Journal of Applied Polymer Science

Property and Storage Stability of Whey Protein-Sucrose Based Safe Paper Glue

Guorong Wang,[1] Mingruo Guo[1,2]
[1]Department of Nutrition and Food Sciences, University of Vermont, Burlington, Vermont 05405
[2]Department of Food Science, Jilin University, Changchun 130062, China
Correspondence to: M. Guo (E-mail: mguo@uvm.edu)

ABSTRACT: Whey is a byproduct of cheese making. Disposal of whey may cause environmental pollutions. Whey protein isolate (WPI) is obtained from whey via ion-exchange and ultra-filtration, and contains more than 90% of protein. Most current commercially available paper glue products are made from undegradable synthetic polymers and may be harmful to human. The objective of this study was to develop environmentally friendly and children safe paper glue using WPI and sucrose as major ingredients. Results showed that both desirable bonding strength and viscosity of the safe paper glue were achieved by adding sucrose. The whey protein and sucrose based paper glue had excellent recovered bonding strength (ideal for self-seal envelopes) and bonding strength to different substrates (paper–paper, paper–wood, paper–plastic, and paper–metal). Shelf life tests indicated that the safe glue was stable with addition of low level of preservative and there were no considerable changes in bonding strength and viscosity during the storage at 23°C or 40°C for up to 1 year. © 2013 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2014**, *131*, 39710.

KEYWORDS: adhesives; addition polymerization; biopolymers & renewable polymers

Received 28 February 2013; accepted 27 June 2013
DOI: 10.1002/app.39710

INTRODUCTION

Whey is the watery liquid separated from cheese curd. If it is directly disposed to environment, it may result in environmental pollutions. Utilization of whey has been gaining increasing concerns from both academia and industry during recent decades. Whey mainly contains lactose (44–52 g L^{-1}), proteins (6–8 g L^{-1}) and minerals (2–7 g L^{-1}).[1] Powdered whey and lactose are the most common whey products used for food and feed industries, but these applications are usually low value-added. Whey protein concentrate (WPC) and whey protein isolate (WPI) have excellent nutritive and functional properties and are used as nutritional supplements and functional ingredients (emulsifier, foaming, and gelling agent etc.) in food industry.[2–6] The steady increase in production of cheese keeps putting pressures on whey utilization[7] and there are still growing economical and environmental needs to explore new applications for whey proteins.

Whey protein is comprised of about 500 g kg^{-1} of β-lactoglobulin (β-Lg), 200 g kg^{-1} of α-lactalbumin (α-La), and 80 g kg^{-1} of bovine serum albumin (BSA).[8] Though applications of other proteins (like casein and soy protein) in adhesives have been studied by many,[9–14] the use of whey protein as an adhesive ingredient is still a new subject. Tschabold patented an adhesive based on condensed whey in 1950s,[15] but no other literature related to whey based adhesive was found until the papers on whey protein based adhesive published by Gao recently.[12,16] Unlike casein, whey protein is comprised mainly by compact globular proteins with low molecular weight, and is not considered as an ideal adhesive polymer. However, those structures can be spread out by thermal treatment or solvent polarity change and then exhibits some properties similar to synthetic adhesive polymers.[317–19] At room temperature, the hydrophobic effect dominates thus keep whey protein folded, and the globular protein structures start to be spread out when the temperature is higher than 65°C. Whey protein components are unfolded at different temperature, ranging from 68°C to 89°C.[20] Once unfolded, the hydrophobic parts that contain reactive groups such as thiol groups expose, and the intermolecular networks are formed via the exchange of thiol-disulfide exchange.[21–27] On the other hand, whey protein is amphiphilic, which can be adsorbed by the surfaces of most materials.[17] Adhesive bonding strength comes from adhesion force (between adhesive molecules and substrate molecules) and cohesion force (within adhesive molecules).[28] For whey protein adhesive, the adhesion force can be formed by the interactions between

protein and adherend substrate molecules via hydrogen bonds, electrostatic forces, van der Waals interactions etc., and the cohesion force can be obtained by protein network-forming property.

Whey protein–polyvinyl pyrrolidone (PVP) based paper glue products have been developed in author's laboratory and the functionality of the products were studied.[29,30] But the whey protein–PVP based glue products still contain a synthetic copolymer (PVP). The objective of this study was to develop a safe paper glue using natural ingredients without synthetic polymer. Paper glue products are usually ready-to-use and packed in small quantity with stable shelf life. The major obstacle of using whey protein for paper glue product formulation is that good bonding strength is exhibited only when the concentration of polymerized whey protein is at higher levels (>100 g kg^{-1}), but this could lead to protein gelation during the thermo-polymerization and/or storage.[30] Sucrose is widely available and safe. It can be used for stabilizing protein during thermo treatment.[31–34] Due to its excellent hydration property, sucrose may also have beneficial effects on the bonding strength of paper glue. In this study, the effects of sucrose on the adhesive properties and storage stability of whey protein based paper glue were investigated.

EXPERIMENTAL

Whey Protein Polymerization

WPI contains 924.3 g kg^{-1} of protein was purchased from Fonterra (Auckland, New Zealand). WPI powder was dissolved in distilled water of room temperature at concentrations of 100.0 g kg^{-1} (P100), 110.0 g kg^{-1} (P110), 120.0 g kg^{-1} (P120), and 130.0 g kg^{-1} (P130), respectively (Table I). Polymerized whey protein solution was obtained by denaturing the WPI solutions at 90°C for 30 min. Viscosity and bonding strength of unpolymerized and polymerized whey protein solutions were evaluated.

