Soy-Flour Adhesive Bonding of Low-Moisture Hickory and Red Oak Veneer

Cody J. Wykle
Audrey Zink-Sharp

Abstract

The goal of this research was to test the effect of low-moisture-content veneer on the strength and durability of plywood test specimens constructed with a soy-flour adhesive. Soy-flour adhesive systems offer certain environmental and health advantages but research studies and performance data are lacking currently for a variety of wood types and end uses. Soyad adhesive was used in this study due to its natural, renewable soy flour, a novel cross-linking resin, and lack of added formaldehyde. Test specimens were prepared using heartwood of hickory and red oak and sapwood of hickory. These wood types were used to represent some of the most challenging wood adhesion conditions. Analytical tests included determination of select chemical properties of the adhesive and wood veneer, measurement of strength properties of the adhesive bond, and assessment of delamination tendencies of bonded panels following water soaking.

Results indicate that moisture levels and the different growth regions and wood types had an inconsistent effect on the bond strengths yet percentage of wood failure was uniformly low and at levels considered unacceptable by industry. No statistically significant or consistent trends emerged for the hickory heartwood or sapwood dry and water-soaked/redried shear strength, percentage of wood failure, or delamination tests. The most consistent differences for hickory heartwood and sapwood occurred in buffering capacity.

Production of wood-based panel products has been growing in all regions globally and was reported to be 367 million m^3 in the most recent FAO database (FAO 2021), which is just a few percentage points short of the record high of 408 million m^3 in 2018. Soy-flour adhesives have existed for decades, but previous formulations included added formaldehyde and exhibited very low resistance to delamination and high susceptibility to mold in the presence of moisture (Frihart et al. 2014). Currently, soy-flour adhesives with no added formaldehyde are used in hardwood plywood interior applications. Although these natural, no-added-formaldehyde soy adhesives offer many advantages, research studies and performance data are currently lacking for a variety of wood types, applications, and end use conditions. More information about adhesive bonding of wood using soy-based adhesives will help optimize the systems and provide technological advancements that minimize potential performance complications. This study evaluated dry and water-soaked/redried shear strength and percentage of wood failure of plywood test panels constructed with a soy-flour adhesive and low-moisture-content hickory and red oak veneer. These tests and specimen configurations provide information about engineered wood flooring applications and the influence of a range of low moisture contents typical of levels achieved during rapid veneer drying in plywood production. The wood types included in this study are very common in engineered wood flooring and represent a difficult adhesive bonding situation with hickory compared to red oak, which is reported to be easier to bond (F. Carter, Columbia Forest Products. personal communication, November 2018).

Increasing materials costs, interest in sustainability, and concerns resulting from the reclassification of formaldehyde as carcinogenic to humans has reenergized interest in bio-based adhesives. Soy flour used in adhesives is deemed suitable for modern industrial use due to high production volumes, lower cost compared to other adhesive formulations, ease of processing, and a reduced need for added formaldehyde (Frihart et al. 2014, Vnučec et al. 2016). However, different manufacturing processes such as higher hot-pressing temperatures and longer press times have been required to achieve good bonding strengths and high water resistance (Li et al. 2014, Vnučec et al. 2016). An example of soy flour use in wood composite products is the recent US

The authors are, respectively, Former Graduate Research Assistant and Professor and Associate Department Head (agzink@vt.edu [corresponding author]), Department of Sustainable Biomaterials, 230 Cheatham Hall, Virginia Polytechnic Institute and State University, Blacksburg, Virginia 24061. This paper was received for publication in July 2022. Article no. 22-00044.

Forest Prod. J. 73(1):13–23.
doi:10.13073/FPJ-D-22-00044

patent approved as US 10,266,694 B2for Via et al. (2019) that allows replacement of petroleum-based adhesives traditionally used to manufacture particleboard commonly used for indoor furniture. In the procedure described by Via et al. (2019), soy flour is heated until denatured and then urea is added to the denatured soy flour.

Soy flours are used to create adhesives due to their high protein content compared to other vegetable proteins, but as currently formulated, they have low moisture resistance. Soy-based adhesives are formulated with a natural soy flour that is the byproduct of soybean oil production and is a renewable adhesive feedstock with advantages and disadvantages relative to fossil fuel–based adhesives (Frihart et al. 2014). An extensive review of the technological performance of several formaldehyde-free wood-based composites is found in Solt et al. (2019). One major issue that has slowed application in certain products is the low water resistance of the adhesive bond. Research is currently focusing on investigating improvement of bond strength and water resistance through the addition of cross-linking agents. Cross-linking improves bonding properties and water resistance of soybean proteins by introducing a curing agent to cross-link the soybean protein molecules into an insoluble three-dimensional network and create a faster protein-based adhesive system (Fan et al. 2016, Li et al. 2019, Solt et al. 2019). Addition of lignin has been shown to improve water resistance through creation of more cross-linking networks (Xiao et al. 2013). Lignin-based resin combined with polyamidoamine-epichlorohydrin (PAE) was determined to form a more effective adhesive for plywood (Luo et al. 2015, Zhang et al. 2017). Wet and dry strength improved considerably because the lignin-based resin could penetrate into the wood for effective interlocking as well as forming a denser cross-linking network with the soy flour. Zhang et al. (2017) determined that PAE improved the wet bond strength, whereas the lignin-based resin improved the dry bond strength when compared to results obtained with just the soy flour–based adhesive with no modifiers. Different cross-linking densities when cross-linked by epichlorohydrin-modified polyamide were found by Li et al. (2019) for three typical soybean meal products including low-temperature soybean flour (also known as defatted soybean flake), high-temperature soybean flour, and physical soybean flour. The low-temperature soybean meal had the most promise due to a greater number of reactive groups, higher cross-linking densities, and superior bond strengths. Only the low-temperature adhesive exhibited the needed water resistance ($>$0.8 MPa) (Li et al. 2019). A comparison of plywood panels prepared with defatted cottonseed and water-washed cottonseed meals to those made with a commercial soybean meal indicated that shear strengths were comparable for all panels but the soybean meal panels produced acceptable panels only when a PAE wet strength agent was added to the formulation (Shmulsky et al. 2021).

Soy flour substitution in wood adhesives can provide significant cost savings without adversely affecting adhesion or product performance (Cheng et al. 2019, Via et al. 2019). Chen et al. (2019) report that soy flour substitution can lower total adhesive costs by 13 percent compared to a polymeric methylene diphenyl diisocyante (pMDI) adhesive used in particleboard without degrading board properties with up to 20 percent substitution. Partial substitution with soy flour has been reported to increase cold tack of the resin (Asafu-Adjaye et al. 2020). Cold tack is an important consideration in minimization of distortion of a wood mat prior to insertion into a press. Soy-flour adhesives do not usually exhibit sticky tack properties seen with other wood-bonding adhesives but resemble wet plaster or cement in their flow characteristics. Asafu-Adjaye et al. (2022) determined that cold tack of pMDI increased with partial substitution with soy flour to the level obtained with urea formaldehyde resins while providing significant cost reduction. However, a reduction in platen sticking during hot pressing was found when adding soy flour to the given amount of pMDI as well as when replacing part of the pMDI by soy flour; both addition or replacement were up to 30 percent of the amount of pMDI, and the stronger effect was given with the replacement (Asafu-Adjaye et al. 2022). The reduction in hot tack due to inclusion of soy flour substantially reduced platen sticking during hot pressing of particle mats and decreased the tendency of resonated particles to transfer and stick to the press platens.

