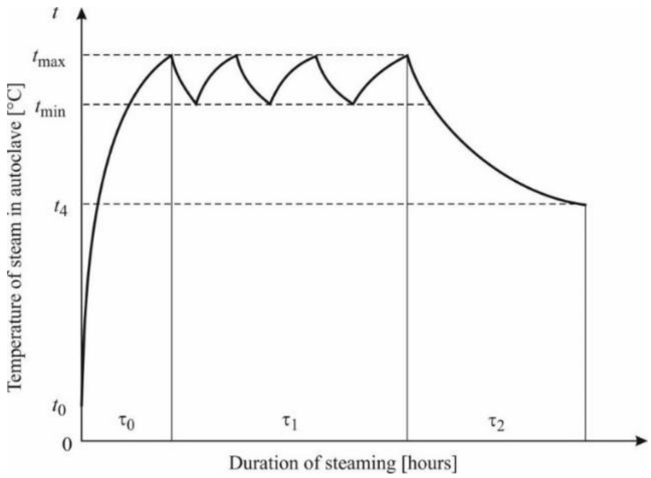


Fig. 1. Mode of color modification of beech wood with saturated water steam

Table 1 Modes of thermal treatment of beech wood with saturated steam

Modes	Temperature of saturated steam [°C]			Steaming time in hours [hours]			
	t_{max}	t_{min}	t_4	τ_0 - heating	τ_1 - phase I	τ_2 - phase II	Total time
Steaming mode I.	107.5	102.5	100		3.5	1.0	≈ 6.0
Steaming mode II.	130.0	125.0	100	≈1.5	4.0	1.0	≈ 6.5
Steaming mode III.	140.0	135.0	100		4.5	1.0	≈ 7.0

It was measured the moisture content and acidity of wet beech wood on the sef of 35 timber samples before the thermal process and after the thermal treatment of timber by each mode after its completion and after cooling of the wood to ambient temperature.

The moisture content of wet beech wood above the fiber saturation point was measured with the resistance humidity meter of type: FMD6.

The measurement of acidity of wet wood was performed with meter SENTRON SI 600 and probe LanceFET+H. The pH probe has a diameter of d = 10 mm and cannot be immersed in a solid material. Because of that reason, the 12 mm hole was created with cordless drill at the measurement point. The drilling sawdust were poured into the formed hole and the probe LanceFET+H of meter SENTRON SI 600 was inserted in to the wet sawdust (Geffert *et al*. 2019).

Subsequently, the thermally modified and unmodified beech timber was dried to a moisture content $W_p = 12 \pm 0.5\%$ in a conventional hot air dryer: KAD 1x6 (KATRES s.r.o). By means of longitudinal and transverse manipulation, wood blanks with dimensions: 27 x 75 x 600 mm were made. All surfaces of blanks were machined on the horizontal plane milling machine FS 200.

The color measurement of both untreated and thermally treated beech wood with a moisture content W_k = 12 ± 0.5% was performed on all surfaces at a distance of 300 mm from the front of the blank. The color of the beech wood blanks in the CIE-L* a* b* color space was measured with a colorimeter Color Reader CR-10 (Konica Minolta, Japan). There was used light source D65 and the diameter of the optical scanning hole was 8 mm. In the CIE L* a* b* and CIE L* C* h° color spaces, the color was considered based on changes in the luminance coordinate L*, the chromatic coordinate of the red color a*, the chromatic coordinate of the yellow color b* and the chroma C*. Chroma C* is the integrated value of the red a* and yellow b* coordinates on the chromatic plane of the cylindrical color space CIE L* C* h°.

$$C^* = \sqrt[2]{a^{*2} + b^{*2}} \qquad (2)$$

Where: a*- value on chromatic coordinate of red color,
b* - value on chromatic coordinate of yellow color.

