THE EFFECTS OF TANNIN AND THERMAL TREATMENT ON PHYSICAL AND MECHANICAL PROPERTIES OF LAMINATED CHESTNUT WOOD COMPOSITES

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The aim of this study was to investigate the effects of tannin and thermal treatment on physical properties such as dimensional stability and moisture content (%), as well as mechanical properties such as bending strength, modulus of elasticity, and compression strength of laminated chestnut wood composites, which are used commonly for shipboard construction in Turkey. The chestnut wood used in boat construction is usually exposed to several treatments in order to achieve better bonding performance and to remove excessive tannins. According to the results obtained, physical properties of laminated chestnut wood without tannin were better compared to samples with tannin. Oven-dry density and air-dry density were found to be higher in samples containing tannins and not exposed to thermal treatment. Moreover, the lowest value of density was observed in samples containing no tannin and exposed to thermal treatment. In terms of mechanical properties, the highest bending strength and modulus of elasticity were obtained from samples containing tannins and not exposed to heat treatment. The lowest values were found in samples without tannin exposed to heat treatment. Regarding compression strength, the maximum and minimum values were found in samples containing tannins and not exposed to heat treatment and samples without tannin and exposed to heat treatment, respectively.

Keywords: Anatolian Chestnut wood; Laminated wood; PVAc–D4; Mechanical properties; Physical properties

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INTRODUCTION

Wood is an organic material that has been used by people for many purposes since the early ages. Wood demand is increasing because of its easy processing, natural texture, good insulation properties against heat and sound, the color and aesthetic properties, a very high fatigue resistance in comparison to steel and concrete, and good fire resistance (Triantafillou 1998). Since the unplanned, unconscious, and excessive consumption of forest areas causes extinction and destruction, it is important to use wood materials more efficiently (Maloney 1996). Therefore, wood materials are used in different forms such as laminated wood, laminated timber, laminated veneer lumber, and many other forms.

One of these forms is laminated wood composites, which can make use of different wood species. Laminated composites consist of two or more layers of wood that are glued together with the grain of all layers, which are referred to as laminations, parallel to the length (Stark et al. 2010). It should not be ignored that the use of wood material incorrectly will cause enormous economic losses in parallel with the increasing importance of wood. Especially from a technical perspective, use of wood as one piece in large and curved elements is not always possible. Sometimes it is not possible to take away the defects completely from wood, noting that defects such as knots, decay, cracks, and spiral grain can harm the value of the piece as a whole. The loss ratio increases in the production of curved elements from a single piece of wood, and it is not economical. Moreover, diagonal grain occurs in the curved cut wood, and it will have a negative impact on strength properties of wood. In order to use wood materials efficiently, to eliminate defects, to prevent diagonal fiber formation during the manufacturing of curve-shaped wood material, and to prevent the decrease on mechanical properties, lamination techniques must be used. Laminated wood materials are used especially in the construction industry (wooden boats, bridges, houses, factories, mosques, churches, silos, sheds, etc.) and in the furniture industry (Örs and Keskin 2002; Nurgul Denizli et al. 2004).

In the case of laminating wood veneers, the amount of moisture content, the surface, the amount of extractives mass and the location where the wood is used have a significant effect on the properties of laminated wood composites. Anisotropic swelling and shrinkage due to fluctuations in wood moisture content causes problems. Besides the problem of dimensional stability, biological attack problems related to moisture can occur (Uysal 2005; Bland 1990). Fungi and bacteria need water to survive. If the moisture content is kept under 20%, the water amount is not enough for the fungi or bacteria to live. So to protect wood material from fungi and bacteria, preservative treatments or impregnation are often used. The disadvantage of such methods is that they require the use of the toxic materials. Wood preservatives can't prevent wood from shrinking or swelling, but they can make the wood chemically stable. In this case, the impregnation process has become too expensive. Recently instead of impregnating wood with substances harmful to the environment, a newly studied method called heat treatment has been applied. Such treatments can provide dimensional stability to wood, and they have been used in many countries (Kandem et. al 2002; Kaygin et. al 2009; Gunduz and Aydemir 2009). According to this method commonly used in Europe, wood materials are heated to 180 ^{o}C under the protection of water vapor. Besides the protection of wood material, water vapor has a significant impact also on the chemical changes.

As a result of this treatment, environmentally-friendly heat treated wood materials will be produced. The color changes in wood during heat treatment. The product is more stable than normal wood under different humidity conditions and has very good thermal conductivity. If the wood is treated at appropriate temperatures, this will also increase the decay resistance significantly (Bekhta and Niemz 2003; Poncsak et al. 2005; Gunduz et al. 2008). Using heat treatment method and lamination method together in the buildings is important for the protection from physical harm caused by fungus and bacteria. In addition, the heat-treated wood material should be preferred because of dimensional stability and avoiding the use of toxic wood preservatives. Thus, wood materials are better for use in application areas such as furniture, solid wood based materials, wooden boats, etc.

