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REVIEW ARTICLE

Protein structures dictate their adhesive performance

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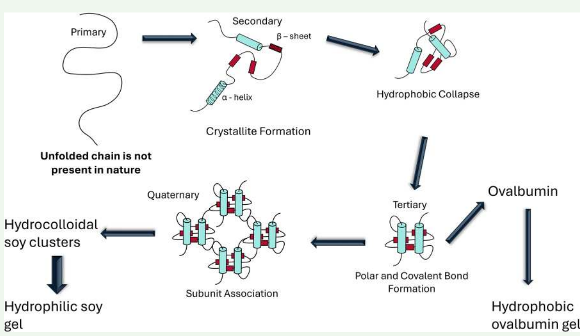
ABSTRACT

Proteins were useful as adhesives for wood bonding, including those derived from animals (collagen, casein, ovalbumin, and blood), and plants (soy, canola, and cottonseed). These proteins have different structures that dictate their wood adhesive performance. Hydrolyzed collagens from animal hides and bones are hot-melt adhesives suitable for paper applications with durability improved by crosslinking. Soy flour adhesives, even with a crosslinker, are amphiphilic, giving moderate wet-strength wood bonds, while with ovalbumin (egg whites) curing exposes hydrophobic surfaces leading to high bond strength, both wet and dry. The presence of carbohydrates in soy flour contributes to lower wet bond strength but oxidizing these carbohydrates to reactive aldehydes leads to greatly improved wet bond strength. Adhesive research has concentrated on expensive commercial soy protein isolates which provide greater wet bond strength, due to jet cooking, compared to affordable soy flour. We propose this strength is primarily due to an increase in the size of the pre-gel protein structures. While soy adhesive literature generally describes soy proteins as elongated, spaghetti-shaped molecules, the model of a hydrocolloidal gel formed from sticky rice-like globular proteins as described in the food and general protein literature is more consistent with the properties of soy adhesives.



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Proteins; adhesives; morphology; wet strength; rheology; hydrocolloid

1. Introduction

Proteins have typically been used as wood adhesives (Adhikari *et al.* 2017). Although most proteins are made using the same 20 amino acids, the proportion and sequence of amino acids vary. This leads to an enormous number of possible protein structures; in addition, many proteins are glycosylated, and some, such as casein and ovalbumin, are also phosphorylated. Compared to other natural polymers, proteins are unique due to the wide variety of side chain groups including hydroxyl, thiol, amino, carboxylic, phenolic, aromatic, and aliphatic groups. Each protein has the correct sequence for its specific role in nature. As an example, enzymes need to have a pocket to let the substrates enter and products exit, and the pocket is lined with amino acid side chains with the proper catalytic properties and orientation to carry out the target reaction. These pockets can be modified by other groups attaching to the enzyme to open or close the pocket for control of the enzymatic activity (Jakubowski and Flatt 2022a, Copeland 2023, Newman 2023). Non-enzyme proteins also have specific structures suited to their utility in nature. Only a few of these proteins have the necessary structure to provide high adhesive strength and adhesion to the substrate, in addition to being readily available in sufficient quantity and price for industrial adhesive applications.

Proteins are an attractive target for wood adhesives because replacing the current fossil fuel-based materials would result in completely biobased products. The many adhesives used in bonding wood are water-borne or solvent-free, which is good environmentally, and fits with the water dispersibility of proteins. The adhesive performance criteria are generally not highly challenging, except for meeting some type of water exposure test. The water exposure causes wood to swell, putting strong forces on the cured adhesive. The most thorough references on protein adhesives for wood are covered by two papers in a series on bio-based adhesives (Dunky 2020a, 2023), and several chapters in a book on "Bio-based Wood Adhesives: Preparation, Characterization, and Testing" (Adhikari *et al.* 2017, Cheng and He 2017, He 2017, He and Wan 2017, Li *et al.* 2017, Qi *et al.* 2017, Shi *et al.* 2017). Because these reviews covered all the numerous

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potential protein sources, we focus here only on those that seem to have commercial viability or have distinct performance attributes. What has been missing from the adhesive literature is a scientifically correct discussion of the protein structure after bonding. The incorporation of information from food processing and general protein literature are important in this paper. The food science literature is especially relevant since the available commercial proteins for wood bonding are manufactured using food application specifications. However, the food literature does not examine the physical and chemical properties relevant to adhesive uses. Some of these issues have also been addressed in two recent manuscript submissions (Frihart and Hunt 2024, Frihart 2025). Unfortunately, it is difficult to reproduce literature procedures for soy adhesives since the specific protein products are often not indicated despite manufacturers offering a variety of flours, concentrates, and isolates with different process histories. For example, Cargill offers soy flours of different protein dispersibility indices and particle size, while ADM offers different soy protein isolates with variations in solubility, water retention, and viscosities. To explain protein adhesive properties, it is important to understand their structure and how this influences their properties when exposed to water, especially for hydrocolloidal proteins.