Glue Prototype Preparation

Prototypes WS0, WS1, WS2, WS3, and WS4 were formulated by adding various amounts (0 g, 10 g, 20 g, 30 g, and 40 g) of sucrose granulates (White granulated sugar, distributed by DZA brands, LLC, NC, USA) respectively to 100 g of Polymerized P120 (120.0 g kg^{-1}). Proxel® BD-20 (Arch Chemicals, Norwalk, CT, USA) and Silicor 1311 FG emulsion (Defoamer.com, Bartlett, IL, USA) were added as the preservative and defoamer at a dosage of 2.5 g kg^{-1} and 4.0 g kg^{-1}. The sample ID and formulations of glue prototypes were listed in Table II. The mixtures were homogenized by using a Eurostar Power Control-Visc digital stirrer (IKA World, Wilmington, NC, USA) at 500 rpm for 10 min, and then defoamed at room temperature overnight. A commercial glue product was used as the control.

Physicochemical Properties

Total solids, protein, and ash contents of the prototypes and the control were evaluated according to the standard methods of ASTM E1756 – 08, AOAC 2001.14, and ASTM D5630 – 06, respectively. The value of pH was determined by Orion 420A pH meter (Jacksonville, FL, USA). Viscosity was tested by Brookfield LVDV-I Prime viscometer (Middleboro, MA, USA) at room temperature. The viscosity of unpolymerized whey protein was tested by using a No. 1 spindle stirring at 50 rpm for 30 sec, and viscosity of polymerized whey protein, prototypes, and commercial control was tested with a No. 3 spindle stirring at 10 rpm for 30 sec. Mass density (the mass per unit volume) was calculated by dividing the mass (g) by the volume (mL) of a given amount of sample at 23°C.

Lap-Shear Bonding Strength

WT 134 Crane cotton paper (Crane & Co., Dalton, MA, USA) was used as adhesive substrates for lap shear bonding strength test according to ASTM D1002 method. Unless indicated otherwise, the phrase "bonding strength" was short for the "paper–paper lap-shear bonding strength." About 0.02 g of glue was applied to the bond area (6.4 mm × 25.4 mm) of a Crane paper strip (101.6 mm × 25.4 mm × 0.5 mm) and lapped by another Crane strip at the bond area (Figure 1). The bonding strength was tested using an Instron 5566 universal testing machine (Instron, Canton, MA, USA) after the glue was totally set. The test speed of crosshead was set at 12.7 mm min^{-1}. The bonding strength was calculated by dividing the load at rupture (N) by the bond area (6.45 cm^2). The values of bonding strength were the averages based on 10 valid tests. The area of paper broken was determined by ImageJ 1.46 software (available for download at http://rsbweb.nih.gov/ij/download.html) and the percentage of paper broken (%PB) was calculated by dividing the paper broken area by the bond area.

Table I. Viscosity and Bonding Strength of the Unpolymerized and Polymerized Whey Protein Solutions (Significance level $P=0.05$).

Sample ID	Whey protein concentration (g kg^{-1})	Viscosity (mPa s)		Bonding strength (MPa)	
		Unpolymerized	Polymerized	Unpolymerized	Polymerized
P100	100.0	2.87 ± 0.12	115.0 ± 8.5	ND[+]	1.15 ± 0.08 a
P110	110.0	3.08 ± 0.20	147.0 ± 8.5	ND	1.07 ± 0.02 a
P120	120.0	2.96 ± 0.18	89485 ± 3978	ND	1.28 ± 0.06 b
P130	130.0	3.04 ± 0.37	NA*	ND	ND

[+] ND, not detected.

*WPI with concentration of 130.0 g kg^{-1} gelled after thermo treatment at 90°C for about 10 min.

Table II. Formulations, Bonding Strength, Viscosity, and % of Paper Broken of the Prototypes and Control (Significance level P=0.05).

	Formulations						
Sample ID	Polymerized P120 (g)	Sucrose (g)	Proxel BD20 (g kg^{-1})	Silicor 1311 (g kg^{-1})	Viscosity (Pa·s)	Bonding strength (MPa)	% of paper broken
WS0	100	0	2.5	4.0	89.51±3.97^{a}	1.28±0.16^{a}	90-100
WS1	100	10	2.5	4.0	3.09±0.30^{c}	1.42±0.03^{bc}	100
WS2	100	20	2.5	4.0	1.52±0.03^{d}	1.37±0.06^{ab}	100
WS3	100	30	2.5	4.0	0.94±0.02^{e}	1.41±0.06^{b}	70-80
WS4	100	40	2.5	4.0	0.81±0.01^{f}	1.35±0.07^{ab}	60-70
Control	----------				3.74±0.41^{b}	1.51±0.03^{c}	100

Glue Setting Time

The bonding strength of the prototypes and control was measured at specific time following application. The increase of bonding strength versus time was plotted. The first time paper broken observed was recorded as glue setting time.[35]

Durability of Bonding Strength

The glued crane paper strips were conditioned in 23°C and 40°C for 12 months, respectively. Bonding strength after conditioning was tested in comparison with that of newly glued.

