Nanocellulose-Reinforced Adhesives for Wood-Based Panels

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1 Introduction

The use of wood panels is increasing in two ways. The first is by limiting the dimensions of the log diameters, by the anisotropy and other natural defects that solid wood possesses and the second by the search for greater use of the wood [93, 107]. Therefore the production of panels of reconstituted wood represents a rational use of this raw material [18].

There are several types of wood panels, which include: laminated wood panels, agglomerated wood panels or wood fiber panels [53]. It is reported that 416 million

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m^3 of wood panels were produced in 2016, of which 42% were wood panels and the remaining 58% were fiber based panels [35]. It is interesting to note that each panel has an application such as internal or external use, in furniture or civil construction. Depending on the environment to which the panels will be exposed, there are different types of adhesives that should be used, the most common ones being urea formaldehyde for indoor use and low moisture content, while phenol formaldehyde resin for external use [54].

Improvement of adhesive bonding is a routine process in the wood industry [29] as it is one of the key steps in the production of panels. Changes in adhesion to wood are desirable in terms of performance improvement and adhesive economy [43]. Among the several opportunities offered by nanotechnology for the forest products industry [15] the reinforcement of adhesives with nanocellulose has been already identified as an opportunity, which has been explored. This has shown improvement in both the physical and mechanical properties of the panels [43].

Obtaining nanoscale cellulose fibers and its application as reinforcement in the preparation of biodegradable composites as well as nanocomposites has attracted great attention during the last years [101, 116]. This is attributed to the unique properties of nanomaterials such as high aspect ratio, crystallinity and surface area, excellent mechanical properties combined with less weight and biodegradability [29, 76].

With this background, this chapter presents an overview of the use of nanocellulose in wood-based panels, with examples, of the use of different types of nanocelluloses as reinforcement in several types of adhesives in the production of different types of panels. This chapter also presents some important concepts and properties of all the raw materials used, viz., adhesives, wood, and nanocelluloses.

2 Wood-Based Panels

The solid wood presents some disadvantages, because it is a heterogeneous and anisotropic product, i.e., It possesses different physical properties in its tangential, longitudinal and radial axes [13, 39, 108]. It should also be taken into account that the dimensions of the wood pieces limit their use besides the natural defects, such as knots, grain inclination, the percentage of juvenile and adult wood and reaction wood, among others, all of which interfere with the rheological behaviour of the wood [54]. Also, it is reported that many times the mechanical properties of wood are unsatisfactory for certain uses [104]. Because of the above-mentioned limitations of solid wood, reconstituted wood products have been produced by gluing of veener, boards, slabs, particles or fibers, and these elements are joined by adhesive bonds [13]; Industrial Research [78]. With the use of glue utilization of the wood has been increasing because the glue allows the use of pieces of small dimensions to obtain products with greater added value.

Reconstituted products, such as particle board, oriented strand board (OSB, also known as flakeboard) and plywood panels, among others, appear as an alternative to solid wood, rendering improvement in the characteristics of the raw material. This is because, they allow greater homogeneity of the physicochemical properties, dimensional stability, full use of wood and residues, thus contributing to the conservation of forests [18]. However, the quality of the final product depends mainly on the adhesion technology [89].

Reconstituted wood panels can be divided into three types: laminated panels (plywood and Laminated Venner Lumber-LVL), particle board (wafer board and OSB), and fiberboard (Medium Density Fiberboard-MDF, High-Density Fiberboard-HDF, and insulation board). 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° [54, 119]. The wood veneer can be of different thicknesses and are always in odd numbers (Finnish along the length of the part, in which the thickness of the blades should not exceed 6.4 mm—0.25 Forest Industries Federation [33]. According to the Standard Specification for Evaluation of Structural Composite Lumber Products [5], LVL can be defined as a structural compound composed by layers of thin wood assembled with adhesives with wood fibers oriented mainly inches. In the bonding of the LVL panels synthetic adhesives used are resistant to humidity; the most commonly used adhesive being the phenol-formaldehyde [75]. Both plywood and LVL are gaining visibility for their benefits in structural and non-structural use [10, 65, 73]. These are already used in applications typically dominated by steel and concrete [83].

Particleboard wood panels may be defined with randomly arranged small particles, agglutinated using adhesives and glued using heat and pressure [54, 87]. 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 [44] with phenol-formaldehyde is recommended for external use and the urea-formaldehyde recommended for internal use [44]. The waferboard used as a structural material is produced with larger wafer type particles of square or slightly rectangular shapes, glued with random particle distribution and consolidated through hot pressing [54]. While in the waferboard the particles are arranged randomly, the particles are used as layers in the OSB with perpendicular directions [87]. In view of this arrangement, the superior structural behaviour is exhibited in OSB-type panels having high dimensional stability compared to the waferboard [53, 93]. Accordingly, the OSB wood panels are used for structural applications, considering the evolution of the waferboard differs from its precursor in the direction of the particle [49].

The fiber panels are dry-fabric panels made of lignocellulosic fibers, combined with an adhesive under pressure and temperature [53, 78, 87]. In such type of panels primary adhesion occurs through the interlacing of the fibers and the adhesive properties of some chemical components of the wood [46]. When fabricated with low density, these panels can be used for insulation purposes [60], called ‘insulation board’, with a mean density between 0.02 and 0.40 g cm^{-3} [54]. Natural fiber insulation is known for the good thermal insulation it promotes, but this material also presents good acoustic insulation [80].

Medium density fiberboard (MDF) panels are normally bonded with urea-formaldehyde adhesive and consolidated with hot pressing [78]. Density of MDF varies from 0.50 to 0.80 g cm^{-3} [7, 54]. This is one of the most well-known wood panels, commonly used as a raw material for furniture, carpentry and building products [70]. Panels having a density varying from 0.80 to 1.10 $g{\cdot}cm^{-3}$, called as high-density fiberboard (HDF), are similar to the MDF [107]. These are used as a panel for structural purposes commonly used as a core of laminate flooring [86].

