Bartın Orman Fakültesi Dergisi
2012, Cilt: 14, Sayı: 22, 96-103
ISSN: 1302-0943
EISSN: 1308-5875

RESPONSE OF COLOUR AND HYGROSCOPIC PROPERTIES OF SCOTS PINE WOOD TO THERMAL MODIFICATION

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ABSTRACT

The effect of heat treatment on wood 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 200 °C, for 4, 6 and 8 hours. The change of equilibrium moisture content and density values of the specimens, due to the thermal treatment were recorded, 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 proved to change proportionally to the treatment intensity. Moreover, the swelling in tangential and radial direction and the absorption of the specimens as well, appeared to be enhanced in great extent by the thermal treatment process. Mean value of Swelling percentage in tangential direction decreased 10.26%, 17.22% and 19.60% for specimens treated for 4, 6 and 8 hours, respectively, referring to the final measurement after 72 hours 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 hours specimens, respectively, after 72 hours immersion, which attests the swelling decrease and the improvement of the hygroscopic behavior of Scots pine wood.
Key words: Colour, Hygroscopic properties, Modification, Scots pine, Thermal treatment

1. INTRODUCTION

The beneficial influence of heat treatment on wood is acknowledged for almost a century, but only 2 decades ago was the time for this environmentally method of wood preservation to find wide acceptance from the industrial world, the consumers and consolidate a position in large scale furniture production. Thermally treated wood gained its place in the wood market and is considered to be an important alternative to chemical treated wood or wood preserved with fungicide substances and naturally durable wood species, as well.

According to previous researches, thermal treatment modifies the chemical composition of wood and therefore, its physical and mechanical properties. As it is evident, the dimensional stability and biological resistance against fungi and microorganisms attacks tend to improve, the equilibrium moisture content (EMC) and density decrease, emissions of volatile organic compounds (VOC)

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Makale metni 25.11.2012 tarihinde dergiye ulaşmış, 07.12.2012 tarihinde basım kararı alınmıştır

decrease, resistance to natural weathering and wettability enhances, while colour uniformity and stability is achieved (Awoyemi and Jones 2010).

The colour of the specimens tends to darken and become more uniform and stable, due to thermal treatment. 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 of high mechanical properties and durability. 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.

There is a high interest in the research field of wood modification and extensive research has been done so far evaluating the influence of thermal treatment on the colour change of wood (Akgül and Korkut, 2012; Sahin et al., 2011; Nuopponen et al., 2003; Johansson, 2005; Stingl et al., 2007; Ahajji et al., 2009; Yongming et al., 2010)

Hygroscopic properties test of thermal treated wood was the subject of many researches (Tremblay, 2007; Esteves et al., 2008; Vukas et al., 2010).

The aim of the current study is to improve the quality of Scots pine wood by heat treatment at 200 °C, understanding the process of thermal treatment and the material's responses to different treatment durations, referring to hygroscopic properties and colour of the wood surface.

2. MATERIALS AND METHODS

Experiment was carried out with Scots pine (*Pinus sylvestris* L.) wood, of Greek origin, obtained from a local wood industry of Drama prefecture (North Greece) and it has been naturally desiccated for 8 months. The boards were cut parallel to grain and the dimensions of the boards, intended for thermal treatment, were of 35 mm thickness x 70 mm width x 400 mm length. Prior treatment, the boards were placed into a conditioned room at 20 ± 2 °C temperature and 60 ± 5% relative humidity and were allowed there to reach a nominal equilibrium moisture content (EMC) of 11.63%. That is a limited enough moisture content that contributes to the protection of wood from stress generation and its 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 treatment process, a laboratory heating unit (80cm x 50cm x 60cm) with 2 different thermometers was used, a conventional zinc one, incorporated in the unit and also, a thermometer of digital indication with temperature sensor inside the drying oven, and therefore, the unit was capable of controlling the temperature within a range of ± 1 °C. The temperature applied during the thermal treatment was constantly 200 °C, while the treatment was implemented under atmospheric pressure environment, in the presence of air. The boards placed in the kiln, were of 11.63% moisture content, as mentioned before, and the interior of the kiln had already reached the chosen temperature of 200 °C. The time periods of thermal treatment of the boards were of 4, 6 and 8 hours and for each treatment 10 boards were used.