From the difference of the values on the color coordinates ΔL*, Δa*, Δb* that were determined on the basis of measurements both thermal treated and untreated beech wood surface, the total color difference ΔE was determined according to the following equation *ISO 11 664-4:*

$$\Delta E^* = \sqrt{(L_2^* - L_1^*)^2 + (a_2^* - a_1^*)^2 + (b_2^* - b_1^*)^2} \qquad (3)$$

Where: L^*_1, a^*_1, b^*_1 - values on the color space coordinates of the surface of dried milled thermally untreated beechwood,
L^*_2, a^*_2, b^*_2 - values on the color space coordinates of the surface of dried milled thermally treated beechwood.

The change of color in wood, except of changes on chromatic coordinates in the CIE L* a* b* color space, was also assessed by changes in lightness ΔL*, changes in chroma ΔC* and hue angle h° in cylindrical color space CIE L* C* h°.
Hue angle h° defines the deviation of chroma C* from the coordinate of red color in the chromatic plane that describe the equation:

$$h^{\circ}_{ab} = arctan\left(\frac{b*}{a*}\right) \qquad (4)$$

Where: a* - value on chromatic coordinate of red color,
b* - value on chromatic coordinate of yellow color.

The consumption of saturated water steam for the realization of the technological process for 1 m^3 of wood was determined by model of heat consumption per 1 m^3 for the thermal treatment of wood in the pressure autoclave APDZ 240 Dzurenda (2016):

$$Q_{ha} = \frac{Q_{hw} + Q_{hf} + Q_{hil} + Q_{he} + Q_{hfv} + Q_{hew}}{V_w} \quad [MJ/m^3] \tag{5}$$

Where: Q_{hf} – heat necessary to heat the construction material of the autoclave, MJ;

Q_{hw} – heat necessary for heating colour modification wood, MJ;

Q_{hil} – heat necessary to heat the autoclave's insulation, MJ;

Q_{he} – at necessary to cover heat losses from the surface of the pressure autoclave, MJ;

Q_{hfv} – heat extracted by condensate from the pressure autoclave, MJ;

Q_{hew} – heat extracted by saturated steam after opening the autoclave, MJ;

V_w – volume of the colour modification wood in the pressure autoclave, m^3.

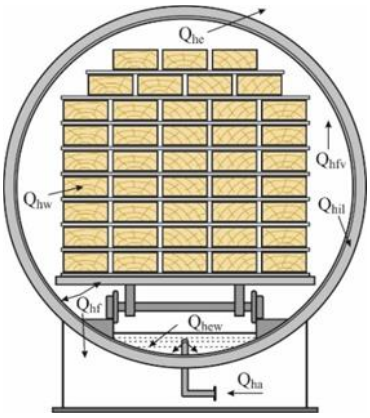


Figure 2. The heat distribution in the autoclave in the process of modifying the color of the wood with saturated water steam.

The consumption of saturated water steam for color modification of $1m^3$ of beech wood is evaluated by the equation:

$$m^{"} = \frac{Q_{ha}}{h'' - h'} \tag{6}$$

Where: Q_{ha} – heat consumption standard for modification of 1 m^3 of beech timber, MJ / m^3;

h'' – enthalpy of saturated water steam at temperature t_{max}, kJ/kg;

h' – enthalpy of saturated water at temperature t_4, kJ/kg.

3. Results and Discussion

The physical properties of wood *Fagus sylvatica L* such as: wood moisture content and acidity of wet wood before the thermal treatment process as well as wood density, color space coordinates CIE L* a* b* and chroma C* in the dry state, are shown in Table 2.