3 Adhesives and Adhesion

3.1 Adhesives

Kinloch [62] defined adhesive as any substance applied to the surface, or both surfaces, of two separate objects that bind them and offers resistance to their separation. On the hand, Peschel et al. [87] added to this concept of the condition of adhesives, these being non-metallic substances with which other materials are solidly bonded together by adhesion and cohesion.

Wood adhesives can be classified according to their origin in natural and synthetic [28, 74]. Natural adhesives can be proteins of animal or vegetable origin, while synthetic ones have the petroleum raw material and, although they resemble the natural adhesives in the physical characteristics, they can be formulated to meet specific requirements and have a higher resistance to humidity [28, 82].

Synthetic adhesives can be classified into two types: Thermoplastic adhesives and thermoset adhesives. The two types differ in their chemical structure and response to heat [81, 88]. Table 1 shows the classification of some adhesives including natural adhesives used in the wood panel industry.

Another type of classification of adhesives that can be made according to their purpose of use involving the environment to which each adhesive would be exposed. Accordingly, Table 2 shows the types of adhesives, the environment in which they are used and the name of adhesives for each of these used in the preparation of wood panels.

Thermoplastic adhesives are liquid adhesives whose aggregate state depends on temperature. The curing and melting are reversible, i.e., if heated after curing they will return to the liquid state since they are adhesives that do not form reticles (net of fibers). These adhesives may also be dissolved in a solvent and then reactivated with solvent evaporation [88]. But, the use of this type of adhesives is limited, i.e., they can only be used in non-structural applications, in low-temperature climate and are not resistant to heat or fire [81]. Of this polyvinyl acetate (PVA) adhesive is the most commonly used in wood glueing [34]. Figure 1 shows the polyvinyl acetate monomer.

PVA is a yellow-white liquid adhesive available in a ready-to-use form and which can be applied directly to the wood and cured at room temperature or through

Table 1 Adhesives used in the wood panel industry

Types of adhesives	Adhesives
Natural	Animal protein derivatives (glutin, casein and albumin)
	Derivatives of vegetable origin (soybean meal)
	Derivatives of starch (wheat flour)
	Cellulose ether
	Natural rubber
Thermoplastics	Polyvinyl acetate
	Polyvinyl/acrylate
	Polyethylene
	Polystyrene
	Synthetic rubber
Thermosets	Urea formaldehyde
	Melanin-formaldehyde
	Phenol-formaldehyde
	Resorcin-formaldehyde
	Tannin-formaldehyde
	Phenol-resorcin-formaldehyde

Table 2 Classification of the use environment of the wood panels according to the type of adhesive (Adapted from [36])

Application area	Name of environment where the adhesive is used	Used adhesive
Structural	Exterior use without any restriction	Phenol-formaldehyde
		Resorcin-formaldehyde
		Phenol-resorcin-formaldehyde
		Polímeros de emulsão/ Isocianato
		Melanin-formaldehyde
	Exterior use with a restriction	Melanin-urea formaldehyde
		Isosyante
		Epoxy
	Interior	Urea-formaldehyde
		Casein
Semi-structural	Exterior use with limitations	Polyvinyl acetate "crosslinking"
		Polyurethane
Non structural	Interior	Polyvinyl acetate (PVAc)
		Construction elastomers
		Contact elastomers
		Hot-melt

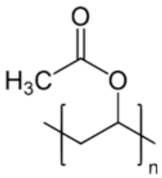

Fig. 1 Polyvinyl acetate (PVA) adhesive monomer. Reproduced from [9] with the kind permission of the publishers

high frequency. After curing this adhesive exhibits high mechanical resistance; however, its use is not recommended in environments with high temperatures and high humidity [36]. Polyvinyl acetate adhesives are fixed by the loss of water mainly by diffusion of water from the adhesive in the wood [34]. This type of adhesive is used for any and all wood glueing operations. Major areas include bonding of corrugated panels, finger-jointing, laminating and assembling [48].

Unlike the thermoplastic adhesives, thermosets are plastics when cured of a soft solid or viscous liquid prepolymer results in a molecule of higher molecular weight and with higher melting point and therefore, will not have the cure reversed by heat [88, 121]. The cure of thermoset adhesives is heat induced, reaching 200 °C. These adhesives generally are stronger than the thermoplastic adhesives and more recommended for high-temperature applications [88] and are more commonly used in wood structures [121]. In spite of a large number of adhesives available for wood panels, the most used are synthetic ones, such as phenol-formaldehyde, urea-formaldehyde, resorcinol-formaldehyde and melamine-formaldehyde [54, 107]. These four adhesives make up approximately 90% of all adhesives used in wood panels and all of these are derived from fossil fuels [14].

Phenol-formaldehyde (PF), applied in a broad spectrum of engineered wood products, is very strong and resistant to dry and humid conditions and exhibits strong adhesion to wood [41, 89, 121]. Figure 2 shows part of a polymer chain of the phenol-formaldehyde adhesive.