Hickory (*Carya* spp.) wood was used for this study due to performance problems reported by industry colleagues with hickory engineered wood flooring bonded with soy-flour adhesive. There are several species included in the *Carya* genus, common name hickory, including *C. ovata, C. laciniosa, C. glabra,* and *C. tomentosa*. Hickory is a ring-porous hardwood in which fiber tracheids range from thin- to thick-walled. Tyloses in hickory are moderately abundant. The low percentage of vessels accompanied by the high percentage of thick fiber cell walls and low lumen volume in hickory can make adhesive penetration difficult and this, in turn, can further limit mechanical interlocking of adhesives to one or two cells deep. Higher-density woods such as hickory exhibit large stresses as they change dimensions with changes in moisture content and the large stresses can also contribute to poor bond performance. Although hickory was the primary focus of this research, red oak (*Quercus* spp.) wood was also used as a face veneer to compare to the adhesion with hickory. Red oak is reported by industry to bond well with soy-flour adhesives (F. Carter, F. Carter, Columbia Forest Products. personal communication, November 2018). Several species are included in the *Quercus* genus, common name oak: *Q. rubra, Q. velutina, Q. shumardii, Q. coccinea, Q. palustris,* and *Q. phellos*. Red oak is a ring-porous hardwood in which fiber tracheids and libriform fibers are medium-thick to thick-walled. Tyloses are absent or sparse. Yellow-poplar (*Liriodendron tulipifera*) wood was used for the core of the three-ply plywood tested in this study. Engineered wood flooring is made with a lower-density wood such as yellow-poplar or some type of particle or fiber composite as the core. Yellow-poplar is a diffuse porous hardwood with fiber tracheids varying from thin-walled to moderately thick-walled. Tyloses are absent or sparse. Shrinkage expressed as a percentage of the green dimension from green to ovendry moisture content for hickory averages 7.4 percent in the radial direction and 11.4 percent in the tangential direction (Forest Products Laboratory 2021). Shrinkage values of red oak average 4.4 percent in the radial direction and 10.1 percent in the tangential direction and for yellow-poplar, 4.6 percent in the radial direction and 8.2 percent in the tangential direction (Forest Products Laboratory 2021).

Drying of wood veneer for use in plywood products is required to make the veneer suitable for adhesive bonding. Industrial practice is to remove the moisture in veneer as rapidly as possible using continuous-type, high-temperature (>100°C) conveyer driers (Irle et al. 2013). Drying can deactivate the veneer surfaces, which can impair adhesive wetting and can have negative effects on the bond-line performance. This effect is the more pronounced the lower the final moisture content is. When wood is subjected to high temperatures in veneer drying, extractives found in the heartwood of the wood materials migrate to the surface and physically block adhesive and reduce the surface wettability (Kumar and Pizzi 2019). The drying process can also affect the chemical nature of the extractives. High temperatures imposed during drying may convert hydrophilic extractives into hydrophobic substances (Kumar and Pizzi 2019) and consequently, migration of extractives to the surface may result in thermal deactivation of the surface (Roffael 2016); the extractives migrate to the wood surface in water and form what is a thin or weak boundary layer (Gao 2010, Kumar and Pizzi 2019). Extractives can affect the pH or buffering capacity, which also affects curing and setting of the adhesive system (Roffael 2016, Bockel et al. 2019, Kumar and Pizzi 2019) and adhesive spread and penetration (Mirabile and Zink-Sharp 2017). With a deactivated surface, a water-based adhesive such as Soyad might not readily penetrate the surface and this in turn might cause the bond line to have a slower cure due to the excessive water at the bond line. In addition, the amount of moisture in wood can greatly influence the wetting, flow, penetration, and cure of aqueous wood adhesives (Dunky et al. 2002, Frihart and Beecher 2016, Hänsel et al. 2021) and the machining of the raw materials (Baharoğlu et al. 2012). With low-moisture-content wood adherends, the dry wood can absorb water from the adhesive so quickly that adhesive flow and penetration into the bulk becomes inhibited due to an increase in solids content on the surface and reduction of solvent at the bonding interfaces (Frihart 2013).

Materials and Methods

Test materials preparation

The adhesive used in this research was Soyad adhesive, a commercially available adhesive produced by Solenis. Soyad was of particular interest due to the use of natural, renewable soy flour, a novel cross-linking resin, and lack of added formaldehyde. Soyad is a water-based thermoset adhesive formulated with a proprietary cross-linking resin (a PAE) with a solids content of 45 to 60 percent and a pH value of 5.5 (Birkeland et al. 2010). Soyad is used to construct decorative plywood, particleboard, medium-density fiberboard, and engineered wood flooring. Fresh batches of adhesive were prepared according to manufacturer's instructions just prior to bonding of the plywood test panels. The exact formulation is proprietary, but in general terms, the adhesive is made of soy flour, water, a proprietary cross-linking PAE resin, a defoaming agent, and a pH modifier. The adhesive was prepared by first mixing the water, PAE, and defoamer with a fan-blade mixer. Then one half of the soy flour was added, followed by the pH modifier, followed by the other half of the soy flour. The components were mixed for 5 minutes with the fan blade at 1,000 rpm. The final step was to check the pH of the adhesive batch to ensure it matched the industry recommended pH of 5.5.

Three-ply test panels were constructed using hickory heartwood, hickory sapwood, red oak heartwood, and yellow-poplar core veneer. Radial sheet average thickness of the hickory and red oak veneer was either 0.62 mm (hereafter referred to as ''thin'') or 2.06 mm (''thick''). All yellow-poplar veneer averaged 2.06 mm thick. Veneer sheets were sized to 15.24 by 15.24 cm (6 by 6 in) and moisture conditioned to 2, 4, 6, or 8 percent moisture content using a Russells Technical Products GD-8-105 moisture and humidity chamber prior to further processing. This range of moisture content approximates the range wood veneer obtains upon exit from the veneer dryers. After complete equilibration at the respective moisture contents, a few sheets were subjected to oven-drying at 103°C for 24 hours for moisture content determination.

Prior to application of the adhesive, veneer sheets were hand-sanded using 220 grit sandpaper with three passes forward and three passes back until the full veneer face was sanded. Sawdust particles were blown from the surfaces with compressed nitrogen gas. Veneer sheets were stored in the moisture conditioning chamber when not being processed to minimize any moisture content deviations from the final target moisture content. Without an extensive chemical pretreatment of the veneer, better bonding conditions were not possible. Although the best method for preparing wood surfaces for adhesive bonding is to use sharp planer blades, hand-sanding is reported ''acceptable'' in the absence of sharp planer blades because it causes less damage to the cells at and near the surfaces (Frihart 2013, Cool and Hernandez 2011). Since the thermal history of the veneer provided to us by industry was unavailable, and because it was not practical to plane the surfaces due to the thickness of the veneer, bonding surfaces were hand-sanded just prior to adhesive application to achieve the best possible bonding. Sanding also standardized the specimen preparation technique to provide consistency in surface preparation procedures.

Three-ply plywood panels were prepared with veneer sheets conditioned to the respective moisture contents (2%, 4%, 6%, or 8%). Face plys were hickory heartwood, hickory sapwood, or red oak heartwood. All core plys were yellow-poplar. Hickory heartwood and sapwood were used to explore any bonding differences that might manifest with the soy-flour adhesive due to the extractive accumulation in the heartwood. Plywood panels were prepared according to specifications provided for the Soyad adhesive. The adhesive was applied to the inner surfaces of the face veneer using a soft rubber roller at the rate of 200 g/m^2 per veneer face for a three-ply lay-up. After laying the three plys, the panel was put under a 2-kg weight for 10 minutes of stand time. The panels were then taken to a cold press for 5 minutes and 690 kPa of pressure. Panels were then placed in the laboratory Carver press for 3 minutes at 116°C and 1,034 kPa. Panels designated for the dry shear test were conditioned to 9 percent moisture content and those designated for the water-soak shear test and the water-cycling delamination tests were conditioned to 6 percent moisture content to coordinate with industrial testing practices (F. Carter, Columbia Forest Products, personal communication, November 2018).

