



The role of lignin in wood working processes using elevated temperatures: an abbreviated literature survey

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Abstract

The lignin, cellulose and hemicelluloses in wood are polymers that behave similarly to the artificial polymers and are bonded together in wood. Lignin differs from the other two substances by its highly branched, amorphous, three-dimensional structure. Under appropriate conditions, the moist lignin incorporated in the wood softens at about 100 °C and allows the molecules of it to deform in the cell walls. There are many advantages and disadvantages to this phenomenon. If we know this process accurately and the industrial areas where it matters, we may be able to improve these industrial processes. This article provides a brief theoretical summary of lignin softening and the woodworking processes where it plays a role: wood welding, pellet manufacturing, manufacturing binderless boards, solid wood bending, veneer manufacturing, and solid wood surface densification.

1 Introduction: thermoplastic behavior of lignin

Wood is actually a mixture of polymers, composed of partially crystalline cellulose microfibrils and large amorphous hemicellulose and lignin molecules. In lignin, phenyl propane units create chains, which are crosslinked in an amorphous, three-dimensional structure, linked to the cellulose fibrils via hemicelluloses (Sakakibara 1991; Zandersons et al. 2004; Rowell et al. 2005). Lignin can be classified based on the chemical structure of its monomer units. Three major groups can be distinguished: grass lignin, softwood lignin and hardwood lignin. Depending on the configuration of guaiacyl (G), syringyl (S) and p-hydroxyphenylpropane (H) units, grass lignin is classified as a GSH lignin, softwood lignin is classified as G lignin and GS in case of hardwoods (Sarkanen 1975; Stelte et al. 2011b; Sakakibara 1991). The amount of lignin varies between species, and also between individual tissues such as bark, earlywood, latewood, normal wood and compressed wood, branch wood, wood from the roots; also by cell types (parenchyma or fibers), and cell wall layers, for example middle lamella, primary and secondary wall layer, and cell corners. Lignin and its chemical

composition can be examined by various techniques, such as ultraviolet microscopy (Lange 1954; Scott et al. 1969; Adler 1977; Fergus and Goring 1970a, b) densitometric analysis (Scott et al. 1969), interference microscopy (Donaldson 1985), SEM-EDXA (Westermarck et al. 1988), transmission-electron-microscopy (Fromm et al. 2003), and by confocal Raman microscopy (Gierlinger and Schwanninger 2006). The greatest concentrations of lignin were found in the compound middle lamella and in the cell corners; lower concentrations were found in the secondary cell walls (Fergus et al. 1969; Scott et al. 1969; Fergus and Goring 1970a; Donaldson 1985; Saka and Goring 1985; Westermarck et al. 1988; Fromm et al. 2003; Gierlinger and Schwanninger 2006). Although the lignin concentration in the middle lamella and in the cell corners is high, these regions have a lower tissue volume than the secondary cell wall layers, so these regions accommodate only about one quarter of the total amount of lignin; indeed, most of the lignin is located in the secondary walls (Fergus and Goring 1970a, b; Adler 1977; Saka and Goring 1985). The lignin concentration of the vessel wall is higher than that of the fiber, while ray parenchyma cells have a lower lignin concentration than the fibers (Saka and Goring 1985).

A polymer can be amorphous or partially crystalline, determined primarily by the chemical structure of the polymer. If the molecule has large side groups and/or many branches, or it is irregular in structure, it cannot crystallize

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when the melted condition cools down. It remains disordered. These are the amorphous polymers.

The state of the polymer can be glassy, elastic and melted. In the glassy state, parts of the polymer macromolecule can only oscillate. By increasing the temperature, the internal energy is increased, the individual parts and segments of the molecules can move, and deform, but the relative position of the center of the mass of the molecular does not change; the polymer has a high degree of reversible deformation. This is a flexible state. As a result of further internal energy growth, a plasticized condition occurs where the molecules move relative to one another and the polymers flow. The transitions between the individual states occur at temperatures typical of the particular polymer (Bánhegyi 2005; Hargitai 2011; Miskolczi 2012). Since polymers do not have a specific molecular weight, but a density distribution within a range, the conversion temperature also influences the range. It is known that many amorphous polymers change from a glassy state to a rubbery, elastic state above a certain temperature, as they plasticize (Hatakeyama and Hatakeyama 2010; Medved 2010). The temperature range where the glassy to rubbery, plasticized state transformation occurs is the glass transition temperature (T_g) (Back and Salmén 1982; Bánhegyi 2005; Hargitai 2011; Miskolczi 2012). The glass transition temperature can be examined by several methods (both static and dynamic): differential scanning calorimetry (DSC), differential thermal analysis (DTA) and thermogravimetric analysis (TGA), dynamic mechanical analysis (DMA/DMTA), dielectric thermal analysis (DETA), and torsional pendulum analysis (Hatakeyama et al. 1972; Kelley et al. 1987; Wolcott 1989; Lenth and Kamke 2001; Miki et al. 2007; Hatakeyama and Hatakeyama 2010; Miki et al. 2007; Gellerstedt 2015; Kong et al. 2017).

