Scientific Research Publishing

Green and Sustainable Chemistry, 2023, 13, 237-253
https://www.scirp.org/journal/gsc
ISSN Online: 2160-696X
ISSN Print: 2160-6951

Environmentally Friendly Chitosan-Based Wood/Wood Composite Adhesive: Review

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How to cite this paper: Dhawale, P., Gadhave, S. and Gadhave, R.V. (2023) Environmentally Friendly Chitosan-Based Wood/Wood Composite Adhesive: Review. *Green and Sustainable Chemistry*, **13**, 237-253.
https://doi.org/10.4236/gsc.2023.134013

Received: September 4, 2023
Accepted: November 3, 2023
Published: November 6, 2023

Abstract

Due to the world's energy issues and dependency on petroleum resources, focus has switched to finding new, sustainable raw material sources for wood adhesives. Renewable biopolymers would gradually replace petroleum and natural gas as the primary raw materials used in wood adhesives. Chitosan is a biomass substance having a lot of reserves. Chitosan is one of the most fascinating biopolymers in the adhesive sector because of its potential qualities for adhesive applications, such as biodegradability, biocompatibility, and non-toxicity. Chitosan and its derivatives have so garnered considerable interest in a wide range of adhesive applications. However, its adhesive strength is insufficient to glue wood under normal, humid conditions. There has been a lot of study done on how to make chitosan-based adhesives more cohesive and water resistant. In order to effectively use chitosan-based wood glue in wood/wood composite adhesive that gives comparable performance to synthetic adhesives, numerous new ways have been developed. It has been modified by the addition of various cross-linkers, including aldehydes like glyoxal glutaraldehyde etc., epoxy compounds, blended with other polymers, different acids and chitosan grafted onto vinyl acetate. In the production of wood composites, chitosan can also scavenge formaldehyde. This review of chitosan-based adhesives focuses on various cross-linkers for chitosan modification in order to improve the properties of chitosan-based wood adhesives.

Keywords

Chitosan, Wood, Composite, Biopolymer, Adhesive

1. Introduction

Every year, a lot of wood adhesives are made and utilised for things like load-bearing structures, flooring, furniture, windows, doors, plywood, glulam beams, and particleboards [1]. Historically, natural polymers like starch and proteins from milk or blood were used to make wood adhesives. However, due to their superior qualities regarding bonding performance, ease of handling, and advantageous cost development, fossil-derived polymers displaced natural polymers during the 1960s [2].

The market for wood adhesives has been valued at $4.6 billion in recent years, with 4.7% predicted annual growth through 2025 [3] [4]. Since their introduction in the middle of the 20th century, synthetic adhesives have outperformed their bio-based equivalents in terms of bonding performance, usability, and affordability. Petroleum derivatives, which are used in fossil wood adhesives, perform well today, but petroleum resources are finite and not renewable [5]. The years following World War II were an ideal time for the synthesis and development of numerous different synthetic polymers. Some were particularly effective as wood adhesives. Polyvinyl acetate (PVAc), isocyanates, and polyurethane were the most common ones. Others included building adhesives, aliphatic resins, and epoxies. Most of the petrochemical materials used to make the adhesives for furniture (such as PVAc, phenol formaldehyde (PF), urea-formaldehyde (UF), melamine-urea-formaldehyde (MUF) and polyurethane (PU)) are non-renewable. Toxic chemicals including formaldehyde and volatile organic compounds are used to make these adhesives. Finding natural and renewable molecules with strong binding capabilities has proven to be extremely difficult due to the development of wood adhesives with the objectives of energy conservation, low cost, no pollution, adequate viscosity, and being solvent-free [6]. Currently, bamboo composites are mainly bonded with PF [7] [8] [9], MUF, polyurethane (PUR), PVAc [10]. However, the continued release of formaldehyde from adhesives containing formaldehyde poses a risk to human health and the environment. Due to their toxicity, the formaldehyde-free adhesives have issued such high cost, challenging application processes, poor occupational health, and safety [11].