Analytical methods

Methods used for characterization and comparison of bonding low-moisture hickory heartwood and sapwood were selected to evaluate the adhesive or veneer properties, bond shear strength, and bond durability. The overall analytical structure is shown in Table 1. Statistical significance of the test variables was evaluated using a 1-way analysis of

Table 1.—Test methods, materials, and analytical methods structure.[a]

Test completed	Technique	Materials used	No. of tests or specimens
Adhesive and veneer properties			
Adhesive viscosity	Parallel plate rheology; varying shear rates	Mixed adhesive	35 data points for each up and each down ramp, 5 samples
Veneer buffering capacity	Titration with added acid or base	Veneer ground into wood powder	4 per wood type (HS, HH, and ROH)
Bond shear strength and percentage of wood failure			
Dry strength	ASTM D906-98 (ASTM 2017)	Specimens cut from plywood panels and notched	15–23 per HSO, HSC, HHO, and HHC at each MC
Water-soaked/redried strength	Immersion in water	Specimens cut from plywood panels and notched	18 HSO, 18 HSC, 18 HHO, 18 HHC; 6% MC
Percentage of wood failure	ASTM D5266-13 (ASTM 2013)	ASTM D906-98 specimens	15–23 per HSO, HSC, HHO, and HHC at each MC
Bond durability			
Delamination	ANSI/HPVA HP-1-2020 (3-cycle soak test)	Specimens cut from plywood panels	18 HH; 18 HS; 18 ROH

[a] HH = hickory heartwood; HS = hickory sapwood; ROH = red oak heartwood; O = lathe check pulled open; C = lathe check pulled closed; MC = moisture content.

variance (ANOVA) comparison and a Tukey's honestly significant difference test at $\alpha = 0.05$ for comparisons using JMP software. Statistical analyses were completed within and across each moisture content and according to heartwood or sapwood to determine significant differences.

Viscosity measurements of the Soyad were completed to provide a description of the mixture's flow characteristics since adhesive viscosity has been shown to be a key factor during application, open time, closed time, and pressing (Frihart 2013). Viscosity of the prepared adhesive was assessed using a TA Instruments Advanced Rheometer AR-2000 and parallel plate rheology. A 25-mm steel plate was used with a gap of 1 mm. The process involved a ramp up in shear rate followed by a ramp down from 0.005 to 500 1/s. Thirty-five points were taken for each up and down cycle for a total of 70 data points for one adhesive sample. This process was repeated for five different adhesive samples. A shear stress versus shear rate chart through several up and down ramps is shown in Figure 1 for the Soyad adhesive. A change in the shear stress behavior near the 10 cycles per second shear rate indicates that the liquid structures in the adhesive have likely been modified at higher shear rates. A temperature profile performed when testing viscosity to monitor the decrease in viscosity with increasing temperature in the bond line before the curing reaction starts might provide more information. The behavior of various adhesive mixtures at high temperatures could explain different test results for test panels. With a higher temperature in a bond line before the curing process starts, different penetration behavior can occur which can have an effect on bonding performance.

Buffering capacities of the woods used in this study were determined using a Metrohm 905 Titrando instrument. Wood veneer specimens were ground into powders and evaluated with the 905 Titrando set to measure pH and assess buffering capacity. A sample of approximately 0.5 g was placed into a four-neck, round-bottom flask to which 5 mL of 0.06 M NaCl and 300 mL of distilled water were added. The mixtures were titrated to pH 3 with 0.02 N HCl for alkalinity and to pH 10 with 0.02 N NaOH for acidity. The acid or base was added at 0.02 mL every 2 seconds until the desired pH was reached for assessment of the buffering capacity. Two replicates were tested for each wood studied.

Dry shear strength was determined following procedures in ASTM D906-98 (reapproved in 2017) (ASTM 2017). Test specimens were cut from the plywood panels into 82.6-mm-long by 25.4-mm-wide strips with cut inlays being two-thirds of the way through the core layer, which created a shear area of 25.4 by 25.4 mm. Specimen configurations are shown in Figure 2. Bond shear strength tests were conducted using a MTS Sintech10GLtest frame, with a cross head speed of 0.508 mm/min. Shear strength after water soaking and drying was determined from specimens configured the same as the ASTM D906-98 specimens and included the first water immersion step in pretreatment described in EN 314-1 (BSI 2004). Test specimens were immersed in a water bath at 20°C ± 3°C for 24 hours and subsequently allowed to dry at room temperature for 48 hours. The water-soaked/redried specimens were then tested using the MTS Sintech10GL test frame with a cross head speed of 0.508 mm/min. In accordance with specifications in ASTM D906, half of the specimens were tested so that the lathe checks were being pulled closed, and the other half so that the lathe checks were being pulled open as illustrated in Figure 2.

Percentage of wood failure was assessed following ASTM D5266-13 (ASTM 2013). This process involves a visual approximation of the amount of wood remaining on the bond line of the failed dry and water-soaked/redried shear test specimens according to ASTM D5266-13 specifications and instructions (ASTM 2013). To minimize error and inconsistencies with visual estimation, a technique was developed in which one person performed all measurements and specimen surfaces were viewed with a magnification lens of ×10 connected to a lamp with a 60W bulb for clear and consistent lighting. A 25.4 by 25.4-mm grid overlay printed on copier transparency film was created and placed on the surface of the failed test specimens as illustrated in Figure 3. The grid overlay had lines that divided the grid area into 25 equal-size squares and completely covered the entire area of delamination. The area within each blocked square on the overlay grid was evaluated for percentage of the amount of wood failure as defined in ASTM D5266-13, and then all measurements were combined for a final evaluation of the percentage of wood failure for each specimen.

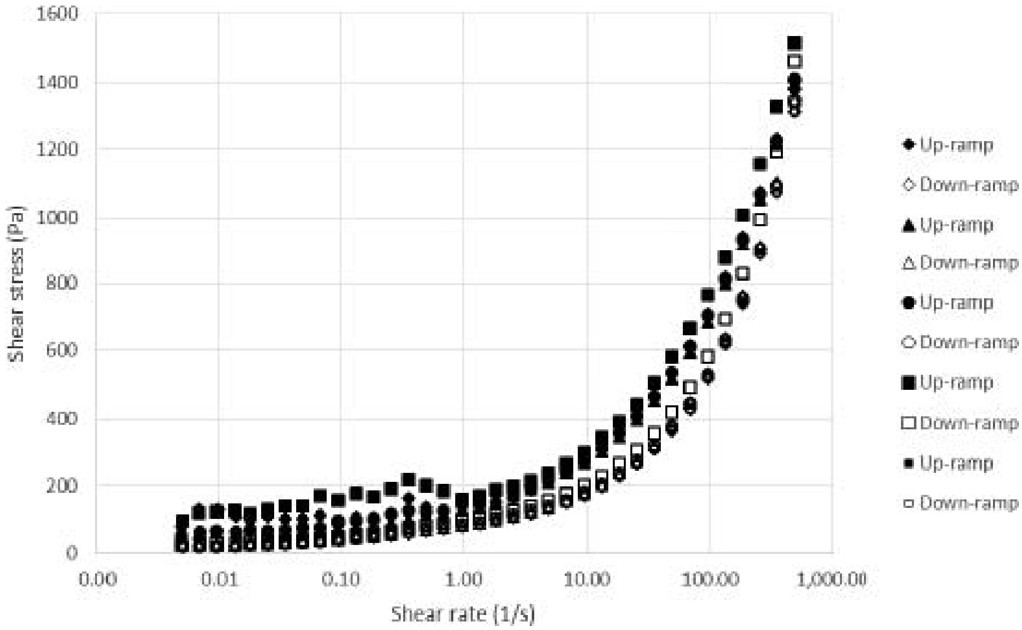

Figure 1.—Shear stress versus shear rate for Soyad adhesive, five measurements.

