

Effects of pH and storage time on the adhesive and rheological properties of cottonseed meal-based products

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ABSTRACT: To increase the basic knowledge of cottonseed meal (CSM)-based adhesives and optimize the operational parameters for practical applications, in this study, we investigated the effects of pH and storage time on the adhesive performance, water resistance, and rheological properties of CSM, water-washed cottonseed meal (WCSM), and cottonseed protein isolate (CSPI). We found all products possessed the highest dry, wet, and soaked adhesive strengths with the adhesive slurries prepared at pH 6.0. The effects of pH were smaller on WCSM than on CSM and CSPI slurries. Storage time (up to 8 days) did not greatly impact the adhesive performance of WCSM slurries prepared at pH 6.0, 7.5, and 9.0, but slightly reduced the adhesive strength of CSPI slurries with the same pH. The viscosity of WCSM slurries increased with storage over 8 days, but did not for CSPI slurries. © 2016 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2016**, *133*, 43637.

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INTRODUCTION

Adhesive bonding is a key factor for efficiently utilizing timber and other lignocellulosic resources. As synthetic adhesives are mostly derived from depleting petrochemical resources and have caused increasing environmental concern, natural product and byproduct-derived adhesives have attracted much attention in the last couple of decades (e.g., refs. 1–7). With this trend, our group has demonstrated that cottonseed meal (CSM)-based products can be used as wood adhesives.^{8–10} Whereas untreated CSM-based adhesives showed poor water resistance, water-washed cottonseed meal (WCSM) and cottonseed protein isolate (CSPI) possess much better water resistance,^{9,11,12} even better than the more widely studied soy protein isolate (SPI)-based adhesives.^{10,13}

Varying the pH of adhesive suspensions is an important parameter affecting protein solubility and surface charges, thus affecting the adhesive strength and the rheological properties.^{14–16} Wang *et al.*¹⁷ evaluated the effect of pH from 1.6 to 9.6 on dry, wet, and soaked strengths of SPI. They reported that the highest wet strength was reached at pH 4.6, the isoelectric point (pI) of SPI (i.e., neutral net charges on the protein surface). Dry and soaked strength were high in the pH range of 3.6–7.6. They attributed the observations to the decreasing solubility and increasing hydrophobicity of SPI adhesive slurries when its pH was equal to or near the pI of SPI. Whereas the adjustment of

the pH of SPI slurries could be a powerful approach for enhancing the adhesive performance of SPI, SPI is mostly insoluble at the pI. Thus, suspension additives (e.g., xanthan gum) were applied to improve the flowability of SPI at the expense of adhesive strength. On the other hand, Vnucac *et al.*¹⁸ reported that after thermal modification in a vacuum chamber, SPI adhesives without pH adjustment (i.e., at pH 6.0) had a viscosity below 28 mPa and exhibited no adhesive penetration. The adjustment of pH to 10 increased the viscosity and adhesive penetration, which improved the adhesive bond strength. In an earlier work, Hettiarachchy *et al.*¹ reported complicated observations. First, an SPI water suspension was modified to various pHs, that is, 8.0, 9.0, 11.0, and 12.0, shaken for 1 h at various temperatures and then freeze-dried. They found an increase in the adhesive strength of SPI at higher pH modifications. However, the readjustment of the pH 10-modified SPI preparation to pH 7.0, 8.0, and 9.0 had no significant effect on the adhesive strength of SPI, even though the hydrophobicity was affected.

Storage time (or pot life, shelf life) could also affect the application of natural product-based wood adhesives.^{19,20} Per the adhesive strength on wood veneers, Qi *et al.*²¹ claimed that sustainable shear adhesion strength of SPI was obtained within 3 months using NaHCO₃ modification although no specific data were shown. Khan *et al.*²² conducted the shelf-life study of a bagasse lignin (50%) substituted phenol formaldehyde wood

adhesive. The viscosity of the adhesive increased from an initial 2 to 15 mPa s after 42 days of storage. The adhesive strength of the adhesive was basically unchanged during the 42 days of testing. The authors further concluded that the shelf-life of the lignin phenol formaldehyde wood adhesive was comparable to the control of pure phenol formaldehyde adhesive.

