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RELATIONSHIP BETWEEN COLOUR CHANGE AND SURFACE HARDNESS IN THERMALLY MODIFIED SESSILE OAK WOOD

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Abstract

The aim of this paper is to investigate the relationship between the surface hardness (Brinell's hardness) and colour change of thermally modified Sessile oak (*Quercus sessiliflora* Salisb.) wood. The material was taken from the coppice forests of the National Park Đerdap in eastern Serbia. Two groups of samples were selected from sapwood and from heartwood. The samples were heat treated at 170ºC, 190ºC and 210ºC for 4 hours. Radial hardness and CIELab (L*a*b*) coordinates of wood colour were investigated before and after the thermal treatment. The obtained results indicate that thermal treatment causes a reduction of CIElab coordinates and radial hardness whereas colour change (ΔE) increases with the rise in temperature. All the measured coordinates of the CIELab system have positive values in all the analyzed samples. The positive correlation between lightness (L*) and hardness was determined, whereas colour change has negative impact on radial hardness of oak wood. The results presented in this paper indicate that the colour can be used for assessment of hardness of thermally modified Sessile oak wood.

Key words: CIELab, thermally modified wood, Sessile oak, colour change, hardness.

Introduction

Thermally modified wood (TMW) is obtained by intensive treatment at extremely high temperatures. Thermal treatment of solid wood permanently changes large number of its properties (Jamsa and Viitaniemi 2001, Bekhta and Niemz 2003, Tjeerdsma and Militz 2005, Johansson and Moren 2006, Shi et al. 2007, Gonzales–Pena and Hale 2009).

This type of modification causes a change in hardness, which is a very important indicator of TMW quality and usability. Jamsa and Viitaniemi (2001) showed that hardness is most often an indicator of the quality of TMW surface. Shi et al. (2007) reached the conclusion that hardness in most thermally modified types of wood is higher than in the untreated ones e.g. by 7% in spruce wood, by 52% in pine wood, by 22% in birch wood and by 39% in cottonwood wood. The above listed research results do not give a clear picture of the impact of high temperatures on wood hardness. One of the possible reasons for that may lie in the fact that in some cases and in some types of wood the surface of wood hardens extremely, so that the results obtained using the Brinell method do not reflect the true state of hardness in the whole volume of the investigated sample (Popović et al. 2008).

Besides the change in hardness, heat treated wood obtains a darker tone of colour and the intensity of darkening increases with the rise in temperature. The colour of TMW is an indicator of its chemical properties (Esteves et al. 2007), equilibrium moisture content and shrinkage (Patzeit et al. 2003), as well as hardness (Johansson and Moren, 2006). So far there have been few researches of the relationship between colour and mechanical properties of TMW. Bekhta and Niemttz (2003) found strong correlation between colour change (ΔE) and bending strength in spruce wood thermally modified at 200ºC. Gonzales–Pena and Hale (2009) showed that in beech and pine wood colour change ΔE proved as the best parameter for the prediction of wood hardness, and in spruce wood that was the coordinate of wood lightness L^*. The obtained results indicate that quantitative assessment of colour can serve as a good indicator of some mechanical properties of TMW.

Taking into account that fact that Sessile oak (*Quercus sessiliflora*) wood is wood the most significant hardwood industrial species in Serbia, besides beech, the main aim of this paper was to investigate the relationship between its colour and surface hardness (Brinell's hardness) after thermal modification at high temperatures and to obtain data which can be useful for industrial production and application. The relationship between colour and hardness was investigated for heartwood and sapwood. The investigation of the properties of such wood can contribute to the enhancement of the percentage of sapwood in the final product and consequently the enhancement of utilization in the production of thermally modified Sessile oak wood.

Materials and Methods

The material was taken from the coppice forests in the National Park „Đerdap" in east Serbia. In the forest management unit „Porečke šume", department 54, section „a", three trees were felled (D = 50 cm at 1.3 m). 18 logs with the length of 50 cm were cut, at height from 0.3 m to the first green branches, at every 2 meters. Logs at breast height (1.3 m) and in the middle of stem (5.3 m) were selected for this research (Figure 1).