Chestnut wood is commonly used to make wooden boats in Turkey. The elasticity and strength of chestnut wood has been judged to be sufficient for this purpose. But chestnut wood contains a large amount of tannins; for this reason chestnut wood is soaked in water for the removal of tannins prior to using the wood, and thus water resolves tannin and removes it from wood. Tannins consist mainly of gallic acid residues that are linked to glucose *via* glycosidic bonds. There are a lot species of tannin, and the structures of the tannins are different from each other. Tannins contain mainly gallic acid residues that are linked to the glucose via glycosidic bonds as shown in Fig. 1. Tannin hydrolyzes easily when it is boiled in aqueous acids. Tannin is an amorphous mass that can be colorless or light-yellow. Tannin can be effectively removed from wood with washing in warm water during 1 or 1.5 weeks (Hagerman 2002; Jones 2007).

Tannins are polyhydroxypolyphenolics that occur in many plant species, but they have a high enough concentration to be worth isolating from only a few species. Chestnut wood is one of those few species. Tannins are used because they are more reactive than phenol, but they are also more expensive than phenol (Frihart 2005).

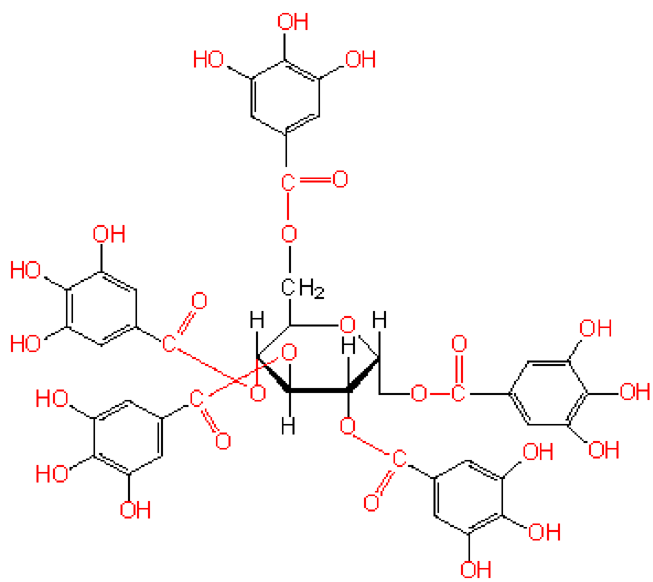

Figure 1. The Chemical Structure of Tannin (Sengbusch 2003)

Chestnut wood is in the water during removal of tannin, and therefore physical and mechanical properties of wood can change. The changing in the laminated composites can have important related to its subsequent uses.

Mantanis et al. (1994a) studied the effect of extractives on swelling-shrinkage of spruce, maple, and aspen. They determined activation energies for swelling of extracted wood in water. The rate of swelling in water considerably increased after removal of the extractives, and the maximum tangential swelling also generally increased by approximately 5 to 10%. It was additionally found that removal of extractives caused a large decrease in the activation energy of wood swelling, E_a.

In this study, the feasibility of using heat-treated and laminated Anatolian chestnut wood for the construction of boats and yachts in Turkey was investigated. Chestnut wood contains a high proportion of tannin, a phenolic substance. So, when the tannin was leached, effects of mass loss and physical properties improved due to heat treatment. Effects of tannin removal on mechanical properties were also determined.

MATERIALS AND METHODS

Anatolian chestnut (*Castanea sativa* Mill.) wood used in this study was obtained from Zonguldak, Turkey. Chestnut logs were cut to dimensions of 25 x 70 x 780 mm, and the dried lumber pieces were sliced to veneers having a 4 mm thickness. Half of the veneers obtained were washed under water to remove tannin compounds (these veneers were called veneers with non-tannin) and other veneers weren't exposed to any process (other veneers were called as veneers with tannin). Both non-tannin and tannin samples were equilibrated to moisture content of 12% in an oven at 103±2 °C. After completing the equilibration process, all of samples were thermal-treated at 180 °C for 2 hours. As a result of all the modification processes, veneers for test samples were obtained as tannin/non-thermal treated, tannin/thermal-treated, non-tannin/non-thermal treated, and non-tannin/thermal treated. According to TS EN 386, the adhesive used in this study was poly vinyl acetate based PVAc-D4 adhesive. The adhesive was applied to veneers with a brush at an application rate of 180±10 g/m^2. This process was done at room temperature (23±2 ^{o}C and 65±5% relative humidity. Samples with adhesive were combined with each other and the samples obtained were pressed in a hydraulic press (cold press at 20±5 ^{o}C) for 90 minutes at room temperature (23 ± 2 °C) at a pressure of 1.0 MPa. Loading speed of press was 5 mm/sec, and the press was used to achieve better bonding of samples during manufacturing process. The four groups mentioned above were bonded as five layers. 30 replicates for each group were prepared.

