SHEAR STRENGTH OF HEAT-TREATED TALI (*ERYTHROPHLEUM IVORENSE*) AND IROKO (*CHLOROPHORA EXCELSA*) WOODS, BONDED WITH VARIOUS ADHESIVES

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The aim of this study was to evaluate the effect of heat treatment on the shear strength of tali (*Erythrophleum ivorense*) and iroko (*Chlorophora excelsa*) woods, bonded with some structural adhesives. Shear strength of untreated and heat-treated woods bonded with phenol-formaldehyde (PF), melamine-urea-formaldehyde (MUF), melamine-formaldehyde (MF), and polyurethane (PUR) adhesives was studied. An industrial heat treatment method (ThermoWood) was used. The timbers were thermally modified for 2 hours at 180 ºC. Laminated samples having two sample sets were prepared from untreated and heat-treated wood for the shear strength test. The results of the tests showed that the heat treatment affected shear strength of laminated wood negatively. Although there was a considerable difference in adhesive bond shear strength between untreated and treated wood, both wood species bonded with the adhesives fulfilled the required value for shear strength of the adhesive bonds. PF, MUF, MF, and PUR adhesives performed in a rather similar way for both wood species.

Keywords: Heat treatment of wood; Adhesive; Shear strength; Phenol-formaldehyde; Melamine-urea-formaldehyde; Melamine-formaldehyde; Polyurethane

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INTRODUCTION

Wood is thermally treated in order to make it a biologically durable construction material, as well as to reduce equilibrium moisture content, to decrease water absorption, and to increase the dimensional stability (Kartal et al. 2007), but sometimes thermal treatment is used to change the aesthetic properties of wood. Wood colour becomes darker depending on treatment temperature, time, and techniques. This has opened new markets. Unfortunately, the mechanical properties, e.g., strength, hardness, and stiffness are reduced at the same time.

Chemical modification of wood components, especially hemicelluloses and lignin, occurring during heat treatment is mainly responsible for these new properties and could confer new reactivity to the material. As a consequence of chemical changes in wood's structure, the bonding performance of wood can change. The adhesion between wood and adhesive depends on a number of factors, including the wettability of the wood surface, the roughness, the penetration behavior, the moisture content, the presence of extractives, the hygroscopicity, and the chemical composition as well as the pH of the

wood. Wood, which is originally hydrophilic, becomes hydrophobic after heat treatment (Paul et al. 2007). This situation can alter the distribution of the adhesive on the wood surface and the penetration of the adhesive into the porous wood structure (Sernek et al. 2008). Also, the low hygroscopicity of heat-treated wood might affect the curing of water-borne glues (Boonstra et al. 1998). The wettability of heat-treated wood with water is decreased (Petrissans et al. 2003), which might hinder waterborne adhesives from adequately wetting the surface. Also, heat treatment results in a decrease in pH, which probably affects the curing process, depending on adhesive type used. On the other hand, the improved dimensional stability of heat-treated wood contributes to a better glue-ability of wood and at the end a better functionality of the construction with finger joints, because the stresses acting on the adhesive bond due to shrinking or swelling are reduced (Boonstra et al. 1998).

In the last decade several research groups developed heat treatment methods suitable for industrial applications. The temperatures and durations for heat treatment generally vary from (180 to 280) °C and 15 min to 24 h, depending on the heat treatment process, wood species, sample size, moisture content of the sample, and the desired mechanical properties, resistance to biological attack, and dimensional stability of the final product (Korkut et al. 2008). The "ThermoWood Process" was developed at the Technical Research Centre of Finland (VTT) in the early 90's. The ThermoWood process is based on heating the wood material for a few hours at high temperatures above 180 °C under normal pressure while protecting it with water vapour (Viitaniemi 1997). Water vapour protects the wood from burning and cracking, and it also affects the chemical changes taking place in wood (Viitaniemi 2000).

The heat-treated wood has a growing market in outdoor applications such as exterior cladding, window and door joinery, garden furniture, and decking. There are also many indoor applications for heat-treated wood such as flooring, paneling, kitchen furnishing, and interiors of bathrooms and saunas (Viitaniemi 2000). As a consequence of the loss of strength properties, heat-treated wood is not recommended for use in load-bearing construction (Viitaniemi 1997).