The temperature required by lignin at glass transition (T_g) is influenced by a number of factors, such as the presence of rigid phenolic side groups on the main chain, the presence of crosslinking, the number of bonds between chains, hydrogen bonds, its molecular weight, isolation method, species, lignin conformation, and thermal prehistory, etc. (Chow and Pickles 1971; Hillis and Rozsa 1978; Olsson and Salmén 1992, 1997; Lenth and Kamke 2001; Windeisen and Wegener 2008; Gellerstedt 2015; Furuta et al. 2008, 2010; Hatakeyama and Hatakeyama 2010). When dry, the differences in the glass transition temperature of cellulose, hemicellulose and lignin are not big: 200–250 °C for the amorphous region of cellulose, 150 to 220 °C for hemicelluloses, and 205 °C for lignin (Goring 1963; Back and Salmén 1982). These values are difficult to validate because hemicellulose and lignin degrade near this temperature (Schaffer 1973; Back and Salmén 1982). Lignin seems to be the most thermally stable component of wood, but various changes occur below 200 °C (Sehlstedt-Persson 2005). Lignin decomposes over a broader temperature range (200–500 °C)

than cellulose and the hemicellulose. Degradation studies performed on different types of lignin showed an endothermic peak at 100–180 °C, as a result of the elimination of humidity, followed by two exothermal peaks, the first from 280 to 390 °C and the second one around 420 °C (Brebou and Vasile 2010).

Many researchers have studied the effect of moisture content on the T_g of extracted hemicelluloses and lignin (Goring 1963, 1971; Back and Salmén 1982; Irvine 1984) or in situ hemicelluloses and lignin (Irvine 1984; Kelley et al. 1987; Morsing and Hoffmeyer 1998). Moisture dramatically affected the T_g (Goring 1963; Becker and Noack 1968; Sakata and Senju 1975; Back and Salmén 1982; Kelley et al. 1987; Wolcott 1989; Wolcott et al. 1990; Bouajila et al. 2006; Miki et al. 2007; Hatakeyama and Hatakeyama 2010; Sen et al. 2013). The structure of lignin is rich in phenolic hydroxyl and it offers the possibility to create intermolecular hydrogen bonds. Water molecules can break these hydrogen bonds and segmental motion can occur easily (Hatakeyama and Hatakeyama 1998, 2010). The T_g of lignin decreases with increasing moisture content until the wood or the lignin reaches its water saturated point. The number of hydroxyl groups influences the amount of water bound to lignin (Hatakeyama and Hatakeyama 2010). The difference between the results could stem from the equilibrium and non-equilibrium moisture conditions. For example, Salmén (1984) maintained strict moisture control during the temperature changes, while Kelley et al. (1987) and Irvine (1984) conditioned the samples to an initial moisture content and then provided no explicit control for moisture content during the tests (Wolcott et al. 1990) (Table 1).

Softwoods contain more lignin than hardwoods, and there also are structural differences between softwood and hardwood lignin (Sehlstedt-Persson 2005). Softwood lignins have higher glass transition temperatures (138–160 °C) while hardwood lignins have a lower T_g range 110–130 °C under dry conditions (Sen et al. 2013). The main structural differences between the two types of lignin is a lower content of free phenolic hydroxyl groups. A substantially higher content of methoxyl groups is found in hardwood and less in softwood lignins, and the hardwood lignins are less cross-linked than the softwood lignins (Olsson and Salmén 1992). The result of Horváth et al. (2011) contradicts though this argument above. They examined the thermal softening behavior of genetically modified aspen trees with reduced lignin content and/or increased S/G ratio, and their results suggest that the higher content of methoxyl groups and thus less cross-linked lignin did not alter the softening behavior of lignin. The modification of lignin can shift the T_g : sulfonation of the lignin lowered the T_g depending on the degree of sulphonation (Back and Salmén 1982) while lignin esterification decreases T_g (Thiebaud and Borredon 1995; Lisperguer et al. 2009). Finally, yellow poplar juvenile wood

Table 1 Moisture content effect on the glass transition temperature of lignin

| References | Moisture content | T_g (°C) |
|---------------------------------------|------------------|------------|
| Urakami and Nakato (1966) | Sat | 50 |
| Stelte et al. (2011b) | Sat | 53 |
| Nakajima et al. (2008) | Sat | 60 |
| Kelley et al. (1987) | 30 | 60 |
| Östberg et al. (1990) | 25 | 60 |
| Irvine (1984) | 25 | 62 |
| Wolcott (1989); Wolcott et al. (1990) | 10–15% | 70 |
| Hillis and Rozsa (1978) | Sat | 72–128 |
| Gašparík and Barčík (2014) | Sat | 77–128 |
| Becker and Noack (1968) | Sat | 78 |
| Furuta et al. (2000) | Sat | 80 |
| Furuta et al. (1997) | Sat | 80 |
| Sadoh (1981) | Sat | 80 |
| Kong et al. (2017) | 20% | 80–94 |
| Salmén (1984) | Sat | 82–100 |
| Uhmeier et al. (1998) | Sat | 85 |
| Östberg et al. (1990) | 6.5 | 86 |
| Goring (1963) | 27% | 90 |
| Stelte et al. (2011b) | Sat | 91 |
| Bouajila et al. (2006) | Sat | ~ 100 |
| Irvine (1984) | 7 | 108 |
| Sen et al. (2013) | Dry | 110–160 |
| Jonsson (2009) | 8–15% | 110–135 |
| Back and Salmén (1982) | Sat | 115 |
| Kelley et al. (1987) | 5 | 115 |
| Atack (1972) | Sat | 120–135 |
| Koran (1979) | – | 125 |
| Wert et al. (1984) | Dry | 132–192 |
| Ibach (2010) | – | 170 |
| Goring (1963) | Dry | 195 |
| Back and Salmén (1982) | Dry | 205 |
| Sadoh (1981) | Dry | 235 |

exhibited a lower T_g than mature wood. In case of southern pine, this trend was reversed (Lenth 1999).