Recovered Bonding Strength and Bonding Strength to Other Materials

Recovered bonding strength was the bonding strength of a rehydrated adhesive exhibited. In this study, about 0.02 g of glue was applied onto the bond area of a Crane paper strip to allow it dry and condition at room temperature for one week. After that about 0.02 g distilled water was applied back to rehydrate the dry glue and bond it to another unapplied paper strip and the bonding strength tested was referred as recovered bonding strength.

The prototypes were also used to bind Crane paper to metal (aluminum, 101.6 mm × 25.4 mm × 2.2 mm), plastic (polystyrene, 101.6 mm × 25.4 mm × 0.3 mm), and wood (sugar maple veneer, 101.6 mm × 25.4 mm × 1.7 mm) strips, and their bonding strength was determined by using the same method as previously.

Photography of Cured Glue Sample

About 25 g of glue samples were poured in a petridish (diameter ~85 mm) and cured at room temperature for 1 month, and then digitally pictured by Sony® DSC-H20 camera (Sony, Tokyo, Japan).

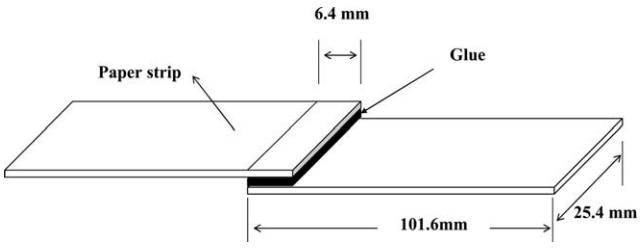


Figure 1. Lap shear bonding strength specimen dimensions.

Storage Stability

The prototypes and control were stored at 23°C and 40°C, respectively for 12 months. The bonding strength and viscosity were tested at an interval of 2 months. The same batches of samples were subjected to total aerobic plates count and yeast/mold count at the end of storage. The aerobics and yeast/molds counts were determined by using 3M aerobic count and yeast/mold count Petrifilm (3M Co., MA, USA).

Statistical Analysis

All values except bonding strength were averages of triplicates. One-way ANOVA analysis was conducted by SPSS 16.0 software at a significance level of 95.0% (SPSS, Chicago, IL).

RESULTS

Effects of Whey Protein Concentrations and Polymerization on the Viscosity and Bonding Strength

Unpolymerized P100, P110, P120, and P130 (with 100.0 g kg^{-1}, 110.0 g kg^{-1}, 120.0 g kg^{-1}, and 130.0 g kg^{-1} of WPI, respectively) were very watery liquid and bonding strength was not detected (Table I). The viscosity of the polymerized (90°C for 30 min) P100, P110, and P120 increased dramatically compared with their corresponding unpolymerized solutions and their bonding strength was detected and paper broken was obtained (Table I). Whey protein concentration positively affected the viscosity and bonding strength of polymerized samples (Table I). Polymerized P100 and P110 were too watery and running for convenient application and additional pressures was needed to hold the adherends until glue cured to bond the paper strips. Polymerized P120 was more a paste than viscous solution and its bonding strength (1.28 ± 0.06 MPa) was significantly stronger (P<0.05) than that of P100 (1.15 ± 0.08 MPa) or P110 (1.07 ± 0.07 MPa) (Table I), but it gradually gelled after stored for couple weeks. P130 with 130.0 g kg^{-1} of WPI gelled at about 10 min during the polymerization and no bonding strength was detected.

Effects of Sucrose on the Viscosity and Bonding Strength

Sucrose was added to the polymerized P120 slurry to decrease and stabilize its viscosity. The formulations, viscosity, bonding strength, %PB of the prototypes and control were listed in Table

II. In general, Addition of sucrose decreased the viscosity and increased the bonding strength. WS0, which was right the polymerized P120, was a thick paste; however, WS1, WS2, WS3, and WS4 with addition of sucrose were viscous flowable liquid. The viscosity decreased as addition of sucrose increased (Table II). Although the viscosity of WS2 (1.52 ± 0.03 Pa s), WS3 (0.94 ± 0.02 Pa s) and WS4 (0.81 ± 0.01 Pa s) were lower than the control (3.74 ± 0.41 Pa s), they were still viscous enough in appearance and had no defect in applications. The bonding strength of WS1 (1.42 ± 0.03 MPa) and WS3 (1.41 ± 0.06 MPa) was significantly higher ($P < 0.05$) than WS0 (1.28 ± 0.16 MPa), while WS2 (1.37 ± 0.06 MPa) and WS4 (1.35 ± 0.07 MPa) was higher than WS0 but no significance was observed ($P > 0.05$). Meanwhile, there was no significant difference in bonding strength detected among WS1, WS2, WS3, and WS4 (Table II). Although all prototypes had significant lower ($P < 0.05$) bonding strength than the control (3.74 ± 0.41 MPa), they were all strong enough to obtain full or partially paper broken (Table II). WS0 obtained an average %PB of 90–100, while full paper broken was achieved by WS1 and WS2 which contained 91.0 g kg^{-1} and 166.7 g kg^{-1} of sucrose, respectively. However, if the sucrose amount was further increased as in WS3 and WS4, partial paper broken was observed (Table II).