2. Biobased Wood Adhesive

In addition to using up petroleum resources, using formaldehyde-based adhesives inevitably emits formaldehyde both during production and use. Sustainable development has received exceptional attention in recent years due to the worry about non-renewable fossil fuel resources and human pursuit of an ecological environment. This emphasis has led to research on environmentally friendly wood adhesives using biomass as a raw material [12]. So-called "nature-inspired adhesives" have gained popularity in recent years because of their ability to resemble the structure of natural adhesives [13] [14] [15] [16] [17]. Our continued research for environmentally acceptable and sustainable materials is advancing the use of biomass-based adhesives. According to a study by Narayan, it is necessary to develop bio-based polymers that have features similar to those of conventional chemicals or to add new, beneficial properties, preferably at no additional expense [18]. Among the interesting biopolymers that have been suggested as potential adhesives are protein, tannin, lignin, and polysaccharides [19] [20] [21] Sustainable regeneration of natural biomass adhesives, including tannic acid, protein, polysaccharide, chitosan, and lignin, has drawn interest [22]. However, they are typically not utilised by themselves. They are derivatives of wood and have mechanical qualities, low water resistance, alteration, and complicated preparation issues [23]. Therefore, it is important to develop a straightforward chemical crosslinking modification to transform renewable biomass energy into high-performing, dimensionally stable, and formaldehyde-free medium density fiberboard (MDF) wood adhesives [24] [25] [26]. The similar behaviour was obtained by Norström *et al.* using locust bean gum dispersion [27]. Wood adhesives made of protein, tannin, chitosan, and polysaccharides can be used [28].

3. Chitosan: Structure and Chemistry

Shrimp, crabs, and insects have crusts that contain chitosan, the most common amino polysaccharide in nature. It is only naturally found in the Mucoraceae family of fungi, although it is easily obtained by deacetylating chitin [29]. Structure of chitosan is shown in **Figure 1**. High biocompatibility, low toxicity, biodegradability, and antibacterial capabilities are just a few of the exceptional qualities of chitosan [30].

Chitosan's distinctive qualities have drawn a lot of interest due to its abundance of natural resources and its great capacity to produce useful commodities [31]. The chitosan polymer's component units (glucosamine) and the glucose units in cellulose share a lot of structural similarities [32] [33]. The only variation is the presence of amino groups in the glucosamine structure, which gives the polymer its distinctive characteristics. The paper and textile industries were paying particular attention to this structural similarity to cellulose polymer as it could strengthen paper's mechanical properties if added to the structure. These characteristics allow chitosan to form chemical bonds with metal ions, lipids, and proteins as well as cellulosic fibres [34].

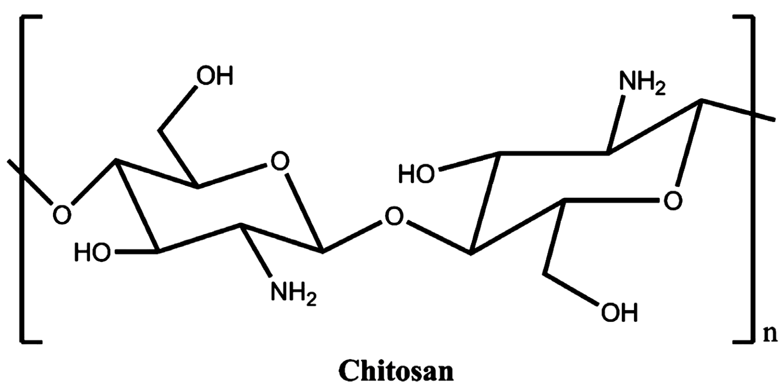

Figure 1. Structure of chitosan.

