# Adhesion Properties of Soy Protein with Fiber Cardboard

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**ABSTRACT:** Adhesion properties of soy protein isolate (SPI) on fiber cardboard and effects of press conditions, pre-pressing drying time, and protein concentrations on gluing strength were investigated. Shear strength increased as press time, press pressure, and/or press temperature increased. The effect of temperature on shear strength became more significant at high press pressure. The shear strength of the SPI adhesive on fiber cardboard decreased by 12–25% after water soaking. Shear strength increased as pre-pressing drying time increased and reached its maximal value at about 10 min. An SPI/water ratio of 12:100 (w/w) gave the highest gluing strength. The specimens showed complete cohesive failure (fiber cardboard failure) except for soaked specimens pressed at low press temperature, low pressure, and short press time. Specimens pressed at 25°C and 2 MPa for 5 min with pre-pressing drying time of 10 min and an SPI/water ratio of 12:100 (w/w) had T-peel strength and tensile bonding strength of 1.15 N/mm and 0.62 MPa, respectively, without water soaking, and 1.11 N/mm and 0.24 MPa, respectively, with water soaking.

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**KEY WORDS:** Adhesive strength, fiber cardboard, protein adhesives, soy protein, soy protein isolate, water resistance, wood adhesive.

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Soy protein-based adhesives were first developed in the 1920s. By the late 1920s, plywood adhesives formulated with soy flour had become a viable industry. The advantages of soy-based adhesives included: (i) their source was inexpensive and plentiful; (ii) they were easy to handle because of their relatively low viscosity; (iii) they could be applied using both hot and cold presses; and (iv) veneer with moisture levels as high as 20–35% could be glued without splitting (1). However, soy protein adhesives had relatively low gluing strength and water resistance. As a result, they were substantially replaced by synthetic adhesives by the 1960s. In recent years, increased demand for adhesives, limited resources for petrochemical products, and increasing concern about environmental problems have prompted the development of environmentally friendly adhesives from renewable, inexpensive resources. Therefore, as a biodegradable adhesive, soy protein gained attention again, and efforts have been made to improve its adhesive strength and water resistance (2–4). Recently, some researchers have explored the feasibility of using soy flour for adhesives because the ingredient costs for adhesives based on soy protein isolate (SPI) are higher than commercial phenol-formaldehyde and urea-formaldehyde adhesives. However, SPI-based adhesive has high adhesion strength (5).

The use of soy protein adhesives was primarily for plywood (6). Generally, soy protein was dispersed in alkali for application. Alkali breaks the strong internal hydrogen bonds of the coiled protein molecules and causes soy protein molecules to unfold so that all their polar groups are available for adhesion to wood (6). In order to reduce the viscosity of soy protein adhesives, salts and disulfide-bond-cleaving agents were used (3). Other chemicals and methods also were used to modify the soy protein structure/conformation to improve the strength and water resistance of the adhesives (2,4,5). Soy protein also was blended with other proteins, such as animal blood and casein, to achieve useful adhesive characteristics (6).

Paperboard has been widely used as packaging materials, containers, tubes, and cartons. The major adhesives used in the paperboard industry have been thermoplastic and thermosetting resins (7). These synthetic resins are difficult to dispose of after use and may result in serious environmental problems. Migration of some components, which may be harmful to humans, from adhesives used in paperboard packaging for foodstuffs was recently observed (8). Although starch- and dextrin-based adhesives also have been used in the paperboard industry, their water resistance needs to be improved in many applications (9). The objectives of this research were to investigate the adhesive properties of soy protein in fiber cardboard application and to examine the effects of press conditions, pre-pressing drying time, and protein concentration on gluing strength and water resistance.

## MATERIALS AND METHODS

*Materials.* SPI (PRO-Fam 970), prepared by acid precipitation and containing more than 90% protein (dry basis), was provided by Archer Daniels Midland (Decatur, IL). Fiber cardboard (V2S) with dimensions of 152 (width) × 89 (length) × 2.5 mm (height) was provided by the U.S. Army (Natick Soldier Center, Natick, MA).