Physicochemical Property of Prototypes

The physicochemical properties of the prototypes were listed in Table III. Total solids content and mass density of the prototypes were increased as more sucrose added. The protein and ash contents and pH were decreased by the addition of sucrose. All prototypes except WS0 had higher total solids content than the control. The ash content, pH and density of the prototypes were higher than the control. The pH value of prototypes was near neutral while the commercial control was more acidic.

Glue Bonding Time

The bonding strength was tested at specific time after application, and the curves of bonding strength versus drying time were plotted in Figure 2. All prototypes and the control exhibited bonding strength 1 min after application, and the first paper broken (glue setting time) was observed at 2 min (WS0), 3 min (WS1), 3 min (WS2), 9 min (WS3), 12 min (WS4), and 9 min (control), respectively, which indicated that sucrose postponed the glue setting time. The bonding strength at the glue setting time of each sample was 0.24 ± 0.03 MPa (WS0), 0.31 ± 0.07 MPa (WS1), 0.27 ± 0.03 MPa (WS2), 0.49 ± 0.02 MPa(WS3), 0.40 ± 0.01 MPa (WS4), and 0.48 ± 0.03 MPa (control). The bonding strength at 30 min were 0.88 ± 0.06 MPa (WS0), 1.13 ± 0.04 MPa (WS1), 1.21 ± 0.01 MPa (WS2), 1.16 ± 0.02 MPa (WS3), 0.82 ± 0.01 MPa (WS4), 0.96 ± 0.06 MPa (control), which were about 69% (WS0), 80% (WS1), 88%(WS2), 82% (WS3), 60% (WS4), and 64% (control) of their final bonding strength.

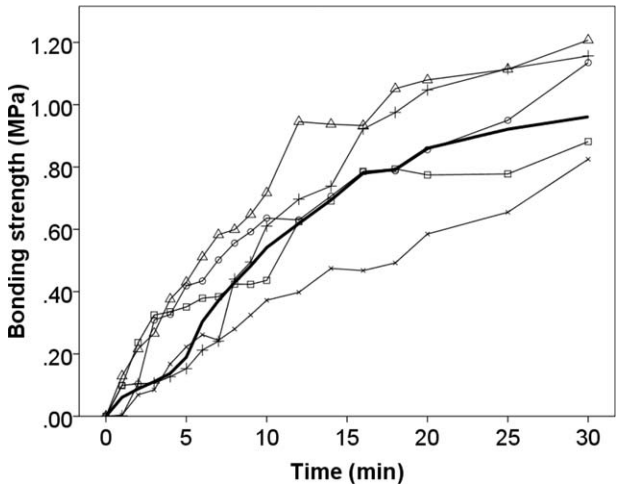


Figure 2. Effects of drying time on the bonding strength of the prototypes and control. WS0 (□), WS1 (○), WS2 (△), WS3 (+), WS4 (×), and control (—).

Durability of Bonding Strength

Durability of bonding strength at 23°C and 40°C were evaluated. The bonding strength of all samples including the control decreased after conditioning at 23°C or 40°C for 1 year (Table IV). The control lost 15% and 23% of bonding strength after conditioning at 23°C and 40°C, respectively. The prototypes WS1-4 lost about 19–26% of bonding strength at 23°C and about 18–29% at 40°C. The percentage of bonding strength decreased for WS0 was lowest among all the specimens, which was 11% at 23°C and 10% at 40°C. However, the decrease of %PB was not as obvious as the bonding strength. The %PB of WS0, WS1 and the control did not change after conditioning, and that of WS2, WS2, and WS4 only slightly decreased after conditioning (Tables II and IV). In general, all prototypes had desirable bonding strength durability.

Table III. Chemical Composition, pH and Viscosity of Prototypes and Control

Prototypes	Total solids (%)	Protein (%)	Ash (%)	pH	Density (g mL^{-1})
WS0	11.84 ± 1.84	11.16 ± 0.93	0.26 ± 0.01	7.21 ± 0.02	1.05 ± 0.02
WS1	20.02 ± 0.18	9.44 ± 0.30	0.22 ± 0.00	7.22 ± 0.01	1.06 ± 0.03
WS2	27.15 ± 0.02	8.71 ± 0.24	0.20 ± 0.00	7.12 ± 0.01	1.07 ± 0.01
WS3	34.13 ± 0.24	8.10 ± 0.13	0.19 ± 0.01	7.09 ± 0.03	1.10 ± 0.02
WS4	41.94 ± 0.68	7.79 ± 0.19	0.18 ± 0.01	7.03 ± 0.01	1.13 ± 0.02
Control	13.38 ± 0.02	ND	0.11 ± 0.00	5.16 ± 0.06	1.01 ± 0.01

Table IV. Durability of Bonding Strength of the Prototypes and Control

		After conditioned at 23°C for 12 months			After conditioned at 40°C for 12 months		
	Newly glued bonding strength (MPa)	Bonding Strength (MPa)	% of bonding strength decreased	%PB	Bonding Strength (MPa)	% of bonding strength decreased	%PB
WS0	1.28 ± 0.05	1.13 ± 0.14	11	90-100	1.15 ± 0.07	10	90-100
WS1	1.42 ± 0.03	1.15 ± 0.09	19	100	1.16 ± 0.06	18	100
WS2	1.37 ± 0.06	1.01 ± 0.20	26	90-100	0.97 ± 0.17	29	90-100
WS3	1.41 ± 0.06	1.05 ± 0.12	25	60-80	1.11 ± 0.13	21	60-80
WS4	1.35 ± 0.08	1.06 ± 0.05	21	50-70	1.04 ± 0.39	23	60-70
Control	1.51 ± 0.03	1.27 ± 0.14	15	100	1.15 ± 0.05	23	100