The logs were transported to the sawmill where their conversion into radial planks of 27 mm thickness was carried out. The planks were transported to the laboratory where they were conditioned. They were cut into two groups of samples (heartwood and sapwood) with dimensions 12 x 22 x 35 mm (radial by tangential by longitudinal). The samples were cut along the longitudinal direction. The heat treatment was done in an oven for 4 h at 170°C, 190°C and 210°C. Three replicates were used for each combination of time/temperature of treatment (B, C and D). After treatment, the samples were cooled down in a dry environment and weighed. Untreated samples (A) were used as the control.

The density was determined in the samples using SRPS D.A1.044 standard and moisture content using SRPS EN 13183-1 standard. The moisture content of thermally treated samples was determined according to Akyldiz and

Ates (2008). Mass loss, as a significant indicator of the quality of thermally modified wood, was determined in all the examined samples (Esteves et al. 2007a).

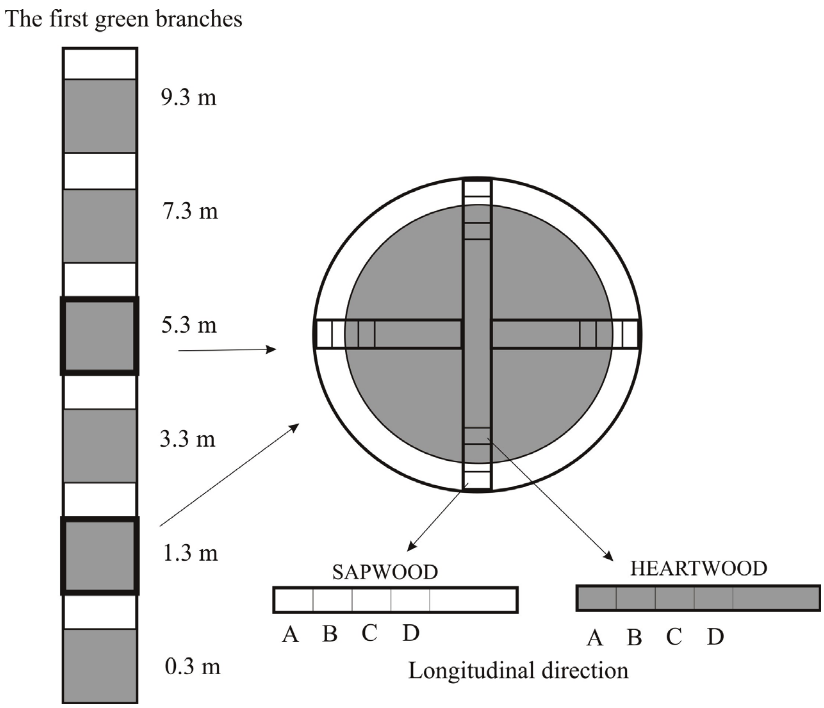

Fig. 1. The way samples were cut.

Wood colour was determined in the CIELab system with the use of a BYK (BYK Gardner GmbH) colorimeter. Settings of the device were the following ones: light source D65 and 10º standard observers. The samples were measured with elliptical aperture, 8mm in diameter. The colour meter was calibrated using white calibration plate which is delivered with the device by the producer. During the measurement each sample was positioned on a flat plate for the sake of measurement stability and constant position in relation to the measuring device. Colour coordinates were measured on a tangential surface before and after the treatment at two measuring points and their average value was used. The following CIEL*a*b* coordinates were measured: L* (black-white), a* (green-red), b* (blue-yellow). Colour change in the CIELab system was determined according to the following formula:

$$\Delta E = \sqrt{(L_0 - L_t)^2 + (a_0 - a_t)^2 + (b_0 - b_t)^2}$$

where:

L_t, a_t and b_t – CIELab coordinates of the samples after treatment,

L_0, a_0 and b_0 – CIELab coordinates of the samples prior treatment.

Wood surface hardness was determined using the Brinell's method (EN 1534) with the direction of the force parallel to wood rays (radial hardness) prior (untreated samples) and after heat treatment (heat treated samples). Radial hardness was determined at the same points as colour (Figure 2). After that, average values of these two measurings were determined.