In this study, we investigated the effects of pH and storage time on the dry and wet adhesive strengths, and rheological properties of three CSM-based products (i.e., CSM, WCSM, and CSPI). Our purposes were (1) increasing the basic knowledge of CSM-based adhesives and (2) optimizing the two parameters for their practical application in wood bonding.

EXPERIMENTAL

Materials

Three CSM products—CSM, WCSM, and CSPI—were prepared in-house from glanded CSM as described previously.^{8,23} These products were stored in a desiccator at room temperature (22–23 °C) until use. Maple wood veneer (1.59 mm thick) was purchased from Certainly Wood, Inc. (East Aurora, NY, USA). Veneers were cut into strips 25.4 mm wide by 88.9 mm long, with the wood grain parallel to the long side, and stored in sealed plastic bags at room temperature (22–23 °C) and room humidity (i.e., 50–60% relative humidity) until use.

Preparation of Bonded Wood Specimens

Adhesive slurries were prepared by mixing a desiccated CSM product with deionized water (3:25 w/w; 10.7% solids) and stirred at room temperature before application to maple wood veneers. For practical preparation, 3 g of CSM product was added to 12.5 g of deionized water and stirred for 2 h at 300 rpm. The starting or optimal pH of the dispersion in deionized water was recorded: 6.5 for CSM and WCSM and 4.8 for CSPI. Each dispersion was adjusted to the preparative pH by slowly adding 1 M NaOH or 1 M HCl; if needed, more water was added to reach the final total weight of product solids. Finally, the slurry was stirred for an additional 30 min before use.

Using a brush, each adhesive preparation was applied to one end of two wood veneer strips covering 25.4 mm (1.0") length with a total application rate of about 4.5 mg dry solid content per cm². The wet adhesive was allowed to air-dry for 10–15 min or until tacky, and then a second layer was applied on top of the first layer and air-dried again for the same time period. The tacky adhesive coated areas of the wood veneer strips were overlapped and bonded by hot-pressing (Carver Benchtop Heated Press, Model 3856, Carver Inc., Wabash, IN) at 100 °C for 20 min at a pressure of 2.8 MPa. The bonded area between the two strips was 25.4 × 25.4 mm (1.0" × 1.0"). These bonded wood specimens were cooled and set aside for conditioning at normal room temperature (22–23 °C) and relative humidity (50–60%) for 7 days before further experiments were conducted per ASTM D1151-00.²⁴

Lap Shear Strength Measurement and Dry Adhesive Strength

The lap-shear strength of five 7-day preconditioned wood specimens was measured with a Zwick Materials Tester (Zwick GmbH & Co., Ulm, Germany) fitted with 32 × 40 mm fishscale

gridded wedge grips, and operated with a cross-head speed of 1 mm min⁻¹. The shear strength at break (MPa) was reported as the dry adhesive strength of the tested CSM products.

Wet and Soaked Adhesive Strength

The test conditions of the adhesive's water resistance were based on two of the water exposures in ASTM D1151-00.²⁴ Specifically, after 7 days of preconditioning, 10 bonded wood pairs were immersed in tap water at 23 °C for 48 h. The wet adhesive strength of a CSM product was determined by immediate measurement of the shear strength at break of five wet wood pairs. The remaining five soaked specimens were conditioned at 23 °C and 50% relative humidity for 7 days in a humidity and temperature stability test chamber (NorLake Scientific, Hudson, Wisconsin). The shear strength at break of these soaked and dried specimens was measured and termed soaked adhesive strength.

Shelf Life of Adhesive Slurries

Adhesive slurries of WCSM and CSPI were prepared as described above with the pH adjusted to 6.0, 7.5, or 9.0 and applied immediately to wood pairs (day 0). The remaining portion of the adhesive slurry was aliquoted evenly into three screw-capped tubes, sealed, and stored at room temperature. At day 2, 4, and 8, the slurries were stirred for 20 min, and applied for bonding wood pairs as previously described. The dry and soaked shear strengths of these bonded pairs were measured in the same way as described above. The physical changes of the adhesive slurries were visually observed and recorded.

Rheological Measurement

Rheological properties of WCSM and CSPI slurries were determined with an Anton Paar MCR 102 Rheometer (Ashland, VA) using a PP50 Parallel Plate measuring system and gap distance set at 1 mm. About 2.2 mL of prepared adhesive slurry was poured onto the sample plate, and rotational tests were performed under steady shear flow at room temperature (22 °C). Shear rate increased from 1 to 100 s⁻¹ at 1 s⁻¹ increments. Each test per slurry preparation was repeated to ensure reproducibility.