Of course, other wood constituents in a native wood cannot be separated from lignin. The softening temperature of native hemicellulose under dry conditions is high, around 180 °C (Goring 1965; Alfthan et al. 1973; Back and Salmén 1982; Olsson and Salmén 2003). Water works like a plasticizer and decreases the glass transition temperature: at 20% water content it is around 50 °C, at 30% it is around room temperature (Back and Salmén 1982; Olsson and Salmén 2003; Placet et al. 2008; Navi and Sandberg 2012). The amorphous regions of cellulose behave similarly. During veneer production and bending, the wood is saturated with water, and during pellet production, it also contains a significant amount of water. During these processes, hemicellulose

is already above its T_g , so raising the temperature does not cause a change in the hemicellulose structure. On the other hand, as the wood dries, the lignin softening is reduced, and the hemicellulose softening becomes more important. Most of these processes take place at elevated temperatures, which will dry wood. However, with wood welding, binderless board manufacturing or surface densification, the temperature is usually above 180 °C, which softens both hemicellulose and lignin. Because there is more than one parameter involved, the plasticization does not occur at a fixed temperature, the T_g is also affected by pressure, and as certain operations (e.g. pellet production) take place at high pressure, the increase in the T_g is not negligible. Based on previous works, a linear relationship can be established between the T_g and pressure (Ichihara et al. 1971; Zoller 1982; Zoller et al. 1989; Andrews and Gruclke 1999; Krevelen 2009). All in all, the importance of lignin is highlighted in the literature, so we can also study these wood industry processes examining lignin.

This type of wood softening—mainly the lignin—is used in several industrial processes, namely: pellet manufacturing, binderless panel manufacturing, wood welding, wood bonding, wood surface compacting, and veneer manufacturing by peeling. Of course, during these industrial processes, in addition to softening, other physical and chemical reactions take place, all of which contribute to the end result of the process. The aim is not to describe these reactions in detail, but rather to provide a summary of the industrial processes where lignin softening may play a role.

2 Wood welding

Frictional welding is a relatively new technology for creating wood joints but this technique is widely used in the plastics industry (Ganne-Chédeville et al. 2006). Sutthoff et al. (1996) made the first efforts to join wood by pressure and frictional heat. During the welding process, no other material is added to the system, the welded pieces of wood are frictioned together to provide the necessary energy. In doing so, the components of wood (lignin, cellulose and other polymers) are melted and partially decomposed by heat and are subject to chemical reactions during the process.

At the beginning of the welding process, Coulomb friction causes the surfaces to heat up. After a few seconds (3–10 s), at a temperature of about 320–350 °C, the wood surfaces start to decompose at the increased temperature. The temperature of 180 °C is reached quickly at the interface/surface but at less than 1 mm below the interface, the temperature is still 20 °C lower (160 °C). The wood next to the rubbed surfaces starts to soften, forming a viscous film. After reaching the maximum temperature about 420–450 °C, the frictional movement is terminated, and the joined parts

are held together. The final cooling down leads to solidification of the interfacial film forming the connection between the wood parts (Stamm et al. 2005b). No preparation of the welded surfaces is required and the time necessary to complete the bond is shorter than one minute (Stamm et al. 2005b).

The mechanism of welding, in addition to the chemical reactions that take place due to the temperature-induced softening, is chemical activation, flowing and solidification of the intercellular material, mainly amorphous polymers: lignin and hemicelluloses (Stamm et al. 2005a; Windeisen and Wegener 2008). This flow of material induces high densification of the bonded interface (Leban et al. 2005; Ganne-Chédeville et al. 2006; Pizzi 2017). The physical entanglement of the fibers interconnected as a result of friction can improve the connection. In the brief pressure-holding phase immediately after welding, chemical reactions occur. The main reactions are the formation and self-condensation of furfural and the cross-linking reaction of lignin with carbohydrate-derived furfural (Pizzi 2017).

Sun et al. (2010) suggested that the chemical changes when applying friction welding are similar to changes during fast pyrolysis at a lower pyrolysis temperature. This is a transformation of a nonvolatile compound into a volatile mixture by heat in the absence of oxygen. During these processes, bonds are broken, free radicals are formed, which re-polymerize and create side-chains (Kawamoto 2017). In these temperature ranges, only the amorphous components of wood are affected, particularly lignin and hemicelluloses, and to a lesser extent, amorphous cellulose. First, hemicelluloses degrade through acid hydrolysis and dehydration, which starts above 100 °C, but the weight loss of hemicelluloses takes place mainly in the temperature range of 220–315 °C. The mono- and oligosaccharides change into furfural and 5-hydroxymethylfurfural as they dehydrate. Xylan is more affected than mannan, because the glycosidic bonds of xylan are more susceptible to acidic attack (Stamm et al. 2005a). The changes of the amorphous cellulose are similar to those of hemicelluloses and levoglucosan and furan and the same kind of compounds produced. The effects of heat on lignin occur within a wide temperature range. The proportion of the C–C and C–H bonds decreases, while those of C–OH, C–O–C, C=O and O–C–O increase. First –OH groups are thermo-oxygenated and aldehydes are formed. Chemical substances which contain oxygenated functionality are also formed and move to the welding zone: terminal hydroxymethyl groups from the G-type lignin side chains leave in the form of formaldehyde, which then can contribute to condensation reactions of lignin fragments generated during the welding. The amount of typical bonds of phenylpropane units decreases, while furfural and furan derivatives react with the lignin (Gfeller et al. 2004; Kanazawa et al. 2005; Stamm et al. 2006; Delmotte et al.