The obtained data were submitted to an analysis of variance (ANOVA) and t-test.

Results and Discussion

Oven dry density, moisture content and mass loss

The average value of Sessile oak density in oven dry state was 0.715 $g.cm^{-3}$ in heartwood with a 5.2%, coefficient of variation and in sapwood 0.590 $g.cm^{-3}$ with a 7.4% coefficient of variation.

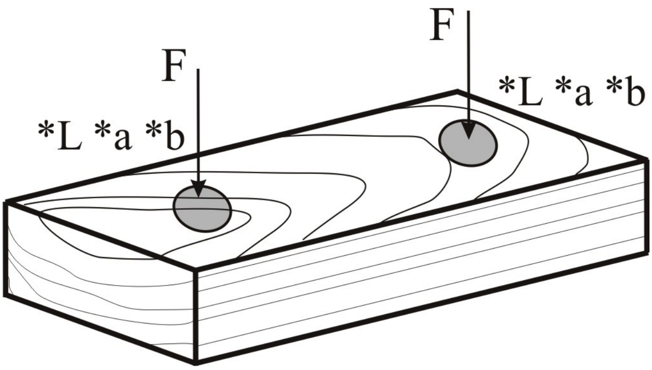

Fig. 2. Place on a sample at which hardness and CIELab coordinates were determined.

The obtained value of density is the approximate value to the one reported by Le Moguedec et al. (2002).

By conditioning the samples at temperatures from 22±2°C and 55±5%, humidity, untreated heartwood samples reached an average value of 10.8%, and sapwood 10.2%. The thermally treated samples were conditioned in the same conditions for 30 days, before colour and hardness recording and they reached the following values of moisture content: 170ºC: heartwood – 6.4%, sapwood – 6.6%; 190ºC: heartwood – 4.4%, sapwood – 4.6%; 210ºC: heartwood – 3.9%, sapwood – 4%. The obtained results show that thermal treatment contributes to the reduction of equilibrium moisture content in wood.

Mass loss increased with a rise in temperature. Mass loss of the heat treated samples was: at 170ºC: heartwood – 1.3%, sapwood – 1.6%; at 190ºC: heartwood – 4.1 %, sapwood – 6.4% and at 210ºC: heartwood – 10.7% and sapwood – 11.7%. The presented results indicate that the losses are higher in sapwood than in heartwood which can also be an indicator of significant changes in the physical properties of this part of oak wood.

Brinell hardness and colour change

The obtained values of radial hardness are shown in Table 1. In untreated samples radial hardness of heartwood was on average 36.1 $N.mm^{-2}$, and for sapwood 27 $N.mm^{-2}$. Variability of the analyzed data was higher for heartwood – 18.4%, whereas it was 12.7% for sapwood.

After thermal treatment there is a reduction in the radial hardness of wood. The analysis of variance revealed that there is significant difference in average values between the untreated and treated samples for both heartwood ($F=9.36$, $p<0.01$) and sapwood ($F=15.73$, $p<0.01$). T-test was used to reveal that at 170ºC the value of hardness does not differ from the untreated samples, as well as that there is no substantial difference among the values at temperatures between 190º and 210ºC. These results confirm the premise that the substantial reduction of wood hardness begins at temperatures over 190ºC.

Table 1. Brinell hardness of thermally modified Sessile oak wood.

Statistical parameters	Heartwood				Sapwood			
	Untreated	170ºC	190ºC	210ºC	Untreated	170ºC	190ºC	210ºC
N	24	24	24	24	24	24	24	24
X	36.1	34.2	31.4	28.4	27.0	24.8	20.9	20.9
σ	6.63	4.20	5.47	5.06	3.42	3.57	4.30	3.52
V	18.36	12.26	17.44	17.83	12.65	14.38	20.56	16.83
Max	48.10	41.00	44.50	41.32	33.18	32.38	34.97	27.00
Min	26.30	25.10	19.30	19.30	21.30	16.87	14.16	14.45

N – number of samples; X – mean, $N.mm^{-2}$; σ – standard deviation, $N.mm^{-2}$; V – variation,%; Max – maximum of values, $N.mm^{-2}$; Min – minimum of values, $N.mm^{-2}$.