2. Animal proteins

Animal by-products have always been a good source of proteins for adhesives, with wood as a common substrate. Early humans used many parts of the animal killed for their meat, and processed bones and hides to make collagen adhesives (Fay 2021).

2.1. Collagen

Wood and other substrate bonding started out in pre-recorded history, and protein-based adhesives were some of the bonding agents used at that time (Lambuth 2003, Fay 2021). Nature uses the extra-cellular collagen, the most abundant protein in mammals, for structural elements: cartilage, tendons, ligaments, and skin, due to the structural strength of their linear (extended) structure. Adhesives developed by the ancients included collagen-based adhesives, which involved the use of alkali (probably wood ashes) or acids to hydrolyze the collagen from bones and hides into soluble polymers. These proteins are then separated from the other organics and inorganics by filtration of the aqueous fraction (Hubbard 1977, Pearson 1994, Schellmann 2007, Fay 2021). Other collagen sources included fish scales and skin. These are commonly referred to as animal glues, with the taking of old horses to the glue factory having some historic validity. For stability these adhesives were dried and converted to a powder that could be resolubilized by the user just prior to application to avoid further degradation of the protein that occurs in aqueous solutions.

Collagen adhesives were often used as hot melt adhesives by dispersing collagen powder to make a gel using hot water denaturation. The adhesive solidifies on cooling and water loss to the substrate after application (Hubbard 1977, Pearson 1994, Schellmann 2007, Shoulders and Raines 2009). These proteins are linear helical structures caused by high levels of the amino acids proline and hydroxyproline often as three intertwining chains. The prolines, due to their cyclical structure, naturally cause the protein molecule to bend into a helical shape. Even when in solution, the protein retains its helical structure and during setting of the adhesive, the protein forms extended triple helices with overlapping segments. The durability problem with collagen is that the hydrogen bonded triple helix separates in the presence of water unless the proteins are crosslinked (Hubbard 1977, Pearson 1994, Schellmann 2007). This water sensitivity limited their use to mainly paper applications, but they were some of the original wood adhesives until they were displaced by other proteins, such as casein, blood, and soybean. However, they were used in fine furniture and are still used in wood instruments since the glued joints can be disassembled without damaging the wood. Today, collagens are not used in wood or paper products except for restoring antiques and musical instruments (Schellmann 2007). The viscosities of linear collagen adhesives, even after extensive chain cleavage, are much higher than most plant protein adhesives, which when denatured, typically adopt a compact globular shape.

Synthetic adhesives, such as poly (vinyl acetate) and polyurethanes, have replaced most of the collagen market since these are shelf-stable dispersions and do not require heating as many collagen adhesives do. Collagen is still an important protein, but now the research emphasizes cosmetic applications, medical adhesives, food products, and biobased food wrapping (Shoulders and Raines 2009, Streeter and de Leeuw 2011, Avila Rodríguez *et al.* 2018, Mihalca *et al.* 2021).

2.2. Silks and other linearly extended proteins

There are other extended proteins such as silks, found in cocoons and spider webs. A key characteristic of the silks is high tensile strength, but also good flexibility (Iizuka 1985). These fibers are made of parallel protein strands providing excellent mechanical properties; in the case of spider webs, they also have a sticky surface to hold the web together, bond to supporting surfaces, and trap insects (Hu and Kaplan 2011). These silk fibers would be viable adhesives if they could be made into fluids for adhesive application and were available in sufficient quantities at a reasonable price. Considering that their structure makes these proteins strong and flexible while also being good adhesives, their structure may lead to some bioinspired or biomimicry research that overcome the limited availability and high cost of these proteins.

2.3. Globular proteins

Most proteins are globular in shape due to the proteins' hydrophobicity, although they are not uniform spheres and normally have some hydrophobic groups on the surface. This spherical structure allows shielding most of the hydrophobic groups from the aqueous environment in cells and water-borne adhesives. Examples are animal proteins, like milk and egg proteins, and most plant proteins, including soybean and canola. There is one arrangement known as the native structure which is considered to be the most compact configuration. Denaturation (any structure that is not native) continues to be an area of confusion in that the literature does not distinguish well between the transient unfolded structures and refolded lower energy states. Denaturation includes processes as severe as partial depolymerization or changes in the secondary structures of α-helices and β-sheets, to as mild as alteration of the tertiary or quaternary structures. To allow for freedom of movement of the primary chains, the quaternary and tertiary structures need to be disrupted. This ranges from swelling of the protein to altering the alignment of sections of the backbone structure. The unfolding to a more linear, fully hydrated polymer has been taken to be a stable state of the denatured protein, as illustrated in the protein wood adhesive literature. This is incorrect; rather the expanded state is a transient transition state between folded globular structures (Frihart and Hunt 2024, Frihart 2025). These globules can then form surface aggregates into pre-gels that then form larger gel structures upon curing like beads on a chain (Damodaran 1988, Djabourov 1991, Doi 1993, Bhattacharya and Jena 2007).