Statistical Analysis

Five bonded wood specimens were tested for each treatment. The data analysis package in Microsoft Excel 2007 was used for statistical analysis. The Descriptive Statistics Tool Data was used to calculate averages and standard deviations (SD). Single-factor analysis of variance (ANOVA) was used to evaluate the significance levels of the effects of treatments on adhesive properties.

RESULTS AND DISCUSSION

Effects of pH on Adhesive Strength

Among the five pH preparations tested, the slurries of CSM and WCSM prepared at pH 6.0 showed the highest adhesive strength (Table I). With either lower or higher pH, the dry adhesive strength of CSM decreased gradually by 23.1–32.4%. The effect of preparative pH on WCSM's adhesive strength was smaller as the relative decrease from the optimal pH condition ranged from 13.4% to 20.2%. The adhesive strengths of CSPI prepared at pH 6.0 and 7.5 were similar. The CSPI slurries prepared at the other three pH conditions, i.e., 4.5, 9.0, and 11.0, showed

Table I. Dry Shear Strength (MPa) at Break of Maple Wood Strips Bonded with Adhesive Slurries of Cottonseed Meal (CSM), Water-Washed Cottonseed Meal (WCSM), and Cottonseed Protein Isolate (CSPI) Prepared at Five pHs

pH	CSM	WCSM	CSPI
4.5	2.77 ± 0.78	3.11 ± 1.46	1.78 ± 0.74
6.0	3.73 ± 0.49	3.82 ± 0.92	3.76 ± 0.80
7.5	2.87 ± 0.60	3.11 ± 0.79	3.82 ± 0.44
9.0	2.66 ± 0.34	3.31 ± 0.66	2.44 ± 0.65
11.0	2.52 ± 0.22	3.05 ± 0.50	3.04 ± 0.54
Significance level ($P > F$)	<0.014	<0.67	<0.001

Data are presented in the format of average ± standard deviation ($n = 5$).

lower adhesive strength. Unlike CSM and WCSM, lowering pH to 4.5 from 6.0 greatly decreased the CSPI's adhesive strength by a maximum of 53%. The behavior of CSPI adhesive from pH 4.5 to 7.5 cycled up and down, whereas, the behavior of SPI adhesive remained the same through pH range 3.6–7.6.¹⁷ Comparing the data in Table I, the values of WCSM were always higher than the corresponding values of CSM and CSPI at the same pH, except for CSPI with pH 7.5. This indicated that WCSM was a stronger adhesive than CSM or CSPI, which was consistent with our previous reports^{8,12} that the adhesive strength of WCSM was as good as, or even better than, CSM or CSPI.

Effects of pH on Water Resistance

The wet adhesive strength values of CSM, WCSM, and CSPI are listed in Table II. Soaking the bonded maple wood pairs lowered the adhesive strengths of all three products. However, the highest values are again expressed in slurries prepared at pH 6.0 for all products. The pattern of the effect of pH was similar to that of the dry shear strength values in Table I. Under all pH conditions, the WCSM preparations showed higher wet strength than the corresponding CSM and CSPI preparations. The relative strength decrease of wet WCSM specimens, compared to the corresponding dry specimens, were in the general order of WCSM < CSPI < CSM. Compared to all of the dry shear strength values, the relative WCSM % changes (−31.9% and

−38.2%) at pH 4.5 and 6.0 were not only smaller than the decrease of CSPI (corresponding values −51.9% and −42.7%) (Table II), but also lower than the corresponding −52.9% to −63.5% changes of soy protein adhesives made at the same pH range.¹⁷ In spite of the difference in the absolute adhesive strengths between the soy-protein adhesives in literature¹⁷ and the cottonseed products in this work, relative decrease of the adhesive strength due to water soaking suggested the better water resistance of WCSM and CSPI than soy protein. This observation also implied that certain carbohydrate components in WCSM²⁵ were able to improve the water resistance of the plant seed protein-based adhesives. Previously, Chen *et al.*² reported that the improvement of water resistance of soy-protein wood adhesive by using hydrophilic polyethylene glycol additives.