Quantitative characteristics of the colour of Sessile oak TMW were determined according to the CIELab system (not shown). Wood lightness (L*) ranged from 77.7 in untreated sapwood to 27.7 for wood heat-treated at 210ºC. These values were lower for heartwood and were 67.5 for untreated to 24.9 for a treatment at 210ºC. Values of b* were positive but they had the same trend as L* i.e. they decreased with the rise in temperature. Colour change (ΔE), as a result of a thermal treatment increased with the rise in temperature. Colour change was the greatest at 210ºC, and it was 51.4 for sapwood and 46.4 for heartwood. In the other two treatments colour change was also slightly higher in sapwood which is a result of the increased wood mass loss.

Positive linear correlation was determined between hardness and CIELab coordinates in the form $y=b_0+b_1x$. The impact of CIELab coordinates a* and b* on the hardness of thermally modified oak wood was very weak. Coefficients of determination (R^2) ranged from 0.145 to 0.275 for sapwood and from 0.276 to 0.395 for heartwood. The coordinate L* had the greatest impact on hardness which coincides with the results of Gonzales-Pena and Halea (2009). Table 2 shows the values of the coefficients of determination between L* and radial wood hardness. As shown, coefficients of determination for heartwood are higher than for sapwood, with the exception of the treatment at 210ºC. According to the analysis of variance significant difference was found between radial hardness and L* coordinate in all samples.

Taking into account the fact that the values of colour difference are only slightly different between sapwood and heartwood, the impact of colour change was investigated for both parts of wood together. Colour change ΔE had the opposite impact on the radial hardness of oak wood to CIELab coordinates. This trend was recorded in both heartwood and sapwood. Table 3 also shows the coefficients of the linear equation with the form $y=b_0+b_1x$ and Fig. 3 and Fig. 4 the impact of L* and ΔE on the hardness of Sessile oak wood thermally modified at 190°C.

Table 2. Simple linear regression analysis to estimate Brinell hardness of thermally Modified Sessile oak wood using the colour coordinate L*.

Temperature of treatment	Part of wood	n	R^2	F (sig)	b_0	b_1	Std. error
170°C	Sapwood	20	0.425	13.30	- 31.75	+ 1.00	2.59
	Heartwood	20	0.522	19.67	- 34.97	+ 1.40	2.72
190°C	Sapwood	20	0.494	14.68	- 69.84	+ 2.78	3.07
	Heartwood	19	0.565	22.4	- 64.29	+ 3.07	3.84
210°C	Sapwood	20	0.423	11.01	- 135.6	+ 6.65	4.57
	Heartwood	22	0.250	6.35*	- 76.26	+ 4.23	4.79

n – number of samples; R^2 – coefficient determination of model; F – result of Fisher's test, significance at 99% level; model of the form: $y = b_0 + b_1x$; Std. error – the standard error of the estimate; *significance at 95%.

Conclusions

The results obtained in this research show that thermal treatment causes a reduction in radial hardness of oak wood. This change is not significantly different between thermal treatments at 210ºC and 190ºC, as well as between the untreated samples and the ones treated at 170ºC. This was recorded in both heartwood and sapwood.

All the investigated coordinates of CIELab system (a*, b* and L*) were positive in both untreated and treated samples and they mainly decreased mainly with a rise in temperature. In the analysis of the impact of certain coordinates on wood hardness it was determined that only lightness (*L) has significant impact on it. Positive correlation was found between lightness and radial hardness. On the other hand, the calculated colour change (ΔE) had the opposite trend and its values increased with the rise in temperature. The analysis of the relationship between colour change and radial hardness revealed negative linear correlation.

According to the results presented in this paper, colour change of the thermally modified Sessile oak wood can be a

Table 3. Simple linear regression analysis to estimate Brinell hardness of thermally modified Sessile oak wood using colour difference (ΔE).

Temperature of treatment	R^2	F (sig)	b_0	b_1	Std. error	n
170°C	0.609	71.8	57.29	- 1.31	3.86	48
190°C	0.781	163.98	79.94	- 1.27	3.40	48
210°C	0.692	99.12	61.98	- 0.53	2.07	48

R^2 – coefficient determination of model; F – result of Fisher's test, significance at 99% level; model of the form: $y = b_0 + b_1x$; Std. error – the standard error of the estimate; n – number of samples.

