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Nanocellulose Applications in Wood Adhesives—Review

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Abstract

Bio-based materials open a new world of possibilities in every field due to its independence from the petrochemical origin. Moreover, concerns on environmental footprints and toxicity of synthetic adhesives made scientists investigate the utilization of biomaterials for wood adhesives. In this perspective, nanocellulose as a sustainable and cheap bio-nanomaterial provides a better alternative to conventional adhesive based on formaldehyde-containing condensation resins. Property of nanocellulose to act as both binders and as structural reinforcement in various adhesive systems adds to its potential. Besides by reducing the harmful emission of formaldehyde, it also can improve the mechanical properties and enhance performance of adhesives. This review paper aims to point out the potential application of nanocellulose based wood adhesives compared to petroleum-based conventional systems beyond renewability. New functionalities through structural modification in nanocellulose could bring a replacement with the synthetic adhesive systems which will play a significant role in future bio-economy.

Keywords

Nanocellulose, Reinforcement, Binding, Polyvinyl Acetate, Wood Adhesive

1. Introduction

Synthesis of polymers from bio-based renewable resources has received much acceptance due to the increasing environmental awareness and growing need to decrease the usage of conventional petroleum-based sources. In wood adhesive technology, formaldehyde-based synthetic adhesives such as urea-formaldehyde resin, phenol-formaldehyde resin, and melamine-formaldehyde are the predominant adhesives for the manufacture of various wood panels [1]. Formaldehyde emissions are mainly due to residual unreacted formaldehyde and slow adhesive hydrolysis under hot/humid conditions during the production and use of panel [2] [3]. Because of formaldehyde emissions, it has been proved that formaldehyde-based adhesives are not environmentally friendly products [4]. Even though the majority of wood adhesives are still petroleum-based, recent studies on the toxicity of formaldehyde [2] [5] [6] as a harmful substance accelerated scientists for finding an alternate source. In this aspect biopolymer based wood adhesives have a huge potential with reduced formaldehyde emission [7] [8] [9]. Another problem with phenolic resin wood adhesive is its brittleness; studies showed that this problem could be overcome by reinforcement with biomaterial, microcrystalline cellulose [10]. Hence to reduce formaldehyde and volatile organic content emission and for sustainable development, bio-based materials could be one of the alternatives beyond its renewability [11] [12] [13]. Researchers have a great interest in traditionally bio-based binders, such as starch [14] [15] [16] [17] [18], soy protein [19] [20] [21] [22] [23] or renewable rubber [24] [25] [26] [27], use of modified vegetable oils or lignin derivatives [28]-[34], and various cellulosic materials [35]-[44] for application in adhesive field.

Among these biopolymers, cellulose is the most abundant renewable biomaterial [45] and its natural affinity for self-adhesion makes it a potential material in adhesion science. Cellulose is a polysaccharide macromolecule with its advantage of abundant availability, economical production, excellent mechanical properties, possibilities for making biocomposites and its ability for functionalization makes it a unique material [46] [47]. Isolated cellulosic materials with one dimension in the nanometer range are referred to generically as nanocellulose [48]. They may be cellulose nanocrystals (NCC or CNC), nano fibrillated celluloses (CNF) and bacterial nanocellulose [49]. It can be achieved through mechanical or chemical treatments [50]. Nanocellulose shows extraordinary properties compared to the bulk material [51] [52] [53] [54] [55] and the binding properties of nanocellulose have been investigated in various studies [56] [57] [58]. Chemical modification of nanocellulose and its reinforcing properties in polymer matrices adds its application, improving the mechanical properties of nanocomposite [59] [60] [61] [62]. In conventional adhesives, nanocellulose hence finds application as a bio based reinforcing agent [63] [64]. This is also true in the case of wood adhesives, where introduction to nanocellulose made it possible for incorporating an abundant, sustainable, and cheap nano-bio-material for property improvement [65] [66] [67] [68]. In addition to applications of nanocellulose as ecofriendly binder [42] [58] [69] [70] it also finds application as a binder for Lithium battery electrodes [71] [72] [73], excellent barrier properties of cellulosic materials [74] have been made improved properties and extended its reach in various fields such as gas barrier, water barrier [75] and flame retarding applications [76], packaging applications, paper coating industries with nanocellulose and micro cellulose [77].

This review paper focuses the applicability of cellulose nanomaterials in wood adhesive field. The recent studies on binding property and reinforcement with nanocellulose, including nanocrystalline and nanofibrous cellulose in various wood adhesives systems are also been discussed. This work therefore aims to provide efforts to showcase the potential applications of nanocellulose in wood adhesives both as a reinforcing agent and binder, therefore a step to improve mechanical property of adhesive nanocomposite and to replace common carcinogenic adhesive ingredients, particularly formaldehyde in wood-adhesive research.