Table 1. Technical Properties of PVAc-D4 Adhesive

Properties	PVAc–D_4 Dispersion
Density	Component A/B = 20/1
Hardener	Dorus R.397 (Mixing Ratio: % 5)
Ph Value	≈ 3
Applied Amount	180±10 g/m^2
Application Type	Brush
Press Time	15 min at 20 ^{o}C, 5 min at 50^{o}C, 2 min at 80^{o}C

To determine the density, small, clear Anatolian chestnut specimens (20 x 20 x 30 mm) were obtained from a local mill. Density measurements were carried out according to Turkish Standards (TS) TS EN 2472. To evaluate the swelling and shrinkage (dimensional stability), test samples prepared according to TS 4084 and TS 4083 were oven dried to constant weight in an oven at 103±2 °C, and then the dimensions of samples were measured using a micrometer. For determining to swelling ratio of samples, they were put to water bath at 20 °C for 24 hours. After the soaking, the samples were measured, then the samples were dried again, and shrinkage of samples was determined. To determine the moisture content, according to TS 2471, test samples of dimension 20 x 20 x 30 mm were prepared. After the samples were oven dried, they were kept in a climate-controlled room for determining moisture contents of samples at 20 °C for 35%, 65%, 85%, and 95% relative humidity

Bending strength and modulus of elasticity were determined according to TS EN 326 (Formula 1) and TS 2474 (Formula 2), respectively. Samples prepared with the dimensions 20 x 20 x 360 mm were held at 20+2 °C temperature for 65+5% relative humidity until constant weight (%12 MC) was reached. Conditioned samples were tested by using a Zwick mechanical test machine for determining bending strength and modulus of elasticity. Compression test samples were cut into 20 x 20 x 30 mm blocks, which were prepared and conditioned until their equilibrium moisture content reached 12%. The tests were carried out according to TS 2595 standards,

$$MOR\ (\sigma_e) = \frac{3.F_{max}.L}{2.b.h^2}\ N/mm^2 \tag{1}$$

where σ is the MOR of the samples, F_{max} is the force of the maximum bending, L is the length of samples, b is the width of the samples, and h is the height of the samples.

$$MOE\ (E) = \frac{\Delta F.L^3}{4.b.h^3.\Delta f} N/mm^2 \tag{2}$$

where E is the modulus of elasticity of the samples, ΔF is force changes (strain) at elasticity area, and Δf is deformation at elasticity area.

All statistical calculations were based on a 95% confidence level. ANOVA tests indicated that differences between control specimens, samples with tannin, samples without tannin and heat-treated specimens were significant. Also, a Duncan Test was conducted to determine the differences among groups.

RESULTS AND DISCUSSION

Values obtained in the study were assessed by using variance analysis. Data determined according to variance analysis were found to have significant difference at the $p<0.05$ reliability level. The Tukey test was carried out to determine whether there were significant differences attributed to samples in different groups. This study was made among solid control, tannin/thermal-treated, tannin/non-treated, non-tannin/thermal treated, and non-tannin/non treated conditions. The effects of tannin removal and thermal treatment on air dry and kiln dry density of laminated chestnut wood composite are presented in Fig. 1