Table 2 Measured values of density, color space coordinates of CIE L* a* b* of dry wood, moisture content and acidity of wet beech wood

Timber	Dry beech wood					Wet beech wood	
	Wood density	Color space coordinates CIE L*a*b*				Moisture content	Acidity
	ρ_O [$kg \cdot m^{-3}$]	L*	a*	b*	C*	W [%]	pH
Beech	683.5 ± 65.1	76.6 ± 2.3	6.8 ± 1.8	19.8 ± 1.7	20.9	56.4 ± 3.3	5.2 ± 0.1

The measured density values of beechwood in the dry state are the average values of healthy density, fungi or molds of undamaged beech sapwood. Similar values of beech wood density for Central European territory are mentioned in the works: (Požgaj *et al.* 1997; Makoviny 2010; Kurjatko *et al.* 2010). On the basis of this statement, the analyzed changes of characteristics in beech wood achieved by thermal treatment in individual modes can be marked as representative (standard).

Table 3. shows the results of laboratory work determining the density of both untreated and treated beechwood by the individual modes in the dry state.

Table 3 Densities of dry, untreated and treated beech wood

Thermal modification modes	Statistical characteristics of wood density in the dry state			
	ρ_0 [kg·m^{-3}]	s [kg·m^{-3}]	v_x [%]	n [-]
Untreated wood	683.5	65.1	9.5	18
Modification mode I.	671.8	58.5	8.7	16
Modification mode II.	691.9	60.1	8.7	18
Modification mode III.	705.0	67.8	9.6	17

The variations in density of dry beech wood due to the thermal treatment of the individual modes are contradictory. The density of dry thermally modified beechwood by mode I is less about $\Delta\rho_0$ = - 1.74 % and by the mode II. and III. grows up. The density of dry beech wood treated by mode II. about $\Delta\rho_0$ = + 1.21 % and by mode III. about $\Delta\rho_0$ = + 3.05 %. From the presented results follows, that due to the hydrolysis of the polysaccharide fraction of beechwood and due to the extraction of water-soluble substances by the thermal treatment processes of individual modes, the density of beechwood changes partially. The changes in the densities of the color-modified beech wood do not exceed the limits of natural density tolerance of beech wood, which are in the Central European area: ρ_0 = 490-680-880 kg.m^{-3}.

The acidity of wet wood of most tree species from the temperate zone is in the range of pH = 3.3 – 6.4 (Sandermann and Rothkamm 1956; Irle 2012; Solar 2014; Geffert *et al.* 2019). The acid reaction of most wood tree is caused by free acids and acid groups that are in aqueous solutions of dilute sugars, organic acids and water-soluble inorganic substances fed to the tree in the lumen of cells by the root system (Čudinov 1968; Blažej *et al.* 1975; Zevenhoven 2001, Pňakovič and Dzurenda). The measured acidity values of wet thermally untreated beech wood are pH = 5.2 ± 0.2 at moisture content w ≈ 56.4 % and do not differ from the acid values of beech wood with moisture content above the point of saturated fibers reported by the authors: (Solar 2004; Geffert *et al.* 2019). The acidity values of wet thermally modified beech wood by individual modes after cooling to ambient temperature are shown in Table 4.

Table 4 Measured moisture content and acidity values of thermally modified beech wood

Mode	Temperature of saturated water steam	Wood moisture content	pH value
Modification mode I.	t_I = 110 - 115 °C	w ≈ 48.7 %	pH = 4.5 ± 0.2
Modification mode II.	t_{II} = 125 - 130 °C	w ≈ 45.3 %	pH = 4.0 ± 0.3
Modification mode III.	t_{III} = 135 - 140 °C	w ≈ 44.5 %	pH = 3.7 ± 0.3

The decrease in the pH of thermally modified beech wood by the individual color modification modes confirms the known knowledge about the process of hydrolysis mainly hemicelluloses in the wet wood of leafy tree under the influence of heat, as reported by: (Melcer *et al.* 1989; Laurova *et al*. Samešova *et al*. 2018; Geffert *et al*. 2019; Dzurenda and Dudiak 2019).

