

## **CHAPTER 2**

### **Literature Review**

#### **2.1 DENSIFIED WOOD**

##### **2.1.1 Background**

Wood with inadequate mechanical properties can be modified by various combinations of compressive, thermal and chemical treatments. It can be densified by impregnating its void volume with polymers, molten natural resins, waxes, sulfur, and even molten metals, with subsequent cooling to solidify the impregnant. On the other hand, wood can be compressed in the transverse direction under conditions that do not cause damage to the cell wall (Kollmann et al.1975). The compression of solid wood has been done in Germany since 1930 under the trade name of Lignostone. Laminated compressed wood has been made under the trade name Lignofol. Similar materials, Jicwood and Jablo, have been in production in England for some years (Rowell and Konkol 1987). In the United States, patents on methods of densifying wood (such as Sears, Walch and Watts, Olesheimer, Brossman, Esselen) date back to 1900. These patents did not adequately consider plasticization of the wood or stabilization of the final product; for this reason, they have not been adopted by the industry (Kollmann et al. 1975).

Another two methods of densification created in the United States are Compreg (Stamm and Seborg 1941) and Staypak (Seborg et al. 1962a) developed at the Forest Products Laboratory. Compreg is resin-treated compressed wood. It is normally made by treating solid wood or veneer with water-soluble phenol formaldehyde resin and compressing it to the desired specific gravity and thickness. The wood is first dried at a temperature low enough to avoid pre-cure, and then compressed under temperatures which will cure the resin. To obtain the maximum dimensional stability a resin content of about 30 percent and a specific gravity of 1.3 to 1.4 are desired. Compreg, however, can be produced at lower resin contents and specific gravities. The pressure required for producing the panels depends upon the resin and its volatile contents, the degree of pre-cure of the resin, the distribution of the resin throughout the structure, and the species of wood. Compreg can be made from veneer of most species by applying pressures of 1,000 to 1,200 psi at a temperature of 300°F (150°C), when the volatile content of the resin is between 2 and 4 percent. The compression of the panels to specific gravities above 1.4 is not recommended due to increased ability to check (Seborg et al.1962b). The increase in strength properties of Compreg, except for impact strength, is proportional to the increase in specific

gravity. Compreg is much more dimensionally stable than non-impregnated compressed wood. A similar resin-treated compressed wood to Compreg has been made in Germany under the name of Kunstharzschichtholz (Kollmann et al. 1975). The application of Compreg was for aircraft parts, gears, pulleys, shuttles, bobbins and picker sticks for looms, instrument bases and cases, electrical insulators, and musical instruments.

In contrast with Compreg, Staypak is not impregnated with resin. Treating resins harden within a cell wall making wood more brittle. Thus, if a tough, compressed product is desired, a brittle polymer should not be impregnated in the wood. Unfortunately, untreated, compressed solid wood and veneer tend to spring back or recover from their compression when exposed to moisture. Springback, unlike true swelling, is not reversible. It results from the release of internal pent up stresses, caused by the original compression, when the wood was softened. It is believed, that the removal of pent up stresses, and subsequent reduction of springback, is due to a slight flow of the cementing lignin between the fibers (Seborg et al.1962a). To eliminate springback wood should be pressed under conditions that cause sufficient flow of the lignin. This type of stable form of compressed wood has been given the name, Staypak. Staypak is produced by compressing wood at a moisture content equal to or below that which it will have in service. The temperature ranges from 300°F (150°C) to 360°F (180°C). Pressures of 1,400 to 2,500 psi are generally required where a final specific gravity of 1.3 is desired (Seborg et al. 1962a; Rowell and Konkol 1987). One of the problems associated with making of Staypak is that the panels must be cooled to 100°C or less while under the full pressure. Due to the thermoplastic nature of the lignin, and because the moisture content of the wood is only slightly less after compression than prior to pressing, considerable springback will occur if the product is removed while still hot (Kollmann et al. 1975). This necessity and other disadvantages of Staypak prevented this product from being adopted by the industry. Strength properties of Staypak are generally comparable with those of Compreg, except that the impact strength is considerably higher. Staypak is much less dimensionally stable than Compreg.

### **2.1.2 Wood Stabilization**

One of the main problems associated with most of the types of densified wood (except those with high resin content) is the lack of dimensional stability. When soaked in water or exposed to high relative humidity, compressed products tend to exhibit irreversible swelling or springback. This can be a serious problem when densified wood is used in high humidity

environments. Thus, it is important to determine the pressing conditions under which the recovery from compression for untreated compressed wood is minimized. There have been many studies relating to wood stabilization by various treatments. Hillis (1984) reviewed the literature about stabilization of wood by a heating process. The effect of steam pretreatment on wood was investigated by Hsu et al. (1988); Inoue et al. (1993); Inoue et al. (1996) and Kawai et al. (1992). Lately, the effect of heat on the dimensional stability of compressed wood has been evaluated by Dwianto et al. (1996). Tomme et al. (1998) performed thermo-hygro-mechanical treatment in order to produce densified wood with stable deformation.

