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Study on Modification of Green Adhesives Derived from Plant Proteins for Wood Composite Panels

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Abstract: The wood-based panel industry in China is developing rapidly, and the demand for wood is growing day by day. However, due to the insufficient supply of wood under the natural forest protection policy, finding other ways to reasonably and efficiently use wood resources has become an important way to solve the shortage of wood. Adhesives play a key role in the wood-based panel industry. The preparation of plywood, particleboard, fiberboard, blockboard and other wood-based panels cannot be separated from the participation of wood adhesives. At present, the most widely used wood adhesive in China is still "trialdehyde adhesive". However, the "trialdehyde adhesive" is mainly composed of nonrenewable resources such as petroleum, and the adhesives made from formaldehyde and its derivatives will more or less release harmful substances such as formaldehyde in the application process, causing pollution to the production and living environment and endangering human health. With the continuous depletion of petroleum resources and the increasing attention paid to environmental protection, it is of great scientific significance and practical value to develop a new environment-friendly adhesive with renewable biomass as the main raw material. The use of plant proteins such as soybean and cottonseed as the main raw materials for preparing adhesives can not only reduce the cost of adhesives, but also break through the supply and demand constraints of traditional petrochemical raw materials and make use of biomass resources with high added value. Based on the above research background, this paper reviewed the research progress and common modification methods of plant protein adhesives mainly composed of soybean and cottonseed, and put forward opinions and suggestions for the future development of plant protein adhesives.

Keywords: Plant Protein, Wood Adhesive, Cottonseed, Soybean, Wood-Based Panel

1. Introduction

The rapid development of the wood processing industry has led to the increasing demand for wood. The national natural forest protection policy has made the supply of wood insufficient. Therefore, finding other ways to reasonably and efficiently use wood resources has become an important way to solve the shortage of wood. Adhesives are polymer materials that can bond two or more materials together through surface adhesion under certain conditions and have certain adhesive strength after curing. The emergence of wood adhesives makes full use of small-diameter wood, branch wood, thinning wood and wood processing residues. Through bonding and composite technology, plywood, fiberboard, particleboard and other wood industries have developed rapidly.

Adhesives play a key role in the wood-based panel industry. The preparation of plywood, particleboard, fiberboard, blockboard and other wood-based panels cannot be separated from the participation of wood adhesives. According to the survey of the State Forestry Administration, the output of wood-based panels in 2020 was 311 million cubic meters, and the consumption of formaldehyde based adhesives reached 15.42 million tons (100% solid content), accounting for more than 90% of the total wood adhesives [1].

At present, the wood adhesives widely used in the market are "tri aldehyde adhesives", namely urea formaldehyde resin adhesives, phenolic resin adhesives and melamine resin adhesives. "Trialdehyde adhesive" occupies a leading position in the wood adhesive market with its good performance and low price [2]. However, "Trialdehyde glue" takes nonrenewable resources such as petroleum as the main synthetic raw materials, and adhesives made from formaldehyde and its derivatives will more or less release harmful substances such as formaldehyde in the application process, causing pollution to the production and living environment and endangering human health [3-6]. In October 2017, formaldehyde was included in the list of carcinogens released by the International Cancer Research Institute of the World Health Organization. All countries in the world, especially developed countries, attach great importance to the formaldehyde emission from wood-based panels and their products. The International Organization for Standardization (ISO), the European Union, the United States and Japan have continuously updated the formaldehyde emission limit standards for wood-based panels and their products, putting forward higher requirements for the formaldehyde emission limit. In 2017, the revised GB18580-2017 Formaldehyde Emission from Wood based Panels and Their Products as Interior Decoration Materials was implemented, which stipulated that the formaldehyde emission limit value of wood based panels and their products used as interior decoration materials was 0.124mg/m^3, the limit mark was E1, and the original E2 plate was canceled (GB 18580-2017). More and more stringent standards have greater restrictions on the use of "trialdehyde adhesive". With the continuous improvement of people's living standards, the requirements for living environment are becoming higher and higher. More and more people pay attention to non-toxic and environment-friendly bio based adhesives. At present, starch adhesives, tannin adhesives, lignin adhesives, animal and plant protein adhesives are common in the research, among which the research on plant protein adhesives is a hot spot [7].