The soaked adhesive strength values of CSM, WCSM, and CSPI are listed in Table III. Similar to the dry and wet adhesive strength, the greatest soaked strength was observed at pH 6.0 for all CSM, WCSM, and CSPI. Drying wet specimens partly recovered the lost adhesive strength of CSM as the decreases in shear strength at break of the soaked samples (Table III) were about 21–36% less than those of the wet specimens (Table II). Drying wet specimens not only recovered but also increased the adhesive strength of WCSM and CSPI. As a matter of fact, the adhesive strength of WCSM was fully recovered in the 4 lower pH preparations. The full recovery of CSPI's adhesive strength was reached in 2 pH preparations after the bonded pairs were dried. It is noted that there are some data values with more than 100% recovery. Similar data were reported previously in early studies.^{12,14,17,26} One possible cause of this >100% strength increase could be attributed to the high standard deviations in both dry and soaked strength measurements (Tables I and II).^{14,17} On the other hand, Liu *et al.*²⁶ attributed the increase in the adhesive strength of their soy protein–calcium carbonate adhesives to the removal of some soluble components (including alkaline and sodium) during the soaking–drying cycles. It was also notable that the decrease of the dry, wet, and soaked strengths of the three cottonseed products at pH 6.0 was not sharper than the soy-protein-based adhesive slurries affected by pH away from its optimal pH near 6.0 by Zhong *et al.*¹⁴ This difference indicated that cottonseed adhesive products,

Table II. Wet Shear Strength (MPa) at Break of Maple Wood Strips Bonded with Adhesive Slurries of Cottonseed Meal (CSM), Water-Washed Cottonseed Meal (WCSM), and Cottonseed Protein Isolate (CSPI) Prepared at Five pHs, and the Strength Change (%) Related to Their Corresponding Dry Samples

pH	Wet strength (MPa)			Change (%)		
	CSM	WCSM	CSPI	CSM	WCSM	CSPI
4.5	0.98 ± 0.25	2.12 ± 0.21	0.86 ± 0.53	−64.6	−31.9	−51.9
6.0	1.20 ± 0.15	2.36 ± 0.13	2.15 ± 0.58	−67.8	−38.2	−42.7
7.5	0.63 ± 0.21	1.30 ± 0.11	1.24 ± 0.23	−78.0	−58.2	−67.5
9.0	0.36 ± 0.12	1.15 ± 0.11	0.21 ± 0.09	−86.3	−65.3	−91.5
11.0	0.52 ± 0.08	0.64 ± 0.19	1.24 ± 0.23	−79.3	−78.9	−59.3
Significance level ($P > F$)	<0.001	<0.001	<0.001			

Strength data are presented in the format of average ± standard deviation ($n = 5$).

Table III. Soaked Shear Strength (MPa) at Break of Maple Wood Strips Bonded with Adhesive Slurries of Cottonseed Meal (CSM), Water-Washed Cottonseed Meal (WCSM), and Cottonseed Protein Isolate (CSPI) Prepared at Five pHs, and the Strength Change (%) Related to Their Corresponding Dry Samples

pH	Soaked strength (MPa)			Change (%)		
	CSM	WCSM	CSPI	CSM	WCSM	CSPI
4.5	1.96 ± 0.82	3.18 ± 1.20	1.81 ± 0.82	-29.4	2.3	1.9
6.0	2.54 ± 0.48	4.04 ± 0.48	4.09 ± 0.41	-32.0	5.8	8.8
7.5	1.60 ± 0.41	3.44 ± 0.52	2.95 ± 0.58	-44.2	11.2	-22.9
9.0	0.91 ± 0.23	3.61 ± 0.29	0.88 ± 0.54	-65.7	8.9	-64.2
11.0	1.29 ± 0.10	2.52 ± 0.31	2.87 ± 0.20	-48.8	-17.1	-5.7
Significance level ($P > F$)	<0.001	<0.02	<0.001			

Strength data are presented in the format of average ± standard deviation ($n = 5$).

especially WCSM, offer greater flexibility in bonding wood products when pH is a factor.