*Sample preparation.* Ten grams of SPI powder were suspended in 100 mL distilled water at room temperature and stirred for 6 h. Then the SPI slurry was brushed onto one end of each piece of fiber cardboard (152 × 89 mm) (Fig. 1A) until the entire area was completely wet (about 1.5 ± 0.1 mg/cm$^2$, based on dry protein weight). The amount of slurry on each piece was controlled by using a consistent brushing procedure to minimize variation. The area of application on each end was 152 × 25 mm. The two pieces of slurry-brushed fiber

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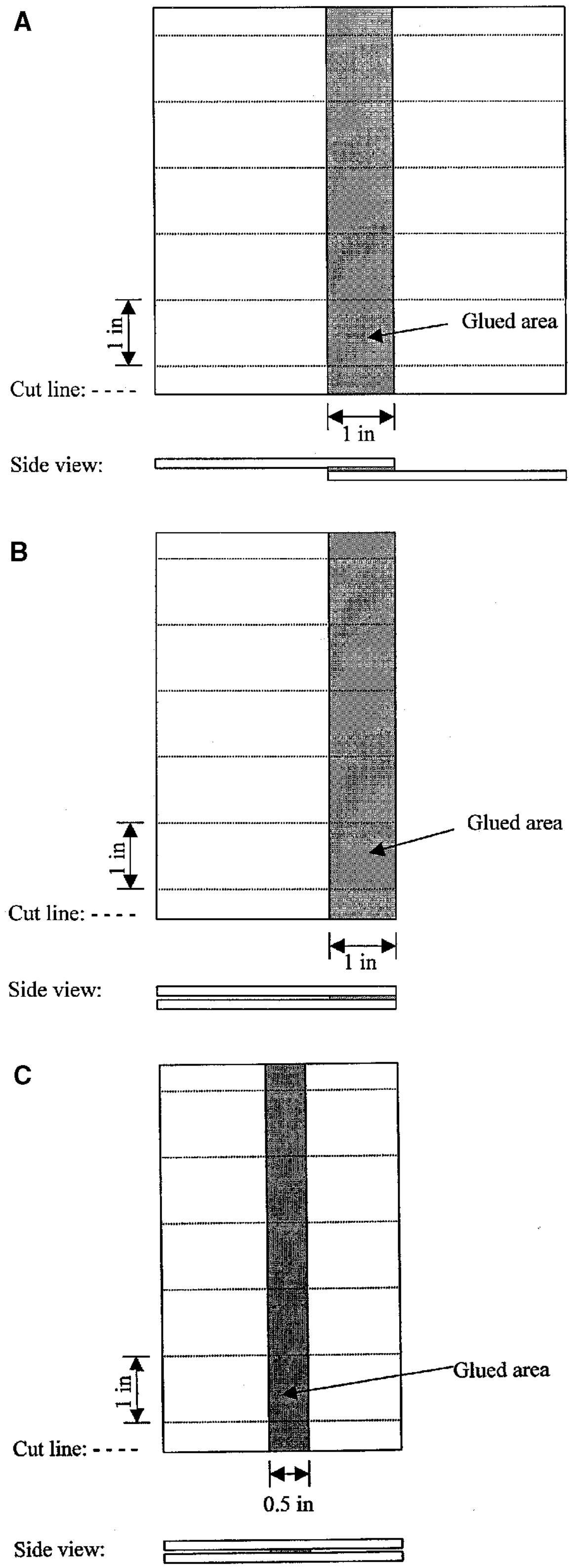


**FIGURE 1.** Fiber cardboard construction for tests of (A) shear strength, (B) T-peel strength, and (C) tensile bonding strength.

cardboard were allowed to rest at room temperature for about 5 min and then were assembled and pressed at various press conditions using a hot press (Model 3890 Auto "M"; Carver Inc., Wabash, IN). Press temperatures were 25, 70, and 100°C; pressures were 0.5, 1, 2, and 4 MPa; and times were 1, 2, 5, and 10 min. Similar procedures were followed for tests of tensile bonding strength, but the SPI slurry was brushed on a strip down the center.