Depending on the temperature and duration of action of the hydrolysis products: acetic acid and formic acid, degradation occur of polysaccharides by carbohydrate and pectin oxidation, dehydration of pentose to 2-furaldehyde and in lignin begin to form free radicals and phenolic hydroxyl groups, resulting in the formation of new chromophoric groups causing wood color change (Fengel and Wegener 1989; Solár 2004; Bučko 2005). These reactions results in the formation of new chromophoric groups that cause color changes in the wood. This is illustrated by the visual change in color of beech wood in Figure 3. and the of changes in the CIE L* a* b* color space of the thermally modified beech wood in Table 5.

The color of the dried, planed, steam untreated beech wood and the color shades obtained by the heat treatment modes – with saturated water steam are shown in Figure 3.

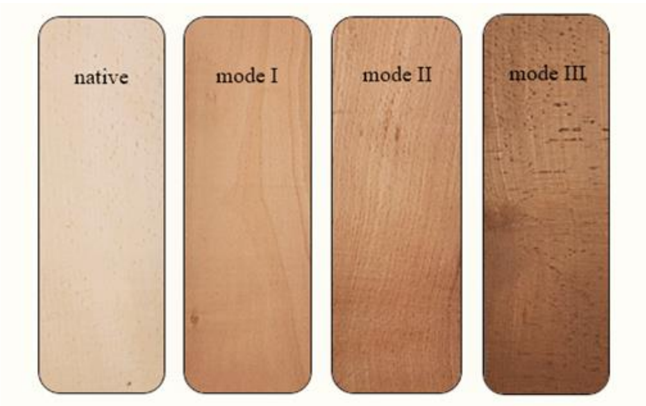


Fig. 3. View of beech wood before and after heat treatment by individual modes.

The coordinate values describing the color of beech wood before and after thermal treatment by each mode in CIE L* a* b*, chroma C* and total color difference ΔE * are shown in Table 5.

Table 5 CIE L* a* b* color space coordinate values describing beech wood before and after starurated water steam treatment with each steam mode

Temperature of saturated water steam	CIE-L* a* b* color space coordinates			Chroma	Total color difference
	L*	a*	b*	C*	ΔE*
Not thermally treated wood	76.6 ± 2.3	6.9 ± 1.8	19.8 ± 1.7	20.9	- - -
t_I = 110 - 115 °C	71.5 ± 2.1	11.1 ± 1.7	21.5 ± 1.5	24.2	6.8
t_{II} = 125 - 130 °C	63.2 ± 2.4	14.9 ± 2.1	23.3 ± 1.7	27.6	16
t_{III} = 135 - 140 °C	54.3 ± 2.4	15.6 ± 1.9	24.1 ± 1.6	28.7	24.4

The color of dry, untreated beech wood identified by coordinates L* = 76.6 ± 2.3; a * = 6.9 ± 1.8; b * =

19.8 ± 1.7 color space CIE L* a* b* is comparable to the color coordinate values reported for beech wood by: (Babiak *et al.* 2004; Dzurenda 2014; Meints *et al*. 2017).

The thermally treated beech wood at temperature of saturated water steam t_I = 105 ± 2.5°C for τ = 6 hours acquire a pale brown-pink-yellow shade. In the CIE color space L* a* b* it is identified by the coordinate values: L_I* = 71.5 ± 2.1; a_I* = 11.1 ± 1.7; b_I* = 21.5 ± 1.5. The total color difference ΔE_I* = 6.8 according to the colorimetric classification of wood color changes during thermal treatment processes of wood processing (Cividini *et al.* 2007) classifies achieved color change into the changes perceptible to the naked eye. The significant color changes of beech wood by thermal treatment are achieved by modes II. and III with a saturated water steam temperature of t_{II} = 127.5 ± 2.5 °C, respectively by temperature t_{III} = 137.5 ± 2.5 °C. Thermal treatment of mode II. by the temperature t_{II} = 127.5 ± 2.5°C for τ = 7.5 hours the beech wood acquires a brown-pink color with the coordinates: L_{II}* = 63.2 ± 2.4; a_{II}* = 14.9 ± 2.1; b_{II}* = 23.3 ± 1.7. Original brown-red color with color coordinate values: L_{III}* = 54.3 ± 2.4; a_{III}* = 15.6 ± 1.9; b_{III}* = 24.1 ± 1.6 the beech wood acquires by thermal treatment conditions at a saturated steam temperature t_{III} = 137.5±2.5°C. The total color difference ΔE_{III}* = 24.4 achieved by thermal treatment of beech wood with saturated water steam at temperature t_{III} = 137.5 ± 2.5 °C, according to colorimetric classification of wood color changes (Cividini *et al.* 2007) belongs to categories of significant color changes.