4. Chitosan-Based Adhesives

Chitosan is a naturally occurring polysaccharide that is produced by deacetylating chitin and is found throughout nature, particularly in the shells of insects, prawns, and crabs. Thus, it ranks as one of the most abundant and sustainable organic resources in the planet [35]. Chitosan has been receiving more and more significant attention as a result of its exceptional qualities, including blood compatibility, biosafety, the capacity to form films, and microbial degradability. Numerous applications in a variety of disciplines, including medicine, food, the chemical industry, cosmetics, water treatment, metal extraction and recovery, and biochemical and biomedical engineering, have resulted from extensive study on it [36]-[42]. Chitosan is the only basic polysaccharide found in natural polysaccharides, and it has numerous distinctive qualities like biodegradability, cell affinity, and biocompatibility. Because of its properties, chitosan has good biological properties and can be modified chemically. The wood adhesive has been treated with modified chitosan to significantly increase the adhesive's waterproof strength, lower production costs, and increase added value [43]. Only a portion of chitin is converted into chitosan, which is a derivative of chitin [44]. It has been widely used in a variety of sectors, including wood, biology, medicine, and others because of its excellent mechanical and adhesive qualities as well as its biocompatibility and biodegradability [45] [46] [47]. In order to offer bonding between materials, the straight chain structure has numerous highly active amino and hydroxyl groups that can react with other chemical groups to produce a superior biomass adhesive [48]. In recent years, not only have chitosan-based adhesives been used to help wood materials achieve good mechanical qualities, but a successful biological adhesive based on chitosan that combines it with other substances has also been developed [49]. A heteropolymer composed of 2-acetamido-2-deoxy-D-glucopyranose and 2-amino-2-deoxy-D-glucopyranose, chitosan is produced from the chitin of crustaceans, insects, and fungi by alkaline deacetylation [50]. Even while acidic solutions of chitosan can be employed as adhesives, either by itself or in combination with other polysaccharidic substances as Konjac glucomannan or xanthan, they have weak bonding characteristics in a moist environment [51] [52] [53] [54] [55]. The many hydroxyl and amine groups in chitosan make up a unique class of sticky polysaccharides that can interact with a wide range of chemical processes. Chitin, an amino polysaccharide deacetylated from chitin that is found in abundance in the shells of marine crustaceans like crabs and prawns as well as in the cell walls of fungi, is the source of chitosan. A number of randomly positioned N-acetyl-glucosamine residues are interspersed between 1,4-linked D-glucosamine residues to form the chemical structure of chitosan. Chitosan can interact with negatively charged molecules because it is soluble in mildly acidic water solutions and exists as a cationic polyelectrolyte. Chitosan has an adhesive quality, to put it another way. Chitosan has attracted a lot of attention as a potential polysaccharide resource in many domains, and it has been thoroughly explored for medicinal and industrial uses;

however, there have only been a small number of studies on the use of chitosan as a wood adhesive. The bonding properties of wood were poor when chitosan alone was utilised as a wood glue, according to an early investigation at FP Innovations. A novel feasibility research on chitosan adhesives was recently undertaken to use specific fungal species to modify chitosan and increase its bonding property in order to improve the adhesive quality of chitosan resin [56].

5. Chitosan-Based Wood Adhesive

Multipurpose wood adhesives with high cold pressing strength, water resistance, hardness, and mildew resistance are still difficult to develop. A multifunctional organic-inorganic hybrid soybean meal (SM)-based adhesive has been developed here, inspired by oysters, by incorporating functionalized silica nanoparticles and dialdehyde chitosan (DCS) to the SM matrix. The stiff nanofillers uniformly dispersed in the SM matrix and the contacts between organic-inorganic phase interfaces were successfully improved by DCS, resulting in energy dissipation that increased the adhesive's tensile strength [57]. A synthesis-free and totally biomass adhesive composed of chitosan and tannin (CST) was successfully developed by a simple process and was inspired by the phenol-amine chemistry of mussels. Testing was done to determine how well CST adhesive adhered to bamboo, wood, and bamboo-wood surfaces. Given its many benefits, including as exceptional water resistance, ease of preparation, full biodegradability, and low cure temperature, CST adhesive has the potential to be an excellent substitute for formaldehyde-based resin for bonding wood and bamboo [58]. The chitosan-tannin (CST) adhesive displayed outstanding bonding strength for wood and bamboo substrates, confirming that the combination of amino-rich chitosan and catechol-rich tannin is a successful technique for imitating the natural phenol-amine adhesive system [59]. Through the use of vanillin (V) crosslinked chitosan adhesive and hot pressing, an eco-friendly MDF was developed. The method of crosslinking and the impact of various chitosan/vanillin addition ratios on the mechanical characteristics and dimensional stability of MDF were investigated. According to the findings, the aldehyde group of vanillin and the amino group of chitosan undergo a Schiff base reaction, which causes them to crosslink to form a three-dimensional network structure. Consequently, MDF that has been V-crosslinked chitosan attached to it can be a potential alternative for wood panels that are friendly to the environment [60].

