# COLOUR STABILITY OF THERMALLY MODIFIED HARDWOOD

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## Abstract

Thermal wood modification has been intensively studied in the recent decades because of the possibility to produce wood with improved biodurability and dimensional stability without use of harmful chemicals. Beside altered physical characteristics, wood colour is changed to lighter or darker brown as a result of thermal treatment. Growth of interest in thermal wood treatment has stimulated numerous researches concerned with discoloration of thermally modified wood which is subjected to light exposure. The objective of this study was to evaluate the colour stability of thermally modified hardwood during storage in the dark where wood discolouration is not photoinduced but rather a result of oxidative ageing. Three thermally modified hardwood species – aspen (*Populus tremula* L.), alder (*Alnus incana* Moench), birch (*Betula pendula* Roth.), where investigated. Wood discoloration was monitored by spectrophotometrical measurements of reflectance spectra and chromaticity parameter calculations using CIELAB colour system where L* is the lightness, and a* and b* are the chromatic coordinates. The colour stability of thermally modified wood as well as of untreated wood of the same species was examined by means of assessment of the colour parameter changes (ΔL*, Δa*, Δb*, ΔEab). All wood specimens under study discoloured during the experiment, but the colour change did not exceed two units that is common and accepted for wood products. Untreated and thermally modified wood showed different trends of discoloration during storage in the dark. The final colour changes that were fixed at the end of the experiment were greater for the thermally treated wood.

**Key words**: hardwood, thermal modification, colour stability.

## Introduction

Thermal wood treatment is a wood modification method that enhances the dimensional stability, hydrophobicity and biodurability of wood without using any harmful chemicals (Ates et al.; 2009, Severo et al., 2012, Andersons et al., 2013). The production of thermally modified wood has been increasing during the last decades (Sandberg et al., 2013). The thermal treatment always results in darkening of wood, which is often attributed to the formation of coloured degradation products from hemicelluloses and extractive compounds (Sundqvist and Moren, 2002; Windeisen and Wegener, 2008) and oxidation products such as the quinones-like substances (Bekhta and Niemz, 2003; Gonzales-Pena and Hale, 2009). The brown colour of thermally modified wood has often been regarded as an additional advantage of it. Sometimes heat treatment is purposely used to change the aesthetic properties of wood and thermally modified wood is used as a substitute for some tropical hardwoods (Miklecic et al., 2011). Wood colour is one of the most valuable characteristics for its use as a decorative material, as it is an important aesthetic component. The colour of solid material is a composite property, the value of which is related to its basic chemical composition. The absorption of visible light for a material is characteristic and is caused by molecules called chromophores, which are able to absorb certain wavelengths region of the visible light. For decorative end-use, the colour stability of wood is a significant prerequisite. Colour stability is usually examined by studying the rate and amount of discoloration that wood surface undergoes when exposed to light. In order to study colour changes, colour has to be measured in an objective way. Most wood colour studies quantify the colour by the CIELAB method developed by the Commision Internationale de l'Eclairage (CIE) (International Commission on Illumination). The CIEALB system is a colour model, which describes each colour by a three-dimensional coordinate system (Pauler, 2002). Numerous studies have been made to establish the effect of light on wood and cellulosic materials. Mainly ultra-violet light (300 to 400 nm) and, to a smaller extension, also the shorter wavelength region of the visible light (400 to 500 nm) are known to be the main reason for the discolouration and degradation of wood surface exposed to light (Kataoka et al., 2007). It has been discovered that the colour of thermally modified wood also is not light resistant and discolours under light exposure (Yildiz et al., 2011). It has also been established, that due to changed chemical structure, the photodiscolouration of thermally modified wood varies from that of untreated wood (Ayadi et al., 2003; Ahaji et al., 2009). However, most of the investigations have been concerned to photoinduced discoloration of thermally modified wood. The objective of this study was to evaluate the colour stability of thermally modified hardwood, which was stored in the dark where wood discolouration is not photoinduced, but it is rather the result of oxidative ageing. Three thermally modified hardwood species – aspen (*Populus tremula* L.), gray alder (*Alnus incana* Moench) and birch (*Betula pendula* Roth.), were studied.