2.4. Casein and blood

Casein protein derived from milk was once a common wood adhesive, which was especially high performing when fortified with blood (Salzberg 1977, Detlefsen 1989, Bye 1990). Casein was derived as a protein precipitate from fat free milk by either biobased or synthetic acid treatment. It was mainly used in glulam products and wooden airplane construction. The longevity of a casein adhesive has been shown by the good strength retention of beams after 75 years of use when a Forest Products Laboratory building was disassembled, and the beams were analyzed for strength and integrity (Rammer *et al.* 2004, Teder and Wang 2013).

The casein proteins are not only glycosylated but also phosphorylated after protein backbone synthesis. Like other protein adhesives, the casein formulations were complex containing a number of additives including caustic and lime (Salzberg 1977, Detlefsen 1989, Bye 1990, Dalgleish 1997, Bhat *et al.* 2016). Two common modifications of casein-adhesive were adding blood to provide greater wet strength and soybean to lower the adhesive cost (Salzberg 1977, Detlefsen 1989, Bye 1990). These adhesives were made by dissolving the sodium caseinate and adding chemicals such as lime, because the divalent calcium ion replaces sodium to crosslink the protein molecules during curing. Like most of the protein-based adhesives, the dispersions needed to be made at the application facility due to poor shelf-stability; storage while wet is limited by curing and biological decay of the protein.

The casein adhesives provided better water resistance than soy adhesives when both were used at high pH conditions. There has been some recent research on casein and blood (Vnucec *et al.* 2017). While caseins are stable powders, blood needs to be carefully dried at low temperatures to prevent formation of a gritty, low solubility material (Lambuth 1977, Detlefsen 1989, Vnucec *et al.* 2017). Although casein and caseinates are commercially available, the cost, limited availability, and human food usage limit future investigation. Likewise, blood has been used for a long time as an adhesive, but supply, cost, biohazard concerns, and need to formulate each lot pose barriers for its use. Its performance also depends on the type of animal, time of year, and time after collection (Lambuth 1977, Detlefsen 1989, Bah *et al.* 2013).

Another protein from milk is the whey isolated as the water-soluble fraction from cheese production. Some work has been done on using whey as a wood adhesive but extensive cross-linking is needed to reduce water-sensitivity (Cayot and Lorient 1997, Gao *et al.* 2011a, 2011b).

Synthetic adhesives, such as phenolics, melamine-formaldehyde, crosslinked poly (vinyl acetate), and polyurethanes, have replaced most of the casein and other protein adhesives due to their greater storage stability and bonded product performance, especially after water exposure. Some synthetic adhesives, such as emulsion polymer isocyanates (EPI) were developed specifically for wood bonding. The phenolics, melamine-formaldehyde, EPI, and polyurethanes are used in structural applications, such as glulam, laminated lumber, and exterior plywood, and phenolics, polymeric diphenylmethane diisocyanate (pMDI) and melamine-formaldehyde are used for oriented strandboard and fiberboard. EPI is widely used for wooden I-joists. There are no protein adhesives that come close to the performance properties of these fossil fuel-based adhesives.

2.5. Ovalbumin

Another animal-derived protein is ovalbumin, the main protein in egg whites. Egg whites have historically not been used as an adhesive, although they were used as additives to paints and coatings. Ovalbumin has all the performance characteristics desired for a wood adhesive, except that the cost and availability are inadequate for large-scale wood production. What we once saw as a simple model for soy proteins, ovalbumin turned out to be totally unlike soy proteins in most properties. Ovalbumin is a monomeric protein, unlike the polymeric soy aggregates of glycinin and conglycinin, and has a low viscosity as the proteins only start to interact when the solids are above 30%. At this concentration soy proteins have a 100 times higher viscosity due to protein–protein interactions (Frihart *et al.* 2014, Frihart and Gargulak 2022). A lower viscosity is important since it allows for a higher solids-content adhesive, because less water removal during bonding speeds production rate and lowers production cost. Most of the studies were done with purified ovalbumin, but unpurified egg whites gave almost equal performance.

Despite the low interaction of the ovalbumin globules in the aqueous adhesive dispersion, on heating the ovalbumin denatures by transforming α-helices to β-sheets, which also increases the surface hydrophobicity of the protein globules for greater protein-protein interaction even in wet environments (Hu and Du 2000). This transformation, which should reduce the water absorption of the cured adhesive in the bondline, explains the very high ovalbumin wet strength compared to other proteins that we have tested (Frihart *et al.* 2014). Autoclaving the ovalbumin dispersion generates solid protein particles while autoclaving soybean dispersions only gives a thicker gel dispersion. Thus, heating the ovalbumin creates a product that is more hydrophobic probably through increased non-polar interactions between the protein globules, but the soy retains its water affinity. As shown in Table 1, the best commercial soy protein isolate (CSPI) has so many associations between protein hydrocolloidal particles that at 15% solids it is more viscous than ovalbumin at 40% solids but does reach as high of a wet bond strength as ovalbumin.

