panels formed with all treatment combinations exhibited acceptable and high shear strength (Fig 1).

No standard strength levels exist for these types of products. As stated in the APA Voluntary Product Standard PS 1–19 (APA 2020), as long as a plywood panel is manufactured using the veneer grades, adhesive, and construction established in the standard's prescriptive requirements, the panel is by definition acceptable. Commercial acceptability is ultimately governed by wood failure and wood strength. As long as the primary location of failure is in the wood and not in the adhesive, then the adhesive is considered to be developing sufficient minimum strength. Although some variation was apparent in individual samples, there were few notable trends among the strength developed for either CS meals or soybean meal. The ANOVA indicated three statistically significant effects (Table 2). A Tukey multiple mean comparison test ($\alpha \leq 0.05$) detected statistical differences in the three tested protein meal types. The direct effect was for protein meal type, as this factor was statistically significant ($p = 0.0429$). The ANOVA also indicated a significant interactions between protein meal type and PAE type ($p = 0.0097$). In addition, there was also a statistical difference detected among panels ($p < 0.0001$).

Most of the CS and soybean-based panels passed the three-cycle ANSI/HPVA water resistance tests (Fig 2). The single combination of treatments that showed some difficulty with this test was the soybean-based meal incorporating the CA1130 PAE product at the lower press times; however, at the 10 min press time, all four plywood panels produced with this combination of protein and PAE treatment passed the test.

By pooling panels by protein type or PAE type, an assessment was made as to the water resistance of the treatment factors under the standard. Both the CS meal and water-washed CS meal passed the three-cycle test with 90.3% and 91.7% of the specimens passing the test. The soybean meal did not pass the test with only 70.1% of the specimens passing after the third cycle. However, if the treatment with CA1130 was neglected, the soybean protein also passed with 100% of the specimens tested. Similarly, the CA1920A PAE treatment also passed with 95.3% of the specimens passing the test. However, the CA1130 treatment did not pass

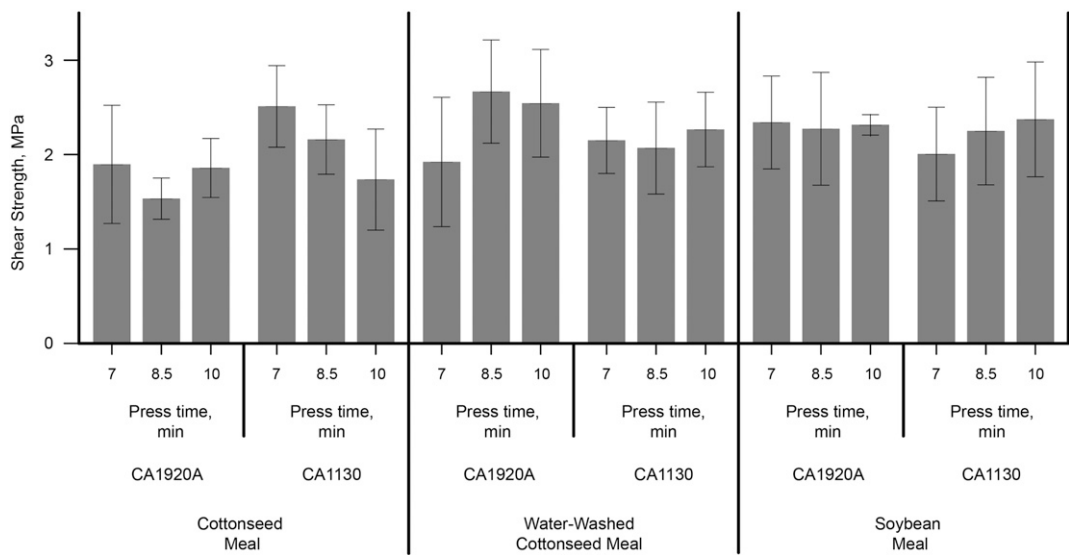


Figure 1. Shear strength of cottonseed meal-based and soybean meal-based yellow poplar (*Liriodendron tulipifera* L.) hardwood plywood adhesives formulated with two different polyamido-amine-epichlorohydrin (PAE) wet strength agents (CA1920A and CA1130) and prepared with three hot-press times (7, 8.5, and 10 min). The bars represent standard deviations (SD).

the standard with only 74.5% of the specimens tested. Again, the difference was due to the failing of the combination of the soybean meal with the CA1130 PAE treatment factor.