Recovered Bonding Strength and Bonding Strength to Other Materials

The recovered bonding strength of either prototypes or control was comparable with the original paper–paper bonding strength (Table V), but WS0 and WS1 caused paper wrinkling when they dried on the paper surfaces, while no paper wrinkling were observed on WS2, WS3, WS4 and the control. Recovered bonding strength may indicate if a glue could be used for self-sealing envelops, which are very popular due to the convenience of use since the dry glue strips on the flaps could be simply rehydrated to exhibit bonding strength.

All prototypes exhibited comparable or higher paper–wood and paper–metal bonding strength than control (Table V). The bonding strength of paper–wood was the closest to that of paper–paper, since both paper and wood are primary composed by cellulose fiber. WS0 failed to bind paper to plastic, but prototypes WS1-4 with the presence of sucrose could, though the bonding strength to plastic was much lower than to other substrates. Sucrose endowed the whey protein adhesive the affinity to plastic. WS1, WS2, and WS3 had comparable paper–plastic bonding strength to the commercial control, and WS4 had the highest paper–plastic bonding strength. In general, the paper–paper and paper–wood bonding strength of the prototypes and the control were stronger than that of paper–metal, and then the paper–plastic.

Photographs of Cured Adhesive Film

The morphologies of cured adhesive prototypes were depicted in Figure 3. WS0 finally fragmented into many small pieces instead of forming an intact film. WS1 did not fragment, but cracks were observed on its fragile film. Compared with WS0 and WS1, the structure of cured WS2 was improved significantly by increasing sucrose content. Cured WS2 was a smooth, homogenous, rigid and fragile film but without crack or sugar crystallization stains observed thereon. However, if the sucrose content further increased, sucrose crystallization presented as observed on WS3 and WS4. More crystallization was observed on WS4 than on WS3 due to the higher sucrose in WS4. The cured control formed a flexible and pliable plastic membrane.

Storage Stability

No microorganism growth was detected in the prototypes at either 23°C or 40°C at the end of storage due to the addition of Proxel® BD 20, which has a broad spectrum of activity against bacteria, fungi and yeasts but low mammalian toxicity and biodegrade in effluent systems. The newly produced prototypes were homogenous yellowish mixtures, which was the typical color that whey protein solutions possessed. No changes in color of the prototypes was detected by naked eyes after stored at 23°C for 12 months, while all prototypes turned brownish after stored for couple months at 40°C, but the changes in color did not affect the bonding strength. Figure 4 shows that the bonding strength of all prototypes was as stable as the control at 23°C or 40°C, neither obvious decrease in bonding strength nor in %PB was detected. WS0, the prototypes without addition of sucrose, became soft gel couple weeks after storage, and its viscosity was beyond the test range of the Brookfield viscometer, but bonding strength did not change considerably throughout

Table V. Recovered Bonding Strength and Bonding Strength to Different Substrates of the Prototypes and Control

Prototypes	Recovered bonding strength (MPa)	Paper–Wood (MPa)	Paper–Metal (MPa)	Paper–Plastic (MPa)
WS0	1.26 ± 0.04	1.39 ± 0.15	0.62 ± 0.15	ND
WS1	1.38 ± 0.04	1.23 ± 0.15	0.87 ± 0.38	0.21 ± 0.07
WS2	1.45 ± 0.04	1.37 ± 0.18	1.08 ± 0.36	0.20 ± 0.04
WS3	1.44 ± 0.08	1.48 ± 0.09	0.59 ± 0.15	0.26 ± 0.06
WS4	1.43 ± 0.12	0.92 ± 0.17	0.72 ± 0.25	0.55 ± 0.17
Control	1.51 ± 0.04	1.08 ± 0.21	0.60 ± 0.22	0.25 ± 0.08

