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ORIGINAL PAPER

Response of colour and hygroscopic properties of Scots pine wood to thermal treatment

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Abstract: The effect of heat treatment on the surface colour and hygroscopic properties of pine wood were investigated in this study. Boards of Scots pine wood (*Pinus sylvestris* L.) were subjected to thermal treatment at 200ºC, for 4, 6, and 8 h. The change of equilibrium moisture content and density values of the specimens in order to facilitate the understanding of the treated material behavior. The colour parameters L^*, a^* and b^*, used to depict the total colour change (ΔE) of wood surface, were shown to change proportionally to the treatment intensity. Moreover, swelling in the tangential and radial directions and absorption of the specimens appeared to be enhanced in great extent by the thermal treatment process. The mean value of swelling percentage in the tangential direction decreased 10.26%, 17.22%, and 19.60% for specimens treated for 4, 6, and 8 h, respectively, referring to the final measurement after 72 h of immersion. In radial direction, mean value of swelling percentage decreased 19.56%, 32.75%, and 34.65% for treated for 4, 6 and 8 h, respectively, after 72 h immersion, which attests the decrease in swelling and improvement in the hygroscopic behavior of Scots pine wood.

Keywords: Colour; Hygroscopic properties; Modification; Thermal treatment; Scots pine

Introduction

The beneficial influence of heat treatment on wood has been acknowledged from the antiquity, but only two decades ago was the time for this environmentally friendly method of wood preservation to find wide commercial acceptance in large-scale furniture production. Thermally treated wood is considered to be an important alternative to chemically treated wood or wood preserved with fungicidal substances and naturally durable wood species, as well.

Thermal treatment modifies the chemical composition of wood and therefore, its physical and mechanical properties. Several changes occur: dimensional stability and biological resistance against fungi and microorganisms attacks tend to improve, equilibrium moisture content (EMC) and density decrease, emissions of volatile organic compounds (VOC) decrease, resistance to natural weathering and wettability enhances, and colour uniformity and stability is achieved (Awoyemi and Jones 2010).

The colour of treated wood tends to darken and become more uniform and stable. These characteristics attach additional aesthetic value to the material, as consumers seem to favor wood species of darker colours, due to their resemblance to tropical species. The phenomenon of darkening is derived mainly from the considerable changes in the chemical composition of wood, such as the degradation of the amorphous carbohydrates and extractives modification.

Some of the main conversions, recorded in wood mass during thermal treatment are the decomposition of hemicelluloses that degrade first among the wood polymers, due to their high reactivity. Intensifying the treatment (by either temperature or duration) softens lignin and tends to start ramification, creating an even more complex network which tightly combines the polysaccharides chains. The amorphous regions of cellulose degrade, increasing the crystallinity of cellulose, which contributes to the emergence of a more hydrophobic character of wood. Additionally, the hydroxyl groups content seems to diminish.

Some of the most crucial properties of wood are, to a great extent, enhanced by the thermal treatment, but some of the mechanical properties are weakened, such as bending strength, impact bending strength, and tensile strength. Therefore, during thermal treatment many factors should be taken into careful consideration, such as the kind of wood species, the maximum treatment temperature, the duration of exposure to the maximum treatment temperature, the heating rate, the treated boards' dimensions and moisture content, kiln atmosphere (gas humidity, existence of shielding gas or oxygen, etc.), economics of the process, potential energy-saving perspective, and desired final properties. The large number of variable factors explains the

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existence of many different treatments, which offer the opportunity to produce a material appropriate for different applications.

Extensive research has been done on the influence of thermal treatment on the physical and hygroscopic properties of wood. Some of the most significant research regarding to colour change are as follows: Akgül and Korkut (2012) examined the effects of heat treatment at 120ºC, 150ºC, and 180ºC for 2, 6, and 10 h on the colour and chemical composition of Scots pine. Uludag fir. Sahin et al. (2011) analysed the colour evolution and changes using small specimens of heat-treated Red-Bud Maple, European Hophornbeam, and Quercus petraea ssp iberica using the CIE L*a*b* colour space. Nuopponen et al. (2003) evaluated the effect of heat treatment on the colour and the behaviour of extractives, referring to fats, resin acids and waxes, of Scots pine studied by FTIR spectroscopic methods. The heat treatment was applied at 100, 120, 140, 160, 180, 200, 220 or 240 ºC under saturated steam, while the time of the maximum temperature was 3 hours. Johansson (2005) studied the strength and also the colour response of wood to heat treatment at 130°C injected into the kiln at atmospheric pressure, for 2–4 h and tried to use colour as a predictor of strength.

