sustainability

Review

A Review of Strategies to Enhance the Water Resistance of Green Wood Adhesives Produced from Sustainable Protein Sources

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Abstract: The health risks associated with formaldehyde have propelled relevant stakeholders to push for the production of non-toxic wood adhesives. Several countries including the USA, Japan, and Germany have implemented policies mandating manufacturers to reduce the emissions of formaldehyde to lower levels. Protein adhesives stand out due to their sustainability, renewable sources, and biodegradability. However, they are limited by poor wet strength and water resistance, which affect their wide acceptability in the marketplace. Researchers have developed multiple strategies to mitigate these issues to advance protein adhesives so they may compete more favorably with their petroleum-based counterparts. This review paper explores these strategies including cross-linking, modified fillers, and the removal of hydrophilic content while providing insights into the methodological approaches utilized in recent literature with a comparison of the resultant protein adhesives.

Keywords: formaldehyde; wood adhesives; protein adhesives; soybean; water resistance

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1. Introduction

The wood adhesive market is estimated to be worth over $6 billion by 2025 [1,2]. Despite the huge market, synthetic adhesives constitute a greater proportion of adhesives due to their superior adhesive performance and low cost [3]. Yet, historically, glues, resins, and composite binders were made using plant- and animal-based proteins until the mid-1900s when petroleum-based products, mainly formaldehyde, urea, and phenolics, became the mainstay feedstocks for industrial adhesives [4].

Urea–formaldehyde resins are the most utilized synthetic adhesives due to their desirable properties, including good bonding strength, clear color, low cost, fast curing, and resistance to moisture [5]. However, the major challenge associated with synthetic adhesives is the emissions of toxic substances including formaldehyde during manufacture and use [6]. Recent years have witnessed an increase in global public sensitivity and awareness about health and environmental concerns [7]. Over time, stricter laws setting new toxic emissions limits for wood panels have been implemented [8,9]. These are incorporated into standards in the United States, Japan, and Europe [7].

This increasingly tightening of regulations and concerns over pollutant volatilization and the manufacturing ecological footprint (sustainability) of these formaldehyde and phenolics-based adhesives creates an opportunity for the development of renewable and “environmentally-friendly” protein adhesives in the market [10]. However, protein adhesives only constitute a small market portion due to a number of challenges preventing their wider application [2,11]. The primary reason protein adhesives cannot be utilized in more applications is their lack of water resistance [4] and wet strength [12,13], yet there is still a growing interest in protein adhesives as evidenced by the increasing rate of publication, which is shown in Figure 1.

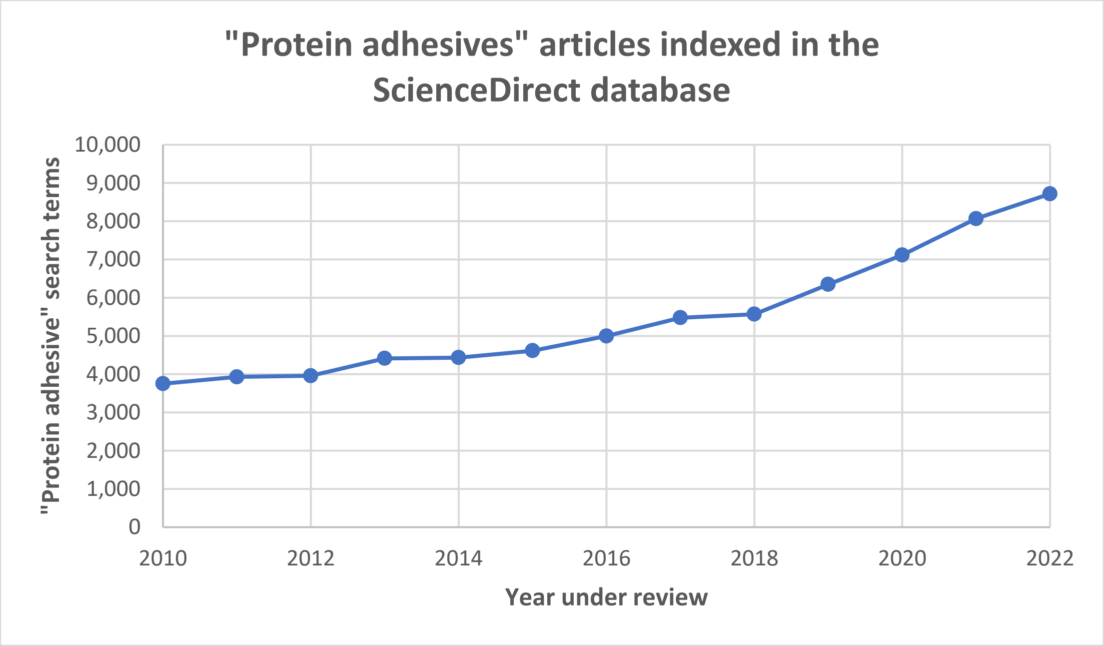

Figure 1. Publication of articles indexed by the ScienceDirect database found under the search term "Protein adhesives" since 2010.

Proteins are the most abundant class of bio-based adhesive feedstocks [4]; they are primarily linear polyamides composed of polypeptides linkages of amino acids [14]. The fundamental structure is determined by the polypeptide structure of the protein [15]. Side chains of amino acids can form bonds with several substances, including cellulose and lignocellulose [16]. Yet, certain potentially reactive side chains are not on the surface of the protein. Moreover, the loss of the adhesive bond under moist conditions occurs within the adhesive itself (cohesive failure) and not between the wood and the adhesive [16]. Proteins have to be denatured to expose the functional groups, facilitate stronger bonding to wood, and enable the aggregation of protein molecules [16].

The overall mechanism to produce protein-based adhesives is a denaturing process that involves an aqueous reaction matrix [17]. The goal of the denaturation process for the formation of adhesives is to dissolve the quaternary, tertiary, and secondary structures of the protein [17,18]. The addition of a strong base increases the pH of the proteins above their isoelectric point [19]. Under this condition, the ions in the solution interfere with the hydrogen bonds and the electrostatic dipole–dipole interactions that allow the proteins to retain their shape and higher-order structures [20]. Many of the natural, covalent cross-linking groups (including cysteine and disulfide bonds) that maintain the tertiary structure in these proteins are also vulnerable to disassociation under alkaline conditions [21]. By splitting the cross-linking groups and interfering with the secondary bonding, this denaturation process exposes the hydrophilic portions of proteins, which allows these adhesive groups to adsorb strongly onto substrates (as a glue) and filler materials (as a binder/resin) [22]. The conditions commonly used to denature proteins, especially temperature and pH modification, also catalyze the hydrolysis of the proteins [23]. This results in lower-molecular-weight protein fragments that suffer a significant loss of shear strength, especially when the chains become too short to entangle with each other [24]. Reaction time, temperature, and pH can be optimized and controlled to achieve denaturation without significant hydrolysis resulting in high-quality adhesives and a chemical foundation for other polymeric substances (such as resins and coatings) [25].

The quantity of amino acid residue and its position along the polypeptide chain are the primary determinants of the physical and chemical properties of proteins [15]. Figure 2 shows how the primary structure of proteins is naturally folded into secondary structures such as α-helices or β-sheets as triggered by the specific amino acid sequences. This process leads to the presentation of some polar groups that may associate with other polar groups to create a stable tertiary structure. Thus, several varieties of sequences can be formed based on the amino acid's number, chaining, and percentage [4]. The functional groups present in the side chains of the polypeptide structure determine the hydrophilicity or hydrophobicity of the amino acid. It offers interactions with carboxyl or hydroxyl groups in wood coupled with its cross-linking sites [26].