DISCUSSION

Early patent literature on protein-based adhesives mentions CS protein among several protein sources as being potentially useful as a glue or wood adhesive (Osgood 1926; Johnson 1935; Galber and Dike

1942; White 1943). The first reported research studies looking at shear strength and chemical resistance properties of defatted CS meal were by Hogan and Arthur (1951a, 1951b, 1952). These reports compared the shear strength properties of wood bonded with defatted CS meal, defatted peanut meal, and casein proteins, and concluded that CS protein could be a potential competitor to these other proteins. However, little follow-up work was conducted at the time, and it was not until the mid-2010s that the topic was re-explored.

Table 2. The ANOVA statistical output test for fixed effects of cottonseed and soybean protein meal-based plywood adhesives.

Effects ^a	DF	Sum of squares	F-Value	p-Value
Protein meal type	2	9.5834	3.34	0.0429*
PAE	1	0.0391	0.03	0.8695
Time	2	0.1400	0.05	0.9524
Protein meal type × PAE	2	6.9808	2.43	0.0097*
Protein meal type × Time	4	9.2319	1.61	0.1855
PAE × Time	2	1.4575	0.51	0.6045
Protein meal type × PAE × Time	4	8.1569	1.42	0.2395
Panel	54	77.4940	32.65	<0.0001*
Residual	360	15.8233	—	—

ANOVA, analysis of variance.

^a PAE, polyamido-amine-epichlorohydrin wet strength agents.

* Denotes statistically significance at $\alpha \leq 0.05$ level.

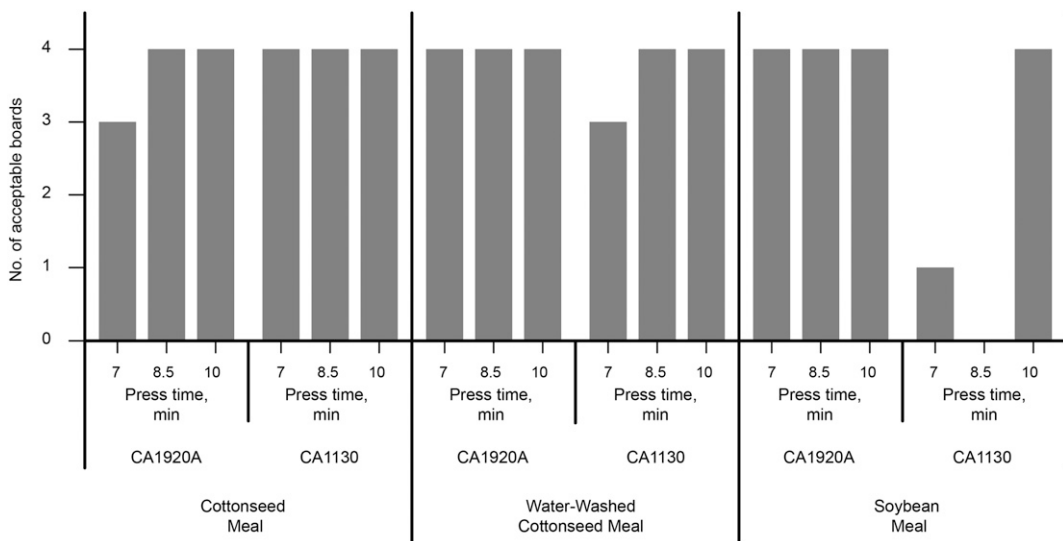


Figure 2. Number of acceptable panels produced from cottonseed meal-based and soybean meal-based yellow poplar (*Liriodendron tulipifera* L.) hardwood plywood panels following ANSI/HPVA HP-1 water resistance testing. Plywood panels were formulated with two different polyamido-amine-epichlorohydrin (PAE) wet strength agents (CA1920A and CA1130) and prepared with three hot-press times (7, 8.5, and 10 min).