## Materials and Methods

Three hardwood species were used in this study - aspen (*Populus tremula* L.), gray alder (*Alnus incana* Moench) and birch (*Betula pendula* Roth.). Wood boards measuring 1000 × 100 × 25 mm were thermally modified in a laboratory experimental wood modification device produced by Wood Treatment Technology (WTT). The modification was carried out in a water vapour medium under elevated pressure (0.6 MPa) at 170 °C for one hour. After modification boards were conditioned at 20 °C and 65% relative humidity for a month. The conditioned boards were cut into specimens measuring 70 × 50 × 20 mm and the surfaces of the specimens were planned. In the same way, specimens from untreated boards of the same species were prepared.

Wood colour and reflectance spectra measurements were carried out using a Minolta CM-2500d spectrophotometer equipped with an integrating sphere, with standard light source D65 at an observation angle of 10 ºC, the diameter of the light spot was 8 mm. The scanning wavelength was from 360 to 740 nm. The reflectance spectra were recorded against a white optical standard. Each reflectance spectrum was the average of three measurements. The spectrum of the reflected light, collected at 10 nm intervals were converted into the CIELAB colour system, where parameter L* describes the lightness (from zero – black to 100 – white) and parameters a* and b* describe the chromaticity coordinates on the green-red and yellow-blue axis, respectively. From the L*, a*, b* parameter values obtained, the differences of the colour parameters ΔL*, Δa*, Δb* and the total colour changes ΔEab were calculated. These values have often been used to trace colour modifications during modification and weathering of woods (Ayadi et al., 2003; Miklecic et al., 2011; Buchelt et al., 2012). The equation used for the colour changes ΔEab calculation was as follows:

$$\Delta E_{ab} = \left( \left(\Delta L^*\right)^2 + \left(\Delta a^*\right)^2 + \left(\Delta b^*\right)^2 \right)^{\frac{1}{2}}, \qquad (1)$$

where: ΔL*, Δa*, Δb* are the changes between the initial and the interval values.

Considering the inherent colour inhomogenity of wood, the measurements were always performed on the same three marked locations and colour of each specimen was described by the arithmetic mean of the three measurements.

Initial colour measurements were performed 24 hours after the specimens were planed. Throughout the experiment the specimens were kept in dark at ambient room temperature and relative humidity and withdrawn only to make colour measurements. Colour measurements for all specimens were made at intervals throughout the test period. The total length of the experiment was 270 days, which was enough time for the colour changes of the thermally modified wood to reach the plateau.

## Results and Discussion

Reflectance spectra of untreated wood of the three studied species noticeably differed from each other (Fig. 1).

The spectra varied both by reflectance rate and pattern. This is in accordance with the CIELAB colour parameter values recorded in the Table 1. The data showed that the three wood species differed from one another by their colour parameter values. The lightness parameter L* and redness parameter a* values showed the greater differences among species. Untreated aspen wood was characterized by the highest reflectance rate throughout the entire visible light spectrum (Fig. 1) as well as the highest lightness and the lowest chromaticity parameter a* and b* values (Table 1). On the contrary, alder wood had the lowest reflectance in the whole range of the visible