At the expiration of each treatment duration, samples were cooled down in desiccators and afterwards stored in climate control room. After a conditioning period of 2 months, at 20 ± 2 °C temperature and relative humidity of 60 ± 5%, EMC and density of the specimens were estimated. Afterwards, 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 properties tests processes 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). For each property test 10 specimens were prepared.

Table 1. Wood properties studied and the respective standards

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

The swelling (in tangential, radial and longitudinal direction) and the absorption percentage measurements were conducted after the immersion of the samples in water of 20 ± 3 °C temperature for 1, 3, 6, 24 and 72 hours, in order to examine the rate of swelling, except the final percentage value.

Surface colour of the specimens was measured using a Minolta Colourimeter, in order to evaluate the colour change owing to heat modification. The Colourimeter specifies the colour as 3 coordinates in 3-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. L^* coordinate describes the lightness and ranges between 100 which represents a perfect reflecting diffuser and 0 which represents black colour, and a^* and b^* describe the chromatic coordinates on the green–red and blue–yellow axes, respectively, without specific numerical limits. The 3 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):

$$\Delta L^* = L^*_{ht} - L^*_{ut} \quad (1)$$

$$\Delta a^* = a^*_{ht} - a^*_{ut} \quad (2)$$

$$\Delta b^* = b^*_{ht} - b^*_{ut} \quad (3)$$

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

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

$$\Delta C^* = C^*_{ht} - C^*_{ut} \quad (6)$$

,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.

3. RESULTS AND DISCUSSION

According to the results, heat treatment of all the 3 durations of time used in this study appeared to enhance the dimensional stability of Scots pine wood specimens (Table 2). Additionally, as the treatment duration was increasing, the swelling percentage value of the specimens tended to decrease, both in tangential and radial direction.

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

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 tangential direction, the 4 hours treated specimens recorded a swelling percentage decrease that ranged between 10.26% and 52.76%, while the 6 hours treated specimens marked swelling decrease of between 17.2% and 56.6% and the 8 hours 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 72 h).

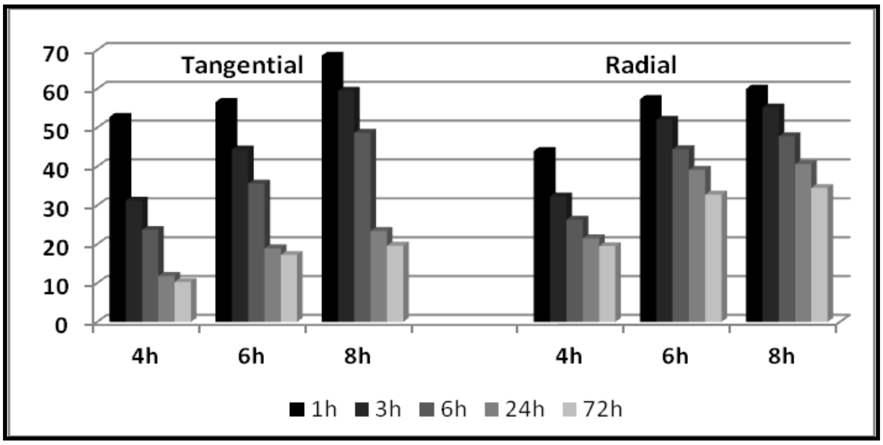

Figure 1 Decrease percentage of Swelling percentage value of tangential and radial direction of the specimens, after 1, 3, 6, 24 and 72 hours

Generally, the percentage values of Swelling in tangential direction were quite higher than the corresponding values of Swelling in radial direction and the higher decrease of Swelling was recorded in radial direction of pine specimens (Fig. 1). Specifically, in radial direction, the 4 hours treated specimens recorded swelling decrease which ranged from 19.5% to 44%, the 6 hours treated specimens marked decrease of between 32.76% and 57.3%, while the 8 hours treated specimens presented a swelling decrease that fluctuated between 34.5% and 60%, taking into account all the measurements values (1, 3, 6, 24 and 72 h). The swelling decrease 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 and this decrease was proportional to the treatment duration increase. Mean value of Absorption percentage after thermal modification was proved to decrease 0.77%, 6.52% and 17.57% for specimens treated for 4, 6 and 8 hours, respectively, which indicates that an improvement of dimensional stability of Scots pine wood can be accomplished using a relatively short thermal treatment duration of 4 to 6 hours at 200 °C.