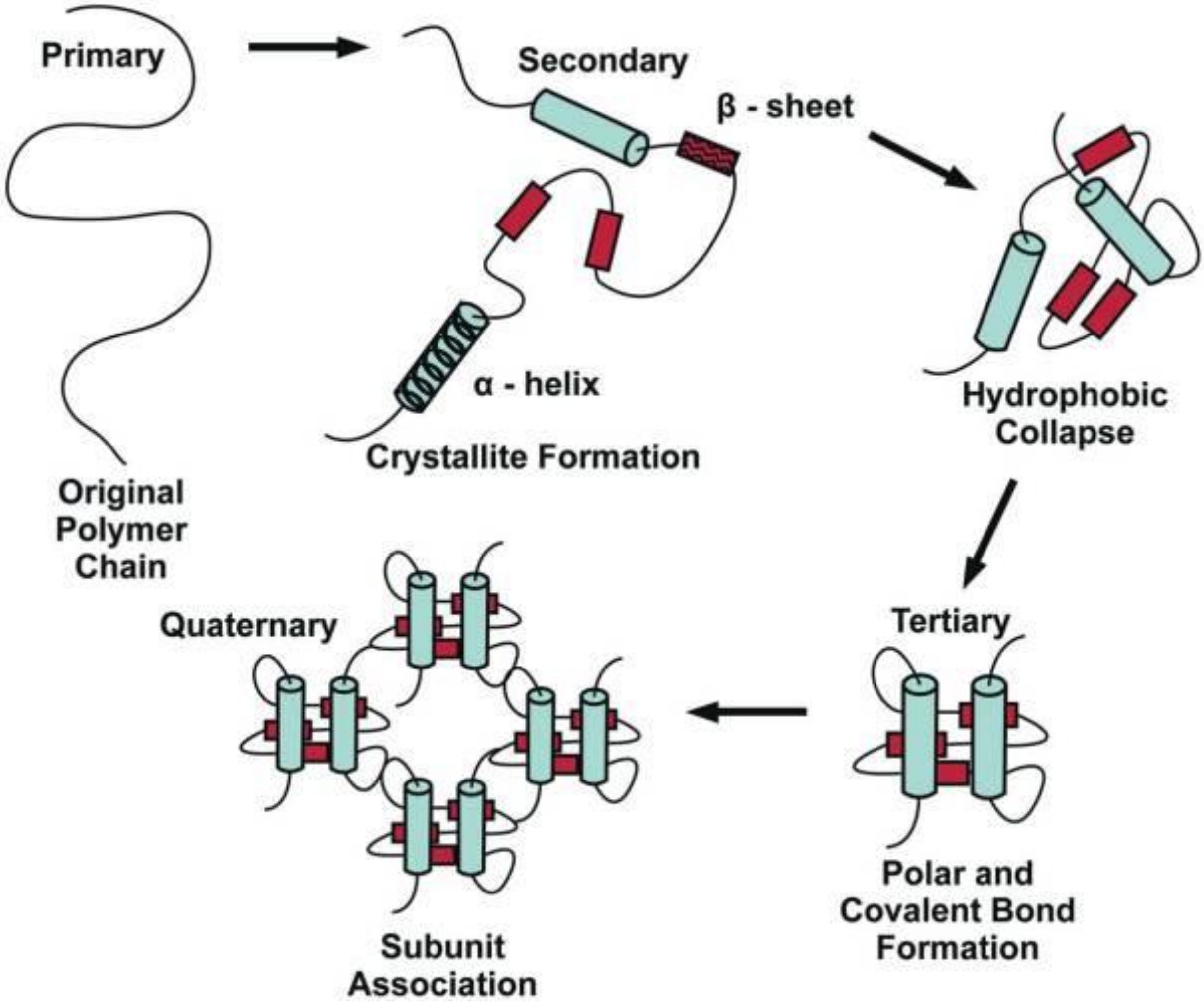

Figure 2. Illustration of levels of protein structure [18] (Reprinted/adapted with permission from Ref. [18]. 2014, American Chemical Society).

Water can degrade protein adhesives by altering the adhesive's physical and chemical characteristics. Protein adhesives are primarily composed of water-sensitive animal or plant proteins that undergo structural changes when exposed to moisture, even after conversion into adhesives, thus the adhesive also suffers from water-exposure-based structural failures [27].

When water interacts with protein adhesives, it can break the hydrogen bonds and other intermolecular interactions that hold the protein structure together [27]. This can result in the adhesive losing its strength, becoming brittle, and losing its ability to adhere to surfaces [27].

The poor water resistance is linked to the multiple polar groups of most of the proteins which make the adhesives absorb water and weaken the bond between the two surfaces [28]. Another author ascribed the reason to the large constituent of carbohydrates in soy flour which contains abundant hydroxyl groups [18]. Fourier Transform Infrared (FTIR) analysis has shown that water quickly forms hydrogen bonds with hydrophilic groups, thereby resulting in poor water resistance [29]. The reduction in the carbohydrate content decreased the hydrophilicity [29]. Protein adhesives especially soy-based adhesives are expected to witness significant growth with a compound annual growth rate of around 7% from 2019 to 2025 [11]. The expansion is due to the environmental benefits offered by the products which is the major factor for the increasing demand [30].

Protein adhesives have the potential to overtake an increased share of the wood adhesives market in the future if the problem of poor water resistance can be mitigated. There have also been some promising results in the use of bio-based adhesives based on tannins [31], lignins [32], and starch [33]. The use of these bio-based materials will also be discussed in this paper, especially as used as additives or modifiers for protein adhesives.

2. Strategies to Improve Water Resistance in Protein Adhesives

Generally, the applications of protein adhesives are limited by their low water resistance. Several strategies are utilized to mitigate or overcome this challenge, including thermochemical treatment, cross-linking networks, and using modified fillers. This paper will review contemporary strategies including bio-based and synthetic treatments.

2.1. Cross-Linking Networks

Cross-linking is a common method for creating or enhancing a network structure, hence improving the protein structural stability and adhesive capabilities of soy protein-based adhesives [34]. These materials are added to protein adhesives during their preparation or before their application [31]. Cross-linking modifications using latex, synthetic resin, polyamide-epichlorohydrin, and isocyanate have been shown to be efficient methods for increasing the water resistance of an adhesive [32,33,35,36].

2.1.1. Synthetic Cross-Linking Agent Sources

There are numerous advantages to synthetic sources including their ease of use, wide range of applications, and low cost [37]. Most of these synthetic sources are derived from petroleum and are known for releasing volatile organic compounds that are likely to contaminate the environment and offer possible health problems [38,39]. These sources also do not offer highly sustainable, green production options which have become a priority in many manufacturing sectors, including the adhesives markets.

The effects of the storage stability of polyamidoamine-epichlorohydrin (PAE) resin on the properties of defatted soybean-flour-based adhesives were explored by Gao et al. [36]. Polyamidoamine-epichlorohydrin is a water-soluble polymeric resin widely utilized in paper sheets as a wet strength enhancer [40,41]. The team explored different modifications of PAE like a polyamidoamine (PA) without incorporating epichlorohydrin and two samples of epichlorohydrin-modified polyamidoamine with different solid contents of 25% (PAE-25) and 12% (PAE-12), respectively [36].