Bond durability testing followed procedures outlined in the ANSI three-cycle soak test (ANSI 2020). Test specimens were sized from the panels to 12.7 by 5.08 cm without any cut inlays made. The prepared test specimens were submerged in water which was 24°C ± 3°C and held there for 4 hours. Specimens were then immediately dried in an oven at a temperature of 50°C for 19 hours. The soak–dry treatment was repeated for a total of three cycles with all testing groups. At the end of the third cycle, delamination was evaluated using a feeler gauge that was 0.08 mm thick and 12.7 mm wide. According to the standard specifications, a specimen was labeled as failed when delamination between two plys was greater than 50.8 mm in continuous length, over 6.4 mm in depth at any point, and greater than 0.08 mm in width.

Results and Discussion

Buffering capacity of a material is a measure of the ability to resist changes in pH (Skoog and West 2013). With wood materials, buffering capacity is quite variable and can have different values across species, as well as within the same tree, at different heights in the tree, and depending on the age of the tree (Hernández 2013). Variations in buffering capacity can have implications for curing and gelation times for adhesives when applied to wood materials because adhesives have narrow pH ranges in which they function effectively (Johns and Niazi 1980, Wang et al. 2010). When an acid or base is added to a slurry or wood flour, the effect on pH change can be large or small, depending on the initial pH and the capacity of the wood materials to resist change in pH (pH is measured on a logarithmic scale). In this study, a titration method was used in which a known volume and concentration of a base or an acid was added to the ground wood being assessed. Buffering capacities obtained for the hickory and red oak powders are shown in Table 2. Standard deviations are shown in parentheses. Alkaline-buffering capacity refers to the volume of acid added and the acid-buffering capacity refers to the amount of base added. As seen in Table 2, the amount of acid required to modify the pH was higher than the amount of base needed for all three woods examined but differences were found in the degree of buffering capacity for the various test specimens. Hickory heartwood alkaline and acid buffering was greater than that of hickory sapwood and red oak heartwood. The acid-

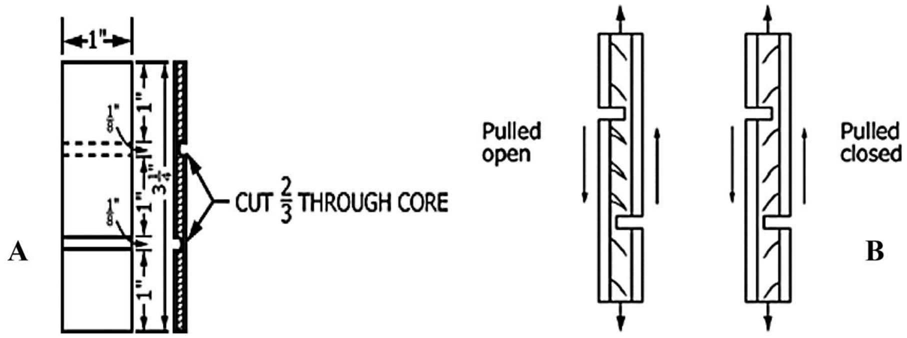

Figure 2.—Specimen configurations for ASTM D906-98. (A) Form and dimension (US Customary Units are shown in the Standard). (B) Lathe and notch orientations (ASTM D906-98) (ASTM 2017).

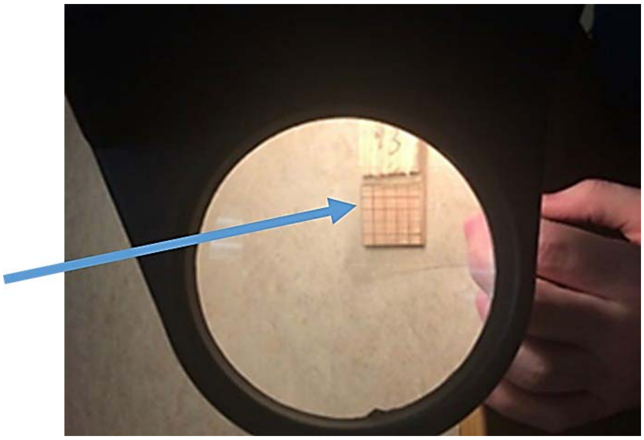

Figure 3.—Example of grid (at arrow) positioned on failed D906-98 test specimen viewed with lighted magnifying lamp.

buffering capacity is of less importance since Soyad functions in the acid range. The alkaline-buffering capacities were determined with two replicates, which makes using a standard statistical test questionable for statistically significance but it is noticeable that alkaline-buffering capacities were different across the three wood types whereas the acid-buffering capacities were quite similar. A buffer resists changes in pH due to the addition of an acid or base though reaction with the buffer. As long as the buffer is not completely reacted, the pH will not change drastically. However, the pH change will increase (or decrease) more drastically as the buffer is depleted. If the buffering capacities of the woods studied in this project were too small or outside the values needed for adequate reactions, there was potential to interfere with adhesion due to altering the adhesive outside the preferred pH range. Significance of these findings and further testing were beyond the scope of this initial study.

Dry shear strength of the bonding with veneer at several low moisture contents and wood regions (heartwood or sapwood) was evaluated using shear-by-tension loading in accordance with ASTM D906-98 (ASTM 2017). Figures 4A and 4B illustrate average bond strength at each of the moisture contents and lathe check test configurations (Fig. 4A pulled open and Fig. 4B pulled closed). In all the figures that follow, identical capital letters indicate no statistically significant differences were found whereas different capital letters indicate there were statistically significant differences using Tukey's ANOVA, $\alpha = 0.05$. Dry shear bond strength values show that Soyad bonding with low-moisture hickory sapwood and heartwood developed reasonable dry shear strength levels for use in hardwood and decorative plywood as specified in ANSI (2004). There was no statistically significant differences found for moisture contents within the range studied, which could imply that any effect was negligible, inconclusive, or constant across this moisture range; however, further testing is needed to confirm whether the effect is measurable and repeatable.

A comparison of dry and water-soaked/redried shear strengths is shown in Figure 5. Error bars indicate ± 1 SD. It can be seen that although variation occurred when comparing sapwood and heartwood, and likewise between closed lathe check test orientations compared to open, there were no statistically significant differences found in sapwood compared to heartwood but statistical differences are noted when comparing closed configuration specimens with open configurations. It is expected that closed lathe check configurations would produce higher strength values for both the dry and water-soaked/redried shear tests since the lathe checks and the adjoining wood cells are being pulled closed in compression, which is a high-strength property for wood materials. Results provided in Figure 5 show that water soaked/redried shear strengths were less than dry strengths, but it is interesting to note that heartwood closed results for dry and water-soaked/redried shear tests were similar when taking data variability into account. Although the water-soaked/redried shear strength results are lower than dry strength in general, the magnitude of the values indicate that sufficient wet strengths had been developed to withstand the intensive water-soaking treatment. Dimensional change that occurred during the wet soak created large strains and stresses at the bond lines, especially in view of the two different wood types each with characteristic shrinkage properties at the interphase of the bond lines. This dimensional change can create deterioration of the bond strength and degradation of the cross-linked adhesive structure at the bond lines, resulting in reduction in bond strength (Li et al. 2019).