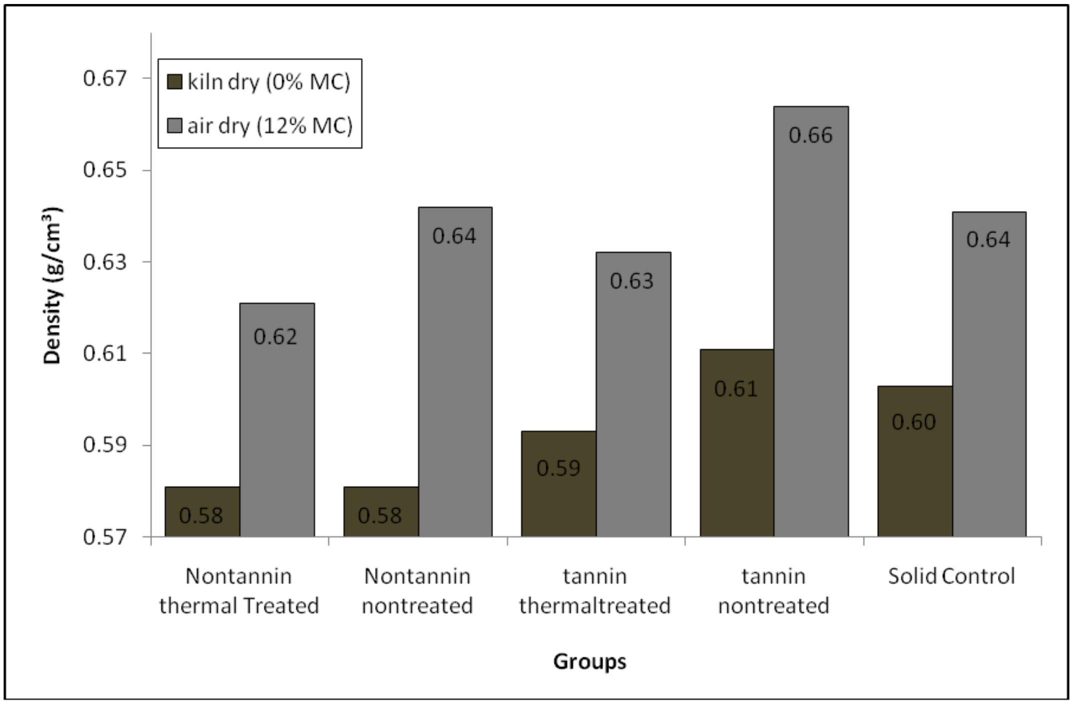

Figure 1. Oven dry and kiln dry of laminated chestnut wood composite

According to Fig 1, both the removal of tannins and thermal treatment affected the kiln-dry and air-dry densities of samples due to degradation of cell wall with heating and removing of tannin. Among the samples, the highest kiln dry density value was 0.61 g/cm³ and the highest air dry density was 0.66 g/cm³ for tannin/non-treated. And the lowest values were observed as 0.58 g/cm³ for both kiln-dry non-tannin/thermal-treated and non-tannin/non-treated, and 0.62 g/cm³ for air-dry non-tannin/thermal-treated samples. Kiln dry density of solid samples also was found 3.12% lower than tannin/non-treated samples, and air-dry density was found the same with non-tannin/non-treated samples.

According to Table 2, the highest shrinkage and swelling ratio of samples at tangential, radial, and longitudinal directions were found in solid control > non-tannin/non-treated > non-tannin/non-treated and solid wood > non-tannin/non-treated > solid wood, respectively. It was determined that the highest and lowest shrinkage ratios of samples were found as 7.60% in solid wood for tangential and 0.74% in non-tannin/thermal-treated for longitudinal direction. For radial directions, the highest and lowest ratio of shrinkage was found in 5.94% in non-tannin/non-treated and 2.70% in tannin/non-treated, respectively. Maximum and minimum values of swelling ratio were found as solid control and tannin/non-treated for tangential direction, non-tannin/non-treated and tannin/non-treated for radial direction, and solid wood and tannin/thermal-treated for the longitudinal direction. Figure 2 shows changes in moisture content (MC) of laminated chestnut wood composites in response to changes in humidity.

Table 2. Swelling and Shrinkage Values of Laminated Chestnut Wood Composite

Samples	Statistic Values	Swelling (%)			Shrinkage (%)		
		Tangential	Radial	Longitudinal	Tangential	Radial	Longitudinal
Tannin/thermal treated	X	***4.28AB***	***3.98B***	***0.74A***	***4.09AB***	***3.39AB***	***0.73A***
	±s	*0.14*	*0.07*	*0.06*	*0.10*	*0.28*	*0.06*
	V(%)	*3.25*	*1.67*	*7.95*	*2.53*	*8.28*	*7.78*
Tannin/ non-treated	X	***3.89A***	***2.74A***	***0.90A***	***3.71A***	***2.70A***	***0.89A***
	±s	*0.19*	*0.24*	*0.06*	*0.17*	*0.26*	*0.06*
	V(%)	*4.78*	*8.95*	*6.52*	*4.60*	*9.71*	*6.46*
Non-tannin/ thermal treated	X	***4.69AB***	***4.18B***	***1.00AB***	***4.53AB***	***4.35B***	***0.87A***
	±s	*0.25*	*0.09*	*0.11*	*0.46*	*0.09*	*0.03*
	V(%)	*5.42*	*2.22*	*11.02*	*10.11*	*2.09*	*3.00*
Non-tannin /non treated	X	***7.81C***	***6.55C***	***1.38BC***	***7.42C***	***5.94C***	***1.83B***
	±s	*0.21*	*0.98*	*0.08*	*0.30*	*0.21*	*0.12*
	V(%)	*2.73*	*14.95*	*5.61*	*4.01*	*3.49*	*6.33*
Solid Control	X	***8.08C***	***4.2B***	***1.5BC***	***7.6C***	***5.1C***	***1.6B***
	±s	*0.3*	*0.27*	*0.19*	*0.25*	*0.23*	*0.16*
	V(%)	*3.73*	*7.1*	*13.24*	*3.3*	*4.6*	*10.12*