The blending of polyamidoamine-epichlorohydrin and defatted soybean flour (DSF) revealed the cross-linking reactions of both constituents, thereby indicating that the azetidinium units of polyamidoamine-epichlorohydrin can effectively cross-link with the amines group and the DSF's carboxyl groups to form three-dimensional cross-linking structures to enhance the water resistance of the DSF-based adhesives [36]. The wet shear strength of plywood panels bonded with DSF adhesive and fresh PAE-25 and PAE-12 is 1.22 MPa and 1.30 MPa, respectively, and reduces by around 30% after 56 days of storage to 0.79 MPa, whereas that of plywood bound with adhesive DSF-PAE-12 decreases by around 17% to 1.07 MPa, thereby demonstrating the effective stability of PAE-12 compared to PAE-25 [36].

Kan et al. explored the possibility of using both polyamidoamine-epichlorohydrin and melamine–urea–glyoxal resins to formulate double-network structures through their cross-linking to soybean-meal-based adhesive with stable and required water resistance for structural use and several analyses, including Nuclear Magnetic Resonance (NMR), Thermogravimetric Analysis (TGA), X-ray Photoelectron Spectroscopy (XPS), and FTIR, which revealed the double-network structure [42,43]. Co-cross-linking between polyamidoamine-epichlorohydrin and melamine–urea–glyoxal resins by aldehyde-amino and azetidinium-amino reactions expanded the three-dimensional networks even further [42]. The advantage is that more than 35% of the expensive polyamidoamine-epichlorohydrin could be replaced by low-cost melamine–urea–glyoxal resin, thereby resulting in an adhesive at a 16% reduced cost [42]. The co-cross-linker and soybean meal adhesive had superior storage stability than melamine–urea–glyoxal and polyamidoamine-epichlorohydrin resins with a wet strength of around 0.95 MPa after 60 days [42].

The impacts of cross-linked phenol–formaldehyde resin and the reaction between glutamic acid and hydroxymethyl phenol (HPF) on the performance of soy-based adhesives were studied by Wu et al. [33]. The addition of HPF increased the dry shear strength from 0.98 MPa up to 1.81 MPa and the wet strength from zero to 1.18 MPa [33]. HPF-containing formulations prepared by the researchers had a higher content of hydroxymethyl groups, which are the main reactive groups that affect the adhesive properties, compared to a normal PF resin [38]. The results from several experiments based on resultant products derived from model compounds revealed that the soy-protein-based adhesive modified with phenol-to formaldehyde-based resin exhibited improved water resistance, indicating a reaction between soy protein and phenol–formaldehyde resin [33]. Although phenol–formaldehyde resin is hazardous, this research gave light on how aldehyde and its derivatives modify soy-based adhesives.

The introduction of wheat gluten protein hydrolysate for the preparation of non-toxic protein-based adhesives was evaluated by Xi et al. [44]. Glutaraldehyde was modified to prepare the adhesive [44]. Also, they explored polyethyleneimine as the cross-linking agent. Thermomechanical analysis showed that the modulus of the adhesive increased due to the addition of PEI, which also increases the wet shear strength from 0.91 MPa to 2.02 MPa. Glutaraldehyde modification also increased the dry strength from 0.76 MPa to 1.14 MPa. The authors attributed the motivation to use these materials to the water solubility properties of polyethyleneimine because it is very reactive and has been discovered to improve wet strength in paper preparation [45,46]. Aside from improving the bonding performance of the adhesives, polyethyleneimine improves its storage modulus and toughness.

Lei et al. studied the effect of several cross-linkers (melamine–formaldehyde, epoxy resin, and their mixture) on the water resistance of soy protein adhesives. Aside from preparing an adhesive product with excellent water resistance, they also intended to improve and optimize the composition of components of multiple cross-linkers [43]. The results showed that all three cross-linkers enhanced the performance of the protein adhesive [43]. The mixture of both epoxy resin and melamine–formaldehyde cross-linker stood out from the individual product and was confirmed by different mechanical tests as having the best water resistance of the formulation systems tested [43]. The improvement of water resistance modified by these cross-linkers might be due to the reaction between melamine–formaldehyde and –NH, as well as the reaction between epoxy and –OH [43]. Thus, this research was able to prepare an adhesive with excellent water resistance as well as effectively optimize the proportion of its components.

Zeng et al. introduced a tailor-made cross-linker containing flexible long chains and multiple epoxy groups [47]. The multiple epoxy groups joined together with the hydrophilic groups to design dense cross-linking networks, thereby improving the water resistance and wet bonding strength of the protein adhesive [47]. Another cross-linker bis(hexamethylene) triamine was synthesized by adding triethylenetetramine and 1, 6 hexadiol glycidyl ether to a flask followed by continuous stirring under controlled temperature [47]. Additionally, the flexible long chains contributed to the enhancement of the toughness of the adhesive, which increased the dry bonding strength and wet bonding strength from 1.11 MPa and 0.22 MPa to 2.79 MPa and 1.12 MPa, respectively [47]. Thus, this research provided a novel strategy for developing environmentally friendly material and bio-based adhesives with excellent water resistance and bonding strength.

Previous research has shown that developing a hyper-branched, cross-linked structure considerably raises the adhesive's cross-link density in the cured adhesive, thereby blocking moisture intrusion [48]. Zhang et al. improved on this research and subsequently built a flexible long-chain biopolymer with a hyper-branched structure and used it to create a unique, high-performance soymeal adhesive with a highly cross-linked structure and wet strength of 1.00 MPa, which is around 300% higher compared to soymeal adhesive with a wet strength of 0.22 MPa [49]. The team synthesized a long-chain dialdehyde starch grafted with hyper-branched polyamide to produce hyper-branched amino starch [49]. Thus, the resulting adhesive was found to be economical with great water resistance and outstanding mechanical qualities, and it has been successfully put to industrial pilot-scale manufacturing with the resulting plywood meeting the standards for interior usage [49]. Wheat flour adhesives can be made more water-resistant through the simultaneous modification of its two primary components, wheat starch and wheat protein, into a water-resistant, three-dimensional, cross-linked network structure [50]. For the most part, raising the temperature causes carbohydrates and proteins to undergo a Maillard reaction, resulting in self-cross-linked network structures [50].

It has been confirmed that thermal treatment of wheat protein is an effective method for unfolding the glutenin protein structure, which enhances the aggregation of the structure and inter-molecular disulfide/sulfhydryl exchange reactions [51]. An anionic surfactant, sodium dodecyl sulfate could be utilized to alter the wheat flour's protein and starch content [51]. Because sodium dodecyl sulfate can react with the hydrophobic groups of the wheat protein to obliterate the hydrophobic reactions and efficiently open up the protein structure, as well as interact with the wheat starch component to form starch–sodium dodecyl sulfate complexes, sodium dodecyl sulfate modification may lead to improved mobility and storage stability of starch-based adhesives [52–54].

Thus, Bai et al. worked on investigating the possibility of preparing water-resistant wheat-flour-based adhesives for wood applications by subjecting wheat flour to thermal–sodium dodecyl sulfate (SDS) or thermal–acid treatment, followed by cross-linking with a reactive polymeric diphenylmethane disocyanate (p MDI) resin coupled with the addition of a polyvinyl alcohol solution as an aqueous polymer [55].

Improved cross-linking density from self-aggregation and inter-change reaction between wheat proteins, the Maillard reaction between wheat starch and wheat protein, and the cross-linking reaction between thermal–SDS-treated wheat flour and cross-linker p-MDI all contributed to the adhesive's increased water resistance, which increased the wet strength from 0.29 MPa to 1.22 MPa [55]. Nevertheless, the thermal–chemical treatment should be performed in the presence of non-toxic chemical substances. It is noteworthy that many crude/raw sources of proteins also contain appreciable levels of carbohydrates, which offers the Maillard reaction enhancement pathway for the resulting adhesives produced from these protein sources.