Starting around 2013; Cheng and coworkers (Cheng et al 2013, 2016, 2017) reported on the use of CS protein isolate, a ~90% protein product, to bond thin wood veneers, both as is and with a series of compounds supplemented to test for an increase either bonding adhesion or water resistance. At the same time, He and coworkers (He et al 2014a, 2014b, 2019) started related work with CS meals, which are lower in protein (~45-60%), but easier to prepare. These studies found comparable adhesive strengths and improved water resistance comparable to that of soybean meal-based adhesives tested as controls.

In the past 2 yr, this research area has been extended to prepare and test plywood panels and other products from CS protein formulated adhesives. Li et al (2019) prepared panels as part of a study on the use of phosphorus and calcium additives to improve the adhesive water resistance. The study found that CS protein isolate fortified with phosphoric acid produced southern yellow pine (*Pinus* spp. L.) three-ply plywood that passed the ANSI/HPVA tests for use as internal hardwood plywood (Li et al 2019). The panels were hot pressed at 170°C for 10 min. Chen et al (2020) produced five-ply panels

from yellow poplar veneer with in-house extracted CS meal. This is the first known study to include a wet strength agent (such as PAE) in the bio-based adhesive formulation with various pressing times and temperatures. Chen et al (2020) also reported that acceptable panels could be produced at temperatures between 120°C and 150°C and at press times greater than 4 min. He et al (2019) also studied the use of a washed CS meal to bond wood pencils, but with relative low press temperature of 40°C and very long press times (60-90 min). They report acceptable pencils that were prepared, but the long press times are a clear disadvantage.

In this work, suitable panels were produced when formulated with two different epichlorohydrin wet strength agents and at press times between 7 and 10 min. The shear bond strength of the panel specimens appeared comparable to those reported in earlier studies, and no obvious trends suggesting that strengths were affected by either wet strength agents or with lower press times were apparent. The results for the two CS protein products indicate that there is no specific improvement for the adhesive strength or water resistant properties for the water-washed CS meal, compared with the

unwashed meal. Hence, there is little added benefit associated with the additional steps used to reduce soluble carbohydrates. While this study was not specifically focused on the production of any specific interior-grade wood product, the results suggest a relatively wide degree in latitude in the choice of conditions and formulations used. Specifically herein, 7 to 10 min achieved results comparable to another bio-based adhesive (soybean).

Many of the published journals and reports shown in Cheng et al (2013, 2016, 2017) and He et al (2014a, 2014b, 2019, 2020) on glued thin veneers used relatively low press temperature (80–120°C) and relatively long press times (10–30 min). In addition to also wanting to prepare and test the properties of the plywood panels, one purpose of our work was to start trying to reduce press times, as this factor directly relates to productivity and the potential for any commercialization. Typical commercial press temperatures range from 107°C to 135°C for hardwood plywood, whereas press times generally range from 2 to 7 min (U.S. EPA 2002; Adhikari et al 2016). However, the time and temperature vary greatly depending on the wood species used, resin used, and the press design as well as panel thickness, ambient temperature, bleed through considerations, etc (U.S. EPA 2002). The choice of 7, 8.5, and 10 min press times and a 135°C press temperature in our study was based on these factors, and in this work these times produced suitable panels for interior applications as specified by ANSI/HPVA HP-1 (2016). Chen et al (2020) also recognizes the importance of press time and tested times from 2 to 6 min at a press temperature of 120°C and found they could press panels to pass the water resistance standard in as little as 4 min. This was impressive as they were preparing 25-mm thick, five-ply panels, and the inner adhesive layers were 10 mm removed from the press surfaces. The results suggest that press times can be reduced to values that would compete with many current industrial press times. Still, a better understanding of the parameter flexibility that exists in this adhesive system is warranted. By increasing press temperature, press times might be further reduced, and optimization of these factors would be useful for different combinations of board ply

number and thickness. Herein, three-ply panels were made, which were very similar in thickness to those used in commercial and industrial production.