To study the effect of pre-pressing drying time of the SPI adhesive on performance, the two pieces of slurry-brushed fiber cardboard were allowed to rest at room temperature for 2, 5, 10, 15, and 20 min and then were assembled and pressed at 25°C and 2 MPa for 5 min. To study the effect of SPI concentration on adhesion performance, various amounts of SPI (5, 8, 10, 12, and 15 g) were suspended in 100 mL distilled water and then applied as described above. The pre-pressing drying time was 10 min, and press conditions were 25°C, 2 MPa for 5 min. After pressing, the fiber cardboard specimens were preconditioned at 23°C and 50% relative humidity (RH) for 48 h and then were cut into five 25 mm-wide specimens (Fig. 1). These were further preconditioned for 5 days in a conditioning room (23°C, 50% RH).

*Shear strength.* Specimens of fiber cardboard for shear strength testing were prepared according to the modified TAPPI UM633 method (10) (Fig. 1A). An Instron universal testing machine (model 4465; Canton, MA) with a crosshead speed of 25 mm/min was used. The shear strength at maximum load was recorded, and each value is the mean of 8–10 specimens.

*Water resistance.* For water resistance testing, 8–10 of the 25-mm-wide specimens for each set of conditions were soaked in tap water at 23°C for 24 h. Then they were dried at 23°C and 50% RH for 5 d, and shear strength was tested as described above.

*T-peel strength.* Specimens of fiber cardboard for T-peel strength testing (glued area is perpendicular to tensile direction) were prepared according to the modified TAPPI UM633 method (10) (Fig. 1B). The Instron universal testing machine with a crosshead speed of 152 mm/min was used. The T-peel strength at maximal load was recorded, and the values presented are averages of 8–10 specimens for each experiment.

*Tensile bonding strength.* Specimens of fiber cardboard for tensile bonding strength testing were prepared according to the modified TAPPI UM559 method (11) (Fig. 1C). The specimens were glued to the grips of the Instron with epoxy resin (Everfix 644; Fibre Glass Evercoat Co., Inc., Cincinnati, OH) and then were tested at a crosshead speed of 1 mm/min. The tensile bonding strength at maximal load was recorded, and the values presented are averages of 8–10 specimens.

*Statistical analysis.* All values of mechanical properties are means of 8–10 replications. In order to avoid crowded graphs, the average standard deviation of the data presented in each figure has been calculated, and the error bar given in the figure is twice the average value.

## RESULTS AND DISCUSSION

*Effects of press conditions on shear strength.* The shear strength test is a widely used method for estimating performance characteristics of adhesives. Shear strength increased with increasing press time and pressure (Fig. 2). The shear strength also increased as press temperature increased (Fig. 3), especially for the sample pressed at 2 MPa.

When the SPI slurry was applied on the fiber cardboard, it first spread and wetted the surface. Then, the polar and apolar groups of SPI molecules interacted with the fiber cardboard surface by physical and/or chemical forces. The SPI molecular chains also may have penetrated into the fiber cardboard surface through the porous structure, and this would have enhanced interactions between the SPI adhesive and the surface. Meanwhile, water evaporated, allowing adhesion between the two pieces of fiber cardboard. Therefore, there are two important aspects for better gluing: (i) intimate contact at the interface between the fiber cardboard and SPI adhesive and (ii) immobilization of the SPI adhesive. As press pressure increased, contact at the interface between the fiber cardboard and SPI adhesive increased, and as press time increased, immobilization of SPI adhesive was enhanced resulting in higher shear strength. In addition, increased press time, as well as any chemical interaction at the interface between the SPI adhesive and fiber cardboard, promoted penetration of SPI molecules into the fiber cardboard surface.

Increased press temperature not only enhanced immobilization of the SPI adhesive but also increased the possibility of chemical reactions at the interface between the SPI adhesive and fiber cardboard. High press temperature also increased the shear strength of the soaked samples (results not shown), and all specimens pressed at 100°C showed complete cohesive failure (fiber cardboard failure).