Tali (*Erythrophleum ivorense A. Chev.*) and iroko [*Chlorophora excelsa* (Welw.) Benth and Hook] woods are used generally for decking in buildings in Turkey. Colour modification is a primary reason for using thermally treated tali and iroko woods. Suppliers will be able to supply wood within a specified range of the colour spectrum, according to one vision of the future (Johansson and Morén 2006). Before this vision can be fulfilled, more research has to be done in the area of how process parameters affect the strength properties, for example shear strength. The bonding of heat-treated wood with adhesives would significantly increase the range of applications for this material, although heat treatment can affect the ability of adhesives to bond the wood (Sernek et al. 2007). With increasing use of heat-treated wood for exterior and interior applications, the concerns about bonding performance of heat treated wood bonded with various adhesives such as phenol–formaldehyde (PF) resin, melamine–formaldehyde (MF) resins, melamine-urea-formaldehyde (MUF), or polyurethane (PUR) has increased. The main objective of this study was to evaluate the shear strength performance of heat-treated tali and iroko woods with exterior structural adhesives that are used widely in woodworking industry.

MATERIAL AND METHODS

Materials

In this study, tali (*Erythrophleum ivorense* A. Chev.) and iroko [*Chlorophora excelsa* (Welw.) Benth and Hook] woods were selected as the materials of study. 10 tali and iroko planks (25 mm × 100 mm in cross section and 3 m long) were obtained from Nova ThermoWood in Gerede, Turkey. The planks had an initial moisture content of approximately 18%-20%. Prior to heat treatment each plank was cut into two 1.5 m long pieces from the middle, and due to this procedure the thermally modified and untreated samples were from the same planks. Then, the one half of these planks that were used for control samples were dried in industrially drying-kiln at approximately at a temperature of 70 ºC to a moisture content of 11%-15%. The other half of these planks were subject to the ThermoWood heat treatment process.

Phenol formaldehyde (PF), melamine-urea-formaldehyde (MUF 3%- MUF 20%), melamine formaldehyde (MF), and polyurethane (PUR) adhesives were used as adhesives. The ready-to-use PF, MF, MUF, and PUR adhesives were supplied from GENTAŞ and POLISAN, producer firms in Turkey, and their characteristics are given in Table 1.

Table 1. Characteristics of Adhesives Used

Adhesives	Density (20 ºC) (g/cm^3)	pH (20 ºC)	Viscosity (20 ºC) (cPs)	Solid (2 h, 120 ºC) (%)
MUF 3%	1.238	9.2	375	55.00
MUF 20%	1.279	9.3	620	64.40
MF	1.245	9.2	60	55.2
PF	1.201	11.0	380	46.32
PUR	1.110	7.0	550	100

Heat Treatment of Wood (ThermoWood)

The heat–treatment was applied according to the method described in the Finnish ThermoWood Handbook (2003). The ThermoWood process involves three distinct process stages: 1. warming up stage, 2. drying stage, and 3. cooling and conditioning stage. The warming up stage is to heat and pre-dry the lumber. The temperature in the kiln is raised rapidly, and a large amount of steam is generated. At the beginning of the drying stage, the temperature is increased steadily, and the timber is dried intensively. At a certain point of the drying stage when the moisture content of the lumber reaches nearly 0%, the temperature is raised rapidly to a range of 185 ºC to 215 ºC, depending on the applications of the treated products. The wood is kept at this temperature for 2 to 3 h. In the cooling and conditioning phase, the temperature of the wood is lowered to 80 ºC to 90 ºC using a water spray system. Conditioning is carried out to moisten the heat-treated wood and bring its moisture content to 4% to 7%.

Tali and iroko planks were heat treated at about 180 ºC under steam. The total time of the heat treatment was 63 h, and the duration time at this high temperature was 2 h. The heat treatment operation was carried out quite slowly in case there might have been a big risk of drying cracks. After heat treatment only the planks that were free of defects were selected for further testing.

Moisture Content and pH Determination

The native (untreated control) and heat-treated planks were cut into sample sets and conditioned in a standard climate with 65% relative humidity (RH) and at a temperature of 20 ºC. The moisture content (MC) of the specimens was determined by the gravimetric method. The MC of the untreated and heat treated rows prior to bonding were 9.6 % and 6.6 % for tali and 11.8 % and 9.7 % for iroko, respectively.

The pH value was evaluated by using an extraction method; 20 g of wood was ground into small particles and soaked in 160 g of distilled water for 24 hours. The extract was filtered and analyzed with a pH meter (Sernek et al. 2008). The pH of the untreated and heat treated rows were 3.90 and 4.04 for tali and 5.45 and 6.42 for iroko, respectively.