The strategies for improving the water resistance of protein adhesives, particularly soy-based product, was investigated by Wang et al. [56]. The team modified defatted soy flour by applying waterborne polyurethane resin and investigated the impact of the waterborne polyurethane on the water resistance of the adhesive through several experimental tests, including tensile tests, thermogravimetric analyzer analysis, scanning electron microscopy, and Fourier transform infrared spectroscopy [56]. Waterborne polyurethane was selected for this research due to its non-toxicity and excellent chemical and mechanical properties; also, waterborne polyurethane is widely used in adhesive, textile, building materials, paint, and leather processing industries [57]. The results showed that the initial denature temperature of the adhesive with waterborne polyurethane modification was lesser than the initial denature temperature without waterborne polyurethane modification [56]. Thus, this implied that the initial denature temperature with waterborne polyurethane modification showed better water resistance and mechanical performance, which increased the wet shear strength from 0.65 MPa to 1.1 MPa [56]. Aside from the fact that waterborne polyurethane reacts with the reducing sugar and active groups in soy protein structures, it also forms a reaction with the active groups in the wood structure, thereby facilitating the significant improvement of the water resistance [56]. The waterborne polyurethane-modified protein adhesive has shown a potential to replace the conventional formaldehyde-based resin, thereby reducing the dependency on petrol-based products.

Xu et al. explored cottonseed meal as the raw material for protein adhesives [58]. They studied the effects of modifier reaction conditions and additions on the bonding properties of the adhesive as well as the modification mechanism [58]. Cottonseed protein has not been fully utilized by other markets (access product resulting in low cost); thus, it can help reduce the cost of adhesive, as it is cheaper than soy protein. At the same time, this excess supply of cottonseed protein can be used to overcome the demand and supply limits of soybean protein, thereby providing an additional high-value usage of cottonseed meal [59]. Figure 3 shows the cross-linking mechanism between soy protein isolate (SPI), polyacrylamide (PAM), and 1,2,3-propanetriol-diglycidyl-ether (PTGE) through which the components form a larger network structure [60].

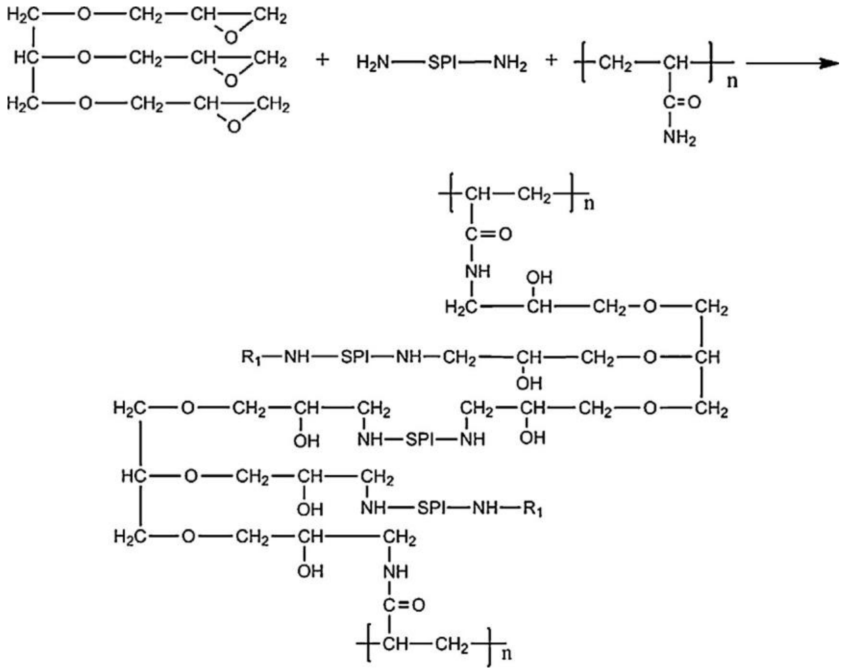

Figure 3. The cross-linking reaction among soy protein isolate (SPI), polyacrylamide (PAM), and 1,2,3-propanetriol-diglycidyl-ether (PTGE) [60]. Reprinted/adapted with permission from Ref. [60]. 2023, Industrial Crops and Products.

The team blended different proportions of isocyanate with cottonseed protein adhesive [58]. The results showed that the solid content and viscosity of the adhesive increased with the addition of isocyanate proportion, and the researchers confirmed that the solid content of adhesive has impacts on the viscosity of the adhesive, thereby affecting the bonding strength, wettability, sizing effect, and amount of glue [58]. The wet bonding strength increased from 0.65 MPa to 1.68 MPa. The increase in the amount of isocyanate translated to a reduction in the application period of the adhesive [58]. Thus, this implies that the preparation process of the constituents (cottonseed protein adhesive and isocyanate) needs to be further explored [58]. Nevertheless, this research provides practical and theoretical guidance and a basis for the preparation of novel low-cost bio-based adhesives and explores a new way for utilizing the waste cottonseed meal.

A summary of the wet strengths found from various proteins combined with cross-linking agents is given in Table 1.

Table 1. Summary of wet bond strengths of proteins with their cross-linking agents.

Protein and Cross-Linking Agent	Wet Bonding Strength (MPa)	Reference
Wheat protein GPPEI	0.44 2.02	[44]
Soy protein EPR + MF/SP	0 0.85	[43]
Soy protein EG/SP	0.22 1.12	[47]
Soymeal SM/TGIC/HD	0.22 1.0	[49]
Wheat flour T-SDS-WF	0.29 1.22	[55]
Soy protein WPU/SP	0.65 1.1	[56]
Cottonmeal CM/Isocyanate	0.65 1.68	[58]

Abbreviations: GPPEI—glutaraldehyde-polyethyleneimine-modified wheat protein; EPR + MF—epoxy resin and melamine–formaldehyde modified soy protein; EG/SP—epoxy groups modified soy protein; SM/TGIC/HD—hyperbranched triglycidyl isocyanurate modified soybean meal; T-SDS-WF—thermally treated sodium dodecyl sulfate modified wheat flour; WPU/SP—waterborne epoxy emulsion modified soy protein; CM/Isocyanate—isocyanate-modified cottonmeal.

2.1.2. Bio-Based Cross-linking Agent Sources

Bio-based sources are synthesized from biomass and renewable materials. Thus, it potentially has the advantages of lower environmental toxicity, renewability, and sustainability.

Pang et al. successfully prepared waterborne epoxy emulsion through graft polymerization to generate an oil-in-water emulsion [61]. As determined by different characterization tests including Fourier Transform Infrared Spectroscopy (FTIR), the synthesized waterborne epoxy emulsion (WEU) had good dispersibility and stability and was deemed an efficient cross-linker for generating multiple stable cross-linking networks with soy protein molecules [61]. The results showed a significant water resistance improvement with a wet shear strength of 0.71 MPa compared to unmodified soy-protein-based adhesive with a wet shear strength of 0.39 MPa and meet the requirements of interior-usage plywood which is either greater than or equal to 0.7 MPa [61].

