The Shades of Color of *Quercus robur* L. Wood Obtained through the Processes of Thermal Treatment with Saturated Water Vapor

Ladislav Dzurenda

This paper presents the shades of color of *Quercus robur* L. wood obtained in the processes of thermal treatment and color modification by saturated water vapor, with temperatures: t_{I} = 112.5 ± 2.5 °C for τ = 5.5 h (mode I), t_{II} = 127.5 ± 2.5 °C for τ = 6.5 h (mode II), and t_{III} = 137.5 ± 2.5 °C for τ = 7.5 h (mode III). The color of oak wood by thermal treatment in mode I changes minimally with mild browning in the CIE-$L^*a^*b^*$ color space: L^* = 65.5 ± 1.7; a^* = 8.8 ± 0.4; b^* = 20.7 ± 0.5. A brown shade with coordinates: L^* = 56.8 ± 1.3; a^* = 9.3 ± 0.5; b^* = 19.4 ± 0.5 is achieved in mode II. Oak wood thermally modified by mode III acquires an original brown-gray color with the color coordinates: L^* = 47.5 ± 2.1; a^* = 9.4 ± 0.5; b^* = 17.1 ± 1.1. The irreversible changes in the color of the oak wood, achieved by some of the color modifications of wood using saturated water steam, extend the possibilities of its use in the field of building-joinery, the artistic field, and the field of design.

Keywords: Wood; Sessile oak; Color; Thermal treatment; Saturated water steam

Contact information: Department of Woodworking, Faculty of Wood Sciences and Technology, Technical University in Zvolen, T. G. Masaryka 2117/24, 960 53 Zvolen, Slovak Republic;
**Corresponding author: dzurenda@tuzvo.sk*

INTRODUCTION

The color of wood is a basic physical property and a typical feature of the sapwood and heartwood of wood species. Wood color of industrially important wood species used as construction material in cabinet making and the furniture industry covers a wide range: from light white-gray-yellow shades in Norway spruce (*Picea excels* (L.) H. Karst.), silver fir (*Abies alba* Mill.), small-leaved linden (*Tilia cordata* Mill.), and European hornbeam (*Carpinus betulus* L.), to red-brown shades of heartwood in English oak (*Quercus robur* L.), European ash (*Fraxinus excelsior* L.), and royal nut (*Juglans regia* L.) (Drapela 1980; Klement *et al.* 2010; Makovíny 2010).

Thermal treatment processes of wet wood, in addition to targeted physico-mechanical and chemical changes of wood, are accompanied by a change in wood color. (Kollmann and Gote 1968; Nikolov *et al.* 1980; Sergovsky and Rasev 1987; Trebula 1996; Deliiski 2003; Dzurenda and Orlowski 2011). In the past, color changes in darkening of the wood in the thermal process have been used to remove the undesirable color differences between sapwood and the dark core, or eliminate color change of wood after mold. Currently, increased attention is given to the targeted change in the color by thermal processes (Molnar and Tolvaj 2002; Matuškova and Klement 2009; Tolvaj *et al.* 2010; Dzurenda and Deliiski 2012; Dzurenda 2013; Barcik *et al.* 2015).

One way to objectively quantify the given color of a wood is by using coordinates in the CIELAB system. This system is based on the measurement of three parameters:

lightness L^* from 100 to 0 for black, chromatic co-ordinate a^* for setting shading between red ($+a^*$) and green ($-a^*$), and chromatic co-ordinate b^* determining the hue between yellow ($+b^*$) and blue ($-b^*$) (ISO 7724-3 1984).

Oak wood has a narrow light brown sapwood and pale brown-yellow core. During steaming, it acquires darker shades of brown (Trebula 1986; Tolvaj *et al.* 2010). There is much interest by individuals involved in floor coverings and restaurateurs in the use of oak wood, particularly shades of darker brown. As a result, the industry has encouraged experimental research on the color modification of oak wood by the thermal process *via* steam of water. The aim of this work was to determine the color of oak wood (*Quercus robur* L.) in the CIE-L^*, a^*, b^* color space after heat treatment with saturated water steam with the following temperatures: t_I = 112.5 ± 2.5 °C for τ= 5.5 h (mode I), t_{II} = 127.5 ± 2.5 °C for τ = 6.5 h (mode II), and t_{III} = 137.5 ± 2.5 °C for τ = 7.5 h (mode III).