By the visual inspection of the wood color on the sides of the beech blanks produced by sweeping and shortening of thermally treated lumber, as well as by measuring the color values at each CIE L* a* b* color space coordinate, beech wood is uniformly throughout colored. The uniformity of coloring of the thermally modified wood throughout the volume makes it possible to use such treated wood for the production of slats for flooring, respectively other 3D processing of the massif without worrying about uneven coloring of wood along its cross-section between surface and center. The above-mentioned facts extend the possibilities of using beech wood with new color shades in the building-joinery, construction-artistic and designing area.

The evaluation of color changes in thermally treated beech wood according to the color space parameters CIE L* C* h° shows, that by the increase in the temperature of saturated water steam in the technological process, the lightness L* decreases significantly and the chroma C* increases slightly. The decrease in the lightness of beech wood achieved by the individual thermal modes ΔL* and the increase in chroma ΔC* of the beechwood achieved by each mode at the temperature of the saturated water steam is shown in Figure 4.

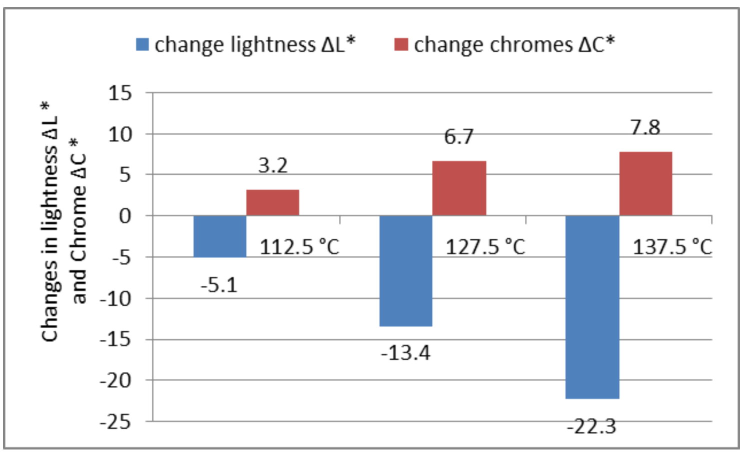
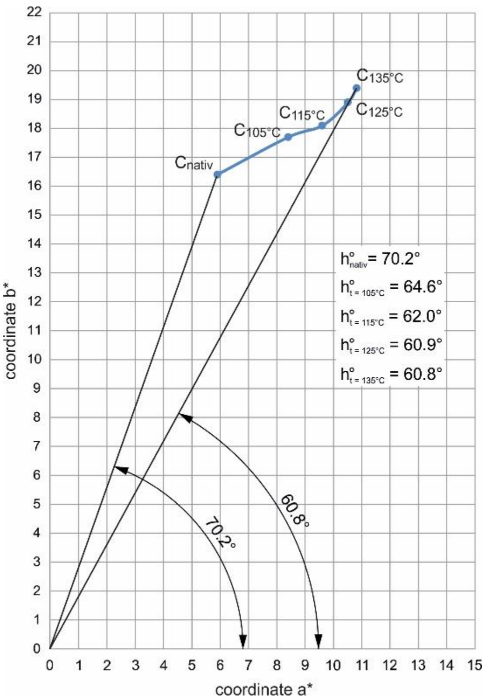


Figure 4. Dependence of lightness changes ΔL* and chroma ΔC* on temperature of saturated water steam in technological process.

