COLOR CHANGE — MASS LOSS CORRELATION FOR HEAT-TREATED WOOD

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Abstract: Heat treatment is renowned as the most environmentally friendly process of dimensional stabilization that can be applied to wood, in order to make it suitable for outdoor uses. It also darkens wood color and improves wood durability. The intensity of heat treatment can be appreciated by means of two parameters: the color change occured in wood due to the high temperature, and the mass loss, which is a measure of the degree of thermal degradation. In order to find a mathematical correlation between these two parameters, an experimental study was conducted with four European wood species, which were heat-treated at 180°C and 200ºC, for 1-3 hours, under atmosheric pressure.

The paper presents the results concerning the color changes and mass losses recorded for the heat-treated wood samples compared to untreated wood. For all four species, the dependency between the color change and the mass loss was found to be best described by a logarithmic regression equation with R^2 of 0.93 to 0.99 for the soft species (spruce, pine and lime), and R^2 of 0.77 for beech. The results of this study envisage to simplify the assessment procedure of the heat treatment efficiency, by only measuring the color – a feature that is both convenient and cost-effective.

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Introduction

Thermal modification, also called "heat treatment," is renowned as the most environmental friendly method of dimensional stabilization, which can be applied to wood. It enables wood to become suitable for exterior uses by reducing its hygroscopicity, without adding any chemical substance into the wood. This is possible due to the modifications, which occur in the chemical composition of wood under the effect of high temperatures (above 160°C). The treating temperature must be chosen so as to produce those modifications of the chemical structure, which favor only the incipient decomposition of the hemicelluloses, but do not affect the cellulose structure. This way, wood becomes more resistant to moisture, without losing its mechanical strengths.

According to literature, the decomposition of the main chemical compounds in wood takes place with maximum intensity at: 180°C for hemicelluloses, 270°C for lignin, and 340°C for cellulose (these results were obtained from poplar and spruce wood, as reported by Bobleter & Binder, 1980). However, changes in the chemical composition of wood, highlighted by mass losses, already occur at much lower temperatures, depending on wood species (Oelhafen, 2005).

In recent past, numerous experimental researches regarding the effects of high temperatures upon wood properties of different wood species have been performed. A comprehensive literature review was published by Esteves & Pereira (2009).

Most experiments were conducted with resinous species, like spruce and pine, which are less sensitive to temperature and are easily treated. The works of Alén, Kotilainen, & Zaman (2002); Allegretti et al. (2012); Bekhta & Niemz (2003); Bal (2014); Borrega & Kärenlampi (2008); Mazela, Zakrzewsky, Grzeskowiak, Cofta, & Bartkowiak (2003); Viitaniemi, Jämsä, & Viitanen (1997) are only several of the most relevant publications in this field. Though not as numerous as for resinous woods, literature also includes studies on the heat treatment of some broad-leaved species, like birch and aspen (Kim, Yun, & Kim, 1998), beech (Gonzáles-Peña & Hale, 2009; Kaymakci & Akyildiz, 2011; Todorovic, Popović, Milić, & Popadić, 2012), ash (Kocaefe, Poncsak, & Boluk, 2008; Zivković, Prša, Turkulin,

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Sinković, & Jirouš-Rajković, 2008), oak (Clauder, Maschmann-Fehrensen, & Seemann, 2009; Korkut, Karayilmazlar, Hiziroglu, & Sanli, 2010; Campean, Gurau, & Olarescu, 2011).

Most tests on heat-treated wood include determinations of the mass loss (ML). This is because the mass loss is the most important indicator of the heat treatment efficiency. It is closely related to the thermal degradation of wood; the higher the mass loss, the more advanced the decomposition of the main chemical wood compounds, which means better dimensional stability, but also mechanical weakening of the wooden material. Viitaniemi et al. (1997) found that ML of 5% represents an important threshold value, at which optimum dimensional stabilization is achieved without significantly affecting the mechanical strengths.

Apart from its aesthetical importance, color is also a crucial indicator of the heat treatment efficiency, i.e. the darker the wood color, the more efficient the heat treatment. Therefore, finding a mathematical correlation between these two indicators of heat-treatment efficiency is the main objective of the present experimental study, which was conducted with four European wood species, heat-treated at temperatures of 180°C and 200°C for 1-3 hours, in air, at atmospheric pressure.

The results of this study envisage to simplify the assessment procedure of the heat treatment efficiency, by measuring only color, which can be done conveniently and cost-effectively, and establishing the mass loss indirectly based on the mathematical correlation as function of the recorded color change.

Material and Methodology

Wooden Material

Four European species were selected for the present study, so as to cover several wood structure categories:

- Spruce (Picea abies) – a resinous low-density species, without heartwood, quite homogenous;
- Black pine (Pinus nigra) – a resinous species with heartwood, denser and more durable than spruce;
- Lime (Tilia cordata) – a soft diffuse-porous broad-leaved species; homogenous; with no spectacular aesthetic value - dull natural color, but silky appearance;
- Beech (Fagus sylvatica) – a diffuse-porous hardwood species, with medium-high density and high internal growth stresses.