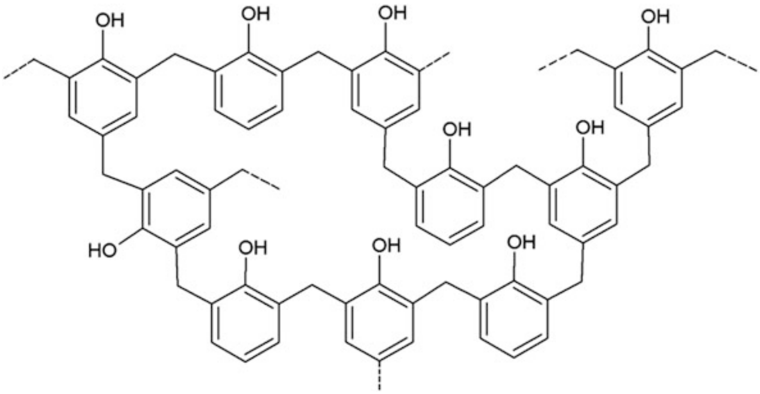

Fig. 2 The molecular structure of the polymer chain of phenol-formaldehyde. Adapted from [124] with the kind permission of the publishers

It can be cured hot or cold; however, for the cold setting process, it is necessary to reduce the pH with the addition of an acid. The PF adhesive is mainly applied to particle or fiberboard, plywood, pressed laminated wood, glued laminated wood, waferboard and OSB [54, 107].

Urea-formaldehyde (UF) is a low-cost, structural and internal-use adhesive [54, 89]. Figure 3 shows part of a polymer chain of the urea-formaldehyde adhesive.

This adhesive is fast hardening, high resistance to dry bonding and presents colourless glue joints. This adhesive can be hardened hot or cold; however, when fast hardening is desired a hardener should be applied [61]. The glue joints of this adhesive are high strength, but brittle and inelastic. Therefore, the stress of the wood, caused by changes in humidity and temperature, impairs glue joints and decreases adhesive performance [41, 121, 74].

Resorcinol-formaldehyde (RF) is a brown, cold-curing, catalyst-requiring adhesive. Figure 4 shows part of a polymer chain of the resorcinol-formaldehyde adhesive. This adhesive is much more reactive than PF [54]. Adhesive bonds of RF

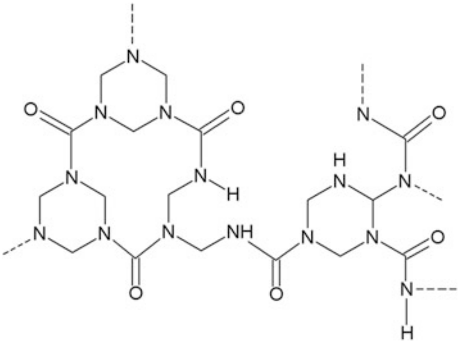

Fig. 3 The molecular structure of the polymer chain of urea-formaldehyde. Adapted from [85], with the kind permission of the publishers

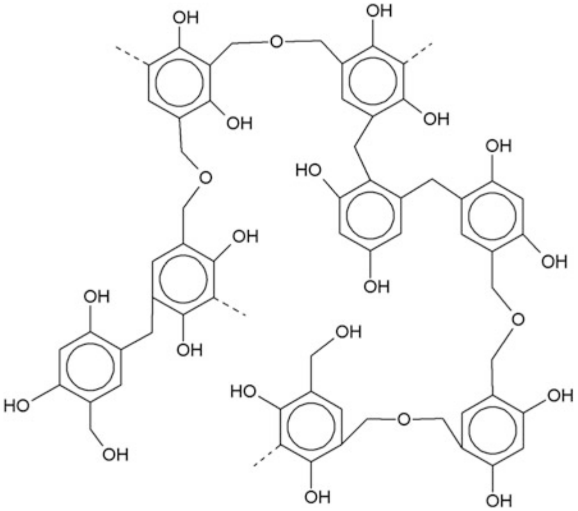

Fig. 4 The molecular structure of the resorcinol-formaldehyde polymer chain. Adapted from [47], with the kind permission of the publishers

are resistant to moisture, boiling water, oil and many other solvents, i.e., an adhesive suitable for exterior use [88]. This adhesive used in the production of rolled beams, shipbuilding, and aviation. However, due to its high cost of production, it is hardly used in pure form. Therefore, it is usually mixed in the same proportion with PF [41, 54, 121].

Melamine-formaldehyde, a product between the condensation of melamine and formaldehyde, is a white-coloured adhesive classified as an intermediate between PF and UF. Figure 5 shows part of a polymer chain of the melamine-formaldehyde adhesive.

RF is a more resistant material than wood [88]. It is a hot curing around (93 °C) structural adhesive Melamine-formaldehyde has the advantages of being more resistant to moisture than UF and curing faster than PF [24, 89]. This can be used outdoors without any restriction [36]. However, its cost is higher than that of these two resins and therefore is commonly used as an additive to improve the performance of UF glue beating, even this form of use is marketed under the nomenclature melamine-urea-formaldehyde [54, 89].

Formaldehyde, present in the four synthetic adhesives mentioned above, is one of the most common chemicals in current use, with the simplest aldehyde of molecular formula H_2CO and the boiling point of −19 °C [97]. It is a colourless gas with pungent, inflammable and highly reactive odour [51] and highly carcinogenic [52]. The use of these resins can lead to the emission of formaldehyde into the atmosphere, generating occupational hazards to the workers involved in the manufacturing process, as well as users of the installations where these panels are used [49]. Exposure to formaldehyde may cause irritation to the mucous membranes of the eyes, nose, nasal cavity, pharynx and larynx, and may also cause drowsiness, nausea, and skin irritation through frequent contact and prolonged exposure [112]. Even coated panels can emit formaldehyde, these emissions are regulated by panel buyers countries [49]. Regulatory standards establish maximum emission limits and analytical methods for gaseous formaldehyde measurements [95].