Dwianto et al. (1996) found that preheating made a great influence on the permanent fixation. According to their results, the permanent fixation of compressive deformation in wood resulted from the release of stresses stored in microfibrils and the matrix substance of the cell wall due to their degradation.

Hsu et al. 1988 developed a steam pretreatment process to produce highly dimensionally stable wood-based composites. They found that steam pretreatment causes partial hydrolysis of hemicelluloses for both hardwoods and softwoods, which greatly increases the compressibility of wood (i.e., reduces the tendency of internal stresses to build up in composites during hot pressing).

Inoue et al. (1993) found that almost complete fixation can be achieved by post-steaming compressed wood for 1 min. at 200°C or 8 min. at 180°C. There was a large increase in hardness and only a slight decrease in MOE and MOR. Inoue et al. (1996) also investigated the effect of pre-steaming. They found that the degree of recovery decreases if the press time and temperature increase. Presteaming increases the compressibility of wood and reduces the amount of stored stress due to the viscous flow of wood substances.

Kawai et al. (1992) produced LVL by steam-injection pressing. They found that MOR and MOE of compressed LVL increased with increasing density. The dimensional stability of LVL has been improved considerably. They also have proposed the mechanism responsible for the fixation of compressive set by steam treatment. They hypothesize that relaxation of the stresses stored in the microfibrils and fixation of the compressive set is due to: rapid hydrolysis of hemicellulose and partial degradation of lignin; partial hydrolysis of cellulose of amorphous and paracrystalline region, and reorientation in the crystalline region by steam treatment.

In conclusion, the stabilization of compressive deformation might be a problem in the application of densified wood; however, this problem can be solved by various treatments of wood.

## **2.2 INFLUENCE OF HIGH TEMPERATURE AND STEAM PRESSURE ON WOOD**

Wood can be characterized as a polymeric cellular material (Wolcott et al. 1990). The properties of cellular materials are influenced by properties of the cell wall and the cellular geometry. The effect of moisture and temperature on the cellular structure is not very significant, whereas properties of the cell wall components are greatly affected by these factors. The dependence of the properties of cell wall material on the moisture and temperature can be characterized through viscoelastic behavior.

### **2.2.1 Viscoelastic Behavior of Amorphous Polymers in Wood**

Viscoelastic behavior of wood has been characterized by many studies (Kunesh 1968; Hillis and Rozsa 1978; Back and Salmen 1982; Salmen 1984; Kelley et al. 1987; Wolcott et al. 1989, 1990; Ostberg et al. 1990). It is known that wood is a composite material, which consists of three structural polymers, cellulose, hemicelluloses, and lignin. Amorphous polymers of wood (lignin and hemicelluloses), as viscoelastic materials, can behave as viscous fluids and as linear elastic solids, depending on the time, temperature and diluent concentration (Wolcott et al. 1990). For isolated amorphous polymers the transition between the glassy and the rubbery state is defined as a glass transition temperature,  $T_g$  (Back and Salmen 1982). Many properties of these polymers, such as elastic modulus, change dramatically when the material passes this softening point.

The effect of moisture and temperature on the individual wood polymers has been investigated in many studies. Moisture has been found to change the glass transition temperature of amorphous polymers (Salmen 1984; Kelley et al. 1987). Plasticization of cell wall compounds by water causes a reduction of the energy required to initiate chain mobility (Kelley et al 1987). Temperature does not have a great effect on mechanical properties when the polymer is below  $T_g$ , because molecular motion is limited. However, when the temperature of the polymer reaches  $T_g$ , stiffness of the material decreases rapidly. This is attributed to the increase in thermal energy available for molecular motion (Cowie 1991; Young and Lowell 1991).

The behavior of whole wood, however, under various conditions can not be explained considering only glass transition points of isolated wood polymers, because the chemical and

physical nature of isolated components may differ from their original form (Hillis and Rozsa 1978; Kelley et al. 1987). Moreover, the combinations between polysaccharides and lignin may influence the behavior. There are some direct analytical methods which are sensitive to the response, on a molecular level, of the whole wood (Salmen 1984; Kelley et al. 1987; Ostberg et al. 1990). They include dynamic mechanical analysis (DMA), dielectric analysis (DEA) and differential scanning calorimetry (DSC). Utilizing these methods for the analysis of the viscoelastic behavior of whole wood, more than one transition can be found (Kelley et al. 1987).

The influence of high temperature and steam pressure on wood has been studied by many researchers (Price 1976; Geimer et al. 1985; Kamke and Casey 1988; Kawai et al. 1992; Kosikova et al. 1993; Ebringerova et al. 1993; Gardner et al. 1993; Inoue et al. 1993, 1996). The strength and stiffness can be either increased or decreased, depending on the conditions under which the material is processed.