Percentage of wood failure is the percentage of wood area remaining on the adherend in the fractured surface test area (ANSI 2020) and is expressed as a percentage graded from 0 to 100 percent. This measure is commonly used in industrial research and practice as an estimate of bond quality. High, or at least acceptable, percentage of wood failure values are said to indicate good adhesive bond quality because failures are within the wood bulk material rather than within the adhesive bond lines. The ASTM D5266-13 evaluation is currently accomplished through a visual assessment by a trained and experienced technician; however, digital and automated systems have been investigated in an attempt to reduce the variability in this test (Zink and Kartunova 1998, Scott et al. 2005, Yang et al. 2008, Daoui et al. 2011, Kariz

Table 2.—Buffering capacities for hickory and red oak wood. Standard deviation in parentheses.

	Hickory sapwood		Hickory heartwood		Red oak heartwood	
	Alkaline-buffering capacity (mL of 0.02 N HCl)	Acid-buffering capacity (mL of 0.02 N NaOH)	Alkaline-buffering capacity (mL of 0.02 N HCl)	Acid-buffering capacity (mL of 0.02 N NaOH)	Alkaline-buffering capacity (mL of 0.02 N HCl)	Acid-buffering capacity (mL of 0.02 N NaOH)
Initial pH average	6.17 (0.01)	6.18 (0.00)	6.41 (0.01)	6.49 (0.03)	5.96 (0.00)	5.94 (0.02)
Average volume (mL) required to reach 3 (acid) or 10 (base) pH	20.17 (0.53)	6.93 (0.76)	22.1 (0.59)	7.1 (0.69)	18.77 (0.46)	6.90 (0.86)

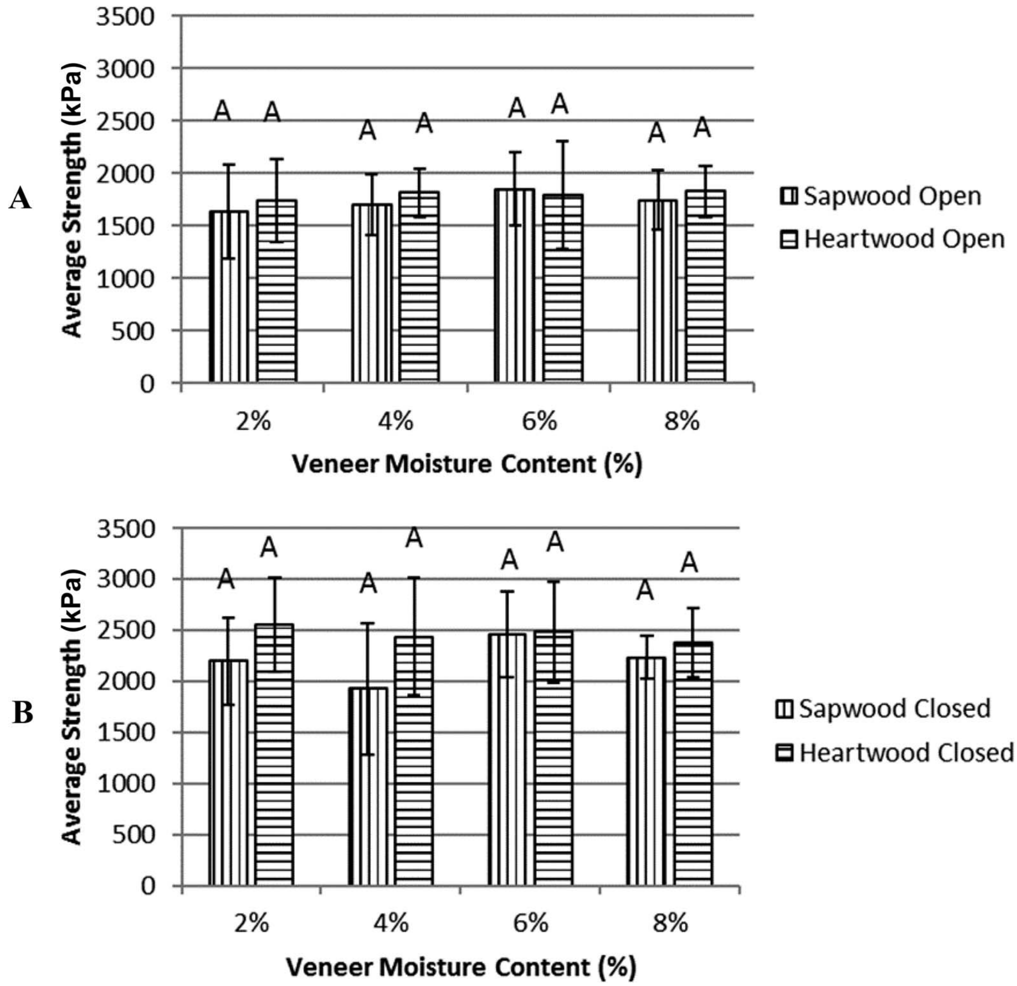

Figure 4.—Average dry shear strength, core lathe check: (A) open orientation, (B) closed orientation.

and Sernek 2014, Lin et al. 2015, Kim and Park 2021, Alade et al. 2022). The grid method developed for this study provided a reliable and repeatable way to increase accuracy and reduce error by measuring detail within small units and then combining that information into the final estimate rather than evaluating the entire surface area as one unit. Figures 6A and 6B show percentage of wood failure results for D906-98 tests.

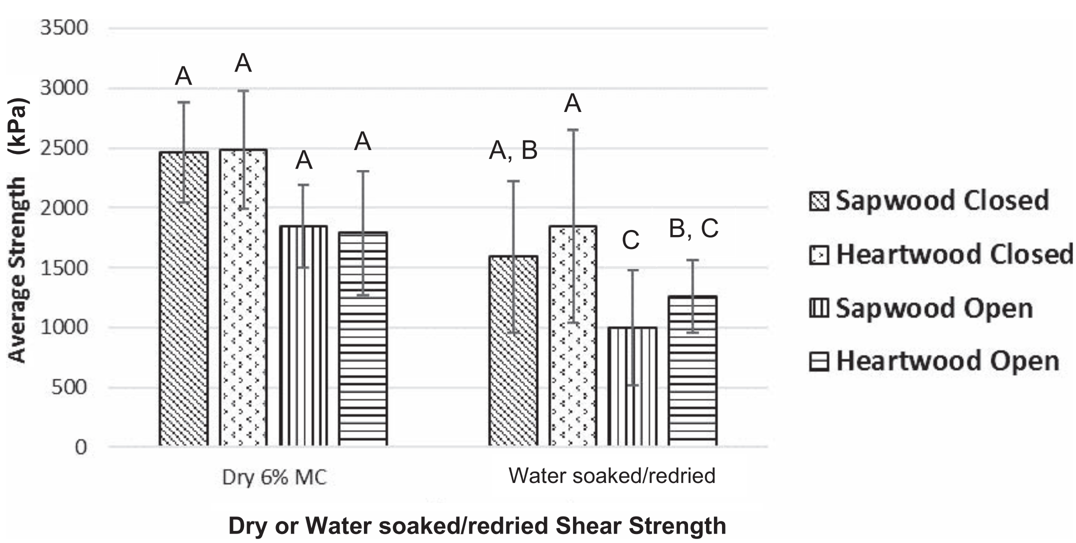

Figure 5.—Comparison of dry and water-soaked/redried shear strength results.