Specimen Preparation and Property Testing

The sample sets were planed to a thickness of 5 mm, then bonded together into small samples. The adhesive was applied at the rate of about 180-200 g/m^2 on a single bonding surface of the rows (single glue line) as recommended by the manufacturer. Glues were spread uniformly on the veneers by manually hand brushing. The press pressure, temperature, and duration were applied as 2 kg/cm^2, 120 ºC, and 15 min for PF, MF, and MUF adhesives, and 2 kg/cm^2, 22 ºC, and 90 min for PUR, respectively. In total, 100 samples were bonded (2 groups of wood, 5 adhesives, and 10 duplicates). The samples were tested after being conditioned for 10 weeks at 20±2 °C and 65±3 % relative humidity.

The measurement of shear strength was carried out in a Zwick/Roel Z50 universal testing machine, according to BS EN 205 and TS EN 12765. A test sample is diagramed in Fig. 1.

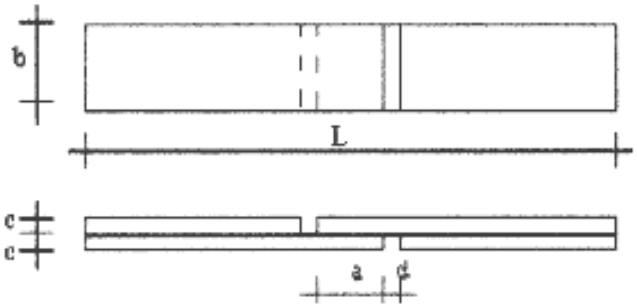

Fig. 1. Shear strength test sample (a, 10 mm; b, 20 mm; c, 5 mm; d, 3 mm; L, 150 mm)

The loading was carried out until a break or separation occurred on the surface of the test samples. The shear strength (σ_k) was calculated using the observed load (F_{max}) and bonding surface area of the sample (A, mm^2) according to the following formula (1),

$$\sigma_k = \frac{F_{\max}}{A} = \frac{F_{\max}}{ab}\left(N.mm^{-2}\right) \quad (1)$$

where a is the width of the glued surface (10 mm) and b is the length of glued surface (20 mm).

RESULTS AND DISCUSSION

Measured equilibrium moisture content (EMC) and density values of untreated and heat treated samples bonded with the adhesives prior to shear test are given in Table 2.

Table 2. The EMC and Density Values of Untreated and Heat-Treated Samples Bonded with the Adhesives Prior to Shear Test

	Adhesives	Untreated control		Heat treated	
		Density (g/cm^3)	EMC[a] (%)	Density (g/cm^3)	EMC[a] (%)
Tali	MUF 3%	0.898	7.4	0.864	6.0
	MUF 20%	0.888	7.8	0.852	5.7
	MF	0.897	7.1	0.841	5.1
	FF	0.893	7.9	0.878	5.3
	PUR	0.895	8.4	0.874	6.0
Iroko	MUF 3%	0.674	8.7	0.559	4.5
	MUF 20%	0.686	8.6	0.545	5.5
	MF	0.716	8.3	0.558	5.3
	FF	0.686	8.3	0.578	4.6
	PUR	0.699	8.3	0.583	5.4

[a] EMC, equilibrium moisture content

Heat treatment caused a decrease in the EMC and density of the samples. The availability and/or accessibility of the free hydroxyl groups of the wood carbohydrates play an important role in the process of water adsorption and desorption. The heat treatment changes the chemical structure of the wood, especially in the case of hydroxyl groups. Hence, reduction of water absorption after heat treatment is most probably due to depolymerisation of the carbohydrates and especially hemicelluloses, resulting in a reduction of the total amount of hydroxyl groups, including the free hydroxyl groups (Tjeerdsma et al. 1998). Therefore the EMC values of the heat-treated samples were less than untreated samples. Due to heat treatment and thermal degradation, wood loses mass (Kortelainen 2006). This causes a decrease in density of wood.

The mean values and standard deviations of the shear strength values of untreated and heat-treated samples bonded with the adhesives and the decrease in the shear strength values with the heat-treatment are shown in Table 3.

According to variance analysis results, the shear strength of tali woods bonded with adhesives was higher than that of iroko wood bonded with adhesives and the Duncan's test showed that the difference was significant. Because of this, the wood species were evaluated separately. The observations confirm the data found in the literature on this subject. In general, it can be said that differences in shear strength between the wood species are due to the inherent shear strength of each wood species and the effect of heat treatment which might differ for each species (Boonstra et al. 2007). We can explain the reason for this result by considering the density of each wood species (Table 2).