A good model to explain the observations in protein wet strength is one related to poly(vinyl acetate) (PVAc) dispersion curing mechanisms. Figure 1 illustrates the desired curing of the PVAc in which the dispersion particles come into contact and become closely packed as the water leaves the bondline. However, the particles do not fuse because the outside of the PVAc colloid is covered with poly(vinyl alcohol) (PVOH) that is added to disperse the vinyl acetate for polymerization. PVOH mainly stays on the surface and allows water to penetrate the matrix, giving poor water resistance. The same is true with most proteins with their hydrophilic surfaces but it is not true with the hydrophobic surface of cured ovalbumin, allowing particle fusion. To reduce the problem of poor water resistance, various crosslinking additives are added to the PVAc (Brown 2003). Although the modification of the soy with polyamidoamine epichlorohydrin (PAE) provides additional crosslinking strength (Allen *et al.* 2010a), it does not solve the problem of the water uptake by the soy that loosens the surface bonds between particles. As the soy proteins come closer together, the bonding of the particles depend on the attraction of the groups on the particle surface, and in addition, the limited protein mobility due to low water content at this stage of curing discourages the optimum fusion of the particles. In the case of soy flour, the carbohydrates also interfere with protein bonding and their low wet strength enhances this moisture problem of separating the protein globules. This is not much of a problem for ovalbumin since the heat during curing substantially increases the number of hydrophobic surfaces, making it hard for the water to separate the protein globules.

2.6. Barnacles and mussels

The toughness of the attachment of barnacles and mussels to various surfaces under wet conditions has inspired much research in trying to understand the chemicals involved in the adhesion of these organisms. Detailed investigations have shown a multistep process involving a wide variety of different materials; unlike the typical industrial application of a uniform polymeric adhesive between two surfaces. Research has been focused on barnacles (Kamino 2006, Liang *et al.* 2019, Li *et al.* 2021) and mussels (Silverman and Roberto 2007, Ahn 2017). Key proteins contain L-DOPA which can crosslink the proteins upon oxidation after secretion into the bondline (Burden *et al.* 2012, Liang *et al.* 2019). However, the final bonded assembly involves other polymers and chemistries to strengthen the bondline. In addition, the cost and premature oxidation leading to crosslinking of L-DOPA are hurdles to its utility in wood bonding applications.

These natural adhesives have led to a variety of research programs but there are different levels of closeness to the natural processes (Dunky 2020a). Replication is doing the same chemistry as in the biological process. Biosimilar or biomimicry is doing a similar process that is used in nature but involves changing the structure of the compounds involved; an example is the Velcro® biomimicry of burrs catching on clothing. Bioinspired is the least close to the natural process. To promote their work, some have claimed that their chemistry mimics barnacle adhesives even when it involves very different chemistries and different mechanisms.

Table 1. Properties of soy and ovalbumin adhesives with the viscosity in Pa·s and ABES lap shear strength in MPa. CSPI = commercial soy protein isolate ADM ProFam 974, PPSPI = FPL pilot plant produced soy protein isolate. The main difference is that the CSPI is jet cooked while PPSPI is not. The bonding and shear strength determinations by ASTM D-7998-15 (ASTM International 2020, Frihart and Lorenz 2020); viscosity data are from Brookfield rotational measurements. Data taken from Lorenz and Frihart (2023).

Adhesive	Viscosity (Pa·s)	Bond Strength (MPa) Dry	Wet
Soy flour, 30% solids	6.4	5.8 ± 0.1	0.3 ± 0.1
CSPI, 15% solids	108.0	9.4 ± 0.5	2.8 ± 0.3
PPSPI, 15% solids	5.7	5.2 + 0.8	1.2 + 0.2
Ovalbumin II, 30% solids	0.25	7.1 ± 0.4	3.4 ± 0.1
Ovalbumin II, 40% solids	7.2	7.8 + 0.9	4.6 + 0.2

3. Proteins from crops

Agricultural crops are a likely source of proteins for wood adhesives since they are available in large quantities and are often inexpensive (Adhikari *et al.* 2017, Dunky 2020a). Obviously, most crops are grown for human and farm animal consumption, and concerns are raised about using them for industrial applications. However, the case of using corn to make ethanol has led to increased planting of this crop to cover the increased demand, which shows that the concern about insufficient supplies has not stopped implementation of new biotechnology.

3.1. Soybean

The dominant crop in wood adhesive research has been soybeans. Soy flour/meal has good availability (348.8 million tons / year) and high protein content at a reasonable price. Also, it was the adhesive of choice for interior plywood prior to the invention of urea-formaldehyde adhesives. The soybean not only has a high protein content but also has a high content of the essential amino acid lysine (Kumar *et al.* 2024). Lysine makes a good human and farm animal food source and with its terminal amine group should be more reactive to crosslinking than other amino acids. The twentieth century soy protein adhesive used mainly soybean flour at pHs greater than 9, achieved with a combination of sodium hydroxide and lime, along with a wide variety of other ingredients added to provide divalent crosslinking (Laucks 1943, Bye 1990). However, this technology cannot compete with synthetic adhesives on performance or cost in the current market.