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 200 °C. More specifically, the average EMC value of untreated pine wood specimens was 11.63%, while after the thermal treatment and a conditioning period of 4 weeks the equilibrium moisture content value of 4 and 6 hours treated specimens were found to be 5.82% and 5.68%, respectively and 5.48% for 8 hours treated specimens. This clearly suggests that thermal treatment affects in great extent the dimensional stability and absorbing capacity of wood and this EMC value reduction is related to the mass loss and the hydroxyl groups loss that have occurred during the thermal treatment.

Furthermore, thermal treatment appeared to cause a decrease in 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 200 °C, to 0.411 g/cm^3 for 6 hours treated specimens and 0.409 g/cm^3 for 8 hours treated specimens, which correspond to decreases of 18.41%, 18.61% and 19%, respectively. The decrease in density is related both to moisture content decrease after treatment that was just mentioned and to 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 physical properties of Camiyanı Black Pine wood and found that density, and Swelling percentage were 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 3 directions of the specimens (tangential, radial, longitudinal) (Fig. 2). This fact indicates that many components absorbing visible light are formed during heat treatment (Yao et al. 2010). Contrarily, *a** parameter records a slight increase during the treatment of 4 hours, while 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 seems to be similar for tangential, radial and longitudinal direction of the specimens.

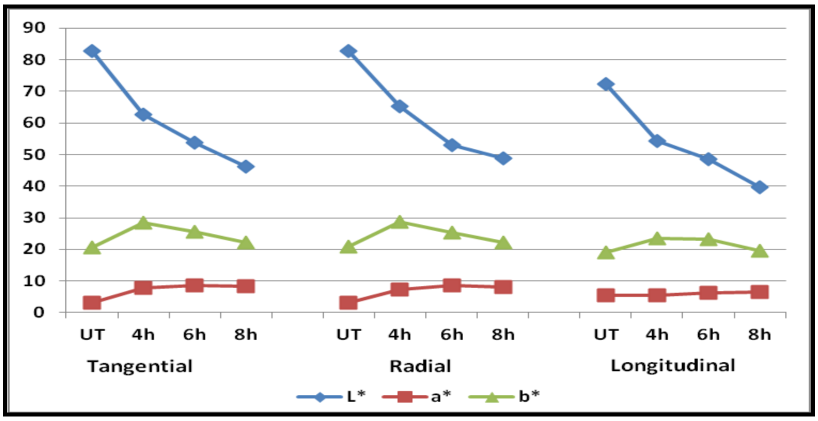

Figure 2 The change of mean values of colour parameters *L**, *a** and *b** in tangential, radial and longitudinal direction of Scots pine specimens

Obviously, a rapid decrease in *L** occurs early in the heat-treatment process, where the largest change can be found between 0 and 4 hours treatment, which indicates that a short period of time is quite enough for the altering of 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 by hot air in an oven for 2, 4 and 8 hours at 150, 175 and 200 °C, by Akgül and Korkut (2012) who measured the change in colour of the Scots pine specimens after thermal treatment at 120, 150 and 180 °C for 2, 6 and 10 hours and by Sahin et al. (2011) who subjected 3 different wood species to thermal treatment of the same conditions.

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

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

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

4. CONCLUSIONS

The main objective of this work was the examination of hygroscopic properties and the colour change of Scots pine wood, thermally treated at 200 °C for 4, 6 and 8 hours in the presence of air. According to the results, as the intensity of the treatment increases, the density and the equilibrium moisture content (EMC) values of wood decrease. Swelling and Absorption percentage values of the specimens appeared to be decreased, which clearly suggests the dimensional stability and the hygroscopic properties enhancement of the treated specimens. As it was evident, thermal treatment of 8 hours at 200 °C resulted in the most severe changes in 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 surface to darken, approximating to 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, enable pine wood to compete other wood species of even higher quality.

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