The average density of the wooden material used within the experiments, also compared to the values given by Wagenführ (2008), is given in Table 1.

The material used in the experiments consisted of 620 x 85 x 30 mm defect-free boards, cut from trees with diameter (Φ) of 15 to 16 cm, resulted from thinning and originating all from the same forest parcel, located in the southern part of Romania (Stroesti – Arges: 45° 8' 0" North, 24° 47' 0" East).

Table 1: Average density of wooden material used within the experiments

Oven-dry density (kg/m^3)	Spruce	Black pine	Lime	Beech
Experimental	371	544	428	684
Reference literature	300–430–640	560	320–490–560	490–680–880

Sources: Authors (experimental values), Wagenführ (2008) (reference values)

Heat treatment

The boards were first air-dried down to a moisture content of 12%, and then conditioned for 2 weeks at 20ºC and 65% relative humidity (RH) in a FEUTRON KPK200 climate chamber. Hereinafter, 20 x 20 x 30 mm strictly radial-tangential samples were cut out of these.

Before being heat-treated, the samples were oven-dried (down to 0% moisture content) constantly at 103ºC in a BINDER electric oven with natural convection and PID controller, in order to avoid fissuring during the heat treatment.

The heat treatment was carried out at two different temperatures (180ºC and 200ºC) in three treatment durations: 1, 2, and 3 hours for spruce, pine, and lime; 2, 3, and 4 hours for beech. The treating conditions were chosen considering the structural peculiarities of the chosen species and previous experiences with their behavior under heat treatment. For each experimented heat treatment condition, as well as for the controls (untreated wood), a set of 10 samples was employed.

Each set of samples was heat-treated in the same BINDER oven, under atmospheric pressure by increasing the temperature to the pre-set values (180°C and 200°C, respectively) and maintained at this condition for a pre-determined period. After each hour of treatment, a set of samples was removed from the oven and cooled down in a desiccator before being tested. Thus, all measurements performed before and immediately after the heat treatment refer to the oven-dry state (0% moisture content) of the samples.

Characterization Tests

Color

Thin color specimens with dimensions 20 x 20 x 0.5 mm were cut out from the middle part of each treated and untreated wood samples. They were scanned immediately after cutting by means of a MUSTEK 1248UB scanner with a resolution of 600x1200dpi, at a resolution of 300dpi.

The three-dimensional L^*, a^*, b^* color space, recommended by CIE (Commission Internationale de l'Eclairage) was used for the quantitative color assessment, in which L^* specifies the lightness, a^* the red-green share, and b^* the blue-yellow share. By analyzing the $L^*a^*b^*$ coordinates obtained for each heat-treating condition, and by comparing them to those of untreated wood, the total color change due to the heat treatment (ΔE^*) was calculated according to equation (1) (McLaren, 1976):

$$\Delta E^* = \sqrt{\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2}} \tag{1}$$

where ΔL^*, Δa^*, and Δb^* are the changes of the three CIE_L*a*b coordinates due to the heat treatment, calculated as the difference between the value for treated and untreated wood, on the same sample.

Apart from its aesthetical importance, color is a critical indicator of the heat treatment efficiency. That is, the darker the wood color becomes, the more efficient the heat treatment can be considered.

Mass Loss

Based on the experimental data, obtained by weighing the samples before and after the heat treatment by means of a KERN electronic scale with 0.001g accuracy, the mass loss (ML) was calculated according to equation (2):

$$ML = \frac{m_0 - m_{tr}}{m_0} \cdot 100 \ (\%) \tag{2}$$

where:

m_0 is the oven-dry mass of a sample before the heat treatment, in g;

m_{tr} is oven-dry mass of the same sample after the heat treatment, in g.

Based on the ML and ΔE* values obtained, for each heat treatment condition and each species, regression equations were established in order to correlate the two indicators of the heat treatment efficiency.

Results and Discussion

Color

Figure 1 presents the color evolution of the samples cut from the four different wood species, due to different heat-treatment conditions.