The effect of steam treatment on structural changes of the lignin-polysaccharide complex has been investigated by Kosikova et al. (1993) and Ebringerova et al. (1993). Beech, aspen and spruce flakes were subjected to various conditions of steaming, such as heating with steam at 200°C for 10 min, heating with steam under pressure of about 4.3 MPa at 255 °C for 55 sec, heating with steam at 115 °C for 5 min and heating with steam at 200 °C for 2 min under steam explosion conditions (4.3 MPa). The changes in structure of wood polymers were characterized using infrared spectroscopy. It was found an apparent increase in cellulose crystallinity in the steamed samples, particularly in the steam explosion wood. The increase was attributed to hydrogen bond destruction as well as to splitting of lignin-hemicellulose linkages in hardwoods and spruce after steam treatment.

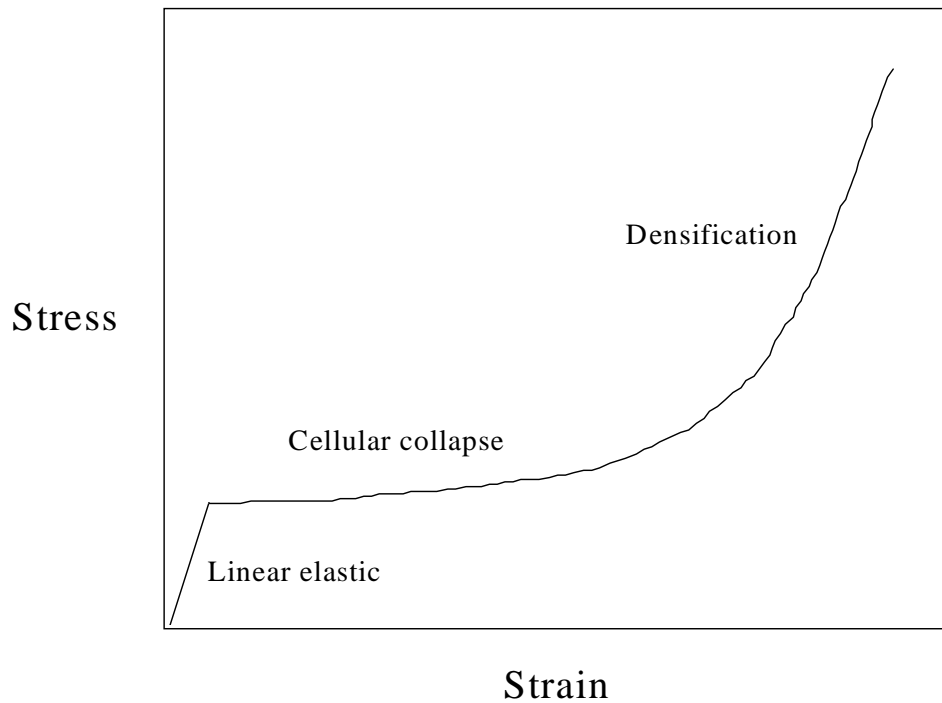
Gardner et al. (1993) investigated the changes in polymer structure of wood flakes under hot-pressing conditions. The data obtained indicate that an increase in cellulose crystallinity with heat and steam treatment results in an increase in elastic modulus. The crystallinity increase is attributed to the transition of amorphous polymers from the glassy to the rubbery state, when the increased mobility of lignin and hemicelluloses can permit reorientation and crystallization of the cellulose microfibrils. They also point out that low pressing temperatures result in a decrease in flake strength and stiffness, whereas, high pressing temperatures result in an increase in these properties.

Kawai et al. (1992) analyzed the effect of high-pressure steam treatment on the properties of LVL. Two steam pressures of 6.3 and 10.2 kgf/cm<sup>2</sup> (160 °C and 180 °C, respectively) were supplied in the pressure vessel for 5 minutes, and the total pressing times were 7 to 9 minutes. The results have shown that the specimens treated at a higher steam pressure (180 °C) had lower MOE values. It was attributed to some degradation to cellulose microfibrils at the higher steam pressure and temperature.

### **2.2.2 Compression of Cellular Materials**

Wood can be characterized as a composite material with cellular structure. The mechanical properties of cellular materials in transverse compression are highly non-linear resulting from the collapse of the cellular structure (Wolcott 1989). The type and amount of the cell collapse makes a great influence on the mechanical and physical properties of densified material. The performance of the resulting product is also influenced by the parameters of the compression process.

All cellular materials loaded in transverse compression exhibit similar behavior, which can be characterized by the stress-strain curve (Gibson and Ashby 1988). A typical deformation goes sequentially through three distinct stages: initial linear elastic cell-wall bending; nonlinear cellular collapse at nearly constant stress level; and the final cell wall densification when the majority of the cells have collapsed (Figure 2.1). Cellular collapse occurs by either elastic buckling, plastic yielding, or brittle crushing, depending on the test conditions and the nature of the cell wall material (Wolcott et al. 1990). Elastic buckling can be defined as buckling without fractures of the cell wall. It occurs when the polymers of the cell wall are in the rubbery phase, and will result in certain recovery of the deformation upon removal of the load. Plastic yielding occurs when the polymer is in transition between glassy and rubbery phases. In this case, permanent deformation remains after the load is removed. Brittle crushing of the cell wall occurs when the polymer is in the glassy phase (Cowie, 1991).



**Figure 2.1** Typical stress-strain curve characterizing a cellular material in compression.

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