X: Average, ±s: Standard deviation, V: Coefficient of variation. Homogeneity groups: Same letters (A, B, C, and D) in each column indicate that there is no statistically significant difference among the samples according to the Duncan's multiple range test at $p<0.05$. Comparisons were done between the control and test samples. Ten replicates were used in each test. All data in variance and one-way ANOVA tests were conducted at a confidence level of $p<0.05$ (95%).

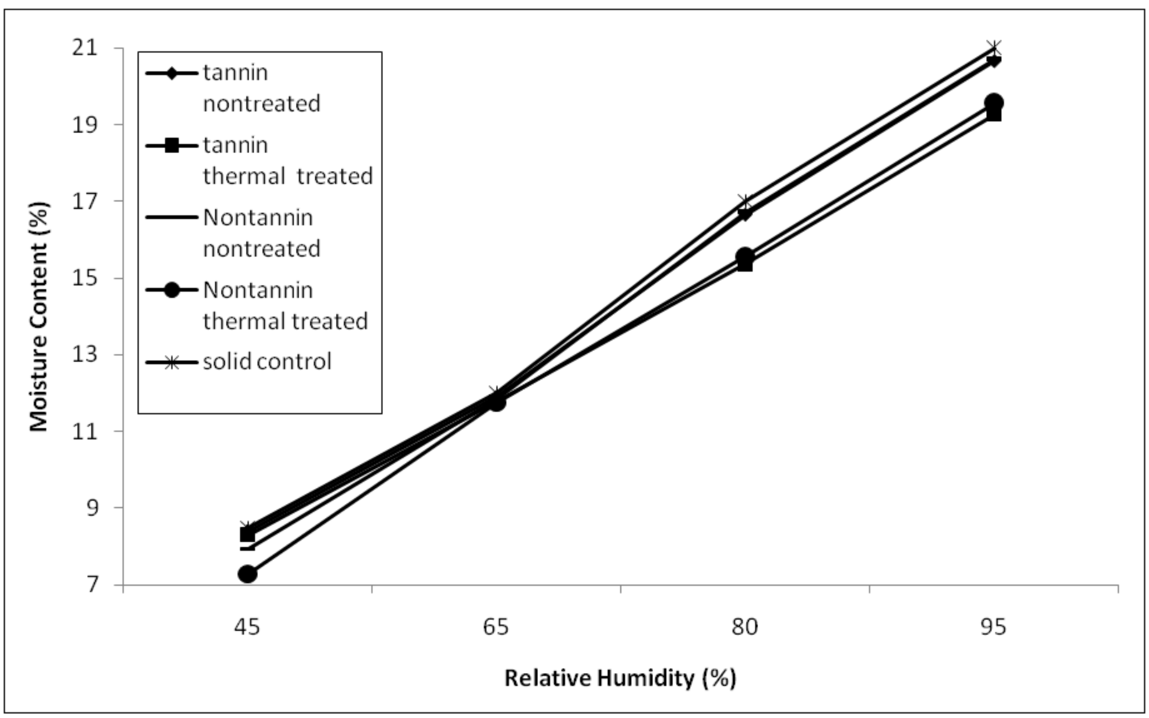

Figure 2. Moisture contents at different relative humidity levels of laminated chestnut wood composite

Choong and Achmadi (1991) found that the shrinkage intersection point ranged from 18.0 to 34.1% when extractives were removed. Also, the linear relationship between volumetric shrinkage and removing of extractives was determined.

Mantanis et al (1995) found that the maximum wood swelling increased after the removal of extractives, especially for sugar maple. Also obvious was the very large increase in maximum swelling for maple and spruce after extraction.

Tannin/thermal-treated, tannin/non-treated, non-tannin/thermal-treated, and non-tannin/non-treated samples were held at 20 ^{o}C and 45%, 65%, 80%, and 95% relative humidity, and their MC values were determined. So, both thermal treatment and removal of tannin had similar effects on moisture content of laminated chestnut wood composites. The moisture content of the solid control was higher than those of the other samples. As a result, MC values decreased with removal of tannin and thermal treatment.