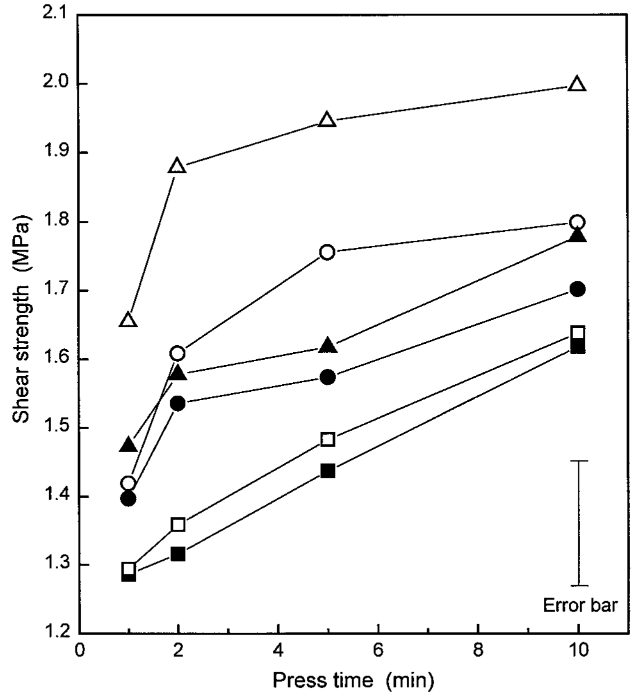


**FIG. 3.** Effect of press time and temperature on shear strength of fiber cardboard specimens glued with soy protein isolate adhesives. Press conditions: 1 MPa at 25°C (■), 2 MPa at 25°C (□), 1 MPa at 70°C (●), 2 MPa at 70°C (○), 1 MPa at 100°C (▲), and 2 MPa at 100°C (△).

*Water resistance.* Water resistance is an important property that determines glue durability. Water soaking specimens for 24 h greatly decreased shear strength (Fig. 4). For unsoaked

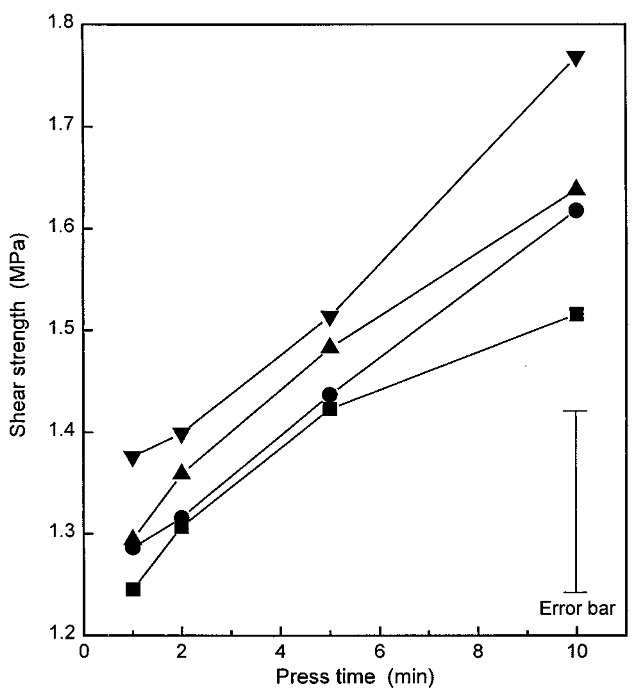


**FIG. 2.** Effects of press time and pressure on shear strength of fiber cardboard specimens glued with soy protein isolate adhesive. Press temperature was 25°C. Press pressure: 0.5 MPa (■), 1 MPa (●), 2 MPa (▲), and 4 MPa (▼).