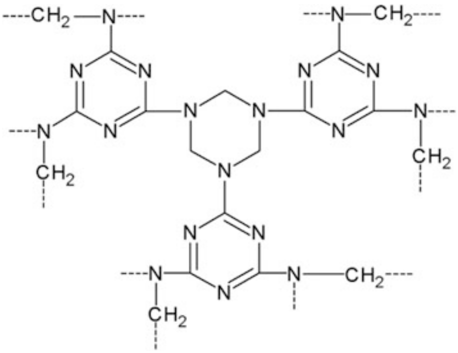

Fig. 5 The molecular structure of the polymer chain of melamine-formaldehyde. Reproduced from [90] with the kind permission of the publishers

3.2 Adhesion

It is understood that adhesion is the force of attraction between molecules of different materials, such as the force between adhesive substance and the junction piece, and cohesion force of attraction between molecules of the same type, such as forces inside a layer of glue [87]. These authors have defined that cohesion is the force of attraction between molecules of the same type, as for example, the forces inside a layer of glue. Therefore, a good adhesive must adhere to the surface of the solid and have sufficient cohesion to ensure the bonding of the solids. According to Schultz and Nardin [100], Petrie [88] and Ebnesajjad and Landroch [27], there are six main adherence theories, viz. mechanical theory, electrical theory also known as electrostatic theory, wetting theory, theory of the diffusion of polymers, also known as diffusion theory, chemical bonding theory and weak boundary layer theory. These are explained below:

- Mechanical theory: According to this, the fluidity and penetration of the adhesive into porous substrates leads to the formation of hooks or a mechanical interlacing of the adhesive tightly attached to the substrate after curing and hardening of the adhesive.
- Electrical theory (Electrostatic theory): According to this, the forces of attraction in adhesion in terms of electrostatic effects at the interface between the adhesive/bonding system is compared to a capacitor, in which the armatures are the two electrical layers formed by the contact of the two substrates. The adhesion results from the forces of attraction developed between the two armatures.
- Wetting theory: According to this, adhesion results from the molecular contact between two materials that develop surface attraction forces. The process of establishing continuous contact between an adhesive and a substrate is called ‘wetting’, which can also be defined as the adhesion of a liquid to a solid.
- Theory of the diffusion of polymers (diffusion theory): According to this, adhesion occurs through the diffusion of segments of polymer chains. Adhesive forces can be visualized as those produced in mechanical adhesion but on a molecular scale. However, the applications of this theory are also limited. The mobility of long polymer chains is very restricted, severely limiting the molecular interpretation proposed in this theory.
- Chemical bonding theory: According to this theory, adhesion occurs through chemical bonds (covalent and metallic ionic). It is currently believed that adhesion at the interface, from the molecular point of view, is due to the action of secondary forces.
- Weak boundary layer theory: This theory proposes the existence of a finite boundary layer composed of absorbed molecules at the interface, which is different in their constitution from the constituent molecules of the adhesive and the adhesive.

The mechanisms related to the adhesion process and wood panels can be explained by the mechanical theories, chemical adhesion and diffusion of polymers

[37]. In the bonding of porous surfaces, such as wood, the initial process is done mechanically [37]. There is penetration of the adhesive at the cellular scale, filling void intercellular spaces, increasing the bonding durability in the wood, especially when the adhesive is diffused into cellulose and hemicellulose molecules. According to this author, deeper the penetration of the more resistant adhesive, greater is the bond, which may even exceed the resistance of the wood. According to the theory of polymer chain diffusion, adhesion occurs through ionic or covalent primary bonds, and/or by secondary intermolecular forces; however, there is no evidence that primary bonds between wood and adhesive occur [37]. After adhesive penetration into the wood, the adhesion is chemically strengthened by attractive intermolecular forces such as Van der Waal forces, dipole-dipole forces and hydrogen bonding [37]. If the polymer chain extends between the molecules of the wood, the adhesion is reinforced by the diffusion theory. Adhesion is a very complex field beyond the reach of any model or theory. In practice, several adhesion mechanisms can occur simultaneously [100].

3.2.1 Factors Influencing the Adhesion Process

The adequate bonding of wood is directly related to various physical-chemical characteristics of the adhesive. These include viscosity, gelatinization time, solids content and pH, and the intrinsic characteristics of the wood, anatomical, physical, chemical and mechanical properties.

Physico-Chemical Characteristics of the Adhesive

The viscosity is one of the most important properties of an adhesive [25], and can be defined as physical property that characterizes the resistance of a fluid to the flow, high viscosity liquids have low fluidity, such as honey, while those already having low viscosity have high fluidity, such as water. According to Peschel et al. [87], this parameter depends on the temperature, decreasing with the elevation of the temperature of the liquid. According to Iwakiri [54] and Gonçalvez and Lelis [45], when the adhesive viscosity is high, the uniform distribution of the adhesive on the wood is difficult, with insufficient penetration into the wood structure, damaging wetting and leaving a thick tail. However, adhesives with low viscosity have higher penetration and their absorption by the wood is also greater, and in extreme situations, can result in excessive absorption of adhesive by the wood [4, 45, 54, 107]. The gel time is important for the quality of the adhesive since it is related to the maximum admissible viscosity for its application. The gel time is measured in seconds, minutes or hours, and corresponds to the period from the preparation of the adhesive to the application to the hardening “point”, or gel phase, when it reaches maximum elasticity [20]. In the industrial scope the gel time is a characteristic foundation, since, from this time on, it is no longer possible to manipulate the resin [22].

In general, the working time of the adhesives should not be very long, as it would require a longer pressing time. However, the short working time results in the difficulty of applying and spreading the adhesive in the wood, due to its rapid polymerization, causing a decrease in the strength of the glue line [22, 54]. The content of solids corresponds to the number of solids contained in the adhesive, which is composed of solid components and volatile liquids. When the panel is subjected to hot pressing, evaporation of the liquid components occurs, which is called "cure", that is, solidification of the adhesive, forming the glue line that is responsible for the bond between the substrates [54]. The pH, hydrogen potential, of an aqueous solution is defined as the concentration of dissociated H^+ ions [97] and its determination is made by direct reading in apparatus called pH meters. In the case of bonding of wood, it is important to consider the influence pH of both wood and resin [54]. According to Wang et al. [114, 115] and Wang et al. [113] the pH of the adhesives should not exceed the range of 2.5–11, because beyond these limits the resin causes degradation of the fibers of the wood.