The MC value of laminated wood with tannin and non-treated was lower than that of solid wood. Both removal of tannin and thermal treatment of wood caused changes in the physical properties such as kiln and air dry density, dimensional stability (swelling and shrinkage), and moisture content (MC). In addition to changing these physical properties, mechanical properties such as bending strength, modulus of elasticity, and compression strength of solid wood and laminated wood were affected due to these changes.

As seen in Fig. 3 and Fig. 4, removal of tannin and thermal treatment affected bending strength and modulus of elasticity of laminated chestnut wood composite.

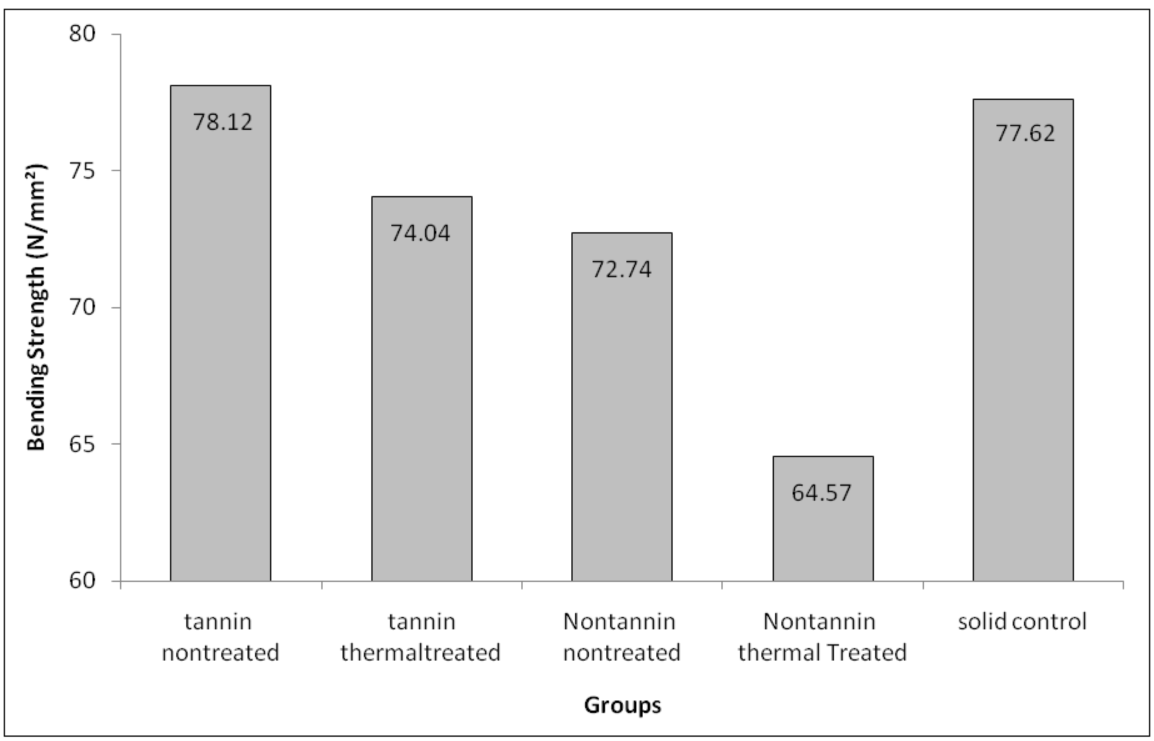

Figure 3. Bending strength of laminated chestnut wood composite

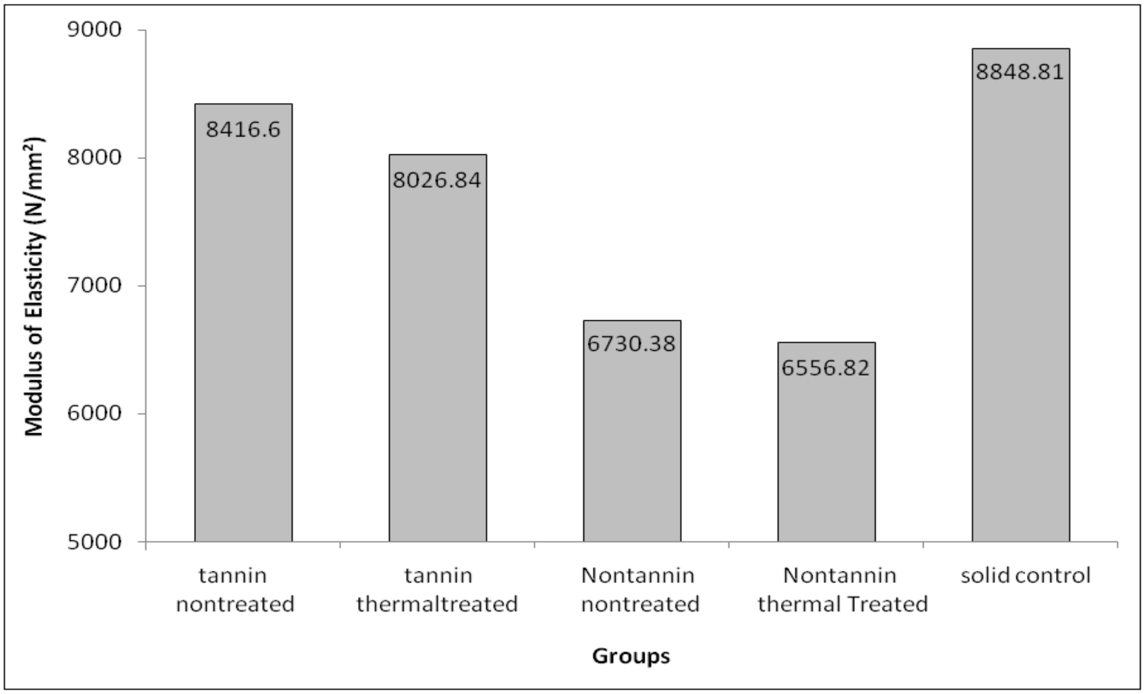

Figure 4. Modulus of elasticity (MOE) of laminated chestnut wood composite

Choong and Achmadi (1991) found that at high humidities, the extracted wood exhibits higher equilibrium moisture content than the unextracted wood. However, the isotherms of extracted and unextracted wood coincide at relative humidities below 70% for both desorption and adsorption. This phenomenon indicates that the removal of the extractives especially affects the sorption characteristic of wood at high relative humidity.

After removing tannin and thermal treatment, the highest and lowest value of bending strength were determined as 78.12 N/mm^2 for tannin/non-treated and 64.57 N/mm^2 for non-tannin/thermal-treated. Both removal of tannin and thermal treatment had an important effect on bending strength. After the laminating process, it was found that bending strength of laminated wood was increased and according to results mechanical properties of the samples was decreased at different ratios after tannin removal and thermal treatment. As compared to solid wood, the bending strength