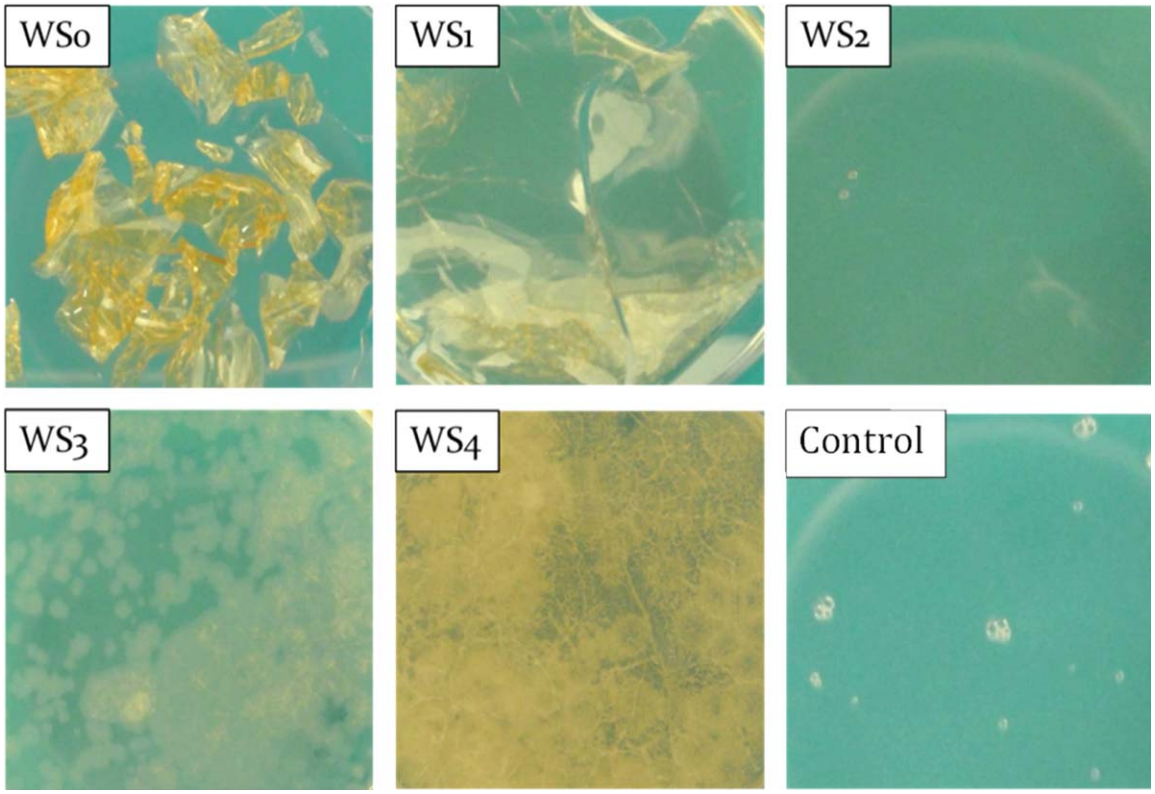


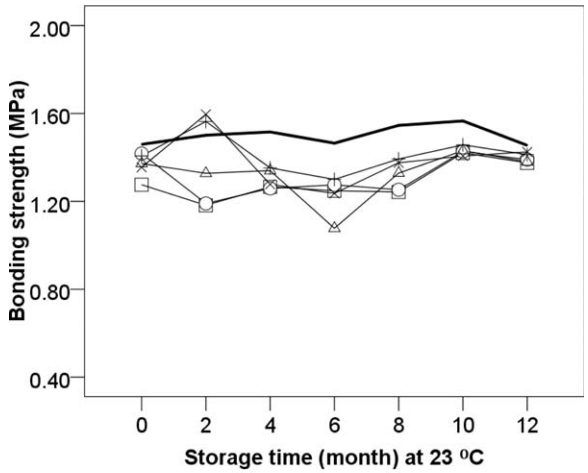
Figure 3. Digital photographs of cured adhesive films. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

the storage of 12 months at 23°C or 40°C. The viscosity of the prototype WS1 increased over the storage at both temperatures (Figure 5), but it was still flowable after one year's storage. The viscosity of WS2 increased over the storage at 23°C, but decreased slightly at 40°C; and the viscosity of WS3 and WS4 was stable at 23°C, and only slight decrease was detected at 40°C (Figure 5).

DISCUSSION

Glue bonding strength comes from two parts, adhesive force (the interphase forces between adhesive and substrate) and cohesive force (the chemical and physical forces keep the mass of adhesive from splitting).[28] Unpolymerized whey protein is comprised by compact globular molecules and does not have gel-forming property,[36] thus, effective cohesive force fail to form. This was verified by the results showed in Table I that the unpolymerized whey protein did not show adhesive strength at all. Polymerization is essential to develop globular whey proteins into adhesives[12,15] by breaking up the compact globular structure into partially linear structures and increasing the aggregate sizes via thiol-disulfide exchange, i.e., the intramolecular disulfide bonds are broken up to form intermolecular disulfide bonds,[18] to enhance the cohesive forces; therefore, bonding strength of polymerized whey protein was detected in this study (Table I). Whey protein polymerization is both temperature and concentration sensitive.[25,37] Our preliminary experiment showed that the highest viscosity of polymerized whey protein (100.0 g kg^{-1}) was obtained by denatured at 90°C for 30 min (data not shown), because that the maximum size of whey protein aggregates were achieved at 90°C according to other studies.[20,38] The viscosity of polymerized whey protein was affected dramatically by its concentration. According to the results, polymerized whey protein solutions with concentrations of 100.0 g kg^{-1} and 110.0 g kg^{-1} were not considered as ideal glue due to the running property. Desirable bonding strength and nearly full paper broken was obtained at the concentration of 120.0 g kg^{-1}. However, the spreadability is not as good as flowable viscous glue, and its viscosity was inclined to increase during storage and made it even difficult to be spread. Therefore, sucrose was added to decrease the viscosity and increase the storage stability.