Intrinsic Characteristics of Wood

The anatomical structure is very diversified, especially in hardwood species, composed of cellular elements that are arranged in various ways to constitute the wood [30]. This cellular organization depends on the botanical species, the age of the plant and the environmental conditions in which it develops [17]. In addition, each cell element has a characteristic of the shape and dimensions being linked to the genotypic characteristics of the species, function of the cellular element and phylogenetic position [110]. The anatomical properties of the wood have a significant influence on the bonding, such as the variability in density and porosity that occurs in early wood (also called spring wood → less dense due to larger cells and thinner cell walls) and latewood (also called summer wood → produced in spring and later), core and sapwood, juvenile and adult wood. Also, the influence of the dimensional instability of the reaction wood, as well as the direction of the grain, in which the penetrability relates to the cutting direction [3] is highlighted.

For the production of particleboards and fiber panels, the wood density is a very important factor since it is related to the compaction ratio of the panel. The compaction ratio and indicates the degree of densification of the wood particles in the panel structure and will affect the properties and qualities of the wood [107]. According to these authors, the compaction ratio should be in the range of 1.3–1.6 so that proper densification and consolidation of the panel in the desired final thickness occurs.

According to Thoemen et al. [107], the characteristics of the growth rings, heartwood and sapwood, tree age, porosity, reaction wood and angle of inclination of the cellular elements are favourably or unfavourably involved, since they may or may not favour the bonding process. In the process of bonding, several adhesion mechanisms occur simultaneously in the wood. The influence of the wood anatomy on the bonding process is related to its structure with respect to the differences of dimensions of the cellular elements, size, disposition, and frequency of the cellular cavities that, in turn, are related to the porosity and permeability of the wood [2]. The interaction between adhesive and substrate occurs mainly by vessels and voids, but there is an effective participation of the rays and, to a lesser extent, the axial parenchyma in this process [2, 103]. Figure 6 shows the glue line adhesive penetration of a plywood panel.

The pH of the wood varies according to the species and is around 3–6, and there may be changes of pH inside a piece of wood as a result of the migration of extractives from lower layers to superficial layers, altering the bonding conditions [54]. The capability is a characteristic of the adhesive and refers to its ability to tolerate contact with more acidic or more basic materials without altering its pH [89]. Some woods may present extractives with pH that inhibit the hardening of the adhesive, impairing the development of resistance and adequate cohesion in the glue line. While in some woods, the pH may favour the pre-hardening of the adhesive, impeding the movement and mobility functions, such as the fluidity, penetration, and wetting of the adhesive in the wood [67]. The most important physical properties of wood in terms of bonding are the density and moisture content of the wood. Antagonistic to porosity and the penetration of adhesives, the density can cause significant effects on adhesion. In low-density woods, there is greater penetration of the adhesive and may result in greater consumption of adhesive. In the case of high-density wood, there are larger dimensional changes

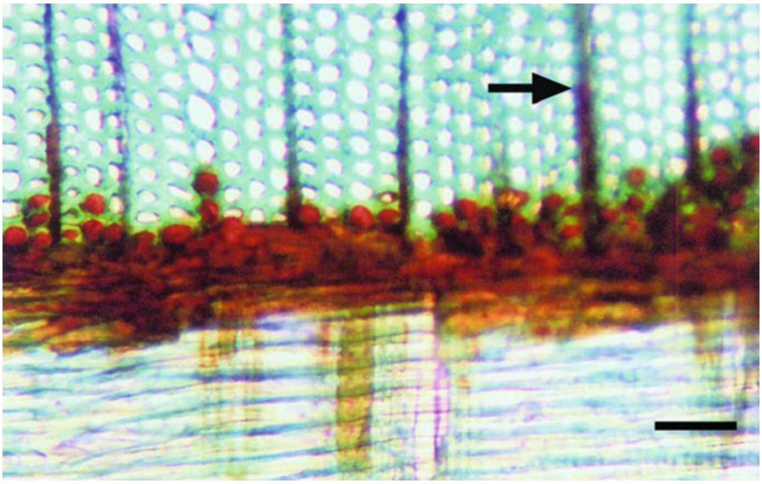

Fig. 6 Penetration of adhesives on the various types of wood cells. Reproduced from [103] with the kind permission of the publishers

resulting from variations in moisture content, generating higher glue line stresses, making the glueing process difficult [54, 77].

The influence on the moisture content of the wood in the cure of the adhesives is related to the amount and the rate of absorption of liquid adhesive by the wood, the lower the moisture content in the wood, the higher the rate of absorption, cure rate and solidification of the wood adhesive [54, 89]. According to Almeida [4], good adhesion between adhesive and wood is obtained provided the moisture content of this adhesive should not exceed 20%. In the wood/glue system, the tensions generated in the bonded product are of extreme importance in the general balance of the resistance. The greater the resistance of the glue line in relation to the strength of the wood, the greater the percentage of rupture or faults in the wood in the interface with the glue line [11]. The tension generated in the glue line is manifested by shear stresses in the plane of the adhesive bond and in the direction perpendicular to it [54].