EXPERIMENTAL

Materials

Oak wood was in the form of hardwood with dimensions of 30 mm x 55 mm x 500 mm and moisture content W_p = 60.2 ± 3.3%. The wood was thermally treated with steam in a pressure autoclave APDZ 240 (Himmash AD, Haskovo, Bulgaria) at Sundermann s.r.o. in the town of Banská Štiavnica.

Methods

The mode of color modification of oak wood is given in Fig. 1. The optimized parameters for each color shade of oak wood, the duration of the technological process, and the heat consumption are shown in Table 1.

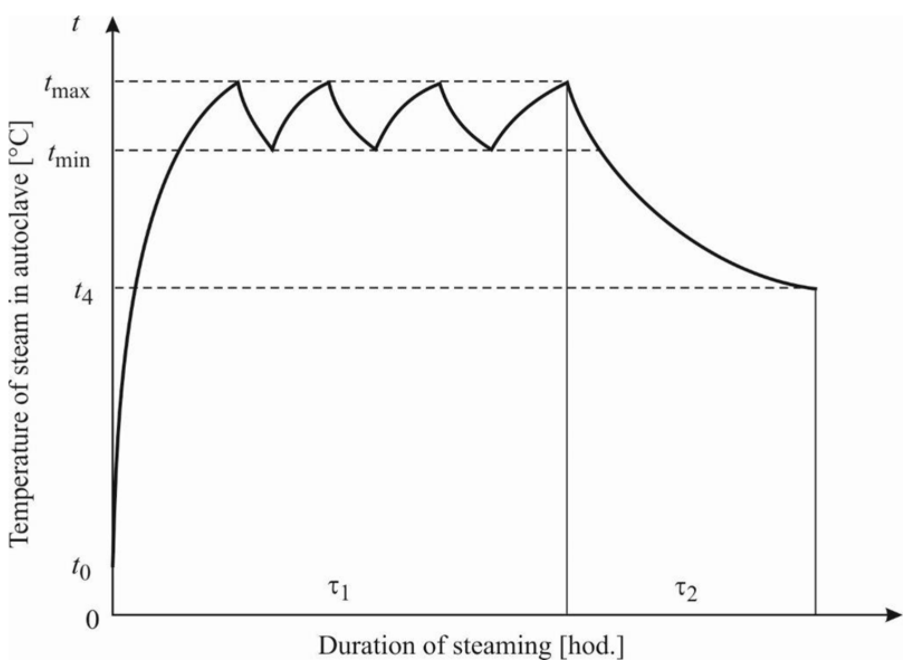

Fig. 1. Mode of color modification of oak wood with saturated water steam

Table 1. Thermal Treatment of the Oak Wood

Thermal Treatment Modes: Color Modification	Temperature of Saturated Water Steam (°C)			Duration (h)			Saturated steam consumption kg/m^3
	t_{min}	t_{max}	t_4	τ_1- stage I	τ_2-stage II	Total $\tau_1 + \tau_2$	
Mode I	110	115	100	4.5	1.0	5.5	205.5
Mode II	125	130	100	5.0	1.5	6.5	206.2
Mode III	135	140	100	5.5	2.0	7.5	206.9

The thermally unadjusted and treated blanks of oak wood were dried to moisture W_p = 12 ± 0.5% in a conventional wood drying kiln and were subsequently planed on horizontal milling cutter.

The wood color of oak blanks in the CIE-$L^*a^*b^*$ color space was determined using a CR-10 Color Reader colorimeter (Konica Minolta, Osaka, Japan). The light source used was D65, and the diameter of collecting area was 8 mm.

The L^*, a^*, and b^* coordinates were measured for 245 oak blanks without heat treatment, 245 oak pieces after mode I treatment, 244 oak pieces after mode II treatment, and 245 oak pieces after mode III treatment. The measurement of color coordinates on dried and planed blanks oak wood was made at the center of the blanks width and at 250 mm from the front of blank. Colored coordinate values were represented by the average measured value and standard deviation. The variance of the color values was determined by the coefficient of variation.