2. Research of Plant Protein Adhesives

2.1. Single Modification

The research on plant protein adhesive has been started in foreign countries for a long time. In 1923, Johnson of the United States successfully prepared the adhesive using skimmed soybean flour and applied it to the production of plywood. From the 1940s to the 1960s, the application of soybean protein adhesive entered the golden age, accounting for 85% of the American market share. During this period, there were other studies on the preparation of protein adhesives using some plant meals as raw materials. In 1950, researchers began to use cottonseed meal as the base material for the preparation of wood adhesives, and compared its bonding strength with the adhesive prepared from peanut meal. It was found that the performance of the adhesive prepared from cottonseed meal was better than that of peanut meal, and it was concluded that the different types of amino acids between the two proteins caused the difference in bonding strength. In the 1970s, with the rapid development of petroleum industry, synthetic adhesives widely replaced protein adhesives with poor water resistance, and occupied the leading position in the wood adhesive market. Since the 1990s, with the improvement of people's living standards, the requirements for the quality of living environment have become increasingly high. The development and use of renewable resources or biomass materials to prepare environment-friendly biomass based adhesives have become a research hotspot. In 1995, it was found that adding sodium hydroxide to soy protein could improve the water resistance of soy protein adhesive [8]. Later, Sun found that the water resistance of urea modified soybean protein adhesive was better than that of alkali modified soybean protein adhesive by comparing the effect of alkali modified and urea modified on the adhesive bond strength.

2.2. Composite Modification

Single modification method has limited effect on improving the water resistance of protein adhesive. Therefore, multiple modification methods are often used in the research, such as combining chemical modification with physical modification or combining multiple chemical modification methods. Liu Yuan used maleic anhydride modified soy protein isolate and polyethyleneimine to prepare adhesive, and the bonding strength and water resistance of the obtained plywood were significantly improved [9]. Tong Ling developed a compound modified soybean protein adhesive with a variety of modifying agents (sodium dodecyl sulfate, urea, acetic anhydride and polyisocyanate) using skimmed soybean flour as raw materials. The strength of plywood tested reached the requirements of the national standard for Class II plywood [10]. Wang pyrolyzed walnut protein under alkaline conditions, and the water resistance of walnut protein adhesive prepared with branched polymer (PEI) as modifier was significantly improved. The chemical bonding between carboxyl and amino groups in walnut pyrolysis liquid protein and active amino and imino groups in PEI promotes the improvement of water resistance [11].

2.3. Selection of Crosslinking Modifier

When modifying protein adhesive, the selection of cross-linking modifier also tends to be environmentally friendly and clean. Cheng added nano cellulose particles as modifiers to the cottonseed protein adhesive, and compared the effects of cellulose nanofibers (CNF) and cellulose nanocrystals (CNC) on the adhesive performance. When CNF was added with 2%, the effect was the best. Compared with the control of cottonseed protein, the dry adhesive strength was increased by 22%. The best addition amount of CNC is 10%, and the strength is increased by 16%. The heat resistance of cottonseed protein isolate can be improved by adding CNF, but not by adding CNC, so CNF is the best environmental modifier [12]. The soybean protein molecule was degraded into small polypeptide chains by bromelain, and then the polypeptide chains were recombined by the biologically derived cross-linking agent triglycidyl amine (TGA). The wet shear strength of the modified soybean protein adhesive reached 1.11 MPa, which was 76.2% higher than that of the unmodified soybean protein adhesive [13].

2.4. Other Protein Resources

In the research of plant protein adhesive, the research on soybean protein adhesive is relatively mature and has achieved industrialization. There are also some preliminary exploration results for other plant proteins, such as peanut protein, cottonseed protein, wheat protein, oilseed protein, etc [14-16]. At present, the plant protein adhesive still has many shortcomings, such as poor water resistance, poor corrosion resistance, short storage period, etc. It will take a long time for the protein adhesive to replace the "trialdehyde adhesive" as a wood adhesive. In order to achieve the goal as soon as possible, many scholars need to continue to explore the research path of the protein adhesive. When the shortcomings of the protein adhesive can be overcome, the plant protein adhesive will have a better development prospect.

3. Modification Method of Plant Protein Adhesives

3.1. Physical Modification

Physical modification refers to a method to change the spatial structure of protein and the bonding mode between protein peptide chains through a series of physical methods such as mechanical treatment, temperature control, etc., so as to cause protein denaturation and improve the performance of protein adhesive. Common physical modification methods include thermal modification, ultrasonic modification, microwave modification, high-pressure homogenization modification, etc.