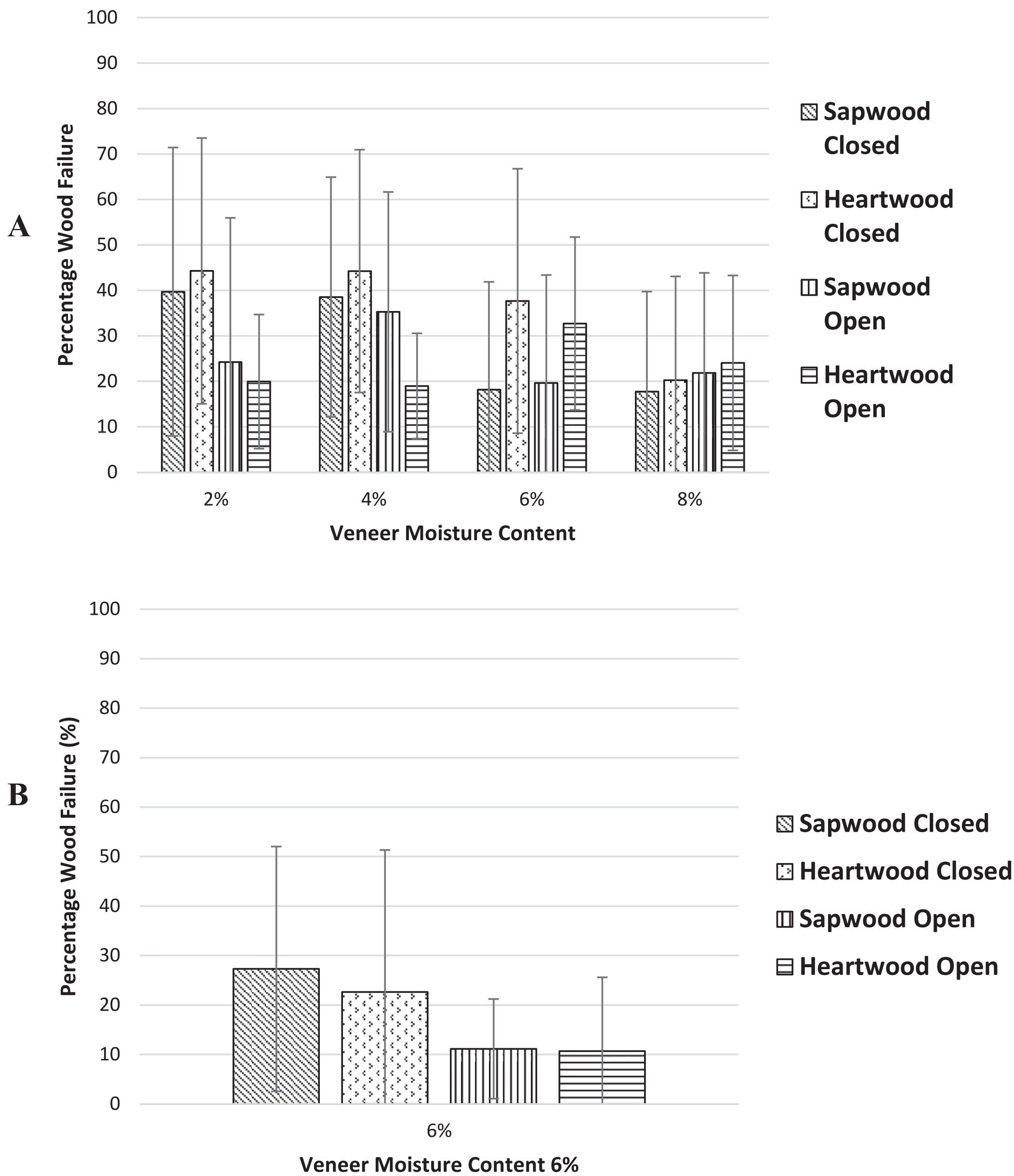

Figure 6.—Percentage of wood failure: (A) dry shear strength test specimens, (B) water-soaked/redried shear test specimens.

High strength and durable bonds have been said to correlate well with high wood failure percentages and, correspondingly, if wood failure is shallow and sparse, bond strength and durability are said to be lacking (Frihart 2013, Kariz and Sernek 2014). Standards indicate that minimum acceptable values for percentage of wood failure vary from 15 percent in some standards to above 85 percent in others, depending on the bond line requirements for a particular product type. For example, wood failure percentage requirements for hardwood and decorative plywood indicate that for technical and Type I plywood bond lines the test piece average must be 50 percent for average failure load less than 1,724 kPa, 30 percent for failure loads between 1,724 and 2,413 kPa, and 15 percent for failing loads over 2,413 KPa (ANSI/HPVA HP-1-2020). In plywood standard EN 314-2 (BSI 1993) the mean apparent cohesive wood failure percentage requirement is 80 percent for mean shear strengths from 200 kPa ti 400 kPa, 60 percent for mean shear strength between 400 kPa and 600 kPa, and 40 percent for values from 600 kPa to 1,000 kPa. There is no mean wood failure percentage requirement for mean shear strength values greater than 1,000 kPa. Data variability can be quite high in this test for certain adhesives due to difficulty in visual assessment of contrast differences between nearly transparent adhesives and the wood background. Nevertheless, this method is commonly used and depended upon to provide a qualitative way to approximate bond quality and was employed in this study at the request of our industry partner and to provide information expected and recognized by industrial laboratories. The magnitude of the standard deviations for some of the configurations in our study would indicate negative percentages of wood failure values, which are not possible, and therefore error lines were not shown beneath the zero line in Figures 6A and 6B.

In industrial situations, it is thought that percentage of wood failure is a good estimate of the bond strength and durability of a bonded wood joint. Results shown in Figures 6A and 6B indicate that the shear strength averages generally were within the 1,724 to 2,413-kPa range and therefore wood failure percentage of 30 percent is needed for acceptable results according to the ANSI/HPVA standards (ANSI 2020). Figures 6A and B show that on average the dry specimens tested in the open configuration for lathe checks did not meet the 30 percent wood failure requirements and averages for the water-soaked/redried specimens did not meet the 30 percent level. This finding indicates that high bond shear strengths were not reflected well in the wood failure percentage measurements. This may not be especially significant since it has been shown that there is less certainty in percentage of wood failure when specimens exhibit a high degree of adhesive failure and a low percentage of wood failure, especially for nearly transparent adhesives as soy flour–based adhesives (Scott et al. 2005). Causes for the almost uniformly low percentage of wood failure as shown in this study could include the low-moisture veneer that pulled water from the adhesive into the wood substrate and led to cohesive failure within the bulk of the adhesive (Frihart 2013). Additional factors could be the difficulty and high degree of variability in visually establishing wood substrate failure versus cohesive failure within the adhesive versus interphase failure due to low contrast between the wood and the transparent Soyad adhesive.

Results from the ANSI three-cycle soak test (ANSI 2020) performed using red oak and hickory veneer and two different veneer thicknesses, 2.06 mm (labeled ‘‘thick’’) and 0.62 mm (labeled ‘‘thin’’) are shown in Table 3. This test subjects the bonded specimens to water soaking and drying to assess resistance to delamination during moisture exposure. As seen in Table 3, all wood combinations passed the delamination criterion set in the standard. Although hickory heartwood thick specimens showed a lower pass rate than did all other combinations, the aggregated values all passed the standard requirements. These delamination results indicate durable bonds that withstood stresses created during water cycling were developed with all combinations of wood regions and types. It is reasonable to say that the wood and adhesive combination levels are sufficient for interior plywood because all specimens passed the water-soak tests and adequate bonding was achieved for this soy-flour adhesive system and these particular test specimens.