The total color difference ΔE^* was determined according to ISO 11 664-4 (2008) (Eq. 1) based on the difference in the color coordinates (ΔL^*, Δa^*, and Δb^*) in the thermally treated and untreated oak,

$$\Delta E^* = \sqrt{[(L_2^* - L_1^*)^2 + (a_2^* - a_1^*)^2 + (b_2^* - b_1^*)^2]} \quad (1)$$

where L_2^*, a_2^*, and b_2^* values at the coordinates were measured before the wood treatment, and L_1^*, a_1^*, b_1^* values were measured in the thermally treated oak wood. The degree of change in the color and shades of wood based on the overall color difference ΔE^* was classified according to the scale shown in Table 2 (Cividini *et al.* 2007).

Table 2. Classification of Δ*E*

Δ*E** Value	Observation
0.2 < Δ*E**	No visible difference
0.2 < Δ*E** < 2	Small difference
2 < Δ*E** < 3	Color difference visible with light quality screen
3 < Δ*E** < 6	Color difference visible with medium quality screen
6 < Δ*E** < 12	High color difference
Δ*E** > 12	Different colors

RESULTS AND DISCUSSION

The heartwood of *Quercus robur* L. has a pale brown-yellow color (Klement *et al.* 2010; Makoviny 2010). Oak wood has been measured with the following values: L^* = 69.9; a^* = 6.5; and b^* = 20.6 (Babiak *et al*. 2004). These findings are confirmed by the present measurements of L^* = 68.1 ± 2.0, a^* = 10.2 ± 0.5, and b^* = 21.4 ± 0.9. The coefficients of variations of the individual sets of measured color coordinate values were as follows: v_{L^*} = 2.9%, v_{a^*} = 4.9, v_{b^*} = 4.2. The color shades acquired through heat treatment with saturated water vapor in modes I, II, and III are shown in Fig. 2. The CIE-*L*a*b** coordinates measured after heat treatment, drying, and planing are shown in Tables 3 to 6.