2008; Belleville et al. 2013). During friction, a new lignin-carbohydrate complex is formed from the condensation reaction between lignin fragments and furfural derivatives from hemicelluloses (Sun et al. 2010). Belleville et al. (2018) found that a higher proportion of lignin seems to be favorable to condensation reactions during the welding process. According to another observation, fatty acids, terpenoids and other extractive compounds form covalent bonds with hydroxyl groups of other compounds. There is a degradation of fatty acids, which deform to carboxyl acids, mainly citric acid, which can improve the mechanical properties through chemical linkages (Belleville et al. 2018). The work of Ganne-Chédeville et al. (2008) summarizes the processes that take place at different temperatures during welding. The main components of the smoke emitted during welding are water vapor, CO₂ and decomposed compounds from wood polymeric carbohydrates and from lignin (Omrani et al. 2008). Delmotte et al. (2009) found that increasing the welding frequency from 100 to 150 Hz, the oxidation of the components decreased due to the much shorter welding time, which improved the joint strength.

Hardly damaged fibres, tracheids were observed immersing in a mass of molten polymer. The welded bondline is then composed of a mass of entangled long wood cells immersed in a matrix of amorphous, fused intercellular material, mostly lignin but also including some hemicelluloses. The bonding line separates into (1) a melting zone, where the wood polymers melt, decompose and partly char; (2) to a fully plasticized zone and (3) a deformed zone, where the polymers are plasticized and the cells deform, and (4) a partially deformed region, where the cells are only slightly affected (Ganne-Chédeville et al. 2006).

The welding processes can be classified as linear, orbital and rotational friction welding (Ruponen et al. 2015). In linear welding, the wood samples are joined to each other with a 1.3–2 MPa pressure. The samples are vibrated with a displacement amplitude of about 2–3 mm and a vibration frequency of 100–150 Hz in the plane of the joint (Leban et al. 2005; Pizzi 2017). Specimens 1.0–1.8 m long can be welded and the welding takes 1.5–5 s and the holding time, still under pressure, after vibration has stopped, is 5 s too (Pizzi 2017). After linear welding, 10 to 11 MPa tensile strength was measured (Leban et al. 2005). With high-speed rotation welding, wooden dowels (usually 10 mm diameter) are inserted at high rotation speed into a smaller diameter pre-drilled hole (usually 8 mm diameter). In dowel welding, generally, cylindrically fluted beech dowels 10 mm in diameter are used (Pizzi 2017). For best results, the drill rotation rate must be between 1500 and 1600 rpm. When bonding is achieved (1–3 s), the rotation of the dowel is stopped, and the pressure is briefly maintained.

The quality of a friction welding joint correlates with several welding parameters, such as welding pressure,

frequency, time, holding pressure and time, amplitude or displacement, wood species, orientation of the grains, equilibrium moisture content and specimen dimensions, chemical composition etc. (Kanazawa et al. 2005; Properzi et al. 2005; Ganne-Chédeville et al. 2006, 2008; Delmotte et al. 2009; Omrani et al. 2009; Župčić et al. 2014; Ruponen et al. 2015; Belleville et al. 2018; Zhu et al. 2019).

The limitation of linear welded wood is that the welded joint has low water resistance in other words the joint is more water sensitive. Water resistance can be increased by selecting appropriate welding parameters (Mansouri et al. 2009; Vaziri et al. 2010, 2011; Vaziri 2011). In contrast to linear vibration welding, the rotation-welded dowel joints are completely resistant to water when immersed, which shows that proper geometry of the joint can improve water resistance (Pizzi et al. 2006).

3 Pellet manufacturing

Lignocellulosic materials can be densified by pelletizing, briquette or cube making. During this process, the particles are pressed together by applying a mechanical force to create inter-particle bonding (Tumuluru et al. 2010; Kaliyan and Morey 2010). The product becomes denser, more manageable and usually more durable. The name ‘pellet’ is usually used for products less than 15 mm in diameter, while ‘briquette’ generally refers to larger dimensions. Several researchers examined the compression process of different raw materials such as wood, wood waste and bark (Chin and Siddiqui 2000; Demirbaş et al. 2004; Lehtikangas 2001; Li and Liu 2000; Rhén et al. 2005, 2007), forest residues (Lehtikangas 1999; Acda and Devera 2014), straws, grasses (wheat, barley, corn etc.) (Smith et al. 1977; Wamukonya and Jenkins 1995; Demirbaş 1999; Kaliyan and Morey 2006; Mani et al. 2004, 2006a, b; Ndiema et al. 2002; Olsson 2006; Shaw 2008; Gilbert et al. 2009; Lehmann et al. 2012; Stelte et al. 2012; Lee et al. 2013), alfalfa (Adapa et al. 2002; Tabil and Sokhansanj 1996a, b, 1997; Fasina and Sokhansanj 1995), olive cake (waste) (Al-Widyan et al. 2002; Yaman et al. 2000), palm fiber and shell (Husain et al. 2002; Tenorio et al. 2016; Wattana et al. 2017). Since pellet production has been studied most frequently and in many detail, this is described further below. Thereby, most statements remain true for other densifying procedures (Tumuluru et al. 2010). There are many methods for characterizing the quality of a pellet, often referred to as the durability of the pellet. There are no standards given, but most methods model the impact and the shearing stresses as such (Oveisi-Fordiie 2003).