Furthermore, Stingl et al. (2007) applied spruce wood to 3 hours of heat treatment at 215 °C, in order to test the greying and colour changing of wood surface, durability, and stability and the effects of erosion products of heat-treated wood on other materials. Ahajji et al. (2009) treated beech and spruce wood at 210 ºC, 235 ºC, and 250ºC for 1 hour and then examined the influence of the process on antioxidant properties, colour stability, and their extractives. They used wood extraction, estimation of total phenols, ESR analysis, colour measurement and ageing test. Fan et al. (2010) examined the discolouration and colour responses of black locust to solvent extraction and heat treatment at 120ºC for 24 h, using vacuum drying of the heated samples prior to measurement of colour parameters.

The hygroscopic properties of thermally treated wood have been the subject of much research. Tremblay (2007) applied the Finnish ThermoWood process at three temperature levels, including Thermo-S (190ºC), Thermo-D (212ºC) treatment classes and 230ºC, on Canadian jack pine, in order to examine its dimensional stability, hardness, resistance to fungal decay, impact, and static bending (MOE and MOR). Esteves et al (2008) treated Maritime pine wood in the presence of air at 170–200ºC for 2–24 h. Strength, wettability, mass loss due to thermal treatment, hemicellulose content, and hygroscopic properties were evaluated. The aim of the current study is to improve the quality of Scots pine wood by altering the hygroscopicity and colour of wood surface. The effect of heat treatment at 200ºC on the properties of this wood species were examined, in order to comprehend the process of thermal treatment and the material's responses to different thermal treatment durations.

Materials and methods

The experiment was carried out with Scots pine (*Pinus sylvestris* L.) wood, of Greek origin, obtained from a local wood industry in Drama prefecture (North Greece) that had been naturally desiccated for 8 months. The boards were cut parallel to the grain and the dimensions of the boards intended for thermal treatment were of 35mm thickness × 70mm width × 400mm length. Prior to treatment, the boards were placed into an conditioned room at 20±2ºC temperature and 60±5% relative humidity and were kept there until they reached a nominal equilibrium moisture content (EMC) of 11.63%. That is a sufficiently limited moisture content that it will help protect the wood from stress generation and the resultant splitting and distortion during the treatment. The mean density (mass/volume, measured at 11.63% moisture content) of the pine wood before treatment was measured as 0.505 g/cm^3.

For the thermal treatment process, a laboratory heating unit (80cm × 50cm × 60cm) with two different thermometers was used, a conventional zinc one, incorporated in the unit, and a digital thermometer with a temperature sensor inside the drying oven. The temperature applied during the thermal treatment was constantly 200ºC, while the treatment was implemented under atmospheric pressure environment, in the presence of air. The boards placed in the kiln had 11.63% moisture content and the interior of the kiln was preheated to 200ºC. The time periods of thermal treatment of the boards were of 4, 6 and 8 h and for each treatment 10 boards were used.

At the end of each treatment, samples were cooled down in desiccators and afterward stored in a climate-controlled room. After a two-month conditioning period at 20±2ºC temperature and relative humidity of 60±5%, EMC, and the density of the specimens were estimated. Afterward, the boards were visually evaluated for cracks, twists, and other deformations and only those boards that were free of defects were selected for further hygroscopic property tests and colour change measurement. These boards were cut in final cross section dimensions for the measurement of properties, according to the respective standards (Table 1).

Table 1. Wood properties studied and the respective standards

Property	Dimensions (cm)	Standard
Density (basic)	2 × 2× 2.5	ISO 3131:1975
Moisture content	2 × 2 × 2.5	ISO 3130:1975
Radial and tangential Swelling	2 × 2× 3	ISO 4859:1982

Swelling (in the tangential, radial, and longitudinal directions) and absorption percentage measurements were conducted after the samples were immersed in water of 20±3ºC temperature for 1, 3, 6, 24, and 72 h, in order to examine the rate of swelling.