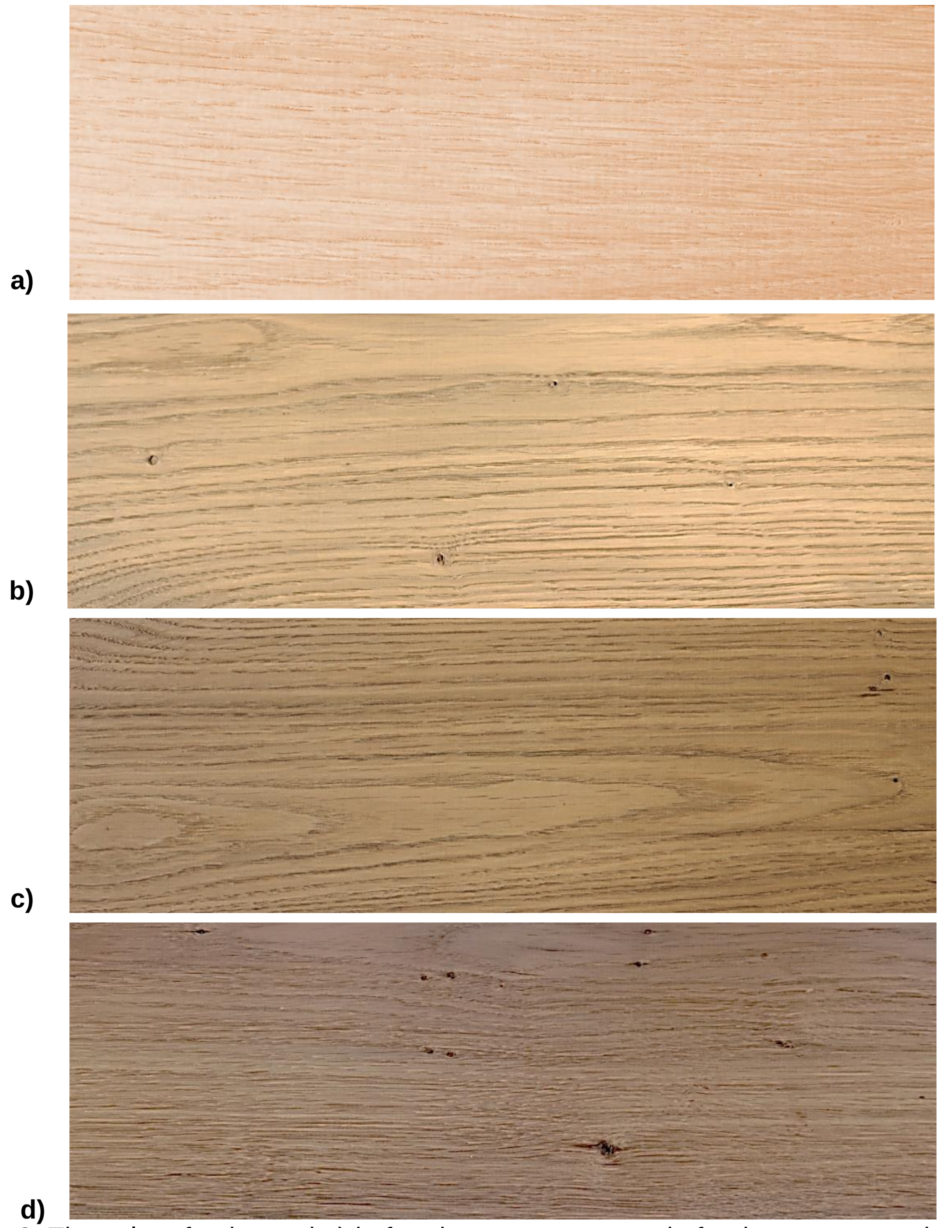

Fig. 2. The color of oak wood a) before heat treatment, and after heat treatment by b) mode I, c) mode II, and d) mode III

The color of oak wood in the process of thermal treatment by mode I changed to a brown-yellow color shade with a highlighted texture of oak wood. Oak modified by regime II acquired a brown color. This color shade of thermally treated oak wood was similar to the color of the wood *Juglans regia*. The original brown color with a gray tint was obtained by oak wood in thermal treatment by mode III. This shade is suitable for imitation of old oak wood.

Table 3. Color Coordinates of Untreated Oak Wood

Parameter	Color Coordinate		
	*L**	*a**	*b**
Number of Measurements	245	245	245
Value	68.1 ± 2.0	10.2 ± 0.5	21.4 ± 0.9
Coefficient of Variation (%)	2.9	4.9	4.2

Table 4. Color Coordinates of Oak Wood Treated with Mode I*

Parameter	Color Coordinate		
	*L**	*a**	*b**
Number of Measurements	245	245	245
Value	65.5 ± 1.7	8.8 ± 0.4	20.7 ± 0.5
Coefficient of Variation (%)	2.6	4.5	2.4

* t = 112.5 ± 2.5 °C

Table 5. Color Coordinates of Untreated Oak Wood Treated with Mode II*

Parameter	Color Coordinate		
	*L**	*a**	*b**
Number of Measurements	244	244	244
Value	56.8 ± 1.3	9.3 ± 0.5	19.4 ± 0.5
Coefficient of Variation (%)	2.3	5.4	2.6

* t = 127.5 ± 2.5 °C

Table 6. Color Coordinates of Untreated Oak Wood Treated with Mode III*

Parameter	Color Coordinate		
	*L**	*a**	*b**
Number of Measurements	245	245	245
Value	47.5 ± 2.1	9.4 ± 0.5	17.1 ± 1.1
Coefficient of Variation (%)	4.4	5.3	6.4