Soybean flours and meals are available in large quantities with the largest being the coarser meal for animal feed, which is denatured to improve digestibility. The advantage of soy is that the infrastructure exists to provide large quantities of consistent flour for wood bonding. It is being commercially used in North American interior plywood with PAE crosslinking

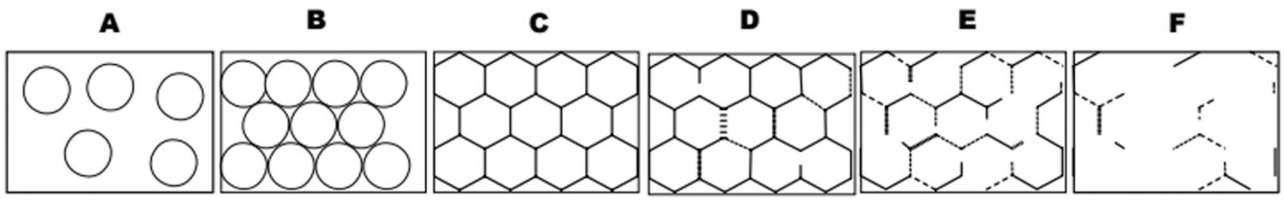

Figure 1. Theoretical steps for coalescence of dispersed adhesives from application to curing (Brown 2003). An applied dispersion (a) loses the water solvent to form a film (b), that becomes a close packing film (c). Hydrophilic surfaces on the dispersion particles hinder the expected ability of the colloid to fully coalesce in d, e, and f by fusion of the particles. This fusion does not generally occur with PVAs or soy adhesives due to the lack of sufficient hydrophobic groups on the colloid's surfaces. Thanks to Prof. Chip Frazier of Virginia Tech for the illustration.

(Allen *et al.* 2010a, Adhikari *et al.* 2017, Vnucec *et al.* 2017, Dunky 2023), but this technology has been slow to be adopted in Europe due to the higher cost for the imported soy flour and a different standard for wet plywood performance; adoption in Asia has also been slow.

Research has concentrated on the most commercially viable route, using PAE as the crosslinker (Allen *et al.* 2010a, 2010b). The commercial PAE has the advantage of being directly added to the adhesive dispersion, along with caustic to bring the pH near neutrality, and separate soy modification steps are not required. In addition, PAE is available in sufficient quantities due to its use in providing crosslinked wet strength to paper towels. Although using PAE is effective, the effectiveness is strong at low concentrations but tails off at increasing concentrations (Frihart and Lorenz 2013). The strength of soy with PAE reaching a limit emphasizes the need for obtaining greater strength from the soy self-curing. Some of the literature emphases how denaturation can lead to improved strength by unfolding the protein (Huang and Sun 2000, Sun 2005). However, other research has contradicted the reported usefulness of these denaturation chemicals (Frihart and Lorenz 2013). Also, some literature implies that the unfolded structure is a stable state rather than just a transition state (Huang and Sun 2000, Sun 2005). This denaturation concept of an extended structure also goes against the food and general protein literature in which unfolding ***and refolding*** are the steps in denaturation (Damodaran 1988, Pain 2000, Sun *et al.* 2008, Schaeffer and Daggett 2011, Dill and MacCallum 2012, Glyakina and Galzitskaya 2020, Jakubowski and Flatt 2022b), with hydrocolloidal gelation being an important property (Kinsella 1979, Doi 1993, Oakenfull *et al.* 1997, Zayas 1997, Monagle *et al.* 2006, Nishinari *et al.* 2014). To unfold, the many internal bonds that provide thermodynamic energy for the folded protein would need to be overcome by increased entropy. Thus, all these bonds (hydrogen, ionic, disulfide, and hydrophobic) in Figure 2 that provide stabilization of the folded protein would need to break to form an unfolded structure (Frihart 2025). From this unfolded transition state, new internal bonds lead to new structures for the refolding of the denatured protein.

In many food products, gelation of the aqueous protein dispersions is important for the proper development of texture (Egbert 2004). Besides the gelling properties, another important food property is good water retention for good chewability. Obviously, water retention by the "cured" adhesive gel is expected to decrease the wet strength due to water's plasticization and swelling of the soy adhesive. For good wet adhesive bond strength, hydrophobic interactions should outweigh water-sensitive polar interactions. The available commercial soy products are marketed according to their use in food products because the companies are reluctant to market a new product without well-defined properties; compared to food applications, adhesives are a small market. This raises the question as to what soy product is the best for adhesive applications. In most commercial soy wood bonds, failure is usually within the adhesive, i.e. cohesive failure in the adhesive layer. With soy containing aggregated globular structures, this means that stronger interactions between the globular particles would improve the bond strength, and that these interactions should not be water sensitive. To answer this strength question, it is important to determine the critical properties needed for making a suitable soy flour product for adhesive applications. At the current time, there are no analytical methods except for dry and wet wood bond strength that answer the performance question.