2. Nanocellulose Application in Wood Adhesives

There are several types of wood panels, including: laminated wood panels, agglomerated wood panels or wood fiber panels. The acceptance of wood in various fields, including furniture manufacturing, construction, and building sector hassled to a high demand for wood adhesive. Among the several opportunities offered by nanotechnology for the forest products industry, the reinforcement of adhesives with nanocellulose [78] has been already identified as a potential opportunity, which has been explored. It has shown improvement in both the physical and mechanical properties of the panels [40]. Reconstituted products, such as particleboard, oriented strand board (also known as flakeboard) and plywood panels, among others, appear as an alternative to solid wood, needs an improvement in the characteristics of the raw material. However, the quality of the final product depends mainly on adhesion technology. The plywood panels are composed of wood overlapping and bonded with adhesives, mainly phenol-formaldehyde and urea-formaldehyde, under pressure and temperature so that they cross their fibers at an angle of 90˚. Particleboard wood panels may be defined with randomly arranged small particles, bonded using adhesives and glued using heat and pressure. The most used adhesives in the production of panels of particleboard wood are the synthetic ones such as urea-formaldehyde, phenol-formaldehyde and melamine-formaldehyde.

The benefits of using nanocellulose as reinforcements in adhesives for the production of reconstituted wood panels include: the possibility of altering the properties of adhesives, gain in mechanical and physical properties of panels and reduction in formaldehyde emissions by using synthetic adhesives. It was observed a variation of viscosity with the increasing percentage of adding cellulose nanocrystals (CNC) in the glue [79]. Addition of cellulose nanofibers (CNFs) in urea-formaldehyde and melamine-urea-formaldehyde adhesives showed reinforcing nature of cellulose nanofibers [68]. Improvements in adhesives fracture energy, and fracture toughness enhanced the mechanical board properties, which could be confirmed by reinforcement of CNF in the adhesive. A similar study in which CNF were investigated as a binder in the formulation of particleboard panels [80]. The modulus of rupture, modulus of elasticity, internal bond, water absorption, and thickness swelling properties were tested. Particleboard panels met the industry requirements in terms of mechanical properties for low-density grades. J. Cui *et al.* [81] conducted a study for investigating the performance enhancement of tannin-based particleboards with cellulose nanofibers. Internal bond strength and viscosity increased on addition of 2% of cellulose nanofibers. The modulus of elasticity and modulus of rupture of the resins was also notably increased while thickness swelling of the panels was not affected. Kojima *et al.* conducted a study on the binding effect of cellulose nanofibers in wood flour board as a reinforcement [70]. The physical and mechanical properties of wood flour boards could be improved with the addition of CNF because of binding effect between the CNF and wood flour particles. A similar study on the evaluation of binding effects in wood flour board containing lingo cellulose nanofibers showed a significant enhancement of physical and mechanical properties of the board [42]. The study focused on reinforcement effects of lingo-cellulose nanofiber on fiberboards made from softwood and hardwood fiber was conducted [82]. In the study reinforcement effects of lingo-cellulose nanofiber on fiberboards was observed, due to close binding between lingo-cellulose nanofiber and wood fibers. Urea-formaldehyde based wood adhesive filled with cellulose nanofibrils were studied for fracture mechanical properties [38]. The highest fracture energy values were observed for urea-formaldehyde bonds filled with untreated nanofibrils, but bonds filled with TEMPO-oxidized fibrils showed inferior properties.

In a study on micro fibrillated-cellulose-urea-formaldehyde adhesives and its effect on the mechanical properties of laminated veneer lumber, the viscosity and gel time of the urea-formaldehyde adhesives increased with an increasing amount of the micro fibrillated-cellulose [41]. Limited amounts of the micro fibrillated-cellulose suspension in the urea-formaldehyde adhesive formulation significantly improved the mechanical performance of urea-formaldehyde bonds. Microfibrillated cellulose enhanced the strength properties of the adhesive and improves ductility of the adhesive.

Srno	Base material	Type of cellulose used	Application	Reference
1	Poly(Vinyl Acetate) and Starch Adhesive	Cellulose Nanofibrils	Wood Joining	[67]
2	Urea-Formaldehyde-Adhesive	Cellulose Nanofibrils	Wood Adhesive	[38]
3	Urea-Formaldehyde and Melamine-Urea-Formaldehyde	Cellulose Nanofibers	Particle Boards and Oriented Strand Boards	[68]
4	Poly(Vinyl Acetate)	Cellulose Nanofibrils	Adhesives for Wood Bonding	[40]
5	Poly(Vinyl Acetate)	Nano crystalline Cellulose	Wood Adhesive	[39]
6	Tannin Adhesives	Cellulose Nanofibers	Wood Adhesives	[81]
7	Urea-Formaldehyde Resin Adhesive	Modified Nanocrystalline Cellulose	Wood Adhesive	[83]
8	Polyethylene Glycol	Cellulose Nano- Whiskers	Plywood Bonding	[84]
9	Wood Flour	Cellulose Nanofiber	Binder in Wood Flour Board	[70]
10	Wood Flour	Ligno-Cellulose Nanofibers	Wood Flour Board	[42]
11	Urea-Formaldehyde	Micro fibrillated Cellulose	Laminated Veneer Lumbers	[85]
12	Sweet gum (Liquidambar styraciflua) & southern pine (Pinus spp.)	Micro Crystalline Cellulose	Particle Board	[86]
13	Wood particles(80:20 softwood: hardwood)	Cellulose Nanofibrils	Particleboard	[87]
14	Adhesive-free fiber-Board	Ligno cellulose Nanofibrils	Adhesive for Medium Density Fiberboard	[88]
15	Southern pine wood particles	Cellulose Nanofibrils	Binder For Particleboard	[80]
16	Cottonseed protein	Cellulose Nanofibers and cellulose nanocrystals	Wood adhesive	[89]