3.3 Adhesive Additives

In the search for improvements in the adhesion process, specific properties in the panels or reduction of the cost or consumption of adhesives in the industry, additives are commonly used in the tailings formulations. Among the main additives is hardening accelerating agents one generally one comes across. Adhesive curing is influenced by the pH of the environment, in that sense pH modifiers may be employed to promote an acid or alkaline environment, depending on the type of adhesive used. Typically, with UF and melamine-urea-formaldehyde adhesives that cure in an acid environment, ammonium sulfate is used because of the fact that in addition to promoting proper pH for a cure, it leads to the formation of a less hydrolyzable microstructure [84]. For the PF adhesive, sodium hydroxide is used as a hardening accelerator because curing occurs in the basic media [32]. Types of the addition of some reagents and their effects on the final characteristics of paper are listed below:

- Paraffin emulsion: This is used to control the dimensional variation of the panels in the short term, preventing the entry of water into the liquid form by capillarity [19, 117].
- Preservatives: The insecticides and fungicides are commonly used to increase the durability of the panels. Some researchers have already employed the use of nanoparticles loaded with biocides, such as tebuconazole or chlorothalonil, aiming at the slow release of these preservatives [26, 69].
- Filler materials: These are non-stickable materials that are added to the adhesive in order to increase the total volume of the adhesive and reduce the cost without affecting the viscosity of the adhesive. These act as penetration controlling agents, avoiding excessive adhesive penetration in the case of the production of laminated panels; however excess extender may impair adhesion. Kaolin, nut shell flour, coconut husk flour are other commonly used filler materials [54].

Improvements in the rheological properties of adhesives increased mechanical and moisture resistance, reduction of formaldehyde emissions and lower production costs of wood panels are some of the goals that can be achieved with the use of additives. In this context research on the use of nanocelluloses as adhesive additives has shown promising results [6, 38, 111]. These are discussed in the next Section.

4 Nanocellulose

Nanocellulosic materials can be extracted by different methods from different plant biomasses [29, 1]. According to Fujisawa et al. [40], the nanocelluloses can be divided into three groups, viz. Nanocrystalline cellulose (CNC), microfibrillated cellulose (CMF) and nanofibrillated cellulose (CNF). The first CNC is a highly crystalline material with free of defects. This is extracted by the hydrolysis of the amorphous regions present along the axis of cellulosic fibers, by means of a chemical process of acid or enzymatic hydrolysis followed by mechanical agitation of the suspension of nanocrystals in water. The most common nanocellulose production process is by acid hydrolysis, while the most commonly used acid being sulfuric acid. Nitric acid, hydrochloric acid, phosphoric acid and hydrobromic acid can also be used in acid hydrolysis, although on a smaller scale [96, 120].

The second one CMF, obtained by a method of the mechanical disintegration of the cellulosic pulp in water. Finally, the third NCFs are extracted laterally in its nanoscale substructural units (nanofibrils) using combined processes of chemical oxidation with the reagent 2,2,6,6-tetramethylpiperidine-1-oxy, followed of mechanical disintegration in water, or only by the method of mechanical disintegration. It may be noted that the process of obtaining CMF and CNF is similar, differing only in the final dimensions after the processing of the cellulose [99]. According to Samyn et al. [99], the CMF is commonly produced by homogenization, where the fiber shear is performed by a strong pressure drop and impact forces inside the processing chamber. A similar effect has been observed by the use of grinding processing [66], where processed suspensions generally contain a heterogeneous mixture of CMF and CNF which are characterized by different diameters and aspect ratios (length/diameter) [99]. The different aspects of the nanocelluloses described above are shown below in Fig. 7.

When isolated and prepared, CNCs have excellent physicomechanical, optical, magnetic, electrical and conductimetric properties, covering a wide range of uses, different from those obtained by materials seen on a macroscopic scale. The advantages of CNC are related to its properties, such as high mechanical strength and stiffness, low density, durability, uniform size distribution, high specific surface area, low coefficient of thermal expansion, high hydrophilicity, optical transparency and self-molding that enable them to be used in a variety of uses [125, 98]. Due to its crystalline arrangement, this form of nanocellulose has a high mechanical resistance, the modulus of elasticity being estimated between 50 and 145 GPa [106, 64] and comparable to the resistance of extremely rigid materials [29] indicating its

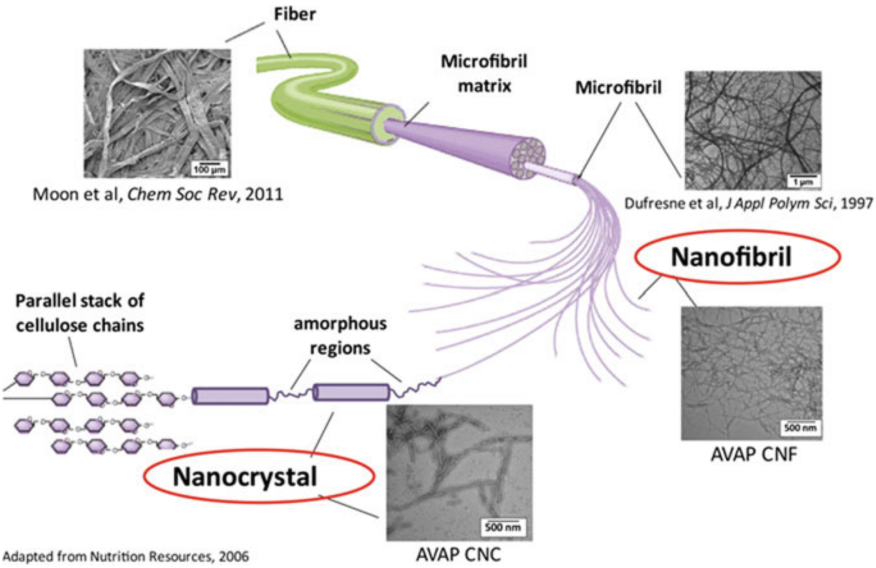

Fig. 7 The mechanism of chemical and mechanical methods for producing CNC, CMF and CNF from cellulose. Reproduced from [79] with the kind permission of the publishers

significant potential as reinforcement material, important, for example, in the automotive industry. However, herein the reported results in the literature are presented in this Chapter.