Crosslinking agents that have low toxicity are frequently utilized in chitosan-based binders due to the issue of formaldehyde emission in conventional MDF. Vanillin, a natural anti-environmental substance, was employed as a crosslinking agent. These outcomes outperform those of chitosan- and wood-fiber composites. The newly developed amide bond and hydrogen bond significantly enhance the mechanical properties, which is the cause of this [61]. Biomass material chitosan has a lot of reserves. In this work, it was used to develop a wood adhesive that is favorable to the environment. Petroleum-based aldehydes were replaced by carbohydrates, including glucose, sucrose, and starch that had undergone specialised oxidation as hardeners because of their unreliability, volatility, and toxicity. It has been demonstrated that oxidised carbohydrates can significantly increase the binding strength and water resistance of chitosan adhesives, with oxidised starch outperforming glucose and sucrose in this regard. When 10% sodium periodate is utilised for the starch's specific oxidation, the best chitosan-oxidized starch adhesive can be made by treating 8 mass% of oxidised starch mixed with a chitosan solution at room temperature heating analysis. It shown that the chitosan-oxidized starch adhesive cures at a lower temperature than the other adhesives in this work, this being one of the reasons for chitosan having a better bonding performance [62]. Therefore, the capacity of suppliers to deliver consistent quality raw materials will be an important consideration in the development of industrial solutions for the use of chitosan as adhesives. This work examined the bonding and modification mechanisms of chitosan modified starch film. The wet and dry shear strengths of plywood are enhanced by the modification of starch with chitosan [63]. The graft copolymerization of vinyl acetate (VAc) onto the chitosan chain was started using a redox initiator, cerium ammonium nitrate, in a dispersion polymerization at 60˚C. After 2 hours of processing, it was discovered that 0.5 - 7.5 g of chitosan based on 50 g of VAc resulted in a monomer conversion of between 70% and 80%. The amount of chitosan added increased the grafting efficiency; nevertheless, the grafting ratio first got and then started to decline. According to the experimental findings, adding polyvinyl acetate to the chitosan chains improved the material's toughness and reduced its water absorption [64].

PVAc is a polymer that is typically used as a binder in wood adhesives. PVAc-based adhesives have the benefits of being simple to use and lack formaldehyde, a dangerous ingredient that is frequently found in commercial adhesives. PVAc adhesives, on the other hand, have a fossil-based composition and can only be used indoors because to their weak water resistance. The purpose of this work is to develop a wood glue with superior characteristics and a higher biobased content by mixing PVAc and the biobased polysaccharide chitosan. In addition to being biobased, chitosan exhibits excellent bonding properties; yet, the high viscosity of chitosan dispersions restricts its use. To combine the promising bonding abilities of chitosan with the practicality of PVAc, VAc was grafted from chitosan in this work [65] using an emulsion polymerization process. The chitosan-graft-PVAc adhesive's increased qualities and continued application, as well as the potential to replace about 20% of the fossil-based material with biobased material, are encouraging aspects for the competitiveness of this type of wood adhesive [66]. Lignin can be altered and its scope of use increased by blending with other biopolymers, which is a promising method. The second-most prevalent polysaccharide on Earth, chitosan, is a perfect biopolymer to mix with lignin to produce wood adhesives [53]. Due to its potential use as a bioadhesive, this polysaccharide, produced via alkaline deacetylation of chitin, has drawn a lot of interest. The biological field and, more recently, the wood building industry were the two main areas where bioadhesives underwent significant growth [67]. Both the molecular weight and the degree of deacetylation of chitosan are factors that affect the bioadhesives attractive features, and numerous research have amply demonstrated that the adhesive capabilities changed when the molecular weight and deacetylation level decreased [68]. Additionally, chitin is mostly extracted commercially from the exoskeletons of prawns and crabs, which are leftovers from the seafood processing industry. Utilising the biopolymer in wood adhesive applications allows for effective handling of chitinous waste due to all of these features. Recent years have seen substantial research into the potential of chitosan-lignin composites for dye and metal ion adsorption [69] [70]. Using chitosan and ammonium lignosulfonate as the primary raw materials, a very effective technique to produce high-performance chitosan-lignin wood adhesives was developed, and the effectiveness of the resulting adhesives was evaluated by producing MDF. The findings showed that chitosan had substantial effects on bonding strength and water resistance, and that the best method significantly increased the performance of the chitosan lignin adhesive: 6% of chitosan-lignin adhesive concentration and a 1:2 weight ratio between the two substances [71].