A group of researchers worked on exploring an enzyme complex (*Aspergillus niger*) by using soybean hulls as a substrate for the development of the protein adhesive. An inexpensive *Aspergillus niger* fermentation broth contains an enzyme complex that performed exceptionally well at hydrolyzing the polysaccharides in defatted soy flour [62,63]. The decreased quantity of water-insoluble materials and the weakened rheological properties of the slurry were evidence that this enzyme complex effectively hydrolyzed polysaccharides in defatted soy flour [64]. In this study, different soy polysaccharides were hydrolyzed into reducing sugars using an enzyme broth made up of *A. niger* which is particularly effective for doing so. The exposed functional groups of the modified soy protein reacted with the reducing sugars, significantly enhancing the soy adhesive's water resistance. The reaction between reducing sugars and soy protein increased the strength and water resistance of soy adhesives, which together helped to increase wet bonding strength [64]. Although the synthesis process is quite complicated, there is a lot of potential for the self-cross-linked soy adhesive created by this practical and affordable technology to replace synthetic wood adhesives.

Due to their catechol composition, marine mussels possess excellent adhesive characteristics [61]. This catechol structure is capable of forming strong hydrogen bonds with the substrate and reacting with amino groups after oxidation to quinones [65,66]. Thus, catechol-structured biomass raw materials appear to offer an excellent option for cross-linking soybean protein adhesives.

Zheng et al. built on previous research by developing a lignin with a catechol structure using a single-step demethylation [67]. Copper ions, lignin, and soybean protein isolate were combined to produce a bio-based adhesive with a triple network structure without the use of an epoxy cross-linking agent derived from petroleum, thereby expanding the potential value of bio-based adhesives in industrial applications and production while increasing the overall sustainability of the potential process [67]. Catechols were randomly oxidized into quinones, and lignin's self-bonding properties made it possible to form both covalent- and hydrogen-bond cross-links with just the addition of lignin [67]. Due to the abundance of metal coordination and hydrogen bonds in the system, which serves as a mechanism for energy dissipation, the adhesive demonstrated the ideal viscosity, outstanding coating characteristics, and tensile strength [67]. The increased number of glue nails and lower viscosity helped create a strong adhesive bond in the wood while the adhesive's water resistance can be attributed to its dense cross-linked structure [67].

Tung oil is extracted from the tung tree's seeds or kernels [68,69]. Most commonly, it is found in inks, coatings, and resins, but it is also used as a natural varnish for wood [68]. About 80% of its fatty acid content is a conjugated, trienoic fatty acid, which gives it its distinctive drying properties [68]. Tung oil's high iodine value, rapid drying time, increased water resistance, and increased hardness as a result of its high level of unsaturation make it a desirable protective coating material [70,71]. He et al. worked on the application of tung oil on adhesives produced from cottonseed protein isolate and water-washed cottonseed meal [72]. The water resistance test was performed by immersing the glued wood specimens in a water bath followed by drying at room temperature for more than 18 h [72]. Water-washed cottonseed meal and cotton-seed protein isolate with tung oil showed improved adhesive strength, compared to tung oil-free adhesive controls [72]. The addition of tung oil to water-washed cottonseed meal and cotton-seed protein isolate also increased their resistance to water by over 40%. Optical microscopic photographs of bond lines of these bonded wood pairs demonstrate that the improvement was mostly attributable to the tung oil's ability to prevent the adhesives from diffusing away from the bond joints [72]. This research has shown that promoting cottonseed protein products as wood adhesives over wheat and soy protein products will alleviate long-term concerns about global food security.

Another bio-based material for cross-linking is tannic acid processed from vegetable tannins [73]. Tannic acid has been widely used as a cross-linking material for different biopolymers like chitin whiskers, chitosan, collagen, casein, albumin, and gelatin [73–79]. Tannic acid is categorized as a tannin which is processed through hydrolysis under mild alkaline or acidic conditions to obtain phenolic acids and carbohydrates [73].

For their research, Ghahri et al. worked on the application of tannic acid for modifying soy flour adhesives, thereby increasing their moisture resistance for wood applications [80]. The tannic acid solution was mixed with the soy slurry and reacted with soy flour to form a cross-linked network, thereby increasing the water resistance and mechanical properties of manufactured wood panels. Figure 4 shows the cross-linking mechanism between amino acids and tannic acids, where the protein–tannin complexes self-associate through hydrogen bonding [80]. The modulus of elasticity was increased from 3213 MPa to 3590 MPa, the modulus of rupture increased from 17 MPa to 18 MPa, and the shear strength increased from 1.43 MPa to 1.96 MPa [31]. The delamination test was successfully completed by plywood manufactured using adhesives produced from tannic-acid-modified soy flour [80]. Adding tannic acid to the soy glue recipe enhanced the plywood's shear strength [80]. Fiberboard tests revealed that the modified soy-based glue improved the resulting board's water resistance and thickness swelling in addition to enhancing its mechanical properties [80]. This has shown to be a good way to bring new eco-friendly and water-resistant soy-based adhesives to the wood composites industry.

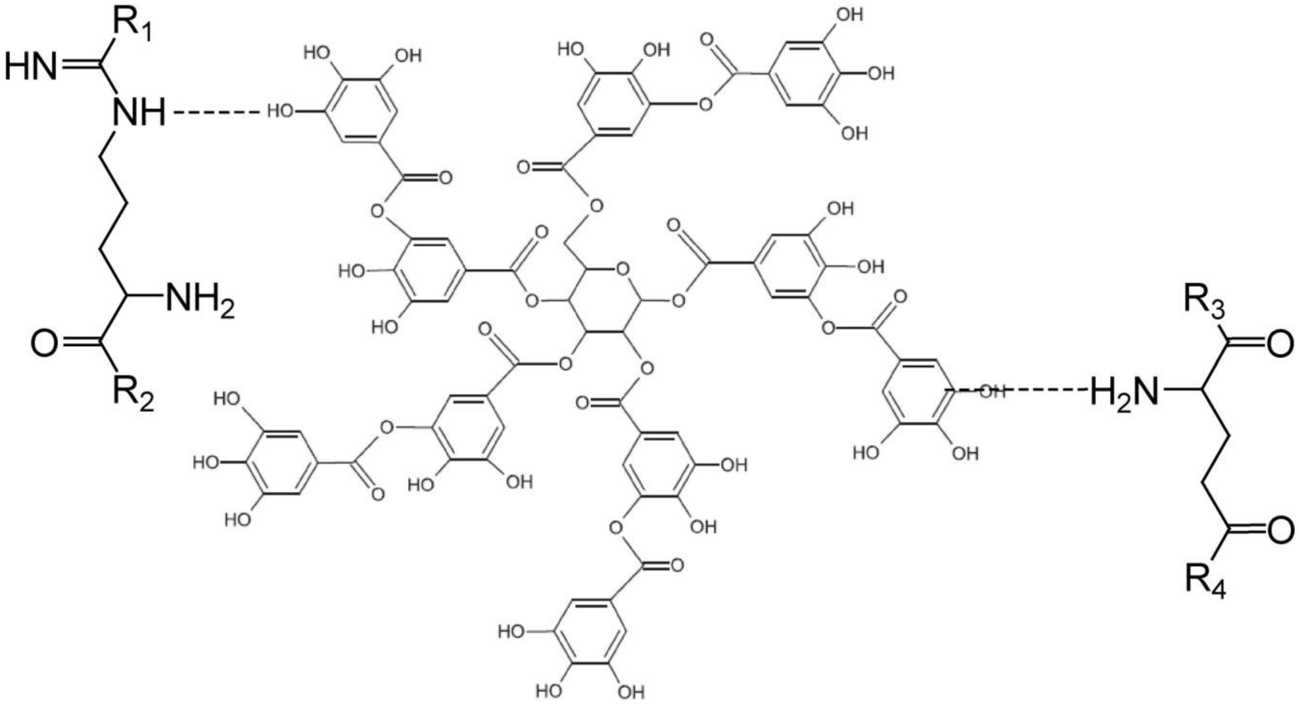

Figure 4. Cross-linking reaction between tannic acids and amino acids [80]. Reprinted/adapted with permission from Ref. [80]. 2023, Journal of Polymers and the Environment.