The decrease in the lightness of beech wood with the increase in the temperature of the saturated water steam in the thermal treatment process is in line with the knowledges of wood darkening in technological processes such as wood steaming declared in the works: (Tolvaj *et al.* 2009, 2010; Dzurenda, 2018 b, c), drying in warm humid air, respectively overheated water steam (Klement and Marko 2009; Dzurenda and Deliiski 2012; Baranski *et al.* 2017) or in thermal treatment of wood in thermo-wood production (Barcik *et al.* 2015, Pinchevskaya *et al.* 2019).



Changes in color saturation chroma C* in the chromatic space of red color a*, yellow color b* and hue angle h° of beech wood due to the increase of wood temperature in the technological process of beech wood color modification with saturated water steam are shown in Figure 5.

Figure 5. Color saturation changes in chroma C* and hue angle h° in chromatic space a*, b*.

The increading distance in chroma C* on the color space a*, b* (Figure 5.) from the center of the red color coordinates a* and the yellow color b* declares the saturation of the beech wood thermal treatment with saturated water steam. This knowledge is important for both wood processors and users of color-modified beech wood products with saturated water steam, because the richer colors of the interior objects are more acceptable for the human eye than the gray shades.

The dependence of total color difference changes ΔE* on wood temperature in technological process of color modification by saturated water steam of beech wood is shown in Figure 6.

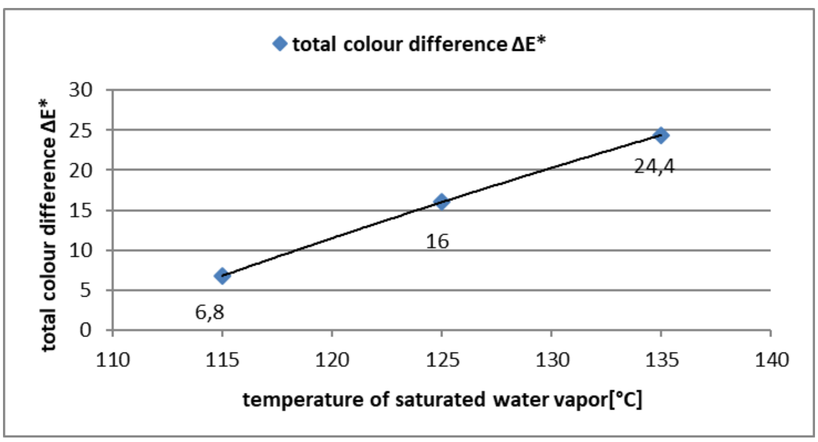


Figure 6. The dependence of total color difference changes ΔE* of beech wood on temperature of saturated water steam in technological process.

The total color differences of beech wood color ΔE* developed by water steam treatment with temperatures from the range 105 °C to 135 °C lies within the range of values: ΔE* = 6.8 ÷ 24.4. The mathematical dependence describing the color change of beech wood, shown in Figure 6., is in decisive extent determined by changes in the luminance coordinate of the thermally treated beech wood and in a lesser extent is determined with changes of red a* and yellow b* chromatic coordinates. According to the above-mentioned, dependence the significant changes in the brown-red color of beech wood occurs according to the categorization of the wood color changes magnitude in the thermal processes reported by Cividini *et al. (2007),* by the water steam temperature above $t \geq 122$ °C.

According to the categorization of physico-mechanical and chemical changes of wood achieved by the technologies of thermal wood processing presented by the authors: Kollmann - Gote 1968; Trebula 1986, the beech wood color changes, achieved by any presented mode of the thermal treatment process, belong to the group of irreversible changes of wood. The irreversibility of color changes in beech wood is confirmed by differences in ATR-FTIR spectroscopy analyzes in the lignin-saccharide complex of untreated and treated wood (Geffert *et al.* (2017), Timar *et al.* (2016), Kučerová *et al.* (2016), as well as the presence of monosaccharides, organic acids and basic lignin units with guajacyl and syringyl structure in the condensate from thermal treatment reported in: (Bučko 1995; Dzurenda and Deliiski 2000; Kačík 2001; Laurova *et al.* 2004; Kačíková and Kačík 2011; Samešova *et al*. 2018).