Hot-pressing was used to produce high-performance wood-based fiberboards with high strength and dimensional stability utilising 2,5-dimethoxy-2,5-dihydrofuran cross-linked chitosan as a sustainable binder. On the mechanical characteristics and dimensional stability of wood-based fiberboards, the impacts of cross-linked chitosan were investigated. The amide connections and hydrogen bonds between the wood fibres and cross-linked chitosan could be responsible for the improvement of the physical and mechanical characteristics of wood-based fiberboards. The eco-friendly wood-based composites that may be made from the high-performance wood-based fiberboards that were developed in this study appear promising [72]. Double-lap shear tests were used to determine the binding strength of several chitosan-based formulations. The adhesive with the highest performance was made up of 6% CS, 1% glycerol, and 5 mmol/L trisodium citrate dehydrate. This formulation's best bond strength was determined to be 6.0 MPa in dried conditions and 1.6 MPa in wet conditions [73].

6. Application of Chitosan as Scavenger

Chitosan was utilised as a biodegradable, ecologically friendly scavenger that reduced MDF's formaldehyde emission. Liquid C-NMR measurements revealed that the amount of free formaldehyde in the urea formaldehyde adhesive containing chitosan powder was significantly reduced. Chitosan powder had a greater capacity for formaldehyde adsorption than chitosan solution. Preparing an adhesive Chitosan performance as an eco-friendly scavenger has been evaluated using chitosan in various solution and powder kinds with and without hardener in the glue [74]. Cross-linking reaction between chitosan and formaldehyde is shown in **Figure 2**.

Aldehyde-based resins selected from the group of UF resin, MF resins, MUF resins, PF resins, and mixtures, combinations, and sub-combinations thereof, have been included in the adhesive composition disclosed. A formaldehyde scavenger chosen from the group of chitosan, nano-chitosan [75]. Chitosan-reinforced UF adhesives are utilized for joining fibrous materials or wood-based composites such as plywood and particleboard. The adhesives are made by combining raw materials containing chitosan that has not been changed with a UF resin to make wood composite adhesive resins [76].

7. Crosslinked Chitosan-Based Adhesives

By mixing chitosan-based adhesives with other biopolymers-based adhesives and making chemical changes to increase their water resistance, chitosan-based adhesives' poor performance in wet conditions can be improved; however, doing so would drastically raise their costs [45]. Chitosan is occasionally employed as an adhesive in the wood sector, though. This could be mostly due to its high viscosity and limited solubility. A more serious downside is that when reactive low molecular weight aldehydes (such formaldehyde, glyoxal, and glutaraldehyde) are used as hardeners, chitosan solutions will quickly solidify in a matter of seconds. Reaction of chitosan and glutaraldehyde is shown in **Figure 3**.

This prevents it from spreading on the wood's surface. According to research on the development of "green" wood adhesives using chitosan, octanal was utilised as a chemical modifier to improve water resistance and produce acceptable adhesive qualities [77] [78] [79] [80]. Although glutaraldehyde, glycidyl ether, and citric acid have been successfully used in polysaccharide-based wood adhesives, some drawbacks remain, including the high cytotoxicity of these substances, the need for higher temperatures, and the need for an additional catalyst in the crosslinking reaction of citric acid [81]. Cross-linking reaction of chitosan with citric acid is shown in **Figure 4**.