Initially, research assumed that the bonding of the affordable soy flour was mainly hindered by its high carbohydrate content, especially since several commercial soy protein isolates (CSPI) gave much better wet strength properties than flour (Frihart and Birkeland 2016). To verify this hypothesis, the native soy protein isolate PPSPI was made in the pilot plant and compared to the soy flour. The PPSPI had better wet strength than flour but fell short of the CSPI (Lorenz *et al.* 2015). As illustrated in Table 1, 15% solids PPSPI has a similar viscosity to flour at 30% solids, indicating that the protein is the biggest contributor to the flour viscosity. Although the PPSPI had improved wet bond strength, it was significantly below the CSPI. This means that wet bond strength of CSPI was dependent on some factors other than just a high protein content. When it was found out that CSPIs were jet cooked, this process was examined and found to be the explanation for the difference (Hunt *et al.* 2023). From the literature and the higher viscosity for the CSPI, it was concluded that jet cooking led to larger pre-gel structures. Thus, the degree of product gelation was related to performance (Damodaran 1988, Doi 1993, Oakenfull *et al.* 1997), but unfortunately the dispersion viscosity is also increased by the pre-gel interaction in comparison with the CSPI and the PPSPI in Tables 1 and 2. With the commercial flour, the solids content of an adhesive was limited to 35% before it became too thick to spread on the wood, and the cured product had low wet bond strength. This information shows that there is not a dramatic change in hydrophobic interparticle interaction in going from the adhesive to the cured bond for soy adhesives. In contrast,

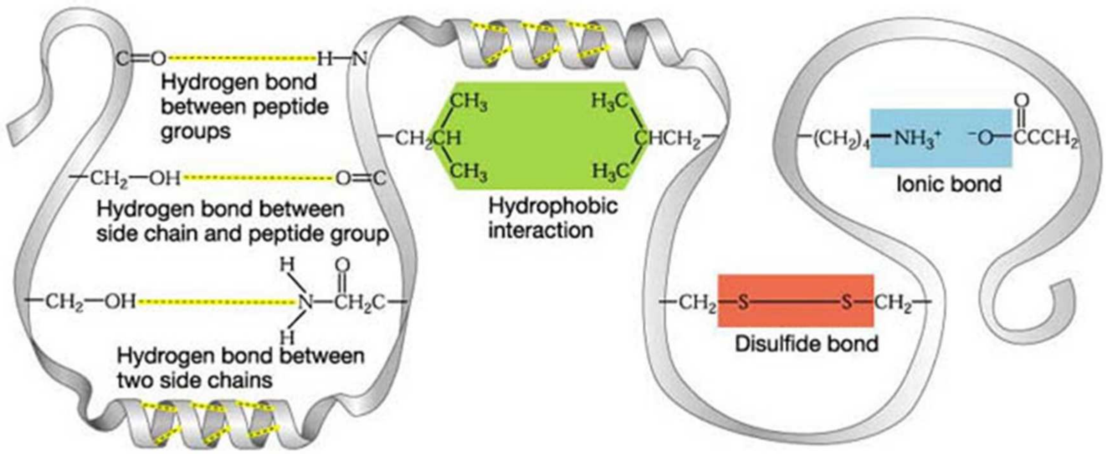

Figure 2. Internal bonding in proteins with the hydrogen and hydrophobic bonds being the most numerous.

ovalbumin had very little interaction in the uncured dispersion state (low viscosity) but had very high particle interaction in the cured product (high wet bond strength). We attribute ovalbumin's remarkable performance to the dramatic increase in surface hydrophobicity of ovalbumin upon heat denaturation (Lorenz and Frihart 2023). This shows that proteins can perform quite well as wood adhesives, if they have the necessary composition and sequence. The ovalbumin goes from having a hydrophilic surface (low particle interaction) to a hydrophobic surface with a strong water resistance bond during curing, while the soy keeps its amphiphilic (both hydrophobic and hydrophilic) surface before and after curing.

While carbohydrates in soy flour had a small but significant effect on the wet strength of the soy flour adhesives (Lorenz *et al.* 2015, Hunt *et al.* 2023), the question arose whether this detriment could be turned into a positive factor by making the carbohydrates reactive with the protein. The use of periodate and a few other oxidants change some of the hydroxyls into aldehydes, known to react with proteins (Feeney *et al.* 1982, Means and Feeney 1998, Barac *et al.* 2004, Frihart *et al.* 2019, Dunky 2020a). Periodate was found to be very effective in converting soy flour (Figure 3) into an adhesive that gave very good wet bond strength (Frihart *et al.* 2019, Frihart and Lorenz 2019). Interesting results of the periodate modification are that (1) the modified soy had no substantial improvement with higher bonding temperature, unlike soy flours, (2) improvements were fairly independent of the protein dispersibility index (a measure of protein solubility), and (3) the process was also effective with other protein sources. The treatment with periodate did not appreciably change the dispersion's viscosity but generated a dark color in the wood by the reaction with lignin. This process is not economically viable due to the cost of the periodate that is not recoverable from the adhesive.

Table 2. Degree of particle-particle interaction for the adhesive dispersion as determined by maximum solids content at working viscosity and cured particle-particle interaction indicated by wet shear strength.