3. Nanocellulose Based Polyvinyl Acetate Adhesives

Polyvinyl acetate (PVA) is an excellent alternative to replace some wood adhesives containing formaldehyde. PVA is a linear and thermoplastic polymer. It is water-soluble, biodegradable with excellent chemical resistance and has no toxic action on the human body. As a wood adhesive, utilization of PVA is effortless [90]. For curing PVA, it does not need high temperatures. Poor performance of PVA towards humid conditions and at elevated temperature contributes to the main drawback and limits the usage. So far two approaches have been used to increase the performance of PVA: 1) Copolymerizing vinyl acetate with more hydrophobic monomers or functional monomers [91], and 2) blending PVA with other adhesives or hardeners [92] [93].

A study conducted by Jiang *et al.* [67] in which commercial polyvinyl acetate and starch adhesives were mixed with dicarboxylic acid cellulose nanofibrils (CNF). On adding optimum amount of CNF, the lap-joint strength increased to 74.5%. Properties were inferior at higher loading. In addition to reinforcing effects, increase in viscosity and shear strength of wood joints with polyvinyl acetate adhesives by CNF, water resistance and enhancement of mechanical properties at wet condition by adding 7% of CNF observed in [94]. Few published studies have done modification of polyvinyl alcohol by cross-linking [95]. Dynamic mechanical analysis (DMA), and wood bonding of polyvinyl acetate and polyvinyl alcohol latex reinforced with cellulose nanofibrils were studied [40]. Increasing the amounts of cellulose nanofibrils (treated or untreated) led to increasing reinforcing effects in the glassy state. Due to the interactions between the cellulose fibrils network and the hydrophilic polyvinyl alcohol matrixled to complete disappearance of the polyvinyl alcohol glass transition for some fibril types and contents. Kaboorani *et al.* [39] used nanocrystalline cellulose (NCC) in the wood adhesive to improve the performance of polyvinyl acetate. The study conducted addition of NCC at different loadings (1%, 2%, and 3%) to polyvinyl acetate and usage of the blends as a binder for wood. The block shear tests demonstrate that NCC can improve the bonding strength of polyvinyl acetate in all conditions. Thermal stability, hardness, modulus of elasticity and creep of polyvinyl acetate film were also enhanced by the addition of NCC.

4. Conclusions

Nanocelluloses (mainly cellulose nanofibrils (CNF) and cellulose nanocrystals (CNC)) represent an abundant natural resource of green and sustainable materials. Depending on the type of nanocellulose, modification of its structure and compatibility between polymer matrixes, performance property of adhesives enhanced even by addition of small amount. The current review highlighted some of the most recent advances and applications of these nanomaterials both as binder and reinforcing agent focusing the wood-based adhesives. Study proves that addition of these biobased nanomaterials could drastically enhance mechanical properties, performance properties and improve adhesive strength of wood adhesives. The benefits of using nanocellulose in the field of wood adhesive open the possibility of altering the properties of adhesives, gain in mechanical and physical properties of wood by its reinforcement and reduction in formaldehyde emissions by using synthetic adhesives.

Although applications of nanocellulose in the field of wood adhesives are at its infancy, it is foreseeable that there is a bright future for application of nanocellulose in the wood adhesive industry. Undoubtedly, there are many challenges for researchers in this field to overcome, and further intensive researches are needed.

5. Futuristic Approaches for the Development of Nanocellulose Based Wood Adhesive

Futuristic research in development and optimization of nanocellulose wood adhesives will be to increase the bonding between nanocellulose and base polymer matrix. This can be achieved in many ways. One such example can be adopting novel functionalizing approaches that permit nanocellulose to extend its applicability in wood adhesives. This includes cross-linking nanocellulose by suitable chemistry, functionalization by Silane coupling agents with adhesive system and nanocellulose grafted wood adhesives. This helps to improve dispersion and redistribution of nanomaterial inside the polymer matrix hence increment in the interaction with nanocellulose, adhesive, and substrate. Another area to be improved is in the water-resistance of nanocellulose based adhesives. One such possibility is functionalizing nanocellulose through chemical modification, cross-linking to get denser network or mixing with synthetic adhesives, and improving its interface for better compatibility.

Conflicts of Interest

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

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