One difference of note between our work and the Chen et al (2020) work is the use of basic or alkaline conditions in the formulations. During initial work for our study, hydrated lime was added to the CS formulations to increase the pH, as is common in reported formulations of soybean-based adhesives (Lambuth 2003), but difficulties were found in preparing and testing the panels, including viscosity issues, plywood panels off-gassing (or burping) during pressing, and frequent delamination after water soaking tests. Given that a base was not used in many of the earlier reports with CS proteins, it was not included in the final formulations for this study. In contrast, Chen et al (2020) reported added sodium hydroxide to their formulations. In our work, tested acceptable panels were prepared at or above pH 11.0, which would suggest that some base addition was necessary. Additionally, the effect of pH was found to be quite variable in a study of the adhesive performance of CS protein fractions used to glue thin maple (*Acer* spp. L.) veneers (He et al. 2020). It is not clear why these differences have been observed in numerous reports. Like most proteins, CS protein is more soluble at basic conditions (eg as is used to prepared protein isolate), and the protein chains may well have more mobility in the formulations during pressing, allowing them to find favorable binding interactions with the epichlorohydrin and other components. Possible mobility of the protein chains can be compensated to a degree by the higher press temperature used in this work, as has been suggested by Li et al (2017) in testing the effect of different meal drying methods on adhesive performance.

For most literature reports on the use of CS protein as a wood adhesive, the tested CS protein isolate or defatted meals were prepared in-house and were not commercial preparations. Current CS oil extraction operations results in a relatively low-protein product (~41%) that has some level of fibrous hull and linter material present. Additionally, this material generally has been exposed to

considerable heating before and after oil extraction. Both factors will likely degrade the protein's adhesive properties. Hence, some modification of the current processing practice would be needed to allow CS protein to best compete as a bio-based wood adhesive. These modifications might include better separation of the hulls from the kernel tissue and the addition of more meal desolventizer-toaster capacity to allow the heaters to run at lower temperatures. Although these are not insurmountable changes, the added costs would need to be compensated by some additional value for the resulting meal.

CONCLUSIONS

Yellow poplar hardwood plywood panels with two different PAE water resin formulations were tested. Defatted CS meal and water-washed CS meal both produced hardwood plywood panels with sufficient adhesive strength and water resistance to be used for interior applications. As panels prepared from both CS meals had comparable properties, there appeared to be little advantage of water-washing the meal to remove soluble carbohydrates and increase the protein level of the CS meal. Panels prepared in this work were made from adhesive formulation prepared without the inclusion of sodium hydroxide, which contradicts some prior literature in this area, and this difference is not yet well understood. Although some modification of the oil extraction processes used to make CS meals may be needed, CS protein appears to be able to compete with soybean protein for use as interior wood adhesives.

ACKNOWLEDGMENTS

The authors would like to thank the US Department of Agriculture (USDA), Research, Education, and Economics (REE), Agriculture Research Service (ARS), Administrative and Financial Management (AFM), Financial Management and Accounting Division (FMAD), Grants and Agreements Management Branch (GAMB), under Agreement No. 58-0204-9-164 for funding of this novel research. Any opinions, findings, conclusion, or recommendations expressed in this

publication are those of the author(s) and do not necessarily reflect the view of the USDA. The authors would also like to thank Mississippi State University (MSU), Division of Agriculture, Forestry, and Veterinarian Medicine (DAFVM), College of Forest Resources (CFR), Forest and Wildlife Research Center (FWRC), and the Department of Sustainable Bioproducts (DSB) for the extramural grant funding to conduct this research. This publication is a contribution of the FWRC at Mississippi State University. The FWRC publication number is SB 1030. Mention of trade names or commercial products in this publication is solely for the purpose of providing specific information and does not imply recommendation or endorsement by the USDA. The USDA is an equal opportunity provider and employer. In memory of Mr. George D. Miller Jr. who passed away on 12 January 2021. You will be forever missed.