* t = 137.5 ± 2.5 °C

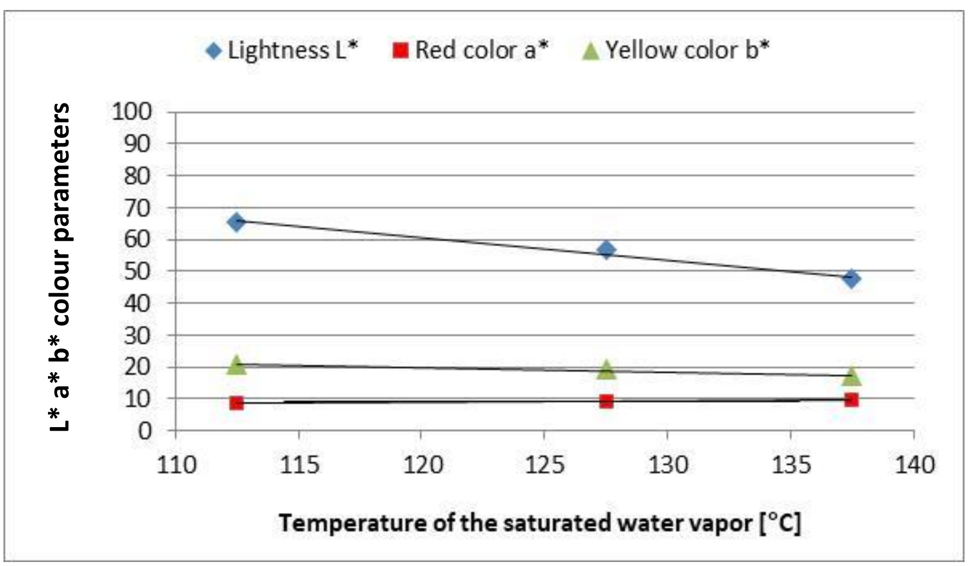

Fig. 3. Dependence of decrease in lightness, red and yellow color of thermally treated oak wood in the CIE *L* a* b** color space on the temperature of saturated steam

Change of the lightness of L^*, red a^*, and yellow b^* color of oak wood on the temperature of the thermal treatment of oak wood blanks with saturated steam in the temperature range t = 112.5 to 137.5 °C is shown in Fig. 3.

The total color differences of oak wood ΔE^* achieved by the individual steam treatment modes are shown in Fig. 4.

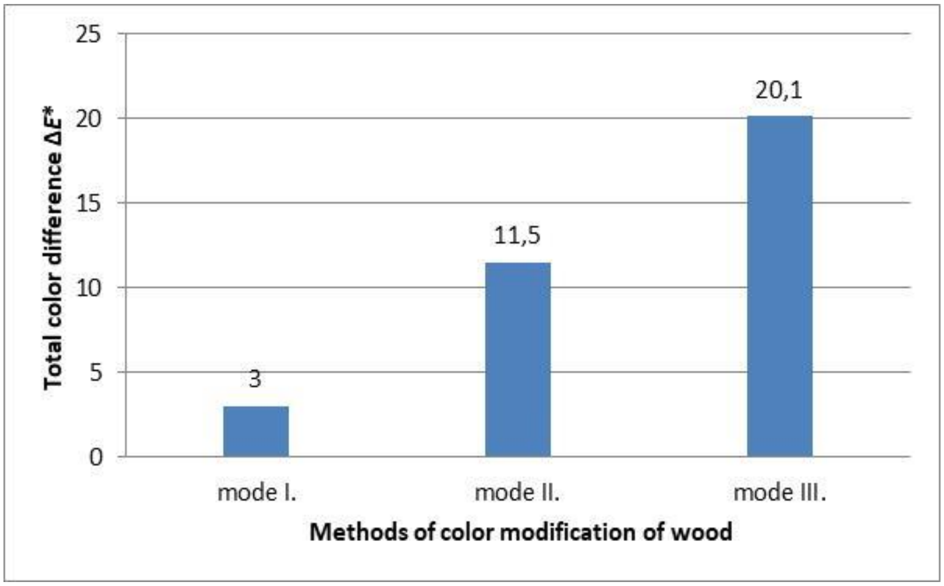

Fig. 4. Values of the total color differences of the thermally modified oak wood achieved by the individual steam treatment modes

The results show that during thermal treatment of oak wood, an increase in the temperature of the saturated steam changed the color of oak wood from a pale brown-yellow color to brown shades, and finally, to a pronounced brown-gray color.

The change in color caused by the individual modes of modification is reflected in changes in the values of the individual coordinates in the CIE $L^*a^*b^*$ color space. The brightness decreased due to a modification of mode I from L^* = 68.1 ± 2.0 to L_I^* = 65.5 ± 1.7, meaning a decrease of ΔL^* = -2.6. The changes in red and yellow chromatic coordinates were due to a decrease of red color value from a^* = 10.2 ± 0.5 to a_I^* = 8.8 ± 0.4, *i.e.* Δa^* = -1.4, and a yellow color decrease from b^* = 21.4 ± 0.9 to b_I^* = 20.7 ± 0.5, which means that Δb^* = -0.7. The decline in whiteness of oak wood and the decrease in red and yellow values is reflected in the mild browning of oak wood quantified by the value of the total color difference ΔE^* = 3.