Sucrose is one of the most common ingredients in food systems. The interaction between sucrose and whey protein during thermo treatment has been extensively studied.[3439–41] In this study, sucrose was added after the WPI solution was polymerized and cooled down. Addition of sucrose decreased the viscosity and increased the bonding strength of the polymerized whey protein slurry, and made it a viscous flowable liquid other than unflowable slurry and paste (Table II). The more sucrose added the more stable of viscosity was obtained during storage (Figure 5). Sucrose, acts as a plasticizer, may replace the water molecules between protein molecules, thus decrease the protein–protein interactions,[42] resulting in decrease in viscosity of polymerized whey protein. As depicted in Figure 3, the network-forming property was improved by addition of sucrose, but excess sucrose interfered with the network-forming and



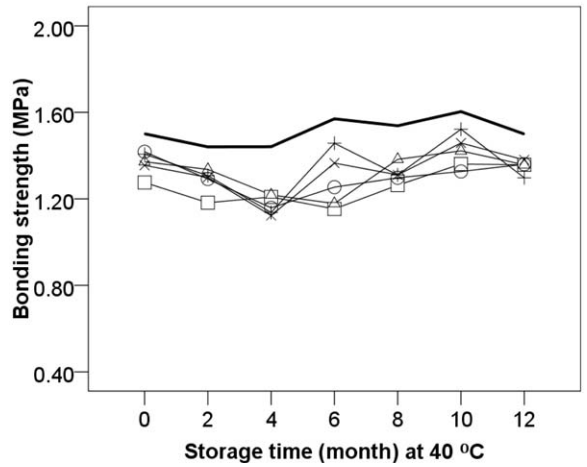
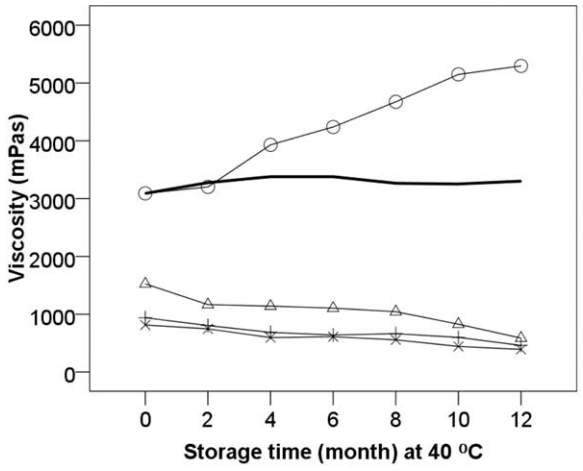


Figure 4. Changes in bonding Strength of the prototypes and control during storage at 23°C and 40°C. WS0 (□), WS1 (○), WS2 (△), WS3 (+), WS4 (×), and control (—).



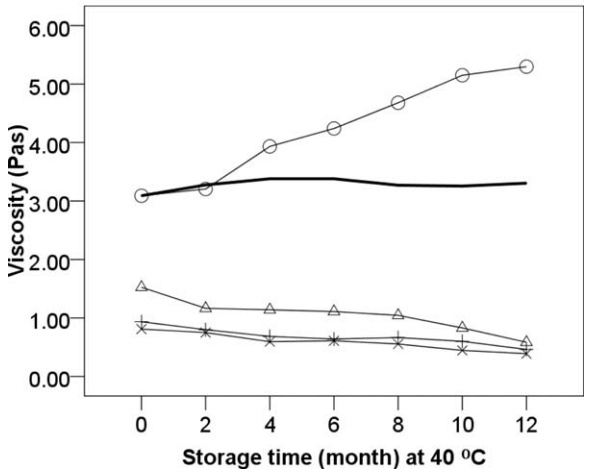


Figure 5. Changes in viscosity of the prototypes and control during storage at 23°C and 40°C. WS1 (○), WS2 (△), WS3 (+), WS4 (×), and control (—).

sucrose crystallization occurred thereon. The hydrogen bonds between sucrose and whey protein may contribute to the improvement of network-forming. Sucrose may also increase the affinity to paper fiber molecules via hydrogen bonds, thus bonding strength of the prototypes with sucrose was stronger than those without sucrose.

CONCLUSIONS

The whey protein-sucrose based paper glue is safe and environmentally friendly. The prototypes exhibited good bonding property and shelf life stable. Addition of sucrose resulted in the polymerized whey protein solution (120.0 g kg^{-1}) flowable and good consistency during storage, and increased the bonding strength of the adhesives. A wide range of viscosity could be achieved by adjusting the amounts of sucrose added. The prototypes containing sucrose are suitable to bond paper to wood, metal, and plastic. The cured glue could be rehydrated to have a fully recovered bonding strength.

ACKNOWLEDGMENTS

The financial support for this project was provided by USDA-NIFA Grant (Project # 023997).

REFERENCES

1. Bonnaillie, L. M.; Tomasula, P. M. In Whey Processing, Functionality and Health Benefits; Onwulata, C.; Huth, P. J., Eds.; Wiley-blackwell: Iowa, **2008**; Chapter 2, pp 15–38.
2. Jost, R.; Maire, J. C.; Maynard, F.; Secretin, M. C. *Int. J. Food Sci. Technol.* **1999**, *34*, 533.
3. Foegeding, E. A.; Davis, J. P.; Doucet, D.; McGuffey, M. K. *Trends Food Sci. Technol.* **2002**, *13*, 151.
4. Boye, J. I.; Alli, I.; Ismail, A. A.; Gibbs, B. F.; Konishi, *Y. Int. Dairy J.* **1995**, *5*, 337.
5. Matthey, F. P.; Hanna, M. A. *LWT - Food Sci. Technol.* **1997**, *30*, 359.
6. Forsum, E. *J. Dairy Sci.* **1974**, *57*, 665.
7. Audic, J. L.; Chaufer, B.; Daufin, G. *Lait* **2003**, *83*, 417.
8. Fox, P. F. In Adanced Dairy Chemistry, Proteins, Part A.; Fox, P. F.; McSweeney, P. L. H., Eds.; KA/PPP Publishers: New York, **2003**; Vol. *1*, Chapter 1, pp 1–48.
9. Southward, C. R.; Walker, N. J. *New Zeal. J. Dairy Sci.* **1980**, *15*, 201.