REFERENCES

- Adhikari BB, Appadu P, Kisilitsin V, Chae M, Choi P, Bressler DC (2016) Enhancing the adhesive strength of a plywood adhesive developed from hydrolyzed specified risk materials. *Polymers (Basel)* 8(8):285.
- ANSI/HPVA HP-1 (2016) American National Standards Institute for Hardwood and Decorative Plywood. Hardwood Plywood and Veneer Association, New York, NY.
- APA (2020) Voluntary product standard PS 1-19 for structural plywood. Office of Standards Services, National Institute of Standards and Technology, American Plywood Association (APA)—The Engineered Wood Association, Tacoma, WA. pp. 1-80.
- ASTM (2017) D 906-17. Standard test method for strength properties of adhesives in plywood type construction in shear by tension loading. American Society for Testing and Materials, West Conshohocken, PA.
- Chen N, Huang J, Li K (2020) Investigation of a formaldehyde-free cottonseed flour-based adhesive for interior plywood. *BioRes* 15(3):5546-5557.
- Cheng HN, Dowd MK, He Z (2013) Investigation of modified cottonseed protein adhesives for wood composites. *Ind Crops Prod* 46:399-403.
- Cheng HN, Ford C, Dowd MK, He Z (2016) Use of additives to enhance the properties of cottonseed protein as wood adhesives. *Int J Adhes Adhes* 68:156-160.
- Cheng HN, Ford C, Dowd MK, He Z (2017) Effect of phosphorus-containing additives on soy and cottonseed protein as wood adhesives. *Int J Adhes Adhes* 77:51-57.
- Conner AH (1996) Urea-formaldehyde adhesives resins. Pages 8496-8501 in JC Salamone, ed. *Polymeric materials encyclopedia*. CRC Press Publishing, Boca Raton, FL.