Lignin-based resin reacted with itself and protein molecules to form an interpenetrating and cross-linking network [81]. Lignin exists in plant materials/products like sorghum, corncob, and wood and has a cross-linked and aromatic structure comprising of several building units [81]. Due to the availability of aromatic compounds derived from plants which are renewable products, the de-polymerization of lignin can be applied to make bio-based polymers [82,83]. Another property of lignin is that some of the compositional structures have similar features to phenol, thus providing the potential to replace phenol in the phenol–formaldehyde resin preparation [81].

The use of lignin-based resin increased the wet shear strength and the water resistance of the resulting soymeal-based adhesive [84]. The researchers attributed the successes to the following reasons: the formation of a cross-linking network due to the reaction between the soy protein molecules and the lignin-based resin; the formation of a smoother fracture surface, thereby preventing the intrusion of moisture; the formation of an interpenetrated network by self-cross-linking lignin-based resin molecules and cross-linked protein molecules; an improvement in the thermal stability of the adhesive; and the resultant appropriate viscosity, thereby benefiting the adhesive distribution and forming a stronger interlock [84].

Pradyawong et al. worked on studying the effects of Kraft lignin on lignin and protein interactions through thermal and rheological properties and the adhesion performance of the protein product [85]. Previous research has shown that Kraft lignin is hydrophobic in nature and has a high content of aliphatic hydroxyl groups allowing for reactive modification and serving as a superb substrate for lignin-derived products [86,87]. Lignin and soy protein were mixed at different weight ratios, and the results showed an improvement in water resistance due to the unfolding of the hydrophobic group to the protein structure [85]. Another advantage is that the lignin increased the thermal stability and dispersal of the soy protein adhesives [85]. The team explored more research on Kraft lignin by modifying laccase/TEMPO lignin to study its adhesion properties and performance [88]. The synthesis process involves depolymerizing kraft lignin by laccase enzyme and TEMPO to enhance the oxidation reaction of both non-phenolic and phenolic compounds [88]. The unique advantage of this process is that it enhances the lignin–protein interaction [88]. The simplified process of mixing the soy protein, kraft lignin, and laccase enzymes increased the wet shear strength by 106% and strengthened the protein structure due to the interaction between the $–NH_2$ and $–COO^-$ group [88]. However, these interactions led to a decrease in the spreadability and flowability [88]. Thus, we can conclude from this research that it revealed a simplified synthesis process for adhesives' preparation and shows that the combination of these materials has great potential to fulfil the massive demand for green products [88].

Another team of researchers led by Zhu also explored depolymerized lignin to improve the water resistance of camelina protein [89]. In this case, the depolymerization of Kraft lignin was completed by H_2O_2-induced oxidation via ultrasound irradiation to reduce the thermal stability and particle size of the lignin and increase the hydroxyl content [89]. The modified lignin contained more hydroxyl groups and smaller particles [89]. Ultrasound-induced oxidation was more effective in inducing the disintegration of the lignin structure and enhancing its reactivity [89]. Thus, the team developed a bio-based wood adhesive from camelina proteins and copolymerized the product with the modified lignin [89]. Also, they compared the product with the most commonly used adhesives like polymeric amine epichlorohydrine, urea-formaldehyde-based adhesives, and $NaHSO_3$-modified soy protein adhesives [89]. The results showed that the oxidation-pretreated lignin-based adhesive produced by this team had the highest water resistance and strongest intermolecular interaction evaluated when compared to the other adhesives [89]. Industrial adhesive uses of the non-edible camelina protein copolymerized with oxidized kraft lignin would be beneficial to both the camelina oil-based energy business and the paper industry [89]. However, the synthesis process should be simplified for large-scale production.

Xiao et al. also worked on improving the water resistance of soy protein adhesives by adding lignin. In this case, the researchers added extruded sorghum lignin and sorghum lignin and investigated the adhesion properties [83]. They added extruded sorghum lignin and sorghum lignin to soy protein adhesives in four different reaction solutions, namely, modified soy protein, soy protein isolate, alkaline solution, and neutral water [83]. The glued wood samples were soaked in tap water at room temperature for 48 h before being air-dried in a fume hood [83]. When the combined amount of sorghum lignin and extruded sorghum lignin grew from 30% to 50%, the water resistance improved [83]. This finding suggests that when sorghum lignin or extruded sorghum lignin is blended into soy protein adhesives at higher concentrations, the advantage of entrapping interaction between lignin molecules and modified soy protein predominates, as opposed to being the driving force behind the blended sorghum lignin and extruded sorghum lignin at lower percentages [83]. Soy protein adhesives combined with extruded sorghum lignin from modified soy protein fared better in water resistance tests than those blended with sorghum lignin, whereas the inverse was true for the range of blending percentages between 30% and 50% [83]. By blending with sorghum lignin, soy protein adhesives' shear strength and water resistance were considerably enhanced, but the improvement with extruded sorghum lignin was more pronounced when the mixed soy protein adhesives were suspended in water [83]. Further tests should be carried out by using other lignin. However, this research further certified the potential of lignin for protein adhesives.

Lignin processed from the waste product of fermenting cellulosic-based sugars into bioethanol was explored for protein adhesive by Xin et al. [90]. The researchers used Fenton oxidation at room temperature in a basic aqueous solution to change the hydroxyl groups in lignin into carbonyl groups (like ketones or aldehydes) [91,92]. Compared to previous research projects, this particular lignin was insoluble in water and most organic solvents due to its original highly branched structure [93]. In organic chemistry, reductive amination is a highly efficient process for obtaining amino substituted products [93]. The team prepared both unmodified lignin and partially depolymerized lignin and used both products for the preparation of lignin amines [90]. The results showed that the amine prepared via the unmodified lignin has a greater effect on the water resistance and adhesion strength of soy protein adhesives [90]. This research demonstrated a novel way of preparing adhesives through the Fenton oxidation and reductive animation method [90]. This process provides competitive advantages of non-toxic, high conversion, environmentally friendly reactions and solvents at room temperatures [90]. The lignin amine might have a potential for usage in other bio-based materials, such as polyurethanes and epoxy resins [90].

There are several issues associated with soy adhesives, such as high viscosity, the requirement of cross-linking modification, and denaturation agent due to its high molecular weight [94]. Due to the use of petrochemical agents, modified soy protein glue is not a completely sustainable product [94]. The unstable performance of the soy protein glue restricts its widespread use in the wood panel manufacturing business [94]. Therefore, a soy protein adhesive with high bonding strength and water resistance, low cross-linker addition, moderate viscosity, and consistent bonding performance must be developed [94].