3.1.1. Thermal Modification

Thermal modification is the most commonly used method in physical modification. Under high temperature, the structure of protein will change, causing the orderly arrangement of protein molecules to be removed. Some non-polar groups originally inside the molecules will be exposed to the surface of the molecules, increasing the interface activity of protein. However, it should be noted that the temperature should not be too high when using the heating modification method. When the temperature exceeds 110 ^{o}C, the protein will be completely denatured, resulting in a decrease in the adhesive strength. The water resistance of soy protein adhesive is limited by simple thermal modification, and the wet bonding strength of the modified adhesive cannot meet the requirements of Class II plate in the national standard. Therefore, most studies combine thermal modification with other modification methods.

3.1.2. Ultrasonic Modification

Ultrasonic modification refers to a modification method that produces high temperature and high pressure effects in solution to decompose proteins under the action of high intensity ultrasound. After ultrasonic treatment, the internal bonds between soybean protein molecules break, the folding and winding structures of protein molecules are unfolded, and the secondary structure changes, exposing the hydrophobic ends of protein to the outside, reducing the binding ability of protein and water, The surface hydrophobicity of the prepared protein adhesive was improved.

Other scholar studied the effect of ultrasonic treatment time on the bonding performance of soy protein adhesive, and found that when the microwave treatment time was 40 min, the water resistant bonding strength of soy protein adhesive increased with the extension of treatment time, because the sulfhydryl group and hydrophobic group contained in the protein were more exposed under the extension of ultrasonic treatment time, and the hydrophobic interaction between protein molecules increased, As a result, the bonding strength and water resistance are improved. When the ultrasonic treatment time was 40 min, the wet bonding strength of soybean protein adhesive could reach 0.77 MPa, which reached the national standard for Class II plywood. The best time for ultrasonic treatment of soybean protein is 40 minutes. When the treatment time is more than 40 minutes, the spatial structure of soybean protein stretch further changes, the protein molecules reassemble, and the hydrophobicity decreases, which reduces the water resistance of the adhesive.

3.1.3. Microwave Modification

Microwave treatment is to treat protein by electromagnetic wave from 300MHz to 300GHz, so that protein molecules can move. Under the dual action of heat and mechanical force formed under high-speed oscillation, the balanced distribution of protein polar molecules is destroyed, and the protein polar molecules are polarized into an orderly molecular structure, so that some nonpolar groups inside the molecules are exposed to the molecular surface, thus effectively improving the adhesion performance of soybean protein. Microwave modification technology was applied to the preparation process of wheat protein adhesive. When the power was 600w for 90min, the prepared wheat protein label adhesive had the best water resistance. In addition, the viscosity of the adhesive prepared by microwave treatment under the same conditions is higher than that of the untreated one. This is because under the action of microwave, the spherical structure of the protein molecule unfolds, exposing the hydrophobic groups inside the protein molecule, which enhances the interaction between protein molecules and the bonding surface with other substances, forming a new network structure, thus increasing the viscosity and bonding strength. The microwave frequency also has an impact on the performance of the protein adhesive. The molecular conformation of the protein molecule changes at a higher frequency, which promotes the improvement of the water resistance of the adhesive. When the frequency is too high, the protein molecules condense and settle, leading to a decrease in solubility and adhesive strength.

3.1.4. High Pressure Homogenization Modification

High pressure homogenization modification refers to a method to improve the performance of soy protein adhesive by destroying the hydrogen bond, ionic bond and other noncovalent bonds in soy protein through extrusion within the high pressure range of 100 MPa to 1000 MPa, and changing the secondary structure of soy protein molecules. After high-pressure homogenization treatment, the particle size of soybean protein becomes smaller, the viscosity of the obtained protein adhesive decreases, and the water resistance is enhanced. This is mainly due to the expansion of the protein molecular structure, the increase of free sulfhydryl content, and the exposure of more hydrophobic groups in the molecule and the hydrophobic residues of the broken small molecular peptide chain. The protein solubility is increased and the viscosity is moderate, and the penetration of the adhesive layer is good, which makes the soy protein adhesive better combine with the bonding surface, and further improves the water resistance of the adhesive.

The physical modification method has the advantages of short processing time and simple operation, which can change the high-level structure and aggregation morphology of protein molecules, but has certain limitations on the modification effect of protein adhesives. Therefore, physical modification is often used as an auxiliary means in combination with other modification methods.

3.2. Chemical Modification

The chemical modification method is more thorough than the physical modification method. On the one hand, it changes the molecular structure of the protein; on the other hand, it enables the protein to form an interaction force with the bonded wood surface, thus improving the adhesive bonding strength and water resistance. In essence, chemical modification is to use the properties of the protein itself to insert or insert new functional groups into its molecular chain to achieve the purpose of modification, improve the functional properties of the protein and improve the physical and chemical properties of the protein adhesive by changing the spatial structure of the protein. Common chemical modification methods include alkali modification, surfactant modification, graft modification and acylation modification [17].