of laminated wood with tannin and non-treated was found to be higher (0.64%), and for tannin/thermal treated, non-tannin/non-treated and were found to be lower (4.61%, 6.28%, and 16.81%, respectively). According to Fig. 4, it was determined that the highest and the lowest value of MOE were 8848.81 N/mm^2 for solid wood and 6556.82 N/mm^2 for non-tannin/thermal-treated. The MOE decrease ratios of tannin/non-treated and non-tannin/non-treated were 4.88% and 23.94%, compared to solid wood and after also thermal treatment, tannin/non-treated and tannin/thermal-treated were found to be 4.88% and 9.28% respectively. Hence, tannin removal was found more effective than thermal treatment on bending strength and modulus of elasticity. Figure 5 shows the effect of removing tannin and thermal treatment on compression strength of tannin/thermal treated, tannin/non-treated, non-tannin/thermal-treated, and non-tannin/non-treated samples. According to the results obtained, both removal of tannin and thermal treatment had significant effects on compression strength. It was found that the highest and lowest values of compression strength were 58.22 N/mm² for tannin/non-treated samples and 46.54 N/mm² for non-tannin/thermal treated samples.

Grabner et al. (2005) found that extractives in the larch heartwood have a significant effect on bending strength and modulus of elasticity. Increasing extractive content goes hand-in-hand with better mechanical properties in the transverse direction. In the lateral direction, increasing of MOE values were observed with increasing extractive content, which is associated with the radially oriented, extractive filled tracheids rows: the rows of extractive-filled tracheids are frequently interrupted by single empty tracheids. Such empty tracheids limit compression strength by the critical buckling load of their cell walls (Grabner et al. 2005).