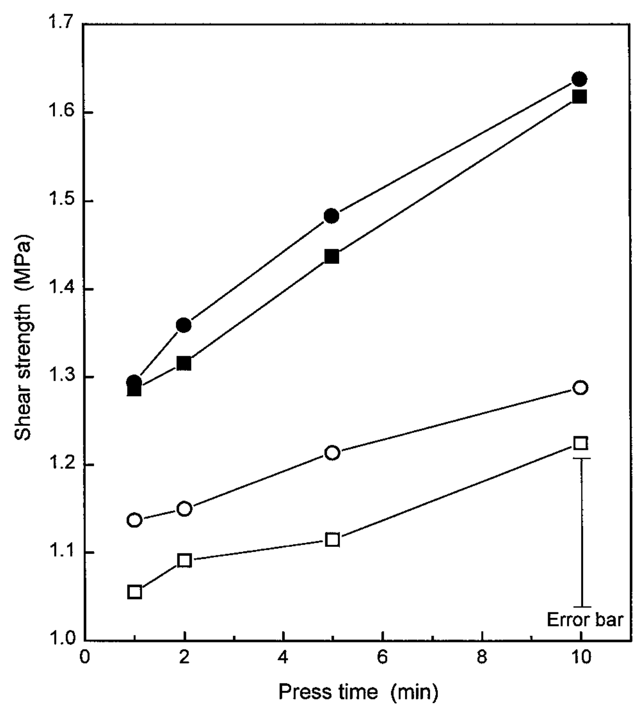


**FIG. 4.** Effect of soaking on shear strength of fiber cardboard specimens glued with soy protein isolate adhesives. Pressed at 25°C and 1 MPa (■ □), 2 MPa (● ○), without soaking (■ ●) and with soaking (□ ○).

fiber cardboard samples, shear strength was generally greater than 1.25 MPa. However, the shear strength of the soaked samples was as low as 1.05 MPa. Press pressure had more effect on the shear strength of the soaked samples than that of unsoaked samples. The difference in shear strength between samples pressed at 1 and 2 MPa was larger with water soaking. All unsoaked specimens showed complete cohesive failure in shear strength testing. However, some soaked specimens showed partial adhesive failure, which meant that the interface between the SPI adhesive and fiber cardboard failed in some of the glued area during the test. The percentage of specimens that showed partial adhesive failure was affected by press conditions and decreased as press time and pressure increased. At 4 MPa pressure for any press time or at 2 MPa for press time longer than 1 min, all soaked specimens had complete cohesive failure. During water soaking, water molecules penetrated into the glued area between the two glued pieces and interacted with SPI molecules, resulting in reduced interaction between the SPI adhesive and the fiber cardboard. However, it was difficult for water molecules to penetrate into the gap in specimens that were pressed at high pressure and for a long time, because the two pieces of fiber cardboard were in closer contact with each other. Pressing at high pressure and for a long time also enhanced the interaction between the SPI adhesive and fiber cardboard and hence reduced the interaction between the SPI adhesive and water molecules.

*Effect of pre-pressing drying time on shear strength.* Pre-pressing drying time is the time that the brushed fiber cardboard rested at room temperature prior to assembling. It is an important parameter affecting the wetting and penetration of the SPI adhesive on the fiber cardboard. The shear strengths of the samples pressed at 25°C and 2 MPa for 5 min (Fig. 5) increased greatly as pre-pressing drying time increased from 2 to 10 min. Further increasing pre-pressing drying time from 10 to 20 min slightly decreased shear strength. Shear strengths of the soaked samples exhibited a similar trend, except that the values were much lower than those of the unsoaked samples. Short pre-pressing drying time resulted in insufficient wetting of the fiber cardboard by the SPI adhesive and less penetration of the SPI molecules. However, long pre-pressing drying time may lead to over-evaporation of water, resulting in precuring and/or difficult mixing of the SPI on the two pieces of fiber cardboard after assembling and consequently may lead to low gluing strength.

*Effect of SPI concentration on shear strength.* The shear strength of SPI adhesive on fiber cardboard was affected significantly by the ratio of SPI/water (Fig. 6) and reached a maximum value at 12:100 (w/w). After that point, the shear strength sharply decreased. The shear strengths of soaked samples showed a similar trend and had maximal values at the same ratio. At lower SPI concentrations, the interaction between the SPI adhesive and fiber cardboard increased as concentration increased. However, at higher SPI concentrations, for example at the SPI/water ratio of 15:100, the adhesive slurry became highly viscous, resulting in poor flowability over the fiber cardboard surfaces.