During the process, the size of the feedstock material is typically reduced by milling or grinding, and conditioned to an appropriate moisture content either by drying or moisturizing. In most cases, the particles of raw material are forced

through a channel. Usually, in a pellet mill, the pressure is 100–150–200 MPa (Kaliyan and Morey 2009). The pelletizing process generates heat that maintains the temperature of the operating die at 110–130 °C (Nielsen et al. 2009). Lignin and extractives in wood and other lingocellulosic materials have been reported to function as natural binding agents during the pressing of the pellets (Bradfield and Levi 1984; Alakangas and Paju 2002; Gilbert et al. 2009; Nielsen et al. 2010; Berghel et al. 2013). During pelleting, in the raw material with 8–15% moisture content, the lignin softens around 110–135 °C (Lehtikangas 2001; Kuokkanen et al. 2011; Kaliyan and Morey 2009), the drier raw material needs higher temperatures (Zhanbin 2003). According to Anglès et al. (2001), the following reactions occur during pelleting: Partial hydrolysis of cellulose and hemicellulose; and then partial hydrolysis of lignin leading to a lower molecular weight material. At high steam temperatures, some low molecular weight lignin melts, flows, and partially coalesces into droplets. The thermally softened lignin contributes to the strength characteristics of pellets and briquettes made of lingocellulosic materials (Granada et al. 2002; Serrano et al. 2011). As the pellets cool, lignin hardens again and the pellet strength increases. Cooling stabilizes the pellets and stiffens the lignin melted on the surface of the pellets, and hence, the shape of the pellets remains unchanged (Alakangas and Paju 2002). There is a positive relationship between pellet durability and lignin content (Lehtikangas 2001; Castellano et al. 2015). The lignin content of different straws is generally lower than in wood, and therefore straws produce less durable pellets (Lehmann et al. 2012; Stelte et al. 2012; Lee et al. 2013).

Macroscopically, two binding mechanisms can be distinguished: solid bridges between particles (Stelte et al. 2011a; Serrano et al. 2011) and other bonding without a solid bridge (Kaliyan and Morey 2010). Without a solid bridge, forces between particles can form bonds by hydrogen bridges, van der Waals’ forces, electrostatic and magnetic forces if the particles are close enough to each other. Due to the application of high pressures and temperatures, solid bridges can develop by diffusion of molecules from one particle to another at the points of contact and be formed by a chemical reaction, hardening of the binders, and solidification of the melted components (Kaliyan and Morey 2010). Bio-based binding materials such as starch, protein, lignin, and pectin in the raw materials under high pressure are pressed out of the raw material, and cause inter-particle bonding. To produce a sufficient bonding area, especially in the absence of a binder, the plasticization of wood polymers above their glass transition temperatures is necessary (Back, 1987). Chung (1991) suggested that at the microscopic level, there are two criteria for bonding between particles: the molecules should be in contact of closer than 9 Å and the maximum attractive force should have a minimum potential energy.

When the maximum attractive force is near the minimum potential energy, chemical bonding is established. Pressure, heat above glass transition temperature may promote adhesion by increasing the molecular contact. Samuelsson et al. (2012) added some further bonding mechanisms, they found that interfacial forces and capillary pressure in movable liquid surfaces, adhesion and cohesion forces, and mechanical interlocking between particles can also strengthen internal bonding.

Several factors were found to influence the process and the result of the densification experiments (Rehkugler and Buchele 1969; Bradfield and Levi 1984; Granada et al. 2002; Shaw 2008; Kaliyan and Morey 2009). Increasing the temperature and moisture content (MC) decreased the energy requirements for pellet manufacturing (Nielsen et al. 2009). A significant part of the energy for the pelletizing process is used to force the compressed sawdust from the surface of the die into the pressing channels. Increasing the temperature of the die increases the pellet density, decreases the dimensional expansion, and increases the tensile strength of the pellets. The application of heat to the aggregated material during its formation confers greater cohesion to it, because at temperatures between 80 and 200 °C lignin becomes softer, and after cooling, it functions as a thermoplastic glue, depending on the materials (Kaliyan and Morey 2006; Relova et al. 2009; Theerattananon et al. 2012).

Moisture acts as one of the binding agents in the pelleting process (Lehtikangas 2001), so the moisture content of the raw material is of high relevance. Decreasing the moisture content increases the pellet density as well as its dimensional stability (Shaw 2008). Increasing the moisture content to between 10 and 13%, the mechanical durability increases; a further increase of moisture causes a decline in durability (Wilfried et al. 2006; Ahn et al. 2014) because of the lignin softening and excessive vapor formation.

Decreasing the particle size increases the pellet density, decreases their expansion, which means higher dimensional stability, and increases the tensile strength of the pellets. A high surface area/volume ratio in each particle allows better penetration of moisture and heat, and consequently improves its strength properties (Lehtikangas 2001).