The surface colour of the specimens was measured using a Minolta Colourimeter, to evaluate the changes owing to heat modification. The Colourimeter specifies the colour as three coordinates in three-dimensional colour space. This system is called CIE *L*a*b**, works according to the CIE standard, and provides a standard scale for comparison of colour values. The *L** coordinate describes lightness and ranges between 100, which represents a perfect reflecting diffuser, and 0, which represents black, and *a** and *b** describe the chromatic coordinates on the green–red and blue–yellow axes, respectively, without specific numerical limits. The three colour coordinates, L^*, a^*, and b^*, were recorded before and after each thermal treatment and the values were used to calculate the total colour difference (*ΔE*), the metric Chroma (*C**) and the Saturation (*ΔC**). The equations used for the determination of these parameters are the following (ASTM D 1536-58 T 1964):

$$\Delta L^* = L^*_{ht} - L^*_{ut} \tag{1}$$

$$\Delta a^* = a^*_{ht} - a^*_{ut} \tag{2}$$

$$\Delta b^* = b^*_{ht} - b^*_{ut} \tag{3}$$

$$\Delta E = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{1/2} \tag{4}$$

$$C^* = (a^{*2} + b^{*2})^{1/2} \tag{5}$$

$$\Delta C^* = C^*_{ht} - C^*_{ut} \tag{6}$$

where L^*_{ht}, a^*_{ht}, b^*_{ht}, and C^*_{ht} refer to the corresponding values of heat-treated specimens, while L^*_{ut}, a^*_{ut}, b^*_{ut}, and C^*_{ut} correspond to the values of untreated specimens (control). Therefore, ΔL^*, Δa^* and Δb^* represent the changes between the untreated and treated specimen values.

Results and discussion

Our results show that heat treatment of all three durations of time appeared to enhance dimensional stability of the Scots pine wood specimens (Table 2). Also, as the treatment duration increased, the swelling percentage value of the specimens tended to decrease, in both the tangential and radial directions.

Table 2 Mean value of swelling percentage in tangential and radial direction after 1, 3, 6, 24, and 72 hours of treatment

Treatment	Tangential swelling					Radial swelling				
	1h	3h	6h	24h	72h	1h	3h	6h	24h	72h
Control	2.35	3.11	3.68	4.23	4.54	1.50	2.23	2.70	3.40	3.54
4h	1.11	2.14	2.81	3.73	4.07	0.84	1.51	1.99	2.67	2.85
6h	1.02	1.73	2.37	3.43	3.76	0.64	1.07	1.50	2.07	2.38
8h	0.74	1.26	1.89	3.24	3.65	0.60	1.00	1.41	2.02	2.32

In the tangential direction, the 4-hour treated specimens recorded a decrease in the swelling percentage that ranged between 10.26% and 52.76%, while the 6-hour-treated specimens marked swelling decrease of between 17.2% and 56.6% and the 8 hour treated specimens presented a swelling decrease of between 19.6% and 68.5%, referring to all the measurements recorded values (1, 3, 6, 24, and 72h).

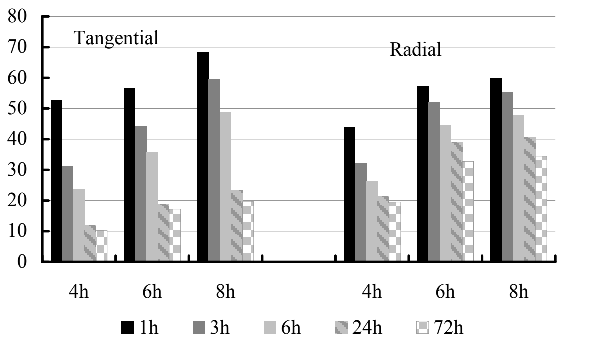

Fig. 1 Decrease percentage of swelling percentage value of tangential and radial direction of the specimens, after 1, 3, 6, 24, and 72 hours of treatment.