This interest in enhancing the sustainability of soy-protein-based adhesives was the basis for the research performed by Xu et al. to improve the qualities of soy protein glues by using the recombination strategy [95]. Molecular recombination is simply the deterioration of a substance followed by a recombination process to achieve excellent properties [96]. Through a process involving both biological and chemical alteration, molecular recombination theory served as a guiding principle for the modification of the soy protein adhesive [96]. Soy protein adhesives of varying molecular weights were manufactured by breaking down soy protein molecules into tiny peptide chains using bromelain of varying concentrations [96]. Using enzymatic hydrolysis, the protein endonuclease-bromelain disrupted the peptide bonds in the soy protein molecules, resulting in the degradation of the protein into short molecular polypeptide chains and a lower molecular weight [95]. Triglycidylamine, a bio-derived cross-linking agent, was used at low concentrations to recombine these polypeptide chains [95]. Using a two-step procedure, the adhesive's performance and bond stability were enhanced by creating a consistent and stable cross-linked network structure [95]. Meanwhile, the adhesive's viscosity was lowered by employing bromelain [95]. By replacing the weak hydrogen bond with a chemical bond during the cross-linking step, water resistance in adhesives is enhanced [95]. After enzymatic hydrolysis, the molecular weight of the soy protein was measured [95]. The resulting soy protein adhesives were analyzed for their viscosity, adhesive layer homogeneity, thermal stability, functional groups, and micro-structure of the fracture surface [95]. Soy protein adhesive plywood samples were fabricated, and their wet shear strength was evaluated [95]. These findings provided evidence for the beneficial effects and underlying mechanism of molecular recombination involving soy proteins in the adhesives [95].

Mussel-inspired polydopamine coating utilizing dopamine chemistry has been demonstrated to be an efficient method for enhancing the surface reactivity of diverse materials, including hydrophilic and hydrophobic surfaces as well as inorganic and organic substrates [97,98]. Also, solutions containing catechols had the highest adhesion energy, indicating that adhesives containing catechols should also exhibit exceptional adhesion energy [99]. The catechol groups of dopamine can oxidize to generate a reactive quinone capable of undergoing Schiff base reactions or Michael-type additions with radical coupling and nucleophiles with other amines or catechol in alkaline conditions without the use of complicated equipment [100,101].

Pang et al. worked on the surface functional modification of mussel proteins to enhance the weak surface reactivity of silkworm silk fiber [102]. The next step was the application of the as-fabricated silkworm silk fiber to improve soy-based adhesives, thereby generating a compact structure [102]. Several experimental analyses confirmed that tannic acid, dopamine, and alkali lignin were successfully coated onto silkworm silk fiber [102]. The soy protein composite had improvements in properties like toughness and wet shear strength. The results showed that the adhesive sample attained a high wet shear strength that is far greater than the interior use standards.

Undecylenic acid (UA) is a green chemical generated from castor oil by pressure-cracking and is extensively utilized in the perfume, cosmetic, and pharmaceutical industries [103]. Undecylenic acid is non-soluble in water and has long hydrophobic chains, thereby preventing water from gaining access into the interfacial surface of the wood and adhesive coupled with a significant improvement of the wet shear adhesion strength in the range of 35–62% [104]. Liu et al. focused on the development and characterization of UA-modified soy proteins to enhance their water resistance for the production of adhesives [104]. The team successfully incorporated undecylenic acid into soy protein via the reaction between NH_2 and COOH, which was confirmed by FTIR [104]. Also, it confirmed the reduction in amino group concentration via the use of the ninhydrin reagent [104]. Thus, this formulation created an effective pathway to develop biodegradable and cost-effective adhesives.

2.1.3. Synthetic and Bio-Based Cross-linking Sources

Zhou et al. produced flame retardant and high-strength adhesive by combining the effect of cross-linking strategy and borate chemistry [105]. Here, oxidized soybean polysaccharide was complexed with borax to form an in situ cross-linking skeleton with periodate oxidation–adipic acid dihydrazide via a Schiff base reaction; chitosan was introduced for the purpose of improving the active sites as well as the water resistance; and the process was inspired by borate chemistry in plants and the periodate oxidation–adipic acid dihydrazide cross-linking strategy [105]. Hydrogen bonding and the covalent bonds of acylhydrazone and borate ester are just two of the cross-linking networks that were built into the soy-protein-based glue [105]. The researchers also aimed to develop an environmentally friendly and effective cross-linking technique of polysaccharides, which is crucial for their use in wood adhesives. The strength of borate-chelated polysaccharide adhesive must be enhanced [105]. In recent years, the periodate oxidation–adipic acid dihydrazide cross-linking technique has been applied successfully to the development of robust polysaccharide-based composites, such as cellulose, starch, and pectin [106]. Also, periodate oxidation–adipic acid dihydrazide is environmentally friendly, and the cross-linked polysaccharide material exhibits outstanding biocompatibility and mechanical strength [107]. Thus, this cross-linking strategy has the potential to develop soy protein adhesive with high cohesiveness, which was greatly enhanced by the double-network structure resulting from multiple cross-linking [105]. The results showed that the wet and dry shear strength increased significantly which was accompanied by a reduction in the moisture absorption rate and an increase in the residual rate, thereby signifying excellent water resistance and bonding strength [105]. The novel soy-protein-based adhesive significantly reduced health threat and environmental emissions [105]. Although the synthesis process is quite complicated, the process of producing excellent polysaccharide-based materials does support its use [105].

Phenol-amine chemistry was applied toward the preparation of novel organic–inorganic protein-based adhesives [108]. Li et al. developed a versatile and green strategy for the development of a robust soy protein adhesive through bio-mineralization reinforcement and phenol-amine synergy [108]. Thus, the researchers co-assembled hydroxyapatite and gallic acid via calcium ion phenolic coordination bonds to synthesize gallic-acid-functionalized hydroxyapatite [108]. In addition, a Schiff base reaction was used to graft ε-polylysine with numerous amino groups to the phenolic gallic-acid-functionalized hydroxyapatite nanoparticles [108]. Several researchers have worked on synthesizing hydroxyapatite to prepare strong adhesives. For instance, Yu et al. incorporated hydroxyapatite and polyvinyl alcohol to develop a composite microfiber with significant improvement in flexibility and toughness [109]. In a similar vein, Jiang et al. developed a superior gelatin-based adhesive via the incorporation of polydopamine-designed hydroxyapatite nanoparticles [110]. The disadvantage is that these methods require tedious and complex processes, the use of harmful chemicals and expensive raw materials. Li et al. have been able to mitigate these challenges by developing a sustainable and simple strategy for preparing organic–inorganic biopolymer materials [108]. The results showed that the synergistic effect of phenolic gallic-acid-functionalized hydroxyapatite with amine-rich ε-polylysine and soy protein chains simultaneously increased the hybrid adhesive's cohesion and adhesion strength [108]. Also, the phenol-amine synergy and solid bio-mineralized structure conferred water resistance and thermal stability to the resulting glue [108]. Thus, the biomimetic approach offers a flexible method for producing biopolymer materials for adhesives, films, coatings, and hydrogels [108].

2.2. Water Resistance from Modified Fillers

The addition of fillers can significantly impact the thermal and mechanical properties of protein adhesive, and the mechanism involves the interaction between the protein matrix and the filler material [111,112]. The incorporation of sodium montmorillonite (Na MMT) into soy protein adhesives at different concentrations was explored by Qi et al. [113]. Sodium montmorillonite is the most extensively utilized kind of silicate clay in polymer nanocomposites with properties, such as thermal and chemical stability, natural abundance, and non-toxicity [114]. The material is widely employed as a reinforcing and nano-filler material to produce nanocomposites owing to its high aspect ratio and unusual layered and nanoscale structure [115]. Thus, Qi et al. worked on developing a novel soy protein and clay system with excellent flowability and strong adhesion at high protein content [113]. Sodium bisulfite ($NaHSO_3$) was used in these formulations, and hydroxyethyl cellulose was used as a suspension agent [113]. $NaHSO_3$ can be utilized as a reducing agent to break the disulfide bonds in protein molecules, thereby resulting in an increase in surface hydrophobicity, solubility, and flexibility [116]. Another researcher, Zhang and Sun, corroborated this claim by using $NaHSO_3$ to break the disulfide bonds of soy glycinin to increase the surface hydrophobicity [117].