The biochemical characteristics of the raw material also are very important. Higher lignin and extract content have a positive effect on the durability of pellets (Bradfield and Levi 1984; Lehtikangas 2001; Stelte et al. 2011a; Serrano et al. 2011; Filbakk et al. 2011). Several studies showed that pellets produced from bark have excellent durability (Lehtikangas 2001; Filbakk et al. 2011; Ahn et al. 2014). Bark usually contains higher levels of lignin than wood (Filbakk et al. 2011), and contains higher levels of extractives. The effect of the bark is unclear; Ahn et al. (2014) found that up to 10% bark reduced the durability of larch pellets. Waxes on the surface of straw produced a weak waxy boundary

layer, resulting in lower strength (Nielsen et al. 2009; Samuelsson et al. 2012; Castellano et al. 2015).

Pretreatment of the raw material generally increases the pellet density and durability.

During the steam pretreatment, the hemicellulose was removed and/or hydrolysed. As a result, a higher relative amount of lignin was present and this lignin was more readily available for binding, thus producing superior pellets (Shaw 2008). Zandersons et al. (2004) suggested that during the steam explosion process, the lignin is activated and the cellulosic structure is changed which facilitates the formation of new bonds. Chemical activation with hydrogen peroxide resulted in a lower mechanical durability and reduced heating value, but thermal activation trials indicated a positive effect on their durability (Wilfried et al. 2006). By pre-treatment with microwave irradiation, the ester bonds between the lignin and hemicelluloses are disrupted, and the free lignin can create additional bonds thus increasing the strength and durability of the densified product (Thomas et al. 1998; Lu et al. 2014).

Pressing aids and adhesives can also increase the durability and strength of pellets (Oberberger and Thek 2004; Wilfried et al. 2006; Kuokkanen et al. 2011; Berghel et al. 2013; Ahn et al. 2014; Lu et al. 2014).

In summary, a high lignin content and optimum MC coupled with a high pelleting temperature tend to improve biomass pellet durability (Relova et al. 2009; Whittaker and Shield 2017).

4 Wood surface densification

Compression is used to increase density and change the surface properties of wood, for example hardness (Donghua et al. 2010; Petrič 2013). Densification of wood can also be achieved by impregnating its void volume with polymers, molten natural resins, waxes, sulphur, and even molten metals (Kutnar and Šernek 2007). Sandberg and Navi (2007) made a summary of these topics. The process of densifying wood by compression requires four steps (Kutnar and Šernek 2007; Rautkari et al. 2011): (1) plasticization of the cell wall, (2) compression perpendicular to the grain in the softened state, (3) cooling and drying in the deformed state, and (4) fixation of the deformed state to eliminate the shape memory effect. The density is increasing, the color of the wood becomes darker and the EMC is reduced markedly (Donghua et al. 2010; Arruda et al. (2015).

Firstly the cell wall should soften. The temperature should be at least 25 °C higher than T_g of lignin, between 80 and 140 °C, and the moisture content near the saturation point. Under these conditions, lignin, hemicelluloses and the semi-crystalline cellulose can be deformed easily. Hemicelluloses in the cell wall are softened (at 54–56 °C), which

decreases the stiffness of wood. This enables wood fibers to adapt their cross-sectional shape to the applied forces. The softening of lignin (at 72–128 °C) in the cell wall and middle lamellae permit further cross-sectional movement within and between the fibers (Kutnar and Šernek 2007; Kutnar et al. 2009; Tu et al. 2014; Rautkari et al. 2011).

Compression changes the wood morphology as the cell walls buckle and the volume of void spaces gets reduced while a non-uniform density profile across the sample is generated as the surface becomes denser. A higher degree of densification is achieved by a larger reduction in the void spaces of the wood and at high pressure, the volume of void spaces in the specimens is drastically reduced, and cell lumens are deformed. Usually the cells are deformed without fracturing the cell walls, while the rays appear to buckle (Kutnar et al. 2009).

When compressed wood is exposed to water, it tends to regain its original dimensions. This effect occurs because internal stresses are introduced into the cell walls during compression. This memory effect or spring back can be completely eliminated by different methods. Heat treatment and steam treatment are the most common ones (Morsing and Hoffmeyer 1998; Wolcott and Shutler 2003; Kutnar et al. 2009). Kutnar and Kamke (2012a, b) suggested that the stabilization of compressive deformation was more successful when wood was compressed under saturated steam at 170 °C. In addition, heat treatment at 200 °C adds some improvement to dimensional stability.

Sandberg et al. (2013) published a review article about thermo-hydro and thermo-hydro-mechanical wood processing, where also other applications of densified wood are discussed.

5 Binderless boards

Due to its environmentally friendly properties, many researchers tried to produce fiber or particle boards without adhesive or using bio-based adhesives such as sugars or lignin. Wood-based binderless fibreboards without synthetic resin binders have been produced for at least 80 years: several types of bio-raw material were used for binderless boards, for example wood bark (Chow 1975; Wellons and Krahmer 1973; Geng et al. 2006; Gao et al. 2011), rice and wheat straw (Zhao et al. 2013; Kurokochi and Sato 2015a, b), kenaf (Xu et al. 2004), banana stems (Nongman et al. 2016), coconut husks (van Dam et al. 2004; 2006), bagasse (Mobarak et al. 1982), oil palm (Hashim et al. 2012; Baskaran et al. 2015), soybean straw (Song et al. 2020) and bamboo (Shao et al. 2009) etc. Recently, a review work on binderless fibreboards from agricultural residues was published summarizing the related studies (Nasir et al. 2019).