More significant changes in the color of oak wood from the heat treatment process are achieved in modes II and III. Oak wood thermally treated using mode II had a brown color, expressed by the value of the total color difference ΔE^* = 11.5. A decrease in brightness of ΔL^* = - 20.6, a decrease in the value of the chromatic color of the red color Δa^* = - 0.8, and a decrease of the value on the chromatic yellow color coordinates by Δb^* = -4.3 are all achieved in mode III, using saturated steam with a temperature of t = 137.5 ± 2.5 °C for the duration of the thermal process τ = 7.5 h. This darkening and browning of the wood creates an original brown-gray color of the blanks of oak wood.

The total color differences of the color of oak wood ΔE^* caused by water steam heat treatment lay in the range of values: ΔE^* = 3.0 to 20.1. Within the colorimetric classification of the color changes (Table 2), the acquired color shades were categorized as medium to significant changes in the color of the wood.

The dependence of the growth of the total color differences ΔE^* of the thermally treated oak wood on the temperature of saturated water steam in the color space is consistent with reported color changes during wood treatment (Molnar-Tolvaj 2002; Dzurenda 2014), as well as high temperature drying in the environment of superheated water steam (Klement and Marko 2009), or thermic processes in the production of thermo wood (Barcik *et al*. 2015).

In terms the physical and chemical properties of wood after thermal treatment, color changes are irreversible (Kollmann and Gote 1968; Trebula 1996). This is due to the partial hydrolysis of hemicelluloses in the lignin saccharide wood matrix and the extraction of water-soluble auxoric substances. These findings are confirmed by the differences in ATR-FTIR spectroscopic analyzes of thermally unprocessed and treated wood (Kučerová *et al*. 2016; Geffert *et al*. 2017), as well as the presence of monosaccharides, organic acids, and basic units of lignin with guajacyl and syringyl structure in the condensate after pressure steaming of wood (Bučko 1995; Dzurenda and Deliiski 2000; Kačík 2001; Laurova *et al*. 2004; Kačíková and Kačík 2011).

The mechanical properties of the color modified oak wood in the dry state did not change significantly. Laboratory tests thermally treated oak wood of mode III showed a slight increase in the compressive strength in the fiber direction by 1.4% and tension perpendicular to the fibers by 3.8%.

The irreversible color change and the creation of new brown shades of oak wood achieved through the thermally modifying of the color of wood by water vapor extends the possibilities of wider use of oak wood in the building-joinery, construction, arts, and design areas.

CONCLUSIONS

1. The paper presents the colors of oak wood obtained in a thermal process using saturated water steam with temperatures: t_I = 112.5 ± 2.5 °C for t = 5.5 h, t_{II} = 127.5 ± 2.5 °C after time τ = 6.5 h and t_{III} = 137.5 ± 2.5 °C for τ = 7.5 h. The color of the oak wood in the process of thermal treatment as per mode I changes from a pale brown-yellow color to a pale brown color with coordinates in the color space CIE-L^*, a^*, and b^*: L^* = 65.5 ± 1.7 a^* = 8.8 ± 0.4; b^* = 20.7 ± 0.5. Thermal treatment as per mode II changes it to a brown color with color coordinates: L^* = 56.8 ± 1.3; a^* = 9.3 ± 0.5; b^* = 19.4 ± 0.5 and color modification using mode III changes it to an original brown-gray shade with color coordinates: L^* = 47.5 ± 2.1; a^* = 9.4 ± 0.5; b^* = 17.1 ± 1.1.

2. By increasing the temperature of the saturated water vapor in the thermal treatment process and by prolonging the heat treatment time, the agate wood loses its brightness (darkening) and by decreasing values on the red color coordinate a^* and on the yellow color coordinate b^* in the color space CIE-L^*, a^*, and b^* new brown shades are achieved.

3. The irreversible changes in the color of oak wood achieved with some of the thermal treatment modes by the saturated water steam and the new color shades gained by these processes, extend the possibilities for its use in the building-joinery, design-art and design areas.

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