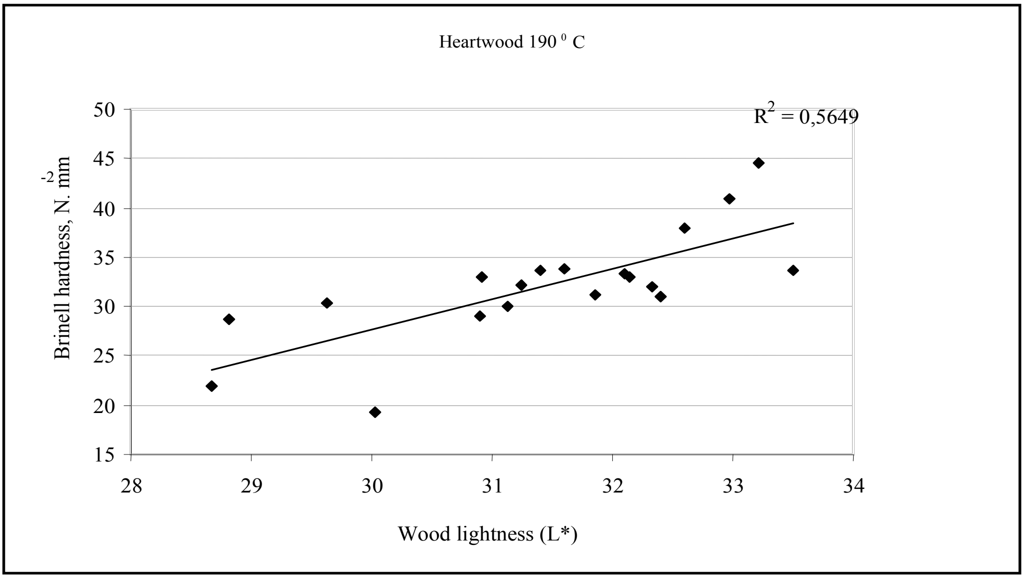

Fig. 3. Models for the prediction of TMW hardness (at 190ºC) using wood lightness (L*).

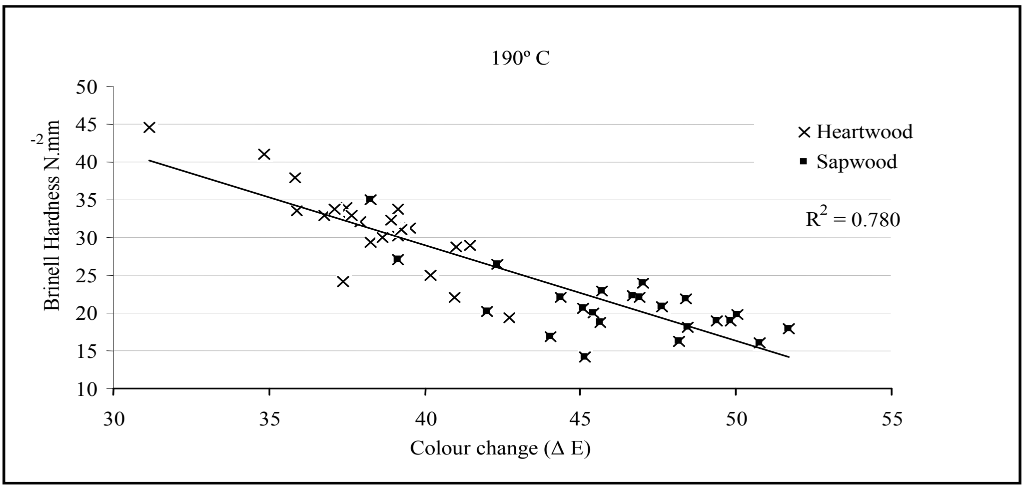

Fig. 4. Models for the prediction of TMW hardness (at 190ºC) using colour change (ΔE).

good indicator of its hardness. The obtained results can be applied in TMW production with mainly visually determined colour, which can significantly influence the application of the final product.

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