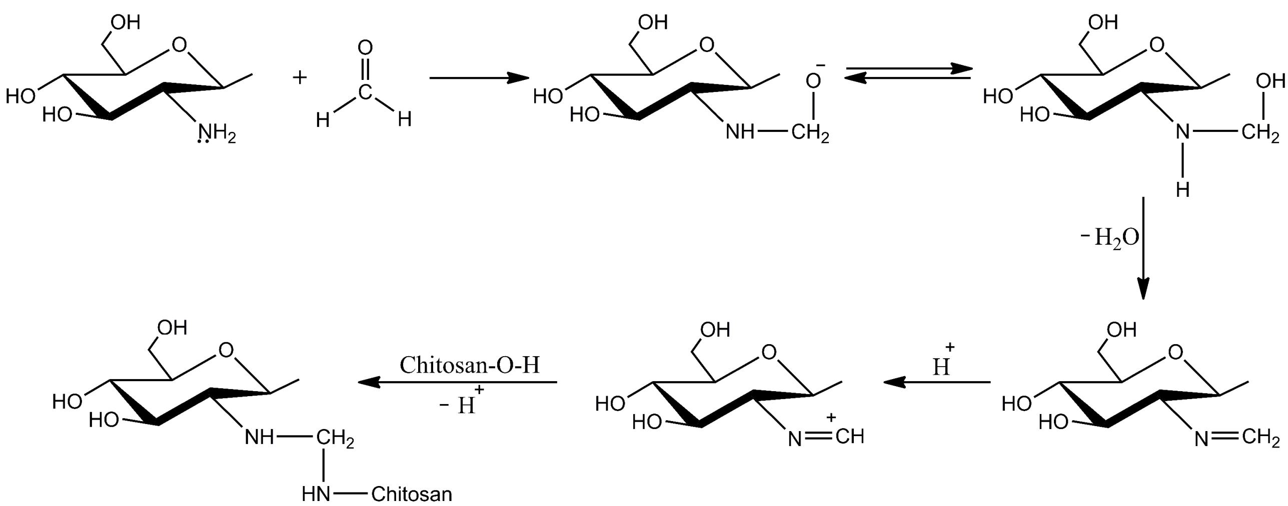

Figure 2. Cross-linking reaction between chitosan and formaldehyde [74].

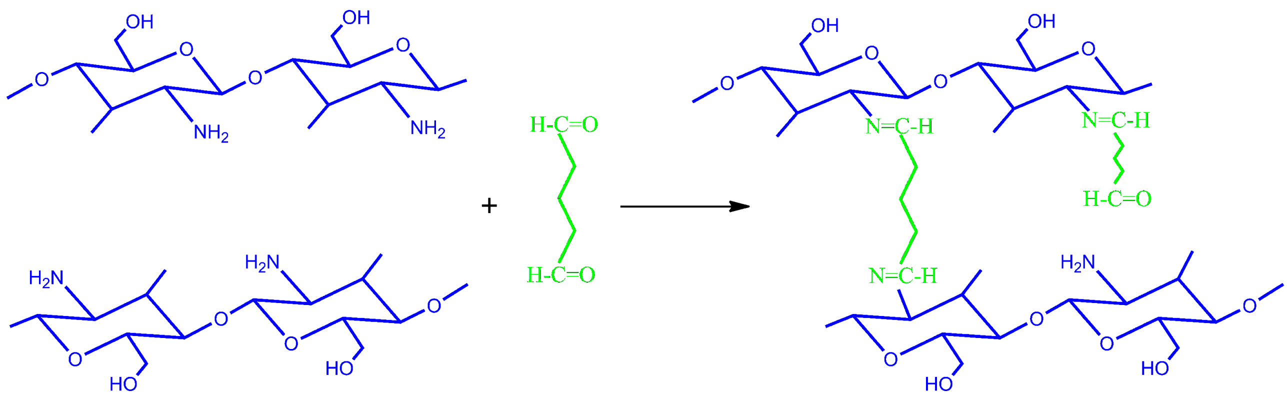

Figure 3. Cross-linking reaction of chitosan with glutaraldehyde [77].