Figure 1: Influence of heat-treatment conditions upon wood color:

Spruce (*Picea abies*):

control	180°C/1h	180°C/2h	180°C/3h	200°C/1h	200°C/2h	200°C/3h

control	180°C/1h	180°C/2h	180°C/3h	200°C/1h	200°C/2h	200°C/3h

Black pine (*Pinus nigra*):

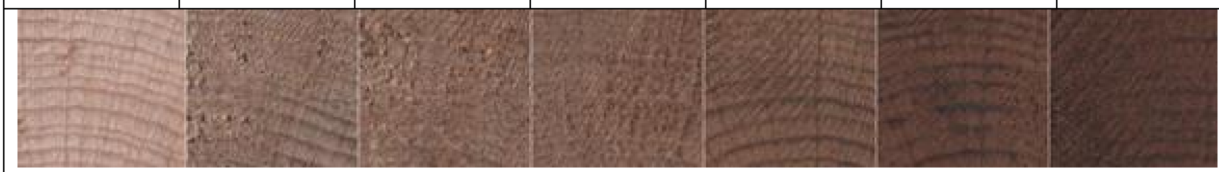
control	180°C/1h	180°C/2h	180°C/3h	200°C/1h	200°C/2h	200°C/3h

Lime (*Tilia cordata*):

control	180°C/1h	180°C/2h	180°C/3h	200°C/1h	200°C/2h	200°C/3h

Beech (*Fagus sylvatica*):

control	180°C/2h	180°C/3h	180°C/4h	200°C/2h	200°C/3h	200°C/4h



Source: Authors

As can be seen from Figure 1, lime wood reacted the fastest to temperature and changed its color significantly, already after 1 hour of exposure to 180°C.

Out of the three color coordinates, the lightness had the greatest decrease with all four species: ΔL^* reached up to -51 for spruce and -50 for lime after the heat treatment at 200°C/3h (Table 2), while the change of the a^* and b^* coordinates was insignificant (< 5). Thus, the greatest part of the total color change ΔE^* (Table 3) was caused by the lightness change.

Table 2: Lightness change (ΔL^*) of heat-treated wood due to different heat-treatment conditions:

ΔL^*	**180°C/1h**	**180°C/2h**	**180°C/3h**	**200°C/1h**	**200°C/2h**	**200°C/3h**
Spruce	-9	-10	-17	-25	-36	-51
Pine	-4	-19	-22	-26	-36	-44
Lime	-16	-23	-24	-37	-45	-50
	180°C/2h	**180°C/3h**	**180°C/4h**	**200°C/2h**	**200°C/3h**	**200°C/4h**
Beech	-11	-13	-17	-15	-24	-34

Source: Authors

Table 3: Total color change (ΔE^*) of heat-treated wood due to different heat-treatment conditions:

ΔE^*	**180°C/1h**	**180°C/2h**	**180°C/3h**	**200°C/1h**	**200°C/2h**	**200°C/3h**
Spruce	10.72	11.58	17.97	25.98	36.47	51.17
Pine	4.47	19.47	22.29	26.02	36.14	44.20
Lime	16.16	23.11	24.27	37.13	45.14	50.17
	180°C/2h	**180°C/3h**	**180°C/4h**	**200°C/2h**	**200°C/3h**	**200°C/4h**
Beech	11.20	13.38	17.15	15.54	24.08	34.12

Source: Authors

Mass loss

The mass loss values obtained for each heat tretament condition and each species are given in Table 4.

Table 4: Mass loss of heat-treated wood due to different heat-treatment conditions (Mean ± Std):

Mass loss (%)	**180°C/1h**	**180°C/2h**	**180°C/3h**	**200°C/1h**	**200°C/2h**	**200°C/3h**
Spruce	0.93±0.1	1.43±0.1	1.63±0.2	1.85±0.2	2.94±0.2	4.21±0.4
Pine	0.96±0.1	1.45±0.2	1.84±0.3	2.14±0.2	3.24±0.4	4.48±0.3
Lime	1.34±0.2	1.75±0.1	2.13±0.2	4.27±1.1	4.96±0.4	6.75±0.4
	180°C/2h	**180°C/3h**	**180°C/4h**	**200°C/2h**	**200°C/3h**	**200°C/4h**
Beech	1.10±0.1	1.36±0.1	1.56±0.1	4.03±0.5	5.38±0.6	8.59±3.0

Source: Authors

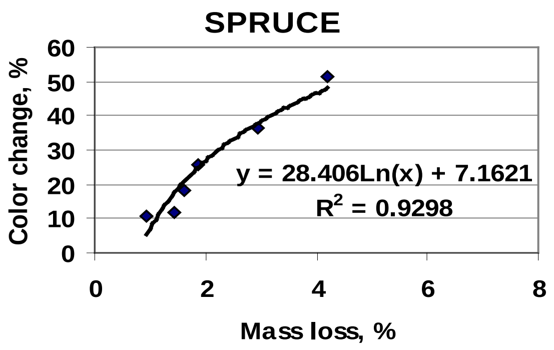
It can be noticed that with all four wood species, the mass loss increased with increasing severity of the heat treatment conditions:

- the raise of temperature from 180°C to 200°C at the same duration (3h) caused a mass loss increase by 61% (for spruce) up to 82% (for beech);
- increasing the treatment duration from 1h to 4h at the same temperature caused a mass loss increase by 43% (for lime) up to 51% (for pine) at 180°C and by 54% (for lime) up to 61% (for pine) at 200°C.