Generally, the percentage values of swelling in the tangential direction were higher than the corresponding values of swelling in the radial direction and the larger decrease in swelling was recorded in radial direction of pine specimens (Fig. 1). Specifically, in radial direction, the 4 hour treated specimens recorded a swelling decrease that ranged from 19.5% to 44%; the 6 hour treated specimens marked a decrease of between 32.76% and 57.3%; while the 8 hour treated specimens showed a swelling decrease that fluctuated between 34.5% and 60%, taking into account all the measurements values (1, 3, 6, 24 and 72h). The decrease in swelling in both the radial and tangential direction of the specimens reveals the improvement of hygroscopic behavior of pine wood.

Table 3 Mean values of absorption percentage of the specimens

Treatment	Absorption				
	1h	3h	6h	24h	72h
Control	18.91	25.21	31.55	45.06	61.24
4h	14.11	21.48	26.84	42.10	60.76
6h	13.35	19.59	24.76	39.79	57.24
8h	10.25	15.03	19.13	33.20	50.48

According to Table 3, the heat-treated specimens demonstrated a decrease in the absorption percentage value, which was proportional to the treatment duration increase. The mean value of absorption percentage after thermal modification decreased by 0.77%, 6.52%, and 17.57% for the specimens treated for 4, 6 and 8 h, respectively. What this shows is that an improvement of dimensional stability of Scots pine wood can be accomplished using a relatively short thermal treatment duration of 4 to 6 h at 200ºC.

The EMC of all heat-treated samples decreased in relation to the initial untreated wood EMC, even for the less intensive treatment of 4 hours at 200ºC. More specifically, the average EMC value of untreated pine wood specimens was 11.63%, while after the thermal treatment and a conditioning period of four weeks, the equilibrium moisture content value of 4 and 6 h treated specimens were found to be 5.82% and 5.68%, respectively and 5.48% for 8 h treated specimens. This clearly suggests that thermal treatment greatly affects the dimensional stability and absorption capacity of wood. The EMC value reduction is related to the mass loss and the hydroxyl group loss that occurred during the thermal treatment.

Furthermore, thermal treatment appeared to cause a decrease in the density of wood specimens. Specifically, the density of the treated specimens was decreased from 0.505 g/cm^3 to 0.412 g/cm^3 for specimens treated for 4 hours at 200ºC, to 0.411 g/cm^3 for 6 hour treated specimens and 0.409 g/cm^3 for 8 hour treated specimens, which correspond to decreases of 18.41%, 18.61%. and 19%, respectively. The decrease in density is related both to the moisture content decrease after treatment that was just mentioned and to the mass loss caused by thermal modification process, which also affects the mechanical properties of treated wood.

Similar results were recorded by Gunduz et al. (2008) who studied the effects of heat treatment on the physical properties of Camiyanı Black Pine wood and found that density and the swelling percentage decreased with increasing heat treatment time and temperature.

Observing the colour parameters measured before and after the heat treatment of the specimens, one can see that *L** parameter ("Lightness") tends to decrease, with the increasing of treatment time period and this applies to the three directions of the specimens (tangential, radial, longitudinal) (Fig. 2). This fact indicates that many components absorbing visible light are formed during heat treatment (Chen et al. 2012). In contrast, the *a** parameter records a slight increase during the treatment of 4 hours, but it tends to decrease again, more or less in the level of untreated specimens, as the treatment duration increases. Parameter *b** demonstrates a slight increase as the duration increases and this tendency appears to be similar for tangential, radial, and longitudinal directions of the specimens.

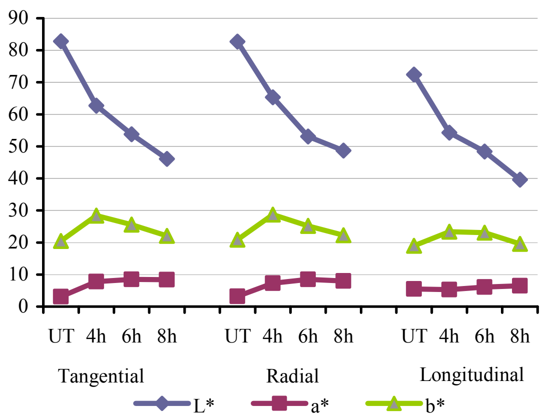

Fig. 2 The change of mean values of colour parameters *L, *a** and *b** in tangential, radial and longitudinal direction of Scots pine specimens**