Several parameters influence the physical and mechanical properties of wood panels: chemical composition (Widyorini et al. 2005; Lui et al. 2018), particle size and geometry (Widyorini et al. 2011; Kurokochi and Sato 2015a, b; Lui et al. 2018; Ahmad et al. 2019; Ferrandez-Villena et al. 2020), pressing temperature (Okuda et al. 2006a; Gao et al. 2011; Hashim et al. 2011; Boon et al. 2013; Milawarni et al. 2019; Song et al. 2020), pressing time (Xu et al. 2006; Gao et al. 2011; Boon et al. 2013; Ferrandez-Villena et al. 2020), pressure (Boon et al. 2013); water content (Widyorini et al. 2005; Xu et al. 2006), and pretreatment (Xu et al. 2006; Takahashi et al. 2010), etc. Trichomes and wart-like protuberances on the epidermis of herbaceous straw might inhibit the bonding between particles. Wax-like substances on the epidermis of rice straw might contribute to the water resistance of the board but inhibited the adhesion of the particles (Kurokochi and Sato 2015a, b).

The chemical composition of the different biomasses is similar and their main components are the same: cellulose, lignin, hemicelluloses. The bonds between the bioparticles are based on chemical and physical interactions occurring during the hot pressing between various components of the particles or their derivatives (Suchsland and Woodson 1987; Yelle 2001; Hubbe et al. 2018). In fiberboard production, during thermomechanical pulping, woodchips are converted to lignin-covered fibers by shearing wood fibers along the lignin-rich middle lamellae (Felby et al. 1997; Kharazipour et al. 1998). Goring (1971), Yelle (2001), van Dam et al. (2004), Bouajila et al. (2005), Okuda et al. (2006a, b), Halvarsson et al. (2009), Wang et al. (2017) and Laine et al. (2019) suggested that lignin has an important role in the production of binderless boards due to the softening of lignin at elevated temperatures and under pressure, fibers with lignin-rich surfaces fuse together as the softened lignin molecules flow from one fiber surface to another one, and possibly form covalent bonds too. If the water content of the raw material is above or near the saturation point, the T_g of the cell wall polymers is relatively low. However, as the temperature of the materials rises during hot pressing, the water content of the particles or fibers decreases and as a result the softening temperature also increases. The T_g of the polymers increases around 200 °C. The polymers of the raw material are in a plasticized state during a limited period and this period depends on their position within the panel (Bouajila et al. 2005). Water leaves the panel near its sides, and the T_g rapidly increases. At elevated temperatures, irreversible softening of the lignin occurs due to dehydration and cross-linking reactions (van Dam et al. 2004), so the role of the softened lignin cannot be separated from the chemical changes.

Several chemical reactions were observed during hot pressing: hydrolysis of the hemicelluloses occurs (Runkel and Wilke 1951; Xu et al. 2006; Zhang et al. 2015), organic

acids, mainly furfural, are formed, but the amount of the resulting furfural depends on the chemical composition of the raw material and its water content, temperature etc. (Rowell and McSweeney 2008; Tshabalala et al. 2012). The monomers generate lignin–furfural linkages or undergo self-polymerization during the pressing, which provide the main self-bonding strength of binderless fiberboards (Suzuki et al. 1998; Zhang et al. 2015).

The surfaces of the particles and fibers can be activated by different pretreatments, which increase the amount of the chemical bonds. Fenton's reagent contains ferrous chloride and hydrogen peroxide; hydroxyl radicals are generated by decomposition of hydrogen peroxide with the assistance of ferrous ions. In the end, reactive components formed in lignin and the properties of the boards improved (Kharazipour et al. 1998; Halvarsson et al. 2009; Zhang et al. 2015).

Enzymatic systems can also activate lignin on fiber surfaces by generating free radicals, and create bonds between the surfaces of fibers and particles without the use of conventional adhesives (Felby et al. 1997; Hüttermann et al. 1998; Kharazipour et al. 1998). Laccase enzyme can oxidize the lignin and stable lignin radicals are formed (Hüttermann et al. 1998; Felby et al. 2004; Pereira et al. 2005; Nyanhongo et al. 2010). This lignin radical reacts with cellulose and other lignin molecules, so it can be used to produce composite materials using lignin and fibers. Wet process boards produced in this manner have shown equal or better bonding properties than boards produced with phenolic adhesives (Hüttermann et al. 1998). The enzymatic activation of lignin for lignocellulosic products such as MDF and particleboard can improve the self-bonding properties of the biomass by oxidation of their surface lignin before being fabricated into boards (Widsten and Kandelbauer 2008).

Panel properties can be improved by steam explosion (Suzuki et al. 1998; Anglès et al. 1999, 2001; Shao et al. 2009; Gao et al. 2011), because lignin droplets were observed on the surface of the fibers (Anglès et al. 1999). Steam injection during pressing has a similar result.

Due to the physical and chemical bonds formed by lignin, the addition of lignin to the raw material can improve the properties of the panels (Anglès et al. 2001; Hemmilä et al. 2013; Milotskyi et al. 2019).

6 Solid wood bending

Wood bending is one of the oldest wood processing techniques. Long experience has evolved from the practice of bending techniques and skilled craftsmen can apply them. Naturally, at normal moisture content and temperature, every wood has some plasticity, but this plasticity is not enough to bend solid wood. Heat and moisture make certain species

of wood sufficiently plastic for bending operations. In general, hardwoods are more readily softened than softwoods, and certain hardwoods more so than others. The purpose of all plasticizing treatments is to soften wood sufficiently to enable it to take the compressive deformation necessary to make a curve (Peck 1957). Hot wood is more plastic than cold wood, and wet wood is more plastic than dry wood. The plasticity of wood can be increased by increasing the moisture and/or temperature of the wood. Together, heat and moisture can produce a degree of plasticity 10 times that of dry wood at normal temperatures (Peck 1957). Hot water and steam are commonly used treatments to prepare wood for bending.