3.2.1. Alkali Modification

Alkali modification is the most common and simplest method to prepare protein adhesives. Under alkaline conditions, the structure of protein molecules is destroyed, the molecular chain expands, the hydrophobic groups and active groups buried inside are exposed, and directly contact with water. While improving the solubility of protein molecules, the water resistance of protein adhesives is also improved. Common alkaline modification reagents include sodium hydroxide (NaOH), calcium hydroxide ($Ca(OH)_2$), borax ($Na_2B_4O_7$), ammonium hydroxide (NH_4OH), etc.

As a strong alkaline modifier, NaOH has better modification effect than weak alkaline reagent. The research shows that the adhesive bond strength of alkali modified adhesive can be more than two times higher than that of unmodified adhesive, which shows that NaOH can significantly improve the bond strength of soybean protein adhesive. The reason is that alkali treatment can improve the solubility of protein and increase the adhesion of protein. At the same time, NaOH can disperse, unfold and partially degrade soy globulin molecules, expose more protein subunits, internal hydrophobic groups and active reaction groups (carboxyl, hydroxyl, etc.), enhance their reactivity during hot pressing, and reduce the content of hydrophilic groups. During hot pressing, the modified protein interacts with the surface of the bonded wood, thereby improving the water resistance of the protein adhesive. The composite alkali modification has better effect than the single alkali modification. However, the alkali modified protein adhesive is easy to cause discoloration of wood. In view of this phenomenon, researchers can effectively improve the discoloration problem by combining acid and alkali modification.

The viscosity of the protein adhesive first increased and then decreased with the increase of the concentration of alkaline reagent added. This is because when the concentration is low, the spherical structure of the protein is destroyed, and the viscosity is increased due to the unfolding and unwinding of the molecular chain. When the concentration is too high, the protein is completely degraded, and the long-chain amino acids are destroyed into peptide chains with smaller molecular weight, which reduces the viscosity. Liu added NaOH to the soy protein adhesive for modification. Under a series of alkali concentration gradients in the experiment, it was found that when the amount of alkali added was 0.9% (accounting for the weight of dry matter of soy protein), the viscosity reached the maximum. Therefore, the proper alkali concentration should be selected when alkali modification is used, regardless of viscosity or adhesive strength.

3.2.2. Surfactant Modification

Surfactant is a kind of amphiphilic molecule with hydrophobic and hydrophilic groups in the molecular structure. By blocking the hydrophobic interaction within the protein molecule, the non-polar group of the protein will interact with its hydrophobic group after exposure to form micelles to increase hydrophobicity. At the same time, more non-polar groups in the protein glue will interact with the wood surface during hot pressing, thus, the water resistance of the modified adhesive is improved. Sodium dodecyl sulfonate (SDS) and sodium dodecyl benzene sulfonate (SDBS), as anionic surfactants, are often used to modify protein adhesives.

The modification effect of surfactant alone is limited, and it is often used in combination with other reagents or modification methods in research. Alkali modification could be combined with surfactant modification, and the wet bonding strength of the prepared protein adhesive after immersion can reach 0.98MPa. Wang used SDS borax composite modified soy protein isolate adhesive. When the amount of SDS added is 0.7% of the mass of soy protein isolate, the amount of borax added is 5% of the mass of soy protein isolate, and the reaction temperature is 55°C, the water resistant adhesive strength can reach 1.97MPa. After the composite modification of borax and SDS, the secondary conformation of the protein has changed, which is the direct reason for the improvement of water resistance [18].

3.2.3. Graft Modification

The principle of graft modification is to react with protein by adding specific chemical reagents, so that a new branch chain or active site can be generated on the protein molecular chain, and then introduce the graft monomer with double bond structure to the active site. The grafted protein has a more active reaction group. During the hot pressing process, the prepared protein adhesive can form a dense three-dimensional network structure with the bonding wood, which prevents water molecules from penetrating into the adhesive layer, thus improving the water resistance of the adhesive [19]. Common graft monomers include polyamide, maleic anhydride, glycidyl methacrylate (GMA), etc.