Pot Life of WCSM and CSPI

Due to the nature of onsite preparation of wood adhesive dispersions for veneer bonding, their stability for 24 h or so would be acceptable “working life” of these adhesive dispersions^{27,28} although some researchers²² evaluated the storage effect up to 42 days. Thus, we conducted the experiment of pot life up to 8 days. The dry and soaked adhesive strength of WCSM and CSPI adhesive slurries prepared at pH 6.0, 7.5, and 9.0 were evaluated over storage time up to 8 days at room temperature (Figure 1). The dry and soaked adhesive strength of WCSM at pH 6.0 were basically constant, showing small fluctuation from 4.5 and 5.1 MPa, respectively, over the storage time. The adhesive strength of the WCSM preparation at pH 7.5 decreased at day 2, and then recovered with the values near the initial levels for day 4 and/or 8. The slurry of WCSM at pH 9.0 showed a decreasing trend in both dry and soaked adhesive strength with increasing storage time. Unlike WCSM, the slurries of CSPI prepared at all three pH conditions showed a trend of decreasing dry and soaked adhesive strengths over the storage time, though there were differences in the decreasing scales. Those data indicated that the adhesive slurries of WCSM prepared at pH 6.0 were the most stable in all the samples tested.

In addition, we examined the color and odor of these slurries over the storage time (Table IV). The newly prepared slurries of both WCSM and CSPI were almost odorless with general brownish color. The color of WCSM slurries gradually became darker as storage time increased. Off odors began appearing at day 4, and became stronger at day 8; off odors can be attributed to the presence of bacteria and/or fungi. The slurries of CSPI seemed more mold resistant as no off odors had appeared by day 4. The off odor was stronger in the storage slurries of both WCSM and CSPI at the higher pH conditions. Whereas 2–4 days should be long enough for general on-site use of stored WCSM and CSPI slurries for wood bonding, adding synthetic reagents^{28,29} or copper preservatives²⁰ into the adhesive slurries has been reported, if a longer storage time is needed.

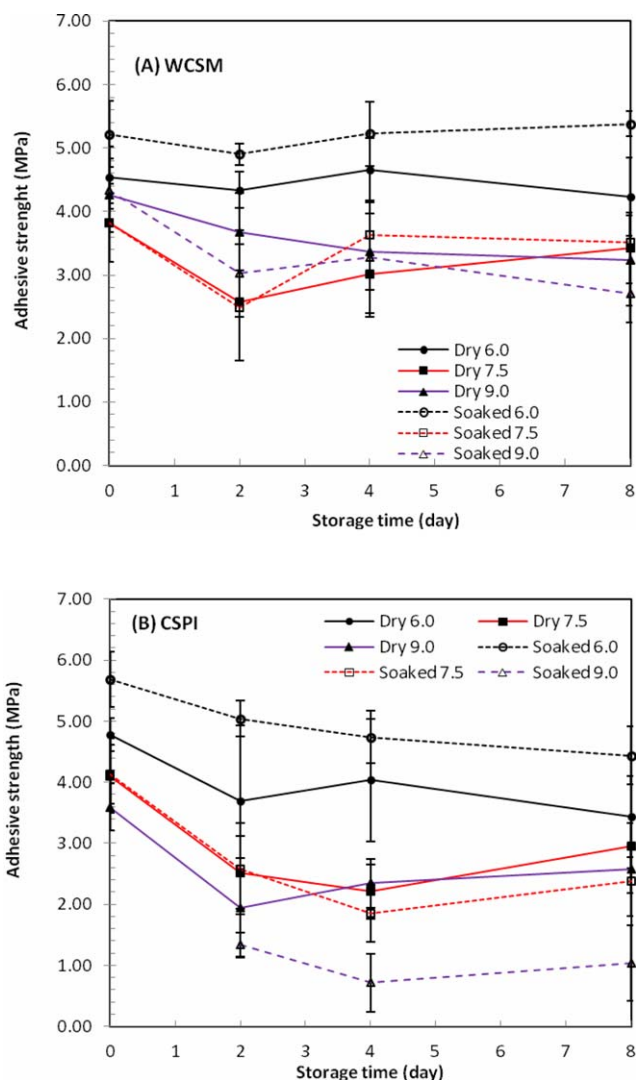


Figure 1. Effect of storage time of adhesive slurries at 22 °C on the dry and soaked adhesive strength of washed cottonseed meal (WCSM) and cottonseed protein isolate (CSPI) with preparative pH of 6.0, 7.5, and 9.0. Values are averages ± standard deviations ($n = 5$). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table IV. Physical Changes of the Adhesive Slurries of Washed Cottonseed Meal (WCSM) and Cottonseed Protein Isolate (CSPI) Prepared at pH 6.0, 7.5, and 9.0 with Storage at 22 °C