10. Imam, S. H., Gordon, S. H., Mao, L.; Chen, L. *Polym. Degrad. Stabil.* **2001**, *73*, 529.
11. Yang, I.; Kuo, M.; Myers, D.; Pu, A. *J. Wood Sci.* **2006**, *52*, 503.
12. Gao, Z.; Wang, W.; Zhao, Z.; Guo, M. *J. Appl. Polym. Sci.* **2011**, *120*, 220.
13. Liu, D.; Chen, H.; Chang, P. R.; Wu, Q.; Li, K.; Guan, L. *Bioresource. Technol.* **2010**, *101*, 6235.
14. Xu, H. N.; Shen, Q. Y.; Ouyang, X. K.; Yang, L. Y. *Eur. J. Wood Prod.* **2012**, *70*, 11.
15. Tschabold, G. L.; Mueller, D. L. **1953**. U. S. Pat. 2,624,679.
16. Wang, W.; Zhao, Z.; Gao, Z.; Guo, M. *BioRes.* **2011**, *6*, 3339.
17. van der Leeden, M. C.; Rutten, A. A. C. M.; Frens, G. *J. Biotech.* **2000**, *79*, 211.
18. Laboure, H.; Cases, E.; Cayot, P. *Food Chem.* **2004**, *85*, 399.
19. Bryant, C. M.; McClements, D. J. *J. Food Sci.* **2000**, *65*, 259.
20. deWit, J. N.; Klarenbeek, G. *J. Dairy Sci.* **1983**, *67*, 2701–2710.
21. Bryant, C. M.; McClements, D. J. *Trends Food Sci. Technol.* **1998**, *9*, 143.
22. Doi, E. *Trends Food Sci. Technol.* **1993**, *4*, 1.
23. McSwiney, M.; Singh, H.; Campanella, O. *Food Hydrocolloid.* **1994**, *8*, 441.
24. Mleko, S.; Foegeding, E. A. *J. Texture Stud.* **1999** *30*, 137.
25. Gezimati, J.; Singh, H.; Creamer, L. K. *J. Agric. Food Chem.* **1997**, *45*, 1130.
26. Gezimati, J.; Singh, H.; Creamer, L. K. *J. Agric. Food Chem.* **1996**, *44*, 804.
27. Hines, M. E.; Foegeding, E. A. *J. Agric. Food Chem.* **1993**, *41*, 341.
28. Schultz, J.; Nardin, M. In Handbook of Adhesive Technology, Second Edition, Revised and Expanded; Pizzi, A.; Mittal, K. L., Eds.; Marcel Dekker: New York, **2003**; Chapter 3, pp 53–68.
29. Wang, G.; Cheng, J.; Zhang, L.; Guo, M. *BioRes.* **2012**, *7*, 5422.
30. Wang, G.; Zhang, T.; Ahmad, S.; Cheng, J.; Guo, M. *Int. J. Adhes Adhes.* **2013**, *41*, 198.
31. Garrett, J. M.; Stairs, R. A.; Annett, R. G. *J. Dairy Sci.* **1988**, *71*, 10.
32. Rich, L. M.; Foegeding, E. A. *J. Agric. Food Chem.* **2000**, *48*, 5046.
33. Dierckx, S.; Huyghebaert, A. *Food Hydrocolloid.* **2002**, *16*, 489.
34. Kulmyrzaev, A.; Cancelliere, C.; McClements, D. J. *J. Sci. Food Agric.* **2000**, *80*, 1314.
35. Gierenz, G.; Klauck, W.; Hoefer, R.; Gruetzmacher, R. **1994**, U. S. Pat. 5,371,131.
36. Sawyer, L. In Advanced Dairy Chemistry, Proteins, Part A.; Fox, P. F.; McSweeney, P. L. H., Eds.; KA/PP Publisher: New York, **2003**; Vol. *1*, Chapter 7, pp 319–286.
37. Hoffmann, M. A. M.; Mil, P. J. J. M. V. *J. Agric. Food Chem.* **1997**, *45*, 2942.
38. Zhu, H.; Damodaran, S. *J. Agric. Food Chem.* **1994**, *42*, 846.
39. Back, J. F.; Oakenfull, D.; Smith, M. B. *Biochem.* **1979**, *18*, 5191.
40. Kamiyama, T.; Sadahide, Y.; Nogusa, Y.; Gekko, K. *Biochim. Biophys. Acta. Protein Struct. Mol. Enzymol.* **1999**, *1434*, 44.
41. Jiang, Y.; Yan, Y. B.; Zhou, H. M. *J. Bio Chem.* **2006**, *281*, 9058.
42. Giles, C. H.; McKay, R. B. *J. Biol. Chem.* **1962**, *237*, 3388.