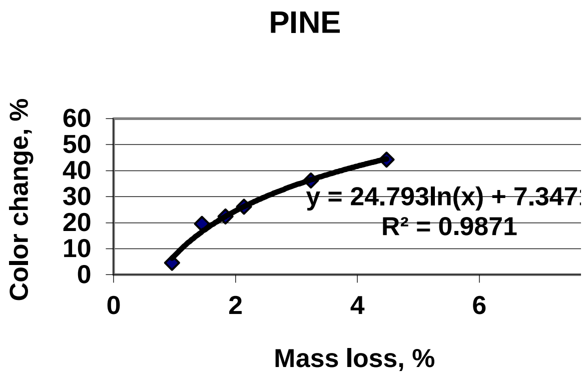
The results concerning the mass loss due to different heat treatment conditions lead to the following conclusions:

- spruce is the least sensitive of the four analyzed species; the heat treatment at 180°C leads to insignificant mass losses; even at 200°C/3h the mass loss is still below 5%, so even more severe conditions (higher temperature, longer duration) can be tested;
- black pine has by up to 18% higher mass losses than spruce; same as with spruce, the heat treatment at 180°C leads to insignificant mass losses, but at 200°C/3h it almost reaches the threshold value ML=5%; fissures begin to occur; therefore, longer treatment is not recommended;
- lime is the most sensitive to temperature of the four analyzed species; the heat treatment at 180°C has no significant effect upon mass loss, but at 200°C/2h it already reaches the threshold value ML = 5%; further maintaining for one more hour at 200°C increases the mass loss up to 6.75%;
- beech is also sensitive to temperature; the heat treatment at 180°C has no significant effect upon mass loss, but at 200°C/3h it already reaches and exceeds the threshold value ML = 5%; fissures begin to occur; therefore, longer treatment is not recommended.

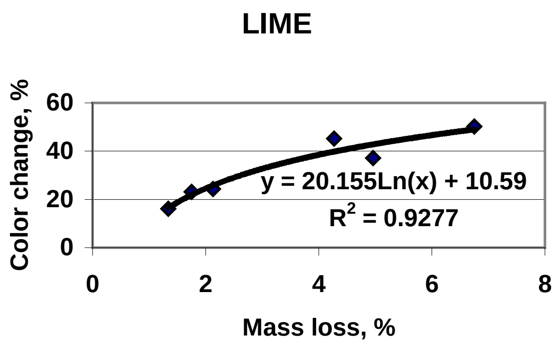
Figure 2: Total color change (ΔE*) vs. mass loss (ML) caused by different heat-treatment conditions at different wood species



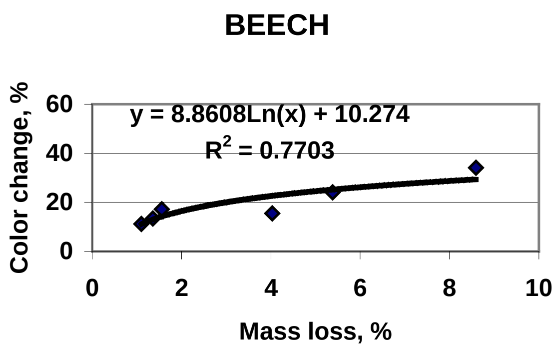
(a) Spruce



(b) Black pine



(c) Lime



(d) Beech

Source: Authors

The correlation between the total color change (ΔE*) and the mass loss (ML) for each species is plotted in Figure 2. All regression equations found are logarithmic, of type: $\Delta E^* = a \cdot \ln(ML) - b$, with coefficients of determination $R^2 = 0.93–0.99$ for the soft species (spruce, pine, and lime), and $R^2 = 0.77$ for beech.

Figure 2 clearly illustrates that the resinous species (a and b) experienced lower mass losses and higher darkening degree than the broad-leaved species (c and d), as their curves are steeper. Beech (d) experienced the lowest degree of darkening ($\Delta E^*_{max} = 30$, compared to $\Delta E^*_{max} = 50$ for spruce and lime).

Conclusion

This paper focuses on establishing a mathematical correlation between two of the most important indicators of heat treatment efficiency—color change and mass loss. The results of an experimental study conducted, with four European wood species, were determined for each species and each heat treatment condition. Based on the statistically processed experimental data, the dependency between the two parameters was shown as a graph for each species and the regression equation were determined. For all species, these curves turned out to be logarithmic of type $\Delta E^* = a \cdot \ln(ML) - b$.

The mathematical correlation found between the color change and the mass loss for each of the four species is a user-friendly tool, which enables one to indirectly determine the expected mass loss in case of heat-treated spruce, black pine, lime or beech wood simply by considering the change in wood color.

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