pH	Storage time (day)			
	0	2	4	8
WCSM				
6.0	Brown	Green/taupe	Dark tan, musty	Greenish brown, musty
7.5	Light brown	Light tan	Tan, musty	Yellowish tan, rotten egg smell
9.0	Brown	Light brown	Tan, sour/sulfury	Yellowish tan, strong rotten egg
CSPI				
6.0	Brown	Light brown	Light tan	Light tan
7.5	Caramel	Light tan	Light tan	Light tan, sour smell
9.0	Dark brown	Light tan	Light tan	Light tan, rotten egg smell

Rheological Properties of WCSM and CSPI

Figure 2 presents the viscosity behaviors of fresh WCSM and CSPI slurries as a function of shear rate. The apparent viscosities of all slurries were shear-rate dependent, indicating shear thinning properties of both WCSM and CSPI. This rheological property of cottonseed-based adhesives was similar to those of other biobased adhesives, such as soy meal³⁰ and protein isolates.^{17,31} The slurry of WCSM showed higher viscosity than CSPI slurry under the same conditions. The viscosities of both WCSM and CSPI slurries increased at higher pH. However, the highest viscosity was observed at pH 9.0 for the WCSM slurry, but at pH 7.5 for the CSPI slurry. Wang *et al.*¹⁷ observed higher viscosity in soy-protein slurry at pH 7.6 than at pH 4.6, and attributed this to lower solubility of soy protein near its pI at lower pH. But, they did not measure the viscosity at pH higher than 7.6.

Generally, viscosity ranges of 5000–25,000 mPa s for most wood laminating purposes (both cold or hot press), over 50,000 mPa s for mastic consistency wood laminating operations, and about 8000–20,000 mPa s for no clamp cold press technique have been reported.³² The quantitative data of apparent viscosity at shear rate 10 s⁻¹ are listed in Table V. The initial viscosity measurements of WCSM slurries were 21 mPa s at pH 6.0, 48 mPa s at pH 7.5, and 134 mPa s at pH 9.0. The viscosity of WCSM slurries prepared at all three pH conditions increased with storage time, and ranged from near 200 mPa s at pH 6.0 to >400 mPa s at pH 9.0. It is reported that the viscosity of lignin phenol formaldehyde adhesive continuously increased during the storage time from initial 15 to 35 mPa s at day 42.²² The initial viscosity of CSPI slurries were 6 mPa s at pH 6.0, 37 mPa s at pH 7.5, and 16 mPa s at pH 9.0. The observation on the increase in viscosity of WCSM slurries with storage may be due to the build-up of internal aggregation structure under rest conditions which probably is destroyed under shear.^{33,34} This thixotropy behavior (decreasing viscosity during time of shearing at constant shear rate) is more extensively in hydrocolloidal starch and protein^{33,34} than in biobased adhesives with higher solid contents.³⁵ In this work, we observed that storage time affected the viscosity of CSPI slurries less than that of WCSM slurries. The viscosity of CSPI slurries prepared at pH 6.0 and pH 9.0 was basically unchanged over storage time; however, the

viscosity of the CSPI slurry at pH 7.5 decreased during the storage period. Those data indicated that WCSM not only possessed the better adhesive performance but also was more flexible in its rheological properties than CSPI and/or soy-protein isolate. For example, adjustment of the viscosity could be obtained by time in storage as a way to increase flowability of WCSM-based adhesives at pH 6.0 with no effect on adhesive strength. In contrast, at a sacrifice of the adhesive strength, increasing pH or addition of denaturants are applied to improve the viscosity and flowability of soy protein.¹⁷ The carbohydrate components in WCSM²⁵ should have contributed to better rheological behaviors than CSPI as Cheng and Arthur²⁷ observed that sucrose increased the viscosity of cottonseed protein dispersion at higher pH; Wang *et al.*²⁸ reported that sucrose made waxy protein a viscous, flowable liquid rather than an unflowable slurry and paste. Asghari *et al.*³⁶ investigated interfacial and foaming characterization of mixed protein–starch particle systems for food-foam applications and proposed that the