According to some researchers working on MFCs, they consist of a material obtained by the disintegration of the cellulose subjected to a mechanical process of homogenization, where it is degraded, promoting the exposure and opening of the surfaces previously located inside fibers, fibrils and microfibrils [109] cited by [63, 99, 105]. This process causes an increase in the external surface, allowing a greater area of contact and better bonding between microfibrils, increasing the resistance properties, with a value of modulus of elasticity of 145–150 GPa [55]. On drying, films with lower opacity, high density and transparency would be produced [56]. Reported definition of MFC is fibers with a diameter between 25 and 100 nm, while CNF is nanocelluloses with a diameter between 5 and 30 nm and of a variable length between 2 and 10 µms [96, 101]. Both CMF and CNF have amorphous and crystalline zones composing their structure. On the other hand, CNC refers to cellulose nanoparticles that underwent hydrolysis under controlled conditions and that lead to the formation of structures in the form of small crystalline cylinders and depending on the source of extraction has a diameter of 3–50 nm [101].

Further detailed information on the production of wood pulp and nanocellulose can be found in the following sections: 3.1 Pulping, 4: Cellulose, 5: Nanocellulose, 5.1 Method of NFC and MFC production and 5.2 Method of CNC and MFC production in the chapter on Nanocellulose in paper making in this volume.

5 Nanocellulose-Reinforced Adhesives Performance and Properties

The possibility of using nanocellulose in the adhesive formulation can be a way of promoting gains in the properties of these adhesives. This is because, the adhesion between wood components, as well as among other materials, depends on a series of parameters related to the physicochemical characteristics of the adhesive and the material to be bonded, besides the operational parameters in the bonding process, the geometric form and the size of the pieces to be bonded [29]. Although modifications in the chemistry of adhesives are a path of optimization of mechanical characteristics, the addition of filler or fibrous particles presents a possible alternative route of modification [43]. By adding fillers, the limitations imposed by polymer chemistry can be overcome, and this approach is common in high- performance adhesives, which can be reinforced with nanoparticles [122, 59, 58, 91, 92, 118].

5.1 *Effects of the Addition of Nanocellulose on Adhesives*

The addition of nanocellulose to the adhesives affects the physicochemical properties of the adhesives, except for the unchanged pH. In general, the percentage increase of nanocellulose in the glue causes an increase in the viscosity and the solids content. Then, the gel time is the property that varies most with this additive, since the partially acidic load in the case of CNC can delay or delay the curing, according to the type of resin to be used.

Damásio et al. [23] evaluated the addition of CNC in the glue mixture of formaldehyde urea glue observed that only the viscosity showed variation, increasing with the increasing percentage of addition in the glue mixture. A similar result has also been reported recently by Ferreira [31], who observed that the increase in the percentage of CNC added (0–8%) in the synthesis of UF adhesives provided an increase in their viscosity, solids content and gelatinization time, in comparison to the synthesis of UF without the addition of CNC. According to the author, the viscosity and solids content gains were marked with a CNC addition up to 6%, although only physical interactions were observed. That is, there was no chemical interaction of the CNC with the other elements present in the adhesive. Similar results were also reported by Liu et al. [68] while evaluating the addition of CNC in lignin PF adhesives. The author has concluded that there was no chemical reaction between the adhesive and CNC since the DSC curves presented only a similar peak at all the compositions and that the addition of the nanocellulose did not affect the energy of the adhesive. Mahrdt et al. [72] found that the addition of CMF in the glue bead of the UF resin caused a delay in the formation of the chemical and mechanical bonding of the resin curing, in addition to increasing the viscosity of the adhesive. However, the addition of CMF allowed better distribution of the adhesive in the wood, with less formation of clots of glue, which do not contribute to the adhesive bond, besides presenting the same penetration in the wood.

Gindl-Altmutter and Veigel [43] contend that the adhesive cure is not excessively prolonged due to the presence of nanocellulose. On the other hand, the severe increase of the viscosity caused by the addition of nanocellulose can represent a serious obstacle for resin spraying and impregnation in the wood. Cardoso et al. [16] have evaluated the addition of CNC produced without the neutralization of the surface charge on the UF glue mixture. They observed that acidic nanocellulose reduced the curing time of the UF resin. Cui et al. [21] produced CNF-reinforced particle boards in tannin-based adhesives. They observed increased gelatinization time and the viscosity of the glue mixture with the addition of CNF (1–3%).

Zhang et al. [123] modified the CNC with 3-aminopropyltriethoxysilane (APTES) and 3-methacryloxypropyltrimethoxysilane (MPS) and evaluated bond strength as well as formaldehyde emission from compensated panels. The results showed higher efficiency for modification with APTES, where 1.5% of modified CNC reduced formaldehyde emission by 53.2% and increased binding resistance by 23.6%, while for MPS modification, the results were 21.3 and 7.0%, respectively.

Ayrilmis et al. [8] used CMF produced from pine sawdust and evaluated the emission of volatile organic compounds (VOC) at different temperatures in LVL panels. The addition of CMF was efficient to reduce the emission up to 35 °C and could be used for furniture for internal use.

Hu et al. [50] have reported that hydrogen bonds between the –OH ends of nanocrystals promote an increase in the frequency and number of hydrogen bonds between UF-nanocrystalline cellulose (UF-CNC) and UF-CNC-wood. In addition, they also promote a higher frequency of the effective hydrogen bonds during the polymerization of the adhesive, allowing gains in resistance of the panels. An American patent [12] claims the use of only nanofibrillated cellulose as an adhesive for the production of particleboard panels. The advantages go beyond the non-emission of VOC, but also advantages in the carbon fixation by the trees producing cellulose used for CNF production.