Table 3. Shear Strength and the Percentage of Wood Failure of Untreated and Heat-Treated Tali and Iroko Woods Bonded with the Adhesives

Wood Species	Adhesives	Shear strength (N/mm^2) Untreated Mean*	SD	Wood Failure (%)	Heat-treated Mean*	SD	Wood Failure (%)	Decrease* (%)
Tali	MUF 3%	5.45	1.46	98	5.03	1.04	80	-7.6
	MUF 20%	6.20	1.27	94	5.72	1.16	75	-7.8
	MF	5.86	1.52	86	5.33	1.02	74	-9.1
	PF	6.41	1.01	90	5.83	1.20	72	-9.1
	PUR	6.54	1.54	100	5.97	1.05	76	-8.8
Iroko	MUF 3%	4.57	0.43	96	4.17	0.55	78	-8.9
	MUF 20%	4.71	0.50	98	4.42	0.86	75	-6.3
	MF	4.67	0.77	85	4.33	0.94	70	-7.3
	PF	4.85	0.74	92	4.46	0.52	72	-8.2
	PUR	5.04	1.01	98	4.73	0.36	78	-6.2

*, Average value of ten replicates; SD, Standard deviation.

The shear strength of laminated wood bonded with all the adhesives used for both wood species was decreased by the heat-treatment. The decrease of shear strength after heat-treatment can be explained by considering a combination of factors. First, heat treatment can alter the distribution of the adhesive on the wood surface and its penetration into the porous wood structure, because the wood, generally hydrophilic, became hydrophobic after heat treatment (Paul et al. 2007). The decrease in hygroscopicity has been related to a decrease in the number of hydrophilic sites in wood, especially the hydroxyl groups of carbohydrates (Nakano and Miyazaki 2003). With the degrading of carbohydrates after heat treatment, the concentration of water-absorbing hydroxyl groups decreases, resulting in slow water uptake and absorption. The plasticization of lignin and the reorganization of the lignocellulosic polymeric component of wood were also proposed as other explanations for increased hydrophobic characteristics of heat-treated wood. Second, the wettability of wood with water is decreased after heat treatment (Sernek et al. 2008; Petrissans et al. 2003; Follrich et al. 2006; Sernek et al. 2004; Gerardin et al. 2007), mainly because the surface of the heat-treated wood is hydrophobic, less polar, and significantly repellent to water. Hakkou et al. (2005) reported that heat-treated wood showed conformational modifications of wood polysaccharide components, leading to lower wettability of wood. Wettability modification during heat treatment could be explained by a modification of conformational arrangement of wood biopolymers due to loss of residual water or more probably to plasticisation of lignin. This might hinder waterborne adhesives from adequately wetting the surface (Sernek et al. 2008). Third, heat treatment reduces the pH of wood. The increase in wood acidity is due to the formation of acetic acid, which is liberated from the hemicelluloses, and which further catalyses carbohydrates cleavage,

causing a reduction of degree of polymerization of the carbohydrates (Tjeerdsma et al. 1998; Windeisen et al. 2007; Boonstra et al. 2007).

According to the results of the present study, the heat treatment reduced the pH from 4.04 to 3.90 for tali and from 6.42 to 5.45 for iroko. Changes in pH affect the hardening process of adhesives, depending on the adhesive type. Low pH conditions might neutralize the alkaline hardener of PF. Decrease of pH might accelerate the condensation reactions of amino resins. This accelerated condensation hinders the penetration of the adhesive into wood. The results of this study suggest that the heat-treatment decreased the pH of wood slightly; therefore the shear strength of adhesives was not affected considerably by decrease of pH.