- Dodds D, Pieralisi BK (2021) Cotton. Mississippi State University Extension, Agriculture, Crops, Cotton. <http://extension.msstate.edu/agriculture/crops/cotton> (19 July 2021).
- Dowd MK, Pelitire SM, Delhom CD (2018) Seed-fiber ratio, seed index, and seed tissue and compositional properties of current cotton cultivars. *J Cotton Sci* 22(1):60-74.
- Dunky M (2003) Adhesives in the wood industry. Pages 872-941 in A Pizza and KL Mittal, eds. *Handbook of adhesive technology*, 2nd edition. Marcel Dekker, New York, NY.
- Galber H, Dike TW (1942) Method of making laminated products. US Patent 2,291,586.
- He Z, Chapital DC, Cheng HN, Dowd MK (2014a) Comparison of adhesive properties of water- and phosphate buffer-washed cottonseed meals with cottonseed protein isolate on maple and poplar veneers. *Int J Adhes Adhes* 50:102-106.
- He Z, Cheng HN, Chapital DC, Dowd MK (2014b) Sequential fractionation of cottonseed meal to improve its wood adhesive properties. *J Am Oil Chem Soc* 91(1):151-158.
- He Z, Cheng HN, Chapital DC, Klasson KT, Ford C, Barroso VAB (2019) Optimization and practical application of cottonseed meal-based wood adhesive formulations for small wood item bonding. *Int J Adhes Adhes* 95:102448.
- He Z, Cheng HN, Nam S (2020) Comparison of the wood bonding performance of water- and alkali-soluble cottonseed protein fractions. *J Adhes Sci Technol* 35(14):1500-1517.
- Hodgson AT, Beal D, McIlvaine JER (2002) Sources of formaldehyde, other aldehydes and terpenes in a new manufactured house. *Indoor Air* 12(4):235-242.
- Hogan JT, Arthur JC Jr. (1951a) Preparation and utilization of cottonseed meal glue for plywood. *J Am Oil Chem Soc* 28(1):20-23.
- Hogan JT, Arthur JC Jr. (1951b) Cottonseed and peanut meal glues: Performance of plywood glue joints as determined by interior and exterior accelerated cyclic service tests. *J Am Oil Chem Soc* 28(6):272-274.
- Hogan JT, Arthur JC Jr. (1952) Cottonseed and protein meal glues. Resistance of plywood bonds to chemical reagents. *J Am Oil Chem Soc* 29(1):16-18.
- Irby JT (2021) Soybeans. Mississippi State University Extension, Agriculture, Crops, Soybeans. <http://extension.msstate.edu/agriculture/crops/soybeans> (19 July 2021).
- Johnson O (1935) Glue and process for manufacturing the same. US Patent 1,992,867.
- Kumar R, Choudhary V, Mishra S, Varma IK, Mattiason B (2002) Adhesives and plastics based on soy protein products. *Ind Crops Prod* 16(3):155-172.
- Lambuth AL (2003) Protein adhesives for wood. Pages 457-477 in A Pizzi and KL Mittal, eds. *Handbook of adhesive technology*, 2nd edition. Marcel Dekker, New York, NY.
- Li J, Li X, Li J, Gao Q (2015) Investigating the use of peanut meal: A potential new resource for wood adhesives. *RSC Adv* 98(5):80136-80141.
- Li J, Pradyawong S, He Z, Sun XS, Wang D, Cheng NH, Zhong J (2019) Assessment and application of phosphorus/calcium-cottonseed protein adhesive for plywood production. *J Clean Prod* 229:454-462.
- Li K (2012) Formaldehyde-free lignocellulosic adhesive and composites made from the adhesives. US Patent 8,268,102.
- Li N, Prodyawong S, He Z, Sun XS, Wang D (2017) Effect of drying methods on the physiological properties and adhesion performance of water-washed cottonseed meal. *Ind Crops Prod* 109(10):281-287.
- Li X, Li J, Li K, Gao Q, Li J (2018) A novel eco-friendly blood meal-based bio-adhesive: Preparation and performance. *J Polym Environ* 26(2):607-615.
- Liteplo RG, Beauchamp R, Meek ME, Chénier R (2002) Concise international chemical assessment document 40: Formaldehyde. United Nations Environment Programme, International Labour Organization, and World Health Organization, Geneva, Switzerland. p. 81. <https://apps.who.int/iris/handle/10665/42430> (19 July 2021).
- Malin N (2005) Columbia forest products launches a revolution in plywood adhesives. *Environ. Building News*, June 1, 2005. <https://www.buildinggreen.com/product-review/columbia-forest-products-launches-revolution-plywood-adhesives> (19 July 2021).
- Marra AA (1992) *Technology of wood bonding principles in practice*, 1st edition. Van Nostrand Reinhold, Springer, New York, NY. 309 pp. ISBN: 0442007973.
- Osgood G (1926) Glue. US Patent 1,601,507.
- Pizzi A (2006) Recent development in eco-efficient bio-based adhesives for wood bonding: opportunities and issues. *J Adhes Sci Technol* 20(8):829-846.
- Santoni I, Pizzo B (2013) Evaluation of alternative vegetable proteins as wood adhesives. *Ind Crops Prod* 45:148-154.
- SAS Institute (2013) SAS Version 9.4. SAS Institute, Cary, NC.
- Trischler J, Sandberg D, Dorn M (2018) Effect of temperature during vital gluten adhesive preparation and application on shear-bond strength. *J Adhes Sci Technol* 32(4):448-455.
- U.S. EPA (2002) *Plywood manufacturing. Air emissions factors and quantification/AP-42, 5th edition, Vol. 1, Chap. 10: Plywood and composite wood products/wood products industry, Section 10.5 Plywood Manufacturing*. Research Triangle Park, NC. <https://www.epa.gov/air-emissions-factors-and-quantification/ap-42-fifth-edition-volume-i-chapter-10-wood-products-0> and <https://www.epa.gov/sites/production/files/2020-10/documents/c10s05.pdf> (19 July 2021).
- White DJ (1943) Seed-meal glue. US Patent 2,312,056.
- Xi X, Pizzi A, Gerardin C, Liao Amirou JS, Abdalla A (2021) Glutaraldehyde-wheat gluten protein adhesives for wood bonding. *J Adhes* 97(1):88-100.