The glass transition temperature (T_g) of the lignin in moist wood is 80–100 °C. Above T_g , the lignin undergoes thermoplastic flow and resets in the modified configuration when cooling (Nakajima et al. 2009; Ibach 2010; USDA 2010).

The temperature of saturated steam at atmospheric pressure, about 100 °C or boiling or nearly boiling water, is generally sufficient to plastify wood for bending. In this condition, the viscosity of the lignin decreases, which makes connections between the cellulose micro- and microfibrils (Angelski 2014). If the raw material has a 20–25% moisture content, no additional moisture is needed, even for severe bends (Peck 1957). Technically these methods belong to the hydrothermal treatment. If thermal treatment is used, water or steam has no impact on the raw material. This method is implemented by contact of hot metal surfaces (Angelski 2014). In another method, high frequency heating is used (Sandberg and Johansson 2005). The high-frequency microwave heating of wood is also used for plasticization (Ibach 2010; Gašparík and Gaff 2013). A longer plasticizing time increases the amount of microwave energy delivered to wood, on the other hand, the higher amount of energy also causes a higher moisture loss. Therefore, a suitable plasticizing time should be chosen (Gašparík and Gaff 2013).

Some chemicals can also soften wood. When chemicals are used for plasticization, the connections between the matrix (lignin) and cellulose and the ties between the cell walls loosen.

The chemical plasticization commonly uses water solutions of ammonia, urea, dicyandiamide, ethylenediamine and liquid ammonia (FPL 1943; Schuerch 1963; Pentoney 1966; Bariska 1969; Davidson and Baumgardt 1970; Bariska and Schuerch 1977; Angelski 2014; Suleman 2015; Šprdlík et al. 2016). Ammonia is a solvent with a similar molecular size but a greater hydrogen bonding capacity than water. Liquid ammonia can swell and soften wood more than water. It breaks hydrogen bonds with all the components of the cell walls and creates new hydrogen bonds, not only with hemicellulose and lignin, but it can also separate the chains of the crystalline cellulose. When the ammonia evaporates,

new hydrogen bonds are formed in new positions and a new shape form.

7 Veneer manufacturing

Several factors have effects on veneer manufacturing and veneer quality (Olufemi 2012). Successful veneer production requires wood softening. The heating of green wood prior to peeling has traditionally been accomplished by soaking—immersing the whole logs in hot water basins—or by steaming them in vats. In both processes, water, as an integral part of wood, makes an ideal medium for heat transfer into wood (Daoui et al. 2007). The energy required for peeling can be reduced if less energy is required to move the cutting knife (Marchal et al. 2009; Dupleix et al. 2012; Dupleix 2013; Xu et al. 2017). If the shear strength of the wood decreases, it is sufficient to exert less pressure on the knife during peeling (Bédard and Poulain 2000). This reduces the power consumption and reduces wear of the tools (Marchal et al. 2004) and the quality of the veneer is improved reducing the formation of lathe checks and other surface quality defects (Lutz 1960; Aydin et al. 2006; Dundar et al. 2008; Marchal et al. 2009; Dupleix et al. 2012; Olufemi 2012; Rohumaa et al. 2016a, b, c; Frayssinhes et al. 2019).

During soaking or steaming the wood softens, and the deformability of wood increases (Baldwin 1975; Bardet et al. 2003; Yamauchi et al. 2005). Under optimal conditions, shallower lathe checks form (Rohumaa et al. 2016a, b, c; Frayssinhes et al. 2019). For this purpose, the temperature should exceed the glass transition temperature T_g of lignin at the MC of green wood across the log. This temperature is lower than the T_g of cellulose, while hemicelluloses are already above their T_g (Engelund et al. 2013; Navi and Sandberg 2012). Therefore, lignin changes have the greatest influence on the behavior of wood. Reaching the T_g of lignin also fluidizes wood resins and softens knots, which increases tool life. Rohumaa et al. (2017) suggested that the logs not only soften by heat, but irreversible changes occur in the wood material.

8 Stress relaxing

Heating logs in water or saturated steam relieves growth stress (Skolmen 1967). The temperatures of *Eucalyptus grandis* logs were determined and correlated with the cracks in the boards during steaming. Thermocouples were inserted in the centers of the logs, registering their temperatures during steaming at 90 °C. It was found that the lengths of the cracks significantly decreased in logs that reached the glass transition temperature (Calonego et al. 2010).

9 Conclusion

We have seen that lignin softening plays a role in a variety of woodworking processes, which are basically divided into two groups. The first group includes processes where lignin softens or possibly liquefies, and the soft lignin penetrates into the gaps of adjacent particles and cell walls, and acts as an adhesive. This includes wood welding, the manufacture of binderless boards or manufacturing pellets. The other group consists of processes where the wood has to undergo some deformation and therefore the cell walls and the materials that compose the cell wall must move relative to one another. This group includes veneer manufacturing, solid wood bending and surface densification.

The referred 257 articles show the great effort of scientists to explore the composition characteristics, and behavior of the components of wood, especially focusing on lignin.

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