The unsaturated double bond structure in maleic anhydride can further modify proteins. Maleic anhydride is connected to soybean protein molecules through amide and ester bonds, reacts with amino groups in proteins to form amide bonds, and reacts with hydroxyl groups to form ester bonds. However, the glued plywood modified by maleic anhydride was delaminated in the boiling water test. However, the water resistance of the plywood was greatly improved after the combination of maleic anhydride and polyethylene imine. This is because the amino group on the polyethylene imine reacted with the amide in the maleic group through Michael addition to produce a highly cross-linked polymer with water insoluble network structure, which enhanced the water resistance of the adhesive [20]. Soybean protein adhesive could be modified with glycidyl methacrylate as graft monomer and ammonium persulfate sodium bisulfite redox system as initiator system. The water resistant bonding strength of the three-layer plywood prepared under the hot pressing temperature of 150 ^{o}C and the hot pressing pressure of 1.0 MPa for 10 min exceeds the corresponding national standards. Spirulina protein isolate was used as the protein raw material, glycidyl methacrylate as the graft monomer, and ammonium persulfate as the initiator to carry out the graft copolymerization. Under the optimal process conditions, the wet bonding strength of the modified adhesive can reach 1.45MPa, and the wood breaking rate is 100%, which is 2.1 times higher than that of the unmodified adhesive.

3.2.4. Acylation Modification

The acylation modification refers to the reaction between the electrophilic group carbonyl group in the acylation reagent and the nucleophilic group amino group and hydroxyl group in the protein molecule, so that the electrostatic force inside the protein molecule is destroyed, the polypeptide chain is stretched, the solubility of the protein is increased, and the hydrophobic group inside the molecule is exposed, promoting the improvement of water resistance. Succinylation and acetylation are the two most commonly used acylation methods. Succinylation is to connect succinic acid hydrophilic groups to the ends of protein molecules, and then long carbon chain lipophilic groups are introduced through catalytic reaction to obtain proteins with bipolar groups; Acetylation is the process of introducing acetic anhydride into the end of protein molecular chain. Soybean protein adhesive was modified with acylation reagent. The wet bonding strength of plywood bonded with modified adhesive prepared by adding 10% acylating agent, 6% degreased soybean powder and reacting for 60 min at 55°C after immersion treatment is 2.63 MPa, which is far greater than the 0.7 MPa required in the national standard. Diacetyl tartaric acid acylating agent was added to the soybean protein pretreatment solution to modify it, and explored the optimal conditions for modifying soybean protein by adding the amount of acylating agent and pH value through orthogonal test. The results showed that when pH was 10.8 and the addition amount of diacetyl tartaric acid was 0.4g, the dry bond strength of the modified soybean protein adhesive reached 2.08MPa, and the wet bond strength was 1.75MPa.

3.3. Enzyme Modification

The biological modification of adhesives can be divided into bioengineering modification and enzyme modification. Bioengineering modification mainly improves the functional properties of proteins by changing the molecular structure of proteins at the gene level through the application of genetic engineering, plant breeding and other transgenic technologies. Enzymatic modification refers to the partial degradation of protein under the action of protease to increase special functional groups that can cross link or connect within or between protein molecules [21]. Enzymatic modification is characterized by mild reaction, fast reaction rate and strong specificity. It can quantitatively control the degree of hydrolysis of protein and effectively improve the modification efficiency. According to the source of enzyme, it can be divided into plant protease, animal protease and microbial protease.

In a study, laccase was added to alkali lignin soybean protein adhesive to modify it. At the ratio of alkali lignin to soybean protein 1:1, the adhesive made of soybean protein with laccase accounting for 60% of alkali lignin achieved a bonding strength of 1.01MPa.

4. Summary

This paper introduces the research progress and common modification methods of plant protein adhesive. Although the protein adhesive has the advantages of non-toxic and environmental protection, it also has the disadvantages of poor water resistance, so it needs to be modified. The previous article introduced many modification methods to improve the water resistance of protein adhesive, such as physical modification, chemical modification and enzyme modification. Among them, the physical modification methods include thermal modification, ultrasonic modification, microwave modification, and high pressure homogenization modification. The chemical modification methods include alkali modification, surfactant modification, graft modification, and acylation modification. However, the wet bonding strength of most of the modified methods meets the requirements of Class II boards, and most of them are only in the laboratory stage. In comparison, what has more practical application value is to use synthetic resin for blending or copolymerization modification. Utilizing the excellent curing performance of synthetic adhesive to provide an excellent three-dimensional reticular framework for protein adhesive, so as to improve the wet bonding strength, which is also the research direction of many researchers at present. With the gradual improvement of the performance of plant protein adhesive, it will inevitably become one of the main wood adhesives to be used in the future.

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