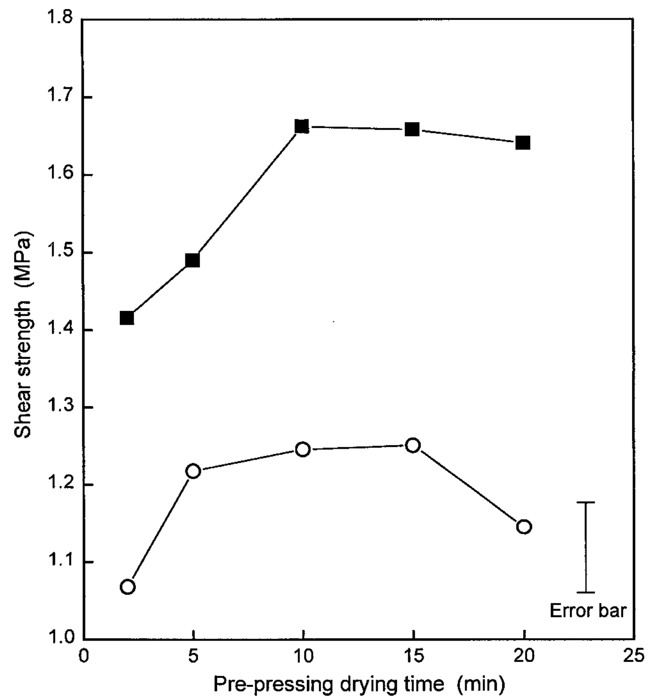


**FIG. 5.** Effect of pre-pressing drying time on shear strength of fiber cardboard glued with soy protein isolate adhesives: unsoaked (■) and soaked (○). Pressed at 25°C and 2 MPa for 5 min.

*T-peel strength and tensile bonding strength.* T-peel and tensile bonding strengths are two other important methods for estimating adhesion properties of adhesives by measuring

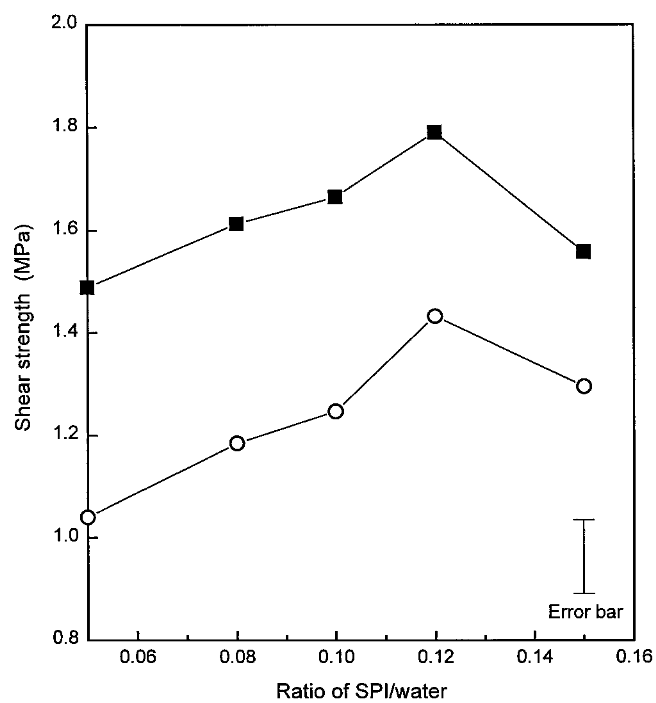


**FIG. 6.** Effect of soy protein isolate (SPI) concentration on shear strength of fiber cardboard glued with SPI adhesives: unsoaked (■) and soaked (○). Pressed at 25°C and 2 MPa for 5 min with pre-pressing drying time of 10 min.

gluing strength in different force directions. The specimens were pressed at 25°C and 2 MPa for 5 min with 10 min prepressing drying time and 12:100 (w/w) SPI/water ratio. The mean T-peel strength of the unsoaked sample was 1.15 ± 0.12 N/mm, and that of the soaked sample was slightly lower (1.11 ± 0.08 N/mm). The mean tensile bonding strength of the unsoaked sample was 0.62 ± 0.09 MPa, whereas that of the soaked sample was much lower (0.24 ± 0.02 MPa). However, all the specimens showed complete fiber cardboard failure in these experiments.

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