The results showed that the addition of Na MMT significantly improved the adhesion strength of the soy protein adhesives due to the adsorption of the soy protein molecules on the surface of the interlayer of Na MMT via electrostatic interaction and hydrogen bonding [113]. Thus, the water resistance of the soy protein/Na MMT increased to 4.3 MPa compared to the 2.9 MPa of control SP at 8% Na MMT and the dry shear strength from 5.7 MPa to 6.38 MPa [113]. This research shows an innovative way to develop an adhesive with excellent adhesion with the incorporation of silicate clay materials.

Ciannamea et al. prepared soy protein concentrate (SPC)-based adhesives and rice husks (RCs) to produce particleboards with the main goal of upgrading the final water resistance and mechanical properties of RH-SPC particleboards via the alkali treatment of soy protein concentrate and rice husks, coupled with bleaching of rice husks with hydrogen peroxide [118]. This facilitated chemical interactions via hydrogen bonds between the more exposed hydroxyl groups of cellulose from rice husks and the polar groups of the unfolded proteins of soy protein concentrate treated with alkali [118]. The particleboards met the mechanical properties requirements for commercial use consideration but failed to achieve the minimum requirements for water resistance as recommended by US Standard ANSI/A208.1 [118,119]. The limitation was linked to the increase in the amorphous content of the cellulose after dispersing rice husk in NaOH for a short period [118]. This drawback is counterbalanced by the adhesive's lack of formaldehyde and the use of complete rice husks in particleboard production, which eliminates milling and screening procedures, resulting in cost savings [118].

2.3. Removal of Hydrophilic Content

Gui et al. centered their research on the preparation of water-resistant soy flour adhesives through the reduction in the water-soluble constituents [120]. Previous research has shown that poor water resistance is mainly caused by feedstock water-soluble components [29]. The design approach involved suspending the defatted soy flour in water followed by the adjustment of the dispersion pH at different temperatures and time points to 4.5 by adding NaOH and HCl solution, respectively [120]. Then, the sample with less water-soluble constituents was separated through a centrifugation [120]. The application of the modified soy flour adhesive on poplar plywood shows that it had a wet strength of 1.02 MPa [120]. The remaining multi-level structures of soy protein contributed positively to soy adhesives' water resistance [120]. Although this is a simple and novel way of developing water-resistant soy adhesives, the product is still limited by low solid content and fluidity compared to formaldehyde adhesives [120]. Thus, further research is required to enhance these properties.

Zhang et al. subjected defatted soybean flour (DSF) adhesive to thermal treatment at different test temperatures to improve its water resistance and investigate the effects of the thermal pretreatment on increasing the water-insoluble content, crystalline degree, and chemical structure [121]. The team also tested the thermal stabilities and bonding qualities of soy adhesives made from thermal treatment DSF (T-DSF) and cross-linker epichlorohydrin-modified polyamide (EMPA) [121]. The test result showed that the thermal treatment facilitated the increase in the acetaldehyde value and water-soluble content of T-DSF [121]. Thus, thermal treatment can enhance protein–carbohydrate Maillard reactions, protein–protein self-cross-linking, and protein–EMPA cross-linking by unfolding the globular form of soy protein and releasing hidden functional groups [121]. Uncertain are the quantitative contributions of protein–protein self-cross-linking, protein–carbohydrate Maillard processes, and protein–EMPA cross-linking, as well as their impact on the water resistance of T-DSF-based adhesives [121].

Qi et al. investigated the effect of liquid 2-octen-1-ylsuccinic anhydride (OSA) on soy protein adhesives [122]. The OSA possesses a long alkyl chain and oily nature coupled with a succinylation reaction that can help enhance protein adhesion strength [122]. Thus, the team studied the adhesive properties of soy protein adhesives modified by OSA at different concentrations as well as characterized its physicochemical properties like morphological, rheological, thermal, and turbidity properties [122]. The OSA modification increased the wet shear strength in plywood samples up to 3.2 MPa with up to 60% wood cohesive failure in comparison to 1.8 MPa of wet strength for the control. They found that the modification of the soy protein adhesives with OSA facilitated the introduction of hydrophobic materials to the protein structures [122]. Due to OSA's hydrophobic properties, hollow cavities could not form since water could not penetrate the interface between the wood surface and the adhesive [122]. The researchers stated that this could be the primary factor for the soy protein adhesives' significantly improved water resistance [122].

Zhang et al. worked on the modification of soy protein adhesives using epoxidized oleic acid and prepared the chemically modified adhesive soy protein and rice straw formulation to produce a fiberboard that might serve as a viable substitute for wood-based fiberboard [123]. The utilized epoxidized oleic acid contains many free epoxy groups that could have a reaction with the functional amino groups in the SPI molecule. Epoxidized oleic acid was utilized to react with the amino groups in SPI molecules to increase the mechanical and water resistance qualities of the adhesive [123]. The team investigated modified-SPI adhesive addition, the effects of NaOH concentration, and fiberboard density on the water resistance and mechanical properties of rice straw fiberboards [123]. The results showed that the fiberboards have optimal water resistance and mechanical performance, which is due to the reaction between the soy protein and the epoxidized oleic acid, and the removal of the wax layer using NaOH with a water resistance value of around 64% [123]. The advantage is that the raw materials are low-cost and easily biodegradable; thus, these fiberboards are an excellent substitute for petroleum-based panels and can be utilized in indoor furniture and decoration.

3. Conclusions

This review paper has outlined recent strategies for improving the water resistance of protein adhesives, which are categorized into cross-linking networks, separation techniques, and modifications with different reagents. In each case, the researchers explored the use of synthetic, bio-based, or a combination of both.

Cross-linking improves the water resistance and adhesive strength of protein adhesives [124]. However, this strategy can increase the complexity of the manufacturing process while some of the agents are harmful to the environment [33]. The use of naturally occurring modified fillers has shown to be effective in improving the water resistance of wood adhesives; however, the challenge is that they may require additional processing steps [113]. The most promising strategy is the removal of hydrophilic content because it does not require a complex synthesis process and is suitable for large-scale applications [120].

The trends have shown that many researchers have started to explore the possibilities of bio-based sources for improving water resistance in protein adhesives due to their sustainability and renewability [125]. However, the researchers should try to simplify the synthesis process for large-scale applications. The combination of both tried to optimize its advantages but came with a more complicated synthesis process and expensive raw materials.

An epoxy cross-linking agent has been shown to be efficient for interior plywood use ($\geq$0.7 MPa) like flooring, ceiling, etc., but its exterior usage has not been fully established [124]. The complete replacement of petrol-based wood adhesives with protein is not feasible unless the limitations of protein adhesives are eliminated. Thus, it is essential for the industry stakeholders to continue to develop viable ways to overcome these challenges to move from a petroleum-based economy to a more sustainable economy and to minimize the use of formaldehyde-based adhesives.

While continuing to improve water resistance and wet strength, future research should also include reducing the complexity and reaction times of these water-resistance amendments because wood composite industries need to make products using fewer steps with short press times in order to be cost-competitive. The combination of proteins with starches, lignin, tannins, and other biomass sources is likely to be a continued area of interest with great potential. Further research should also be conducted to assess the environmental impacts of these novel modifications. One would expect that if the protein adhesives are made more water-resistant, the biodegradability will be affected. The likely environmental fate of the modified proteins, cross-linking agents, and other additives should also be determined before commercializing on a large scale.

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