The heat necessary for the implementation of technological process of beech wood color modification is provided by the intake of saturated steam to the autoclave from the steam boiler. APDZ 240 pressure autoclave heat balance for each beech wood color modification mode of thickness h = 40 mm with moisture content w ≈ 56% and initial wood temperature $t_d \approx 15$ °C for autoclave capacity V_D = 16 m3, shows the Table 5.

Table 5 Heat consumption balance for thermal process of color modification of beech wood of h = 40 mm in APDZ 240

Name of the heat consumption item	Symbol	Mode I.		Mode II.		Mode III.	
		MJ	[%]	MJ	[%]	MJ	[%]
Heating colour homogenised wood	Q_{hw}	3 867.2	70.7	4 860.2	72.6	5 323.9	73.2
Heating the autoclave's construction material	Q_{hf}	540.0	9.9	540.0	8.1	540.0	7.4
Heating the autoclave's insulation	Q_{hil}	82.6	1.5	101.7	1.5	110.4	1.5
Heat losses of the pressure autoclave	Q_{he}	64.9	1.2	85.5	1.3	101.6	1.4
Heat losses from extracted steam	Q_{hfv}	54.9	1.1	54.9	0.8	54.9	0.8
Heat losses from extracted condensate	Q_{hfw}	855.8	15.6	1 049.8	15.7	1 141.6	15.7
Normative Q_{ha} for heat consumption	∑	341.6 MJ·m-3		418.2 MJ·m-3		454.5 MJ·m-3	

The heat consumption expressed in the form of saturated water steam consumption of the pressure autoclave APDZ 240 for individual modes of wood color modification per 1 m^3 of beech wood with thickness h = 40 mm is shown in Table 6.

Table 6 The comsumption of saturated water steam of the pressure autoclave APDZ 240 for individual modes of wood color modification per 1 m^3 of beech wood with thickness h = 40 mm

Mode of color modification	Saturated steam consumption in kg/m^3
Mode I.	155.3
Mode II.	190.1
Mode III.	206.5

The heat consumption of thermally colored beech wood per 1 m^3 depending on the temperature of saturated steam in the technological process is: Q_{ha} = 341.6 - 454.5 MJ/m^3. A positive feature of the above-mentioned technology of thermal modification of wood color is the fact, that from 70.7% to 73.2% of the intake heat from saturated water steam, as shown in Table 5., is used to heat wood to the required technological temperature.

The technological process of color modification of beech wood with saturated water steam is a typical discontinuous process characterized by uneven heat consumption during the technological process. From the analysis of heat consumption of the technological process follows that the highest heat consumption is at the beginning of the technological process at the time $\tau_o \approx 1.5$ hours, when the heat of condensing water steam is used for heating of modified wood and heating the autoclave. Subsequently, the heat of condensing saturated steam is used to cover the losses of the heat in autoclave. In the second stage of the technological process τ_2, steam is not fed into the autoclave and the losses of heat in autoclave is covered by the heat from the isochoric cooling of the saturated steam contained in the autoclave and also by the heat from the cooling of the autoclave construction material. Figure 5 shows the course of saturated water steam consumption at the temperature of t = 137.5 ±2.5 °C for mode III of the thermal modification of beech wood color with thickness h = 40 mm.