Protein	Degree of uncured gelation (dispersion/cured product)	Max solids content	Wet strength by ASTM D7998 (MPa)
Soy flours	Moderate/Lowo	35%	<1
PPSPI	Moderate/Moderate	30%	2–3
CSPI	High/Good	20%	3–4
Ovalbumin	Low/ Very High	50%	>4

3.2. Other oilseeds

With high protein contents, other oil seed crops (Kumar *et al.* 2024) are likely good sources of biobased adhesives, and they fit into three distinct categories: the annual or biennial group, which includes soybeans, sunflower, groundnuts, and rapeseeds; the perennial tree crops, which include coconuts and oil palms; and embryo byproducts, such as cottonseeds and corn germ from starch production. These crops are rich in oil and often have high protein content. The main oilseeds used in human food production include soy, canola, sunflower, sesame, peanut, coconut, and oil palms. Many of these do not have an abundant commercial supply of protein that is available for industrial use, since most are used for human and animal food.

Rapeseed and its safer food version, canola, are widely available in many countries, with Canada and China being the largest growers. Canola is favored in Europe due to the climate and its local availability compared to soy. Canola adhesive research has been a small fraction of the soy research, but some data show that it should be nearly comparable to soy as an adhesive (Li *et al.* 2017, Barzegar *et al.* 2020, 2022, Dunky 2020a).

Although available in smaller volume, one USDA laboratory led by Zhongqi He, has concentrated on examining the flour and purified protein fractions of cottonseed (He *et al.* 2019). The other oilseeds have been studied less thoroughly (He 2017, Dunky 2020a).

3.3 Pulse proteins

Besides soy and other oilseeds, another large protein supply is the pulses, derived from the edible seeds of non-oily legume plants. Dry beans, dry broad beans, dry peas, chickpeas, cow peas, pigeon peas, lentils, bambara beans, vetches, and lupins fall within the pulse group that has 20–25% protein, and a carbohydrate content of 60–65%, with starch constituting the

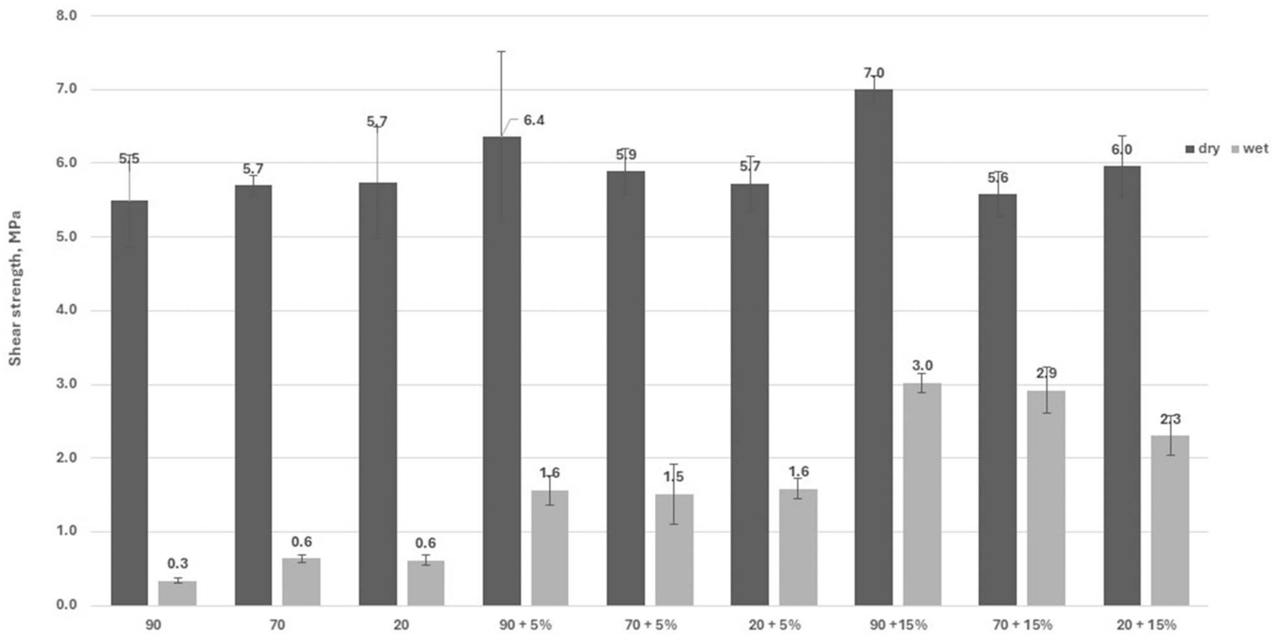

Figure 3. Effect on bond strength with different concentrations of added periodate (% periodate on soy, g/g), and flour type 90, 70, 20 = PDI of flour, with 90 PDI containing native state protein whereas 20 PDI contains denatured proteins and 70 PDI is intermediate. Data taken from Frihart and Lorenz (2019).

major fraction of the carbohydrates. The pulses have been generally lower in protein than the oilseeds, but with the consumer interest in eating more protein, efforts are being made to increase the protein content in some pulses. No substantial research on wood adhesives has been done using pulse proteins.