Of the many and varied factors involved when evaluating wood bonding for creation of a water-resistant, durable composite those that have been reported to have the most significant effect are the wood's ability to distribute stresses away from the bond line and the chemical and physical properties of the adhesive (Frihart 2009). Adhesives can be separated into two groups: in-situ polymerized and prepolymerized (Frihart and Beecher 2016) based on the way the adhesive interacts with the cell wall and the adhesive chemistry. Protein adhesives like Soyad are prepolymerized adhesives that have longer polymer chains and allow more flexibility at the bond line. The longer polymer chains with more flexibility allow for the stress from warping in the wood structure due to moisture change to be distributed through the adhesive rather than at the adhesive–wood interphase. Delamination results show that Soyad adhesive developed sufficiently durable bonds with low-moisture-content veneer of both hickory heartwood and sapwood as well as red oak heartwood.

Summary and Conclusions

Based on the findings from the study it can be said that moisture content within the range studied was not a clear factor leading to poor bonding as tested with the ASTM D906-98 dry shear test. Water-soaked/redried shear strength values were considerably lower than dry shear strengths, but still within acceptable levels for interior hardwood and decorative plywood. However, results for percentage of wood failure indicated that averages were generally below industrial acceptability levels for all moisture contents studied. The perceived low wood failure percentages might indicate that percentage of wood failure measurements did not reflect overall bond quality well.

Heartwood and sapwood chemical differences are known to have strong effects in adhesive bonding of wood, but in this study no robust or consistent trends emerged for the hickory heartwood or sapwood dry and water-soaked/redried shear strength, percentage of wood failure, or delamination tests. The most pronounced differences for hickory heartwood and sapwood occurred in buffering capacity. The significance of differing buffering capacity was not immediately clear, but buffering capacity of wood materials can interfere with resin cure. When hickory was compared to red oak the only differences found were in buffering capacities between the two wood types. This result may provide some insight into why hickory is said to not bond well with Soyad, but this concept must be further explored. Suggestions for further study include investigating the curing behavior of Soyad at different pH levels and a temperature profile experiment when testing adhesive viscosity.

The most intriguing findings from this research were the differences found in buffering capacity for hickory heartwood, hickory sapwood, and red oak heartwood and the contrast between shear strength levels that met expectations for hardwood and decorative plywood, delamination averages that were also within acceptable levels, yet percentages of wood failure that fell almost uniformly below standards levels. Results in this study have provided a broader understanding of adhesive bonding of hickory with soy-flour adhesives when veneer moisture contents were within the range of 2 to 8 percent. Expectations that low

Table 3.—Percentage of specimens with delamination passing three-cycle soak tests (n = 18).

	Thick veneer (average 2.06 mm)			Thin veneer (average 0.62 mm)		
Cycle	Hickory sapwood	Hickory heartwood	Red oak heartwood	Hickory sapwood	Hickory heartwood	Red oak heartwood
First soak	100	89	100	100	100	100
Second soak	100	89	100	100	100	100
Third soak	100	83	100	100	100	100

moisture levels might provide less favorable bonding of hickory and expose problems with heartwood versus sapwood were not confirmed with the combinations of variables and analytical methods used in this study. This important finding indicates soy-flour adhesives can potentially provide adequate bonding across a range of wood types, moisture levels, and radial locations in the tree stem.

Acknowledgments

Authors would like to acknowledge financial and technical support from the Wood-based Composites Center, a National Science Foundation Industry/University Cooperative Research Center (award 1035009), the Department of Sustainable Biomaterials at Virginia Tech, and Columbia Forest Products.

Literature Cited

Alade, A. A., Z. Naghizadeh, and C. B. Wessels. 2022. A new method for estimating wood failure percentage in adhesive-bonded shear specimens. *Int. J. Adhes. Adhes*. 112: 103028. https://doi.org/10.1016/j.ijadhadh.2021.103028

American National Standard Institute (ANSI). 2004. American national standard for hardwood and decorative plywood. ANSI/HPVA HP-1-2004. Hardwood Plywood and Veneer Association, Reston, VA.

American National Standard Institute (ANSI). 2020. American national standard for hardwood and decorative plywood. ANSI/HPVA HP-1-2020. Hardwood Plywood and Veneer Association, Reston, Virginia. 51 pp.

American Society for Testing and Materials (ASTM). 2013. Standard practice for estimating the percentage of wood failure in adhesive bonded joints. ASTM D5266-13. ASTM International, West Conshohocken, Pennsylvania. 4 pp.

American Society for Testing and Materials (ASTM). 2017. Standard test method for strength properties of adhesives in plywood type construction in shear by tension loading. ASTM D906-98 (reapproved 2017). ASTM International, West Conshohocken, Pennsylvania. 4 pp.

Asafu-Adjaye, O., A. O. Alawode, B. Via, and S. Banerjee. 2022. Soy flour substitution in polymeric diphenylmethane diisocyanate resin reduces press platen sticking. *Forest Prod. J*. 72(S1):5–7.

Asafu-Adjaye, O., B. Via, and S. Banerjee. 2020. Increasing cold tack of polymeric methylene diphenyl diisocyanate resin with partial soy flour substitution. *Forest Prod. J*. 70(1):143–144.

Baharoğlu, M., G. Nemli, B. Sarı, S. Bardak, and N. Ayrılmış. 2012. The influence of moisture content of raw material on the physical and mechanical properties, surface roughness, wettability, and formaldehyde emission of particleboard composite. *Compos. Part B: Eng*. 43(5):2448–2451.

Birkeland, M. J., L. Lorenz, J. M. Wescott, and C. R. Frihart. 2010. Determination of native (wood derived) formaldehyde by the desiccator method in particleboards generated during panel production. *Holzforschung* 64:429–433. https://doi.org/10.1515/HF.2010.070

Bockel, S., I. Mayer, J. Konnerth, S. Harling, P. Niemz, C. Swaboda, M. Beyer, N. Bieri, G. Weiland, and F. Pichelin. 2019. The role of wood extractives in structural hardwood bonding and their influence on different adhesives systems. *Int. J. Adhes. Adhes*. 91(2019):43–53.

British Standards Institution (BSI). 1993. Plywood bonding quality part 2: Requirements. EN 314-2:1993. British Standards Institution, London, England.

British Standards Institution (BSI). 2004. Plywood bonding quality part 1: Test methods. EN 314-1:2004 (confirmed 2014). British Standards Institution, London, England.

Cheng, Q., C. Essien, B. Via, and S. Banerjee. 2019. Cost savings from soy flour substitution in methylene diphenyl diisocyanate for bonding flakes and particles. *Forest Prod. J*. 69(2):154–158.

Cool, J. and R. E. Hernandez. 2011. Improving the sanding process of black spruce wood for surface quality and water-based coating adhesion. *Forest Prod. J*. 61(5):372–380.

Daoui, A., C. Descamps, M. Remy, and A. Zerizer. 2011. Influence of veneer quality on beech LVL mechanical properties. *Maderas. Cienc. Tecnol*. 13(1):69–83. https://doi.org/10.4067/S0718-221X2011000100007

Dunky, M., T. Pizzi, and M. V. Leempu. 2002. COST action E13. Wood adhesion and glued products. State of the art report. COST Association (European Cooperation in Science and Technology), Brussels, Belgium. 174 pp.