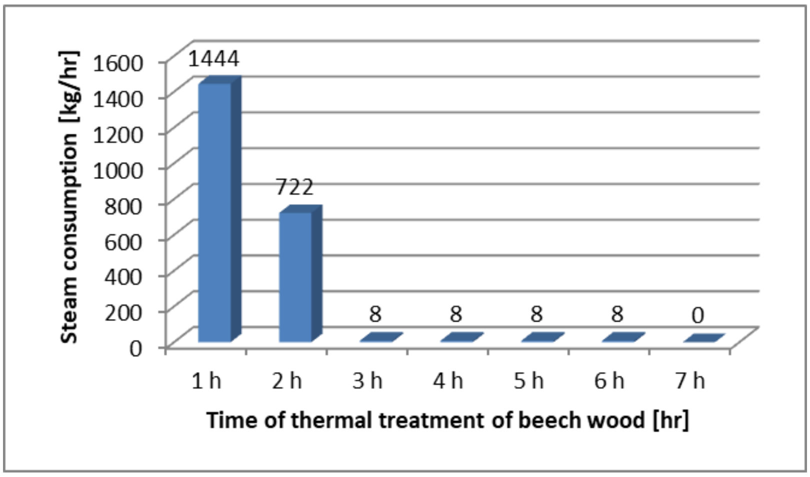


Figure. 5. The consumption of saturated water steam during the thermal modification of beech wood with thickness h = 40 mm by the mode III. in autoclave APDZ 240

4. Conclusions

In this article are presented the changes of beech wood obtained in the targeted process of color modification with saturated water steam with temperatures $t_I = 105 \pm 2.5°C$ for $\tau = 6$ hours (mode I.), $t_{II} = 127.5 \pm 2.5°C$ for $\tau = 6.5$ hours (mode II.), $t_{III} = 137.5 \pm 2.5°C$ for $\tau = 7$ hours (mode III.) and there are also characteristics of the technological process in terms of heat consumption and saturated water steam. These changes of beech wood were analyzed on the basis of targeted experimental work.

- the density of dry beech wood has been reduced due to the thermal modification during mode I. from $\Delta\rho_0 = -1.74$ % to $\rho_0 = 671.8$ kg.m^{-3}, with mode II. increased from $\Delta\rho_0 = +1.21$ % to $\rho_0 = 691.9$ kg.m^{-3} and with mode III. increased from $\Delta\rho_0 = +3.05$ % to $\rho_0 = 705.0$ kg.m^{-3}.
- wet beech wood by thermal process of mode I. changes acidity from pH = 5.2 to pH = 4.5 with mode II. to pH = 4.0 and with mode III. to pH = 3.7.
- the beech wood color changes from a light white-gray color with a yellow tinge to colors from brown-pink-yellow to brown-red hues of brown. With increasing temperature of saturated water steam which realize the process, the brightness of beech wood decreases and the wood stay dark. The color shade of thermally treated wood during mode I in the CIE-L* a* b* color space is identified by the coordinate values: $L_I^* = 71.5 \pm 2.1$; $a_I^* = 11.1 \pm 1.7$; $b_I^* = 21.5 \pm 1.5$, in mode II. with coordinates: $L_{II}^* = 63.2 \pm 2.4$; $a_{II}^* = 14.9 \pm 2.1$; $b_{II}^* = 23.3 \pm 1.7$ in mode III. with coordinates: $L_{III}^* = 543 \pm 2.4$; $a_{III}^* = 15.6 \pm 1.9$; $b_{III}^* = 24.1 \pm 1.6$.
- the range of total color differences ΔE^* of the thermally modified beech wood by the individual modes is in the range of $\Delta E^* = 6.4 – 24.4$ so its in terms of the magnitude categorization of the color changes reported by Cividini *et al.* (2007), beech thermally modified wood belongs to the category of visible and large color changes.
- heat consumption per 1m^{3}of thermal modification of beech wood color in pressure autoclave APDZ 240 for mode I. is $Q_{ha} = 341.6$ MJ.m^{-3}, for mode II. is $Q_{ha} = 418.2$ MJ.m^{-3} and for mode III. is $Q_{ha} = 454.5$ MJ.m^{-3}

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