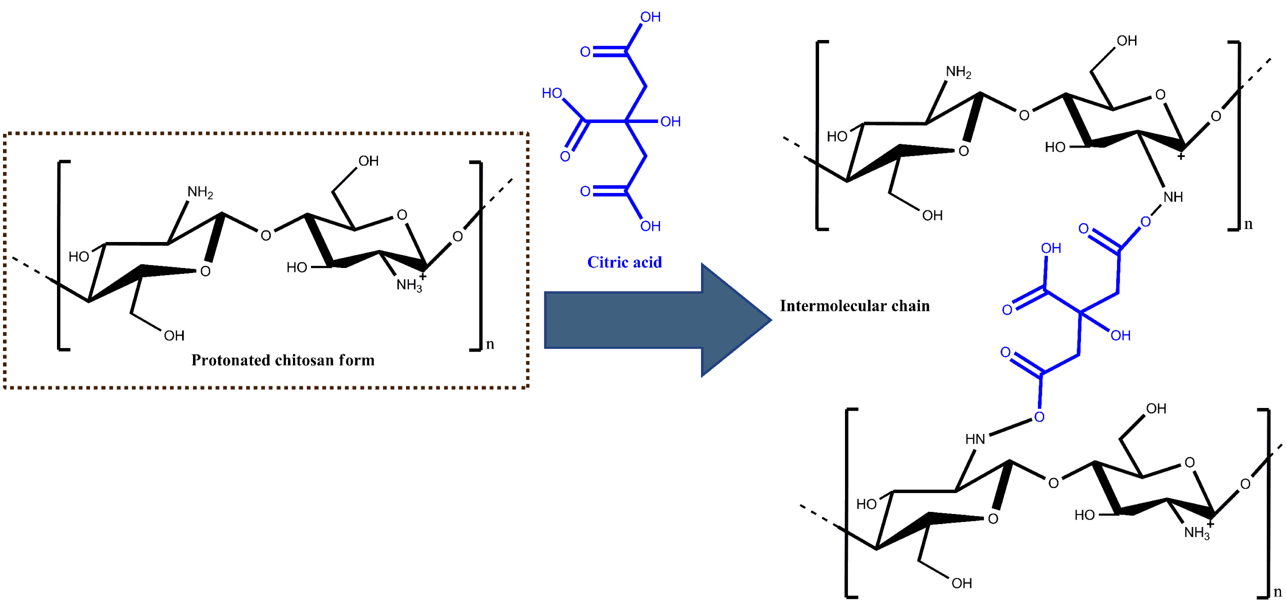

Figure 4. Cross-linking reaction of chitosan with citric acid [82].

However, the chitosan linear structure's weak mechanical qualities limit the scope of its applicability. Chemical crosslinking produces a stable three-dimensional network structure to fix this flaw [82] [83]. A lignosulfonate/chitosan-based adhesive was developed by JI *et al.* [84] using glutaraldehyde as a crosslinking agent. Using the hot-pressing method, Huang *et al.* [85] developed ecologically friendly medium-density fiberboard using 2,5-dimethoxy-2,5-dihydrofuran-crosslinked chitosan as a binder. The findings suggested that crosslinked chitosan might significantly enhance the dimensional stability and mechanical characteristics of wood-based panels. To test the adhesion of an ecologically friendly chitosan adhesive made via selective oxidation modification using glucose, sucrose, and starch as hardeners, Xi *et al.* created three layers of laboratory plywood [86]. The findings revealed that oxidised carbohydrate has good bonding performance and can effectively increase the binding strength and water resistance of chitosan glue. The low toxicity of the crosslinking agent, however, poses a health risk [83].

It is possible to find 4-hydroxy-3-methoxybenzaldehyde both naturally and artificially. It is a perfect harmless crosslinking agent for chitosan and belongs to the phenolic group structurally [86]. Mati-Baouche *et al.* recommended replacing the amine functionalities of chitosan with alkyl groups to boost the water resistance of chitosan formulations. In comparison to native ones, these alkyl-chitosans showed greater water resistance and enough adhesive capabilities on wood. However, the use of amine functions instead produced weak strength resistance. Indeed, Noto *et al.* have demonstrated that the amino groups have a favourable impact on the adhesive strength of chitosan [87].

8. Conclusion

The legislation and growing demand for ecologically friendly adhesives are the key drivers of the rising interest in chitosan-based adhesives. The wood composite industry makes extensive use of the formaldehyde-emitting substances UF, UMF, and PF. The use of synthetic products is accordingly recognised by society as a key contributor to environmental issues, and as a result, the use of renewable natural resources is encouraged. Chitosan is a substance that works well for making wood composite adhesives since it is simple to produce and use. The low water resistance of chitosan-based adhesives can be enhanced by cross-linking the chitosan. The modification and cross-linking of chitosan with synthetic materials has already been the subject of extensive research. Chitosan is crosslinked with acids like citric and boric acid as well as aldehydes like glyoxal and glutaraldehyde to increase the adhesive's strength and resistance to moisture. Chitosan also can scavenge formaldehyde. For the wood/wood composite industries, chitosan is used as an adhesive along with other biopolymers. The feasibility studies and early findings described in this review are encouraging for adhesive producers looking for reasonably priced, environmentally friendly substitutes for hydrocarbons-based materials. In conclusion, chitosan has become more common in the field of wood adhesives as a result of its unique properties in a variety of biopolymers.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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Abbreviations

PVAc: Polyvinyl acetate
VAc: Vinyl acetate
PF: Phenol formaldehyde
UF: Urea-formaldehyde
MUF: Melamine-urea-formaldehyde
PU: Polyurethane
MDF: Medium density fiberboard
SM: Soybean meal
DCS: Dialdehyde chitosan
CST: Chitosan and tannin
V: Vanillin