5.2 *Wood Composites with Nanocellulose-Reinforced Adhesives*

Damásio et al. [23] have evaluated the addition of CNC in the glue mixture of formaldehyde urea glue. They have observed that the strength of the glue line of dry compaction panels increased with increasing percentage of CNC in the adhesive, where the maximum CNC addition (8%) resulted in an increase of 56% when compared to the control. This shows that the nanocrystals increase the wood-adhesive-wood bonding and interaction [50]. For wet strength, all additions increased this property in relation to the control, but the highest gain occurred with the addition of 2% of nanocellulose. Eichhorn et al. [29] found significant gains when 5% of CNF was added to UF adhesive for the production of bonded joints. In another work, the addition of CNF from 0.5 to 5% UF resin allowed a significant gain in stress and strength until composite failure [29]. According to these authors, the UF adhesive belongs to a class of low-priced, widely used wood adhesives, which are well known for their pronounced fragility and their tendency to develop microcracks that limit their mechanical performance. In addition, the UF adhesive is less moisture-resistant due to the reduction of the molar ratio urea: formaldehyde, leaving free urea groups that bind to water molecules [42]. The addition of CNC contributed to the improvement of the two weaknesses of this type of adhesive, both the resistance of the glue line to the dry and its resistance to moisture [23].

Cardoso et al. [16] have evaluated the addition of CNC in the UF resin and observed a reduction in the swelling in thickness, compared to the panel produced with ammonium sulfate only. However, the water absorption was higher for the panels with the addition of nanocellulose.

Eichhorn et al. [29] have stated that the research group led by Wolfgang Gindl and Josef Keckes has been investigating the reinforcement of adhesives with nanocellulose. To this end, the group tested the addition of CMF and hardwood fibers as resin reinforcement. By adding 5% of untreated pulp fibers and found no significant effect on the value of shear strength of 9.9 MPa. In stark contrast, the addition of 5% nanocellulose, which resulted in a significant increase in shear strength to a value of 13.8 MPa. The researchers justify this increase in the absence of cracks commonly observed in UF glue lines. In addition, they claim that the improvement of the properties of UF resin can open doors to panels of structural uses.

The addition of 2% CNF (m/m) in tannin-based adhesives significantly have been found to improve the mechanical properties of the wood panels produced, since the water absorption did not change significantly [21] In adhesives based on polyurethane or isocyanate, the CNF without chemical modification cannot be dispersed due to the high polarity. For addition of CNF in polyurethane-based resin Richter, et al. [94] did not find significant gains, while some others such as López-Suevos et al. [71] and Kaboorani et al. [57] have observed significantly improved binding strengths and durability of the panels by adding CNF and CNC.

In addition, the CNC increased the dry strength [57] and the CNC increased the resistance to wet and high temperature [71]. Cellulose nanofibrils obtained by high-pressure homogenizer mill added to the UF and melamine urea formaldehyde resins used in the production of agglomerate and OSB showed an increase in their mechanical properties. The improvement was significant mainly for OSB, not as pronounced for MDP [111]. However, in the case of physical properties, there was a reduction in swelling after 24 h in contact with water, and significantly increased internal bonding, flexural strength, and rupture, with the most significant results for OSB panels [111]. This result shows that the combination of larger particles with the enhanced MUF resin contributed to this significant gain. For the addition of CNC as reinforcement in lignin PF resins, Liu and collaborators [68] concluded that the best properties of dry and wet tested glue lines occurred with the addition of 0.25–0.5% CNC.

New MDF panels produced by the addition of 1 and 3% nanocellulose produced from old MDF panels using 8 and 12% new fibers some promising results have been observed [102]. The results showed that values of the highest modulus of rupture (14.47 MPa) and modulus of elasticity (1359.09 MPa) for the panels produced with 12% of glue and 3% of nanocellulose, as well as the highest internal bond strength (0.5 MPa), the lowest swelling in thickness (4.72 and 9.86% after 2 and 24 h on water immersion) and the lowest water absorption (33.11 and 80.63% after 2 and 24 on water immersion).

In the production of particleboard panels and bonded sheet joints, Ferreira [31] has stated that due to the best adhesive and resistance properties observed in the particleboard panels prepared using 4% CNC added UF adhesive, these panels have already been used for applications in glue joints. But, the physical and mechanical results were inferior to those obtained for panels produced with commercial UF adhesive.

In addition to CNF as reinforcement in polyurethane adhesives and water-based polyvinyl acetate (PVAc) adhesives, the good rheological stability of the mixture without the CNF sedimentation after a long preparation time has been reported [94]. However, despite the increase in mechanical properties, the authors believed that new research applying the superficial chemical modification of the CNF should be performed to obtain improvements in properties and justify the industrial applications. Similarly, López-Suevos et al. [71] while producing CNF films with PVAc-latex have observed significant improvements in the storage modulus and thermal properties. Atta-Obeng [6] have found improved shear properties of MDP panels with the addition of MCC in the proportions of 0–10% in PF adhesives; however, the static bending strength was impaired. The authors believe that the presence of MCC resulted in a less pronounced spring back effect during hot-pressing. As pressure decreased, the authors have observed the spring effect occurred resulting in an increase in the thickness of the panel, less interaction of wood particles and adhesive, which resulted in the drop in static bending of the panels.

6 Final Considerations

Cellulose is considered the most abundant renewable polymer on the planet, has many advantages such as biodegradability and low cost and the products obtained from cellulose have wide application, especially in paper production. However, because it is a renewable and widely available resource, there is growing interest in the application of cellulose as an additive in activities with more advanced technologies that use nanotechnology for product development. Within the constant search for better performance of adhesives, the use of nanocelluloses appears as a viable option. The benefits of using nanocelluloses 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 panels using synthetic adhesives. However, despite all the advantages mentioned above, there are still some points to be considered. Therefore, it is concluded that more research needs to be done, either in the application of nanocellulose and its modification in different types of resin, as well as application technologies appropriate to the new conditions of the adhesives.

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