3.4. Cereal proteins

Cereals are the world's largest crops, and include rice, wheat, rye, oats, barley, millet, and corn. These tend to be low in protein content and high in carbohydrates, making them less suitable for adhesive bonding. Although the volume of cereals is high, isolated protein fractions are small. Except for a few scattered papers, the amount of adhesive research in this area has been low (Dunky 2020a).

Corn is one of the largest agricultural crops in the United States (Jia *et al.* 2022) used mainly as human and farm animal food, and a major source of bioethanol. There are several ways that it could be used as a wood adhesive. Research has been done on using dried distillers' grains left over from the bioethanol production, but it is not clear that the research has gotten past preliminary laboratory studies (Biancaniello *et al.* 2018, Dunky 2020a). Although there are many factories producing fermentation process residues from ethanol production, there is no standard product for adhesive production and no real characterization of the protein composition, which would be a combination of modified corn proteins and those of the fermentation organisms. Thus, product development would be mainly empirical. A main issue is the high hydrophobicity of corn proteins, which is beneficial for having good water-resistance in the bonded products but makes it hard to make aqueous adhesive dispersions (Oh *et al.* 2019). On the other hand, the corn protein zein is well characterized, and it has been made into a variety of products (Wei *et al.* 2020, Jaski *et al.* 2022, Yuan *et al.* 2022).

3.5. Other proteins

Among the potential adhesive proteins are cultured microorganisms, which would allow the design of suitable proteins for good adhesive performance, but these would be hard to make for a low-cost, high-volume market, such as wood adhesives (Dunky 2020a). On the other hand, there has been a large investment in food research that may lead to suitable ways to modify proteins for adhesive uses.

4. Prospects

Although carbohydrates are more plentiful than proteins, they have been harder to make water-resistant compared to proteins (Dunky 2020b). Most crops are grown to feed people or animals. This means that adhesive uses would have to compete with the current uses or justify additional acreage, which is difficult for a low-cost market like wood adhesives. However, increased production of corn acreage has occurred for biofuel by converting the carbohydrates to ethanol and using the residues containing the protein as animal feed (Jia *et al.* 2022). There have been some attempts at using dry distiller's grain as adhesives, but none have made it past laboratory testing. Biobased adhesives for wood bonding have received some corporate support to use the window of opportunity to replace urea-formaldehyde because of concerns about formaldehyde emissions from its depolymerization under hot and wet conditions (Frihart *et al.* 2012). However, there are many hurdles to overcome in commercializing completely biobased adhesives due the lack of suitable biobased crosslinking chemicals (Dunky 2020a).

Adhesives made from soy flour are used commercially in interior plywood in North America but have not made much progress worldwide. The soy adhesives could benefit from manipulating the soy proteins themselves to develop greater water-resistance to improve plywood performance and use less of the costly PAE. For several technical reasons, the progress in using soy protein adhesives in oriented strandboard has had little commercial progress other than as a reactive diluent for synthetic adhesives (Wescott *et al.* 2006). The technical issues are that the soy brings in too much water in the bondline that make it hard to remove the steam prior to opening the press and delaminating the board, and not enough tack to hold the unpressed board together. The addition of tannin to soy has provided some interesting results (Ghahri *et al.* 2022). Hopefully advances in making protein adhesives can be made if researchers understand the true nature of protein structures to overcome some of their deficiencies.

5. Summary

Although proteins were the original adhesives for wood bonding (soy flour for interior plywood and casein-soy-blood for structural wood products), synthetic adhesives now dominate because of their ease of manufacturing and better performance. Most proteins are made from the same common amino acids, but the bulk of properties are due to their specific sequences. The order of amino acids influences the formation of the secondary structure of α-helices and β-sheets, which greatly influence their larger scale associations such as the tertiary and quaternary structures. Besides the backbone structure, polar and hydrophobic forces also control protein morphology. Surface properties of the protein globules control the interaction with other proteins and carbohydrates. These associations lead to hydrocolloidal pre-gel particles and into gels during curing. These gels have a strong tendency to absorb water leading to poor wet bond strength as an adhesive but good performance in food applications.

Understanding the hydrocolloidal gel properties of proteins is important in understanding adhesive bond strength, especially under wet conditions. Bonding at higher temperatures and for longer times yield greater wet strength, but this is unlikely to be used commercially due to greater production costs. Although autoclaving and jet cooking soy increases the final bond strength, they also greatly increase the viscosity of the adhesive dispersion. Even these treatments are insufficient to provide acceptable wet strength in soy flour. Thus, it is important to use crosslinkers to give the soy flour sufficient wet strength. After examining other protein sources, soy flour is still the dominant commercial protein source for wood bonding. Ovalbumin/egg whites show a very attractive combination of properties, combining low protein–protein interactions when applied to the wood but very strong hydrophobic interactions after denaturation of the protein during cure, leading to high wet bond strength. Unfortunately, this approach is not commercially or economically viable.

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