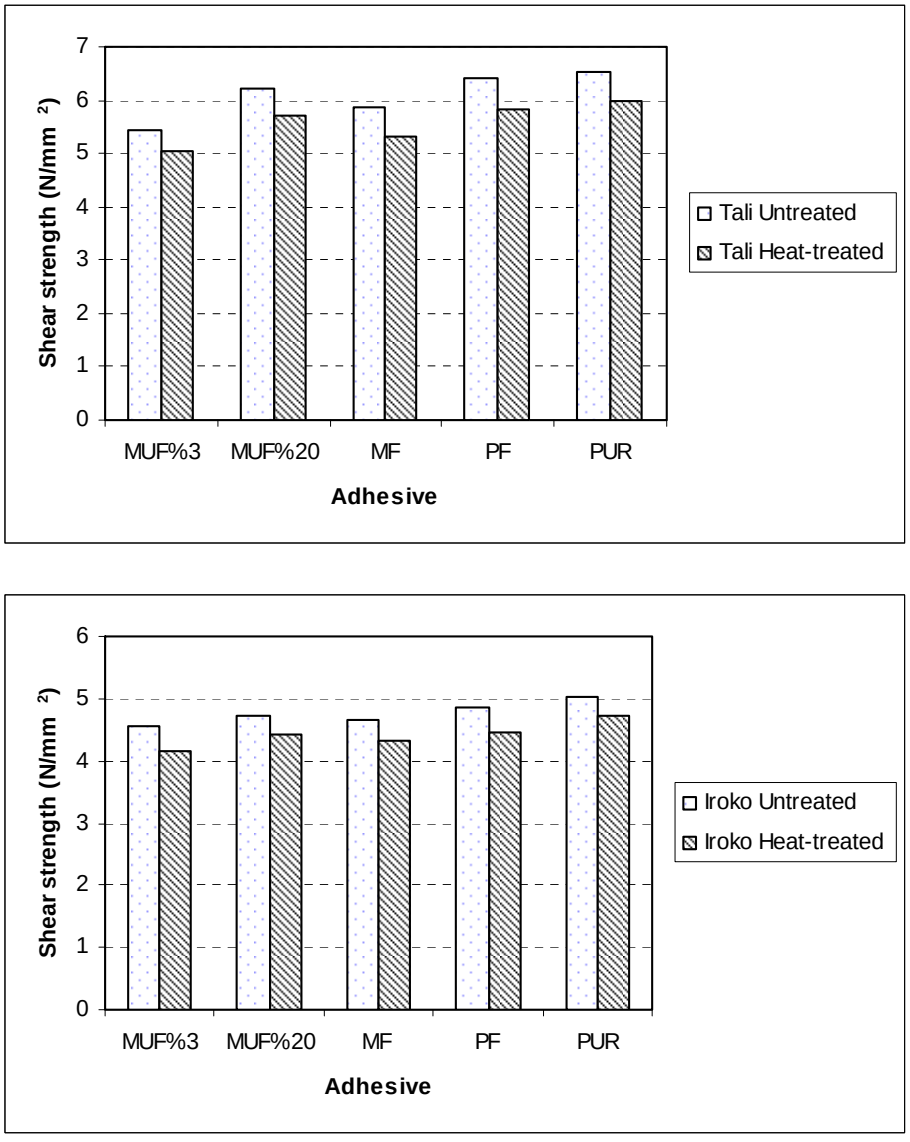

Fig. 2. Shear strength of untreated and the heat-treated tali and iroko bonded wood according to the adhesives

The data were further analyzed using an analysis of variance, followed by application of the Duncan test at the 95% confidence level. When the shear strength of specimens bonded with the adhesives compared, it was obtained that there was no difference between the adhesives for both untreated and heat-treated specimens. In the case of heat-treated specimens, the percentage of wood failure was high, which shows that the wood itself was the weakest part of the system (Table 3). This indicates that heat treatment decreased the shear strength of wood itself rather than that of the adhesive bond. When comparing the shear strength of bonded wood in terms of the adhesives, the highest shear strength values were obtained from the samples bonded with PUR adhesive, and the lowest from MUF 3% adhesive (Fig. 2).

In this study, the decreases in shear strength depending on the adhesives used were relatively similar (Table 3). Sernek et al. (2007) reported a decrease in shear strength of 13% for spruce bonded with PF and UF, each by heat treatment at 210 °C for 2 hours. In another study Sernek et al. (2008) reported that shear strength was 6.28 N/mm^2 for untreated spruce bonded with MUF and 4.86 N/mm^2 for heat treated spruce, a decrease of 23%. The decreases that occurred with heat treatment in our study for iroko and tali woods were relatively low. But these results are not directly comparable, properly because of variation in heat treatment method and especially due to the different wood species used in these studies.

CONCLUSIONS

The results of the tests carried out in this study revealed that the heat treatment reduced the equilibrium moisture of content, density, and shear strength of tali and iroko wood bonded with PF, MF, MUF, and PUR adhesives. The decreases in shear strength according to adhesives used were relatively similar. Considering the all adhesives used, whereas bonding with PUR adhesive resulted in the highest shear strength for the heat-treated tali and iroko woods, bonding with MUF 3% adhesive resulted in the lowest. Because there was no statistically significant difference between the samples prepared with different adhesives, all of them can be used for bonding the heat-treated wood. PF adhesives can be preferred in exterior applications because of their relatively low cost relative to the other exterior grade adhesives. In addition, PUR adhesive can be preferred in exterior applications for assembly purposes due to its ease of use.

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