Obviously, a rapid decrease in *L** occurs early in the heat-treatment process, where the largest change can be found between 0 and 4 h of treatment, which indicates that a short period of time is quite enough for altering the wood surface colour by heat. Using the colour parameters *L*, a** and *b*,* the total colour difference (*ΔE*) was calculated for each direction of the specimen, representing the overall colour changes of the samples in comparison to the same measurements of control samples. The same tendency of parameters *L*, a** and *b** was recorded by Aksou et al. (2011) who treated Scots pine in an oven for 2, 4, and 8 h at 150, 175, and 200ºC.; by Akgül and Korkut (2012), who measured the change in colour of the Scots pine specimens after thermal treatment at 120, 150, and 180ºC for 2, 6 and 10 h and by Sahin et al. (2011) who subjected three different wood species to thermal treatment under the same conditions.

Thermal treatment processes were proven to strongly modify surface colour with overall colour differences (*ΔE*) between the raw and treated specimens that ranged between 18.94 and 37.14 (Table 4). As was expected, *ΔE* increased proportionally to treatment duration increase. This decrease of luminance (darkening) on the wood surface could be justified by the formation of hemicelluloses and extractives thermal degradation products or possibly attributed to lignin polymerization reactions during treatment.

Table 4 Mean value of Total Colour Difference (*ΔE*) and Saturation index (*ΔC) of the treated specimens, measured in tangential, radial, and longitudinal directions**

Treatment	Direction	*ΔE*	*C**	*ΔC**
Control	Tang.	-	20.77	-
	Rad.	-	21.18	-
	Longit.	-	19.80	-
4h	Tang.	22.36	29.51	8.74
	Rad.	19.61	29.64	8.46
	Longit.	18.94	24.03	4.22
6h	Tang.	31.52	27.01	6.24
	Rad.	30.09	26.67	5.49
	Longit.	24.62	23.97	4.16
8h	Tang.	37.14	23.64	2.87
	Rad.	34.39	23.68	2.49
	Longit.	32.82	20.67	0.87

Noticeable is the fact that in tangential direction the higher colour difference values were marked, whereas the radial direction followed with quite lower total colour difference values and the lowest values of colour difference were recorded in longitudinal direction. Heat treatment has also an obvious effect on colour saturation (*ΔC**). As the treatment temperature increases, *ΔC** value demonstrates a decrease and additionally, referring to each of the treatments, the higher *ΔC** values were recorded in tangential direction, followed by the corresponding values of radial and finally, longitudinal direction. The decrease in colour saturation (*C**) values is mainly attributed to the changes of *a** and *b** values owing to thermal treatment.

Pine wood contains large quantities of resin and extractives, that tend to move toward the surface of the wood specimen and spread during the thermal treatment, which attaches an undesirable apearance to the wood surface (Fig. 3). Fortunately, discoloration of the wood surface formed due to resin release proved to be easily removed with sanding, for it is superficial and does not influence the colour of wood in deeper levels or other properties such as hygroscopic properties.

Figure 3. Release of resin on the wood surface during thermal treatment. A. Wood surface with released resin from a resin pocket before sanding, B. The same wood surface after sanding

Conclusions

The main objective of this work was the examination of hygroscopic properties and the colour change of Scots pine wood when thermally treated at 200ºC for 4, 6, and 8 h in the presence of air. Our findings showed that as the treatment intensity increases, the density and the equilibrium moisture content (EMC) values of wood decreases. The swelling and absorption percentage values of the specimens clearly decreased, which demonstrates the enhanced dimensional stability and hygroscopic properties of the treated specimens. Thermal treatment of 8 h at 200ºC resulted in the most severe changes to physical properties, referring to enhancement of hygroscopic properties, colour darkening, EMC, and density loss.

Colour measurements of thermally treated specimens revealed a decrease in L^*, increase in a^* and b^* parameter values, and total colour difference value (ΔE) of the samples, as well. These changes depicted the tendency of wood surfaces to darken, approximating more desirable colour tones and therefore, enhancing the appearance of the final material. Consequently, heat treatment methods may improve some of the most crucial properties of pine wood, like swelling and absorption, widening the application range of the material and thus, enabling pine wood to compete with other wood species of even higher quality.

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