Fan, B., L. Zhang, Z. Gao, Y. Zhang, J. Shi, and J. Li. 2016. Formulation of a novel soybean protein based wood adhesive with desired water resistance and technological applicability. *J. Appl. Polym. Sci*. 133:43586.

Food and Agriculture Organization of the United Nations (FAO). 2021. Forest product statistics FAOSTAT. https://www.fao.org/forestry/statistics/80938/en/. Accessed September 26, 2022.

Forest Products Laboratory. 2021. Wood handbook—wood as an engineering material. General Technical Report FPL-GTR-282. U.S. Department of Agriculture, Forest Service, Forest Products Laboratory, Madison, Wisconsin. 543 p.

Frihart, C. R. 2009. Adhesive groups and how they relate to the durability of bonded wood. *J. Adhes. Sci. Technol*. 23(4):601–617.

Frihart, C. R. 2013. Wood adhesion and adhesives. *In:* Handbook of Wood Chemistry and Wood Composites. 2nd ed. R. M. Rowell (Ed.). CRC Press, Boca Raton. Florida. pp. 255–320.

Frihart, C. R. and J. F. Beecher. 2016. Factors that lead to failure with wood adhesive bonds. *In:* World Conference on Timber Engineering, WCTE 2016, August 22–25, 2016, Vienna, Austria. pp. 1130–1137.

Frihart, C., C. Hunt, and M. Birkeland. 2014. Soy Proteins as Wood Adhesives. Chapter 16. *In:* Recent Advances in Adhesion Science and Technology in Honor of Dr. Kash Mittal. W. Gutowski and H Dodiuk (Eds.). CRC Press, Boca Raton, Florida. pp. 277–294. https://doi.org/10.1201/b16347-21

Gao, T. 2010. Mode-I fracture in bonded wood: Studies of adhesive thermal stability, and of the effects of wood surface deactivation. Master's thesis. Virginia Polytechnic Institute and State University, Blacksburg.

Hänsel, A., J. Sandak, A. Sandak, J. Mai, and P. Niemz. 2021. Selected previous findings on the factors influencing the gluing quality of solid wood products in timber construction and possible developments: A review. *Wood Mater. Sci. Eng*. 17(3):230–241. https://doi.org/10.1080/17480272.2021.1925963

Hernández, V. 2013. Radiata pine pH and buffering capacity: Effect of age and location in the stem. *Maderas: Cienc. Tecnol*. 15(1):73–78. https://doi.org/10.4067/S0718-221X2013005000007

Irle, M. A., M. C. Barbu, R. Reh, L. Bergland, and R. M. Rowell. 2013. Wood composites. In: Handbook of Wood Chemistry and Wood Composites. 2nd ed. R. M. Rowell (Ed.). CRC Press, Boca Raton. Florida. pp. 321–414.

Johns, W. E. and K. A. Niazi. 1980. Effect of pH and buffering capacity of wood on the gelation time of urea-formaldehyde resin. *Wood Fiber Sci*. 12(4):255–263.

Kariz, M. and M. Sernek. 2014. Determination of the percentage of wood failure by 3D scanning of the adhesive bondline. *Eur. J. Wood Prod*. 72:551–553.

Kim, M. and B.-D. Park. 2021. A method of measuring wood failure percentage of wood specimens bonded with melamine-urea-formaldehyde resins using image analysis. *J. Korean Wood Sci. Technol*. 49(3):274–282.

Kumar, R. M. and A. Pizzi. 2019. Wood surface inactivation due to extractives. *In:* Adhesives for Wood and Lignocellulosic Materials. R. M. Kumar and A. Pizzi (Eds.). Scrivener Publishing, Beverly, Massachusetts. pp. 211–222.

Li, H., C. Li, H. Chen, D. Zhang, S. Zhang, and J. Li. 2014. Effects of hot-pressing parameters on shear strength of plywood bonded with modified soy protein adhesives. *Bioresources* 9. https://doi.org/10.15376/biores.9.4.5858-5870

Li, J., B. Zhang, Z. Li, Y. Yi, F. Shi, J. Guo, and Z. Gao. 2019. Effects of typical soybean meal type on the properties of soybean-based adhesive. *Int. J. Adhes. Adhes*. 90:15–21.

Lin, L., S. He, F. Fu, and X. Wang. 2015. Detection of wood failure by image processing method: Influence of algorithm, adhesive and wood species. *Eur. J. Wood Prod*. 73:485–491. https://doi.org/10.1007/s00107-015-0907-z

Luo, J., C. Yuan, W. Zhang, J. Li, Q. Gao, and H. Chen. 2015. An eco-friendly wood adhesive from soy protein and lignin: performance properties. *RSC Adv*. 5(122):1849–1855. https://doi.org/10.1039/c5ra19232c

Mirabile, K. V. and A. Zink-Sharp. 2017. Fundamental bonding properties of Douglas-fir and southern yellow pine wood. *Forest Prod. J*. 67(7/8):435–447.

Roffael, E. 2016. Significance of wood extractives for wood bonding. *Appl. Microbiol. Biotechnol*. 200(4):1589–1596.

Scott, C. T., R. Hernandez, C. Frihart, R. Gleisner, and T. Tice. 2005. Method for quantifying percentage wood failure in block-shear specimens by a laser scanning profilometer. *J. ASTM Int*. 2(8):1–10. https://doi.org/10.1520/jai12957

Shmulsky, R., M. K. Dowd, D. J. V. Lopes, G. D. Miller, Jr., and E. D. Entsminger. 2021. Production of yellow-poplar interior plywood with cottonseed-based protein adhesives. *Wood Fiber Sci*. 53(3):206–215.

Skoog, D. A. and D. M. West. 2013. Fundamentals of Analytical Chemistry. 9th ed. Cengage Learning, New York. 1072 pp.

Solt, P., J. Konnerth, W. Gindl-Altmutter, W. Kantner, J. Moser, R. Mitter, and H. W. G. van Herwinen. 2019. Technological performance of formaldehyde-free adhesive alternatives for particleboard industry. *Int. J. Adhes. Adhes*. 94:99–131.

Via, B., W. G. Hand, and S. Banerjee. 2019. Use of soy flour in adhesive formulations used to manufacture engineered wood composites. US patent 10,266,694 B2.

Vnučec, D., A. Kutnar, and A. Goršek. 2016. Soy-based adhesives for wood-bonding—A review. *J. Adhes. Sci. Technol*. 2016:1–22. https://doi.org/10.1080/01694243.2016.1237278

Wang, X., W. Zeen, H. Paul, W. Yaolin, Z. Romulo, and R. Casilla. 2010. The ability of wood to buffer highly acidic and alkaline adhesives. *Wood Fiber Sci*. 42(3):398–405.

Xiao, Z., Y. Li, X. Wu, G. Qi, N. Li, K. Zhang, and X. S. Sun. 2013. Utilization of sorghum lignin to improve adhesion strength of soy protein adhesives on wood veneer. *Ind. Crops Prod*. 50: 501–509. https://doi.org/10.1016/j.indcrop.2013.07.057

Yang, Y., M. Gong, and Y. H. Chui. 2008. A new image analysis algorithm for calculating percentage wood failure. *Holzforschung* 62(2):248–251.

Zhang, X., Y. Zhu, Y. Yu, and J. Song. 2017. Improve performance of soy flour-based adhesive with a lignin-based resin. *Polymers* 9:261.

Zink, A. G. and E. Kartunova. 1998. Wood failure in plywood shear samples measured with image analysis. *Forest Prod. J*. 48(4):69–74.