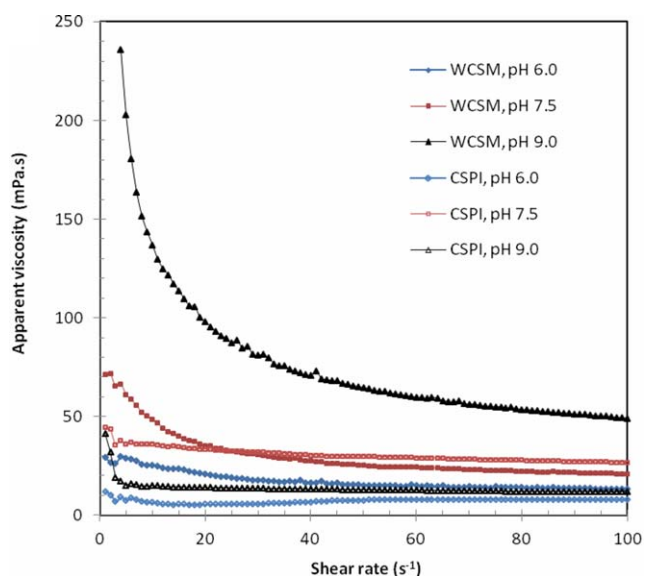


Figure 2. Effect of shear rate on the viscosity of the fresh adhesive slurries of washed cottonseed meal (WCSM) and cottonseed protein isolate (CSPI) with preparative pH of 6.0, 7.5, and 9.0. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table V. Effect of Storage Time on Apparent Viscosity (mPa s at Shear Rate of 10 s^{-1}) of the Adhesive Slurries of Washed Cottonseed Meal (WCSM) and Cottonseed Protein Isolate (CSPI) Prepared at pH 6.0, 7.5, and 9.0

pH	Storage time (day)			
	0	2	4	8
WCSM				
6.0	21.2 ± 5.8	66.0 ± 2.1	88.2 ± 8.8	195.9 ± 8.5
7.5	48.2 ± 0.7	126.6 ± 7.6	165.5 ± 33.5	251.4 ± 30.5
9.0	133.9 ± 4.2	379.0 ± 63.6	379.9 ^a	436.2 ± 44.0
CSPI				
6.0	6.2 ± 0.4	5.8 ± 0.2	5.6 ± 0.7	7.3 ± 2.7
7.5	36.8 ± 0.8	33.6 ± 0.3	26.4 ± 4.8	19.2 ± 0.7
9.0	16.3 ± 1.9	13.8 ± 0.1	46.8 ± 2.1	18.7 ± 1.2

Data are presented in average ± standard deviation ($n = 2$).

^aOnly one data point available.

potential synergy between protein–starch particles systems should depend on protein type and starch physical and surface properties. They also reported more than one mechanism could be responsible for the increased stability the mixed wet foam systems. These findings may be also applicable for the observation of protein adhesives with carbohydrates. Whereas the current work is more on the measurement of the viscosity parameters for practical use, additional advanced analysis (such as FTIR for protein and carbohydrate structure bonding, SDS-PAGE for protein structures) in the future research could contribute to the understanding of the mechanisms of CSM-based adhesives rheological behaviors affected by storage time.

CONCLUSIONS

All three products—CSM, WCSM, and CSPI—showed the maximal adhesive strength and water resistance at pH 6.0 in the tested pH range from 4.5 to 11.0. The general trend of the dry, wet, and soaked adhesive strength was in the order WCSM > CSPI > CSM. Storage of WCSM slurries up to 8 days did not change both dry and soaked adhesive strengths of WCSM slurries prepared at pH 6.0, 7.5, or 9.0. However, the viscosity of these slurries increased with storage time. In contrast, the dry and soaked adhesives strengths of CSPI slurries decreased slightly under the same storage conditions, while the viscosity of CSPI slurries were unchanged or decreased over storage time. Per the observations in this work, the inexpensive WCSM preparation was more feasible and showed a range of acceptable operational parameters than the relatively expensive CSPI preparation when used for wood bonding purposes. The adhesive slurry of WCSM could be prepared at its optimal pH of 6.0, but it was also usable in the pH range of 4.5–9.0.

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