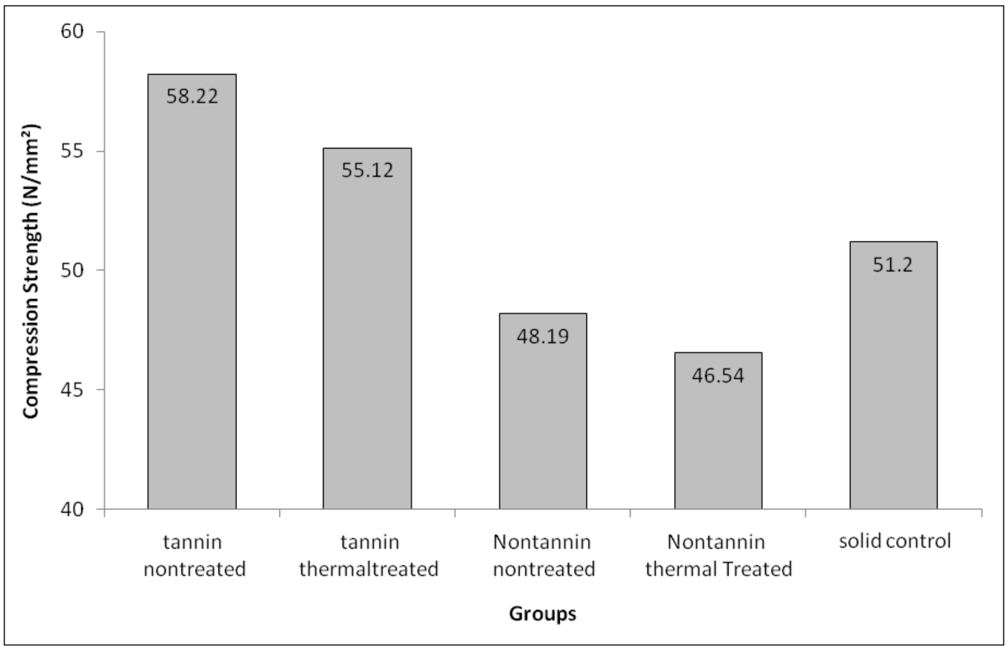

Figure 5. Compression strength of laminated chestnut wood composite

As seen in Fig. 5, compression strength of samples with tannin/non-treated and tannin/thermal-treated was 12.05% and 7.11% higher than solid control. For non-tannin/non-treated and non-tannin/thermal treated, compression strength was found to be 5.87% and 9.10%.lower than solid control specimens. As a result, it was found that the removal of tannin and thermal treatment had a significant effect on mechanical properties. Both heat treatment and removing of tannin decreased the density of chestnut wood. Since mechanical properties of wood depend on density, mechanical properties of laminated chestnut wood were significantly decreased. Compression strength (9.10%) was less affected than bending strength (maximum decrease (%) was 16.81% and modulus of elasticity (maximum decrease (%) was 25.90%) as compared to solid control.

Extractives in the heartwood had a significant effect on transversal compression strength and Young's Modulus. Increasing extractive content was obtained better mechanical properties in the transverse direction. The extractive arabinogalactan in larch has multiple effects on different aspects of wood quality, among which is lateral mechanical enforcement (Grabner et al. 2005).

Kuo and Arganbright (1980) and Grabner (2002) presented evidence for a direct influence of extractives on the modulus of rupture and the modulus of elasticity, in addition to their effect on wood density.

CONCLUSIONS

Chestnut wood, which contains tannin at high levels, has been widely used in making wooden yachts and boats. When chestnut is used in such applications, in order to increase adhesion between woods, tannin removal can be used in preparation of laminates for boat or yacht making. In this study, the effects of removing tannin and thermal treatment on physical and mechanical properties of chestnut wood were investigated. According to the results obtained, it was determined that removal of tannin from chestnut wood caused decrease in both kiln dry and air dry density.

Besides these density decreases, removal of tannin and thermal treatment decreased moisture content and swelling and shrinkage, thus improving the dimensional stability. According to the results obtained, density was affected more with removal of tannin and the maximum decrease of density was found in samples from which tannans had been removed before thermal treatment. The maximum decrease in swelling and shrinkage also were determined in samples in the non-tannin/thermal treated sample condition. But in moisture content, samples with tannin/thermal treated and non tannin/thermal treated were found to exhibit similar changes, with the maximum values decreasing and approaching each other.

After removal of tannin and thermal treatment, mechanical properties such as bending strength, modulus of, elasticity and compression strength decreased at different rates. Ratios of maximum decreasing were approximately 17% for bending strength (non-tannin and thermal treated samples), 26% for modulus of elasticity (non-tannin and thermal treated samples), and 9.10% for compression strength (non-tannin and thermal treated samples) as compared to solid control samples. As seen from the results, it can be said that removal of tannin affected all properties to a greater extent as compared to thermal treatment at 180 ^{o}C. According to the results of mechanical properties tests, improvements obtained with thermal treatment alone is sufficient for manufacture of boats with laminated chestnut wood. Therefore, it can be said that non-tannin and thermal treated chestnut samples are suitable for the construction of wooden yachts or boats. Improvements in physical properties caused by heat treatment and removal of tannins can be a positive factor.

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