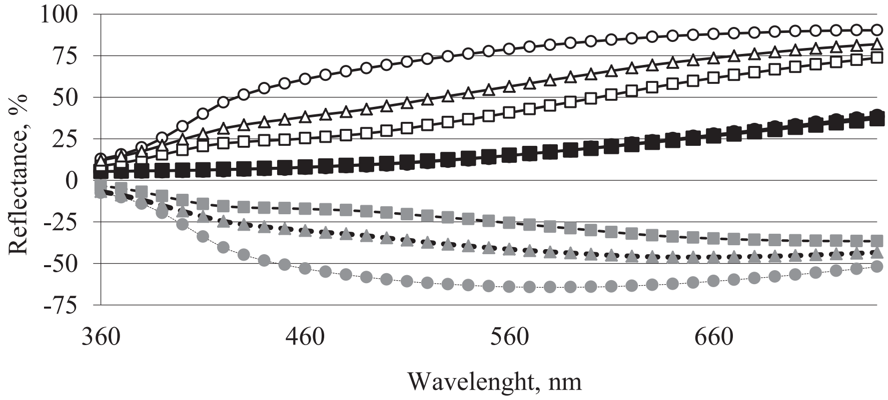

Figure 1. Reflectance spectra and reflectance difference spectra of untreated and thermally modified (170 °C) aspen, alder and birch wood: black symbols – thermally modified wood; white symbols – untreated wood; gray symbols – reflectance difference; ○ (circles) – aspen; □ (squares) – alder; D (triangles) - birch.

Table 1

**Colour parameters and colour changes of untreated and thermally modified (170 °C) aspen, alder and birch wood**

| Type of sample | | L* | a* | b* | DL* | Da* | Db* | DEab |
|---|---|---|---|---|---|---|---|---|
| Thermally modified | Aspen | 45.0 (2.2) | 10.6 (0.3) | 20.7 (2.3) | -45.2 | 9.7 | 4.2 | 46.5 |
| | Alder | 45.2 (0.6) | 9.5 (0.3) | 20.1 (0.2) | -24.0 | -0.6 | -1.1 | 24.0 |
| | Birch | 45.2 (2.1) | 10.5 (0.3) | 20.4 (1.3) | -33.6 | 4.4 | 0.2 | 33.9 |
| Untreated | Aspen | 90.2 (1.4) | 0.9 (0.7) | 16.5 (1.7) | ´ | ´ | ´ | ´ |
| | Alder | 69.2 (1.3) | 10.1 (0.3) | 21.2 (0.8) | ´ | ´ | ´ | ´ |
| | Birch | 78.8 (3.1) | 6.1 (1.1) | 20.2 (1.4) | ´ | ´ | ´ | ´ |

* Values in parenthesis are standard deviations

light spectrum as well as the lowest lightness and the highest chromaticity parameter values.

Thermal modification induced noticeable changes in wood colour of all investigated species and wood with dark brown colour was produced. During the thermal modification, the reflectance of wood obviously diminished in the whole range of the visible light spectrum for all wood species (Fig. 1). It implies that components absorbing visible light were formed during thermal modification. Contrary to untreated wood, the reflectance spectra of the thermally modified wood for all the species were nearly equal. The decrease of the reflectance rates was consistent with the changes in lightness ΔL* (Table 1) which went towards darkening (negative lightness difference ΔL* values) for the all species but in different magnitude. The changes in the chromaticity coordinates a* and b* did not follow one common pattern for the three wood species. For aspen and birch wood both coordinates a* and b* increased and bigger changes occurred to redness coordinate a*.

Inversely, chromaticity coordinates of alder wood decreased and the bigger change occurred to yellowness coordinate b*. The observed differences are in accordance with findings of T. Schnabel et al. (2007) and B.M. Esteves et al. (2008) who have stated, that different wood species undergo dissimilar colour changes during heat treatment even under similar modification conditions.

The reflectance difference spectra (Fig. 1) show that the reflectance decreased in the whole range of visible light for all the wood species as a result of the thermal treatment. There was no distinct maximum on the reflectance difference spectra. This finding indicates that a variety of chromophores with different characteristic absorption wavelengths was produced during thermal modification.

Colour changes during storage in the dark are mainly related to readily oxidizable chemical compounds on the wood surface that undergo oxidative reactions resulting in an alteration of the chromophoric groups. During the experiment, all the studied specimens discoloured but at different rate and magnitude (Fig. 2).

Untreated alder and birch wood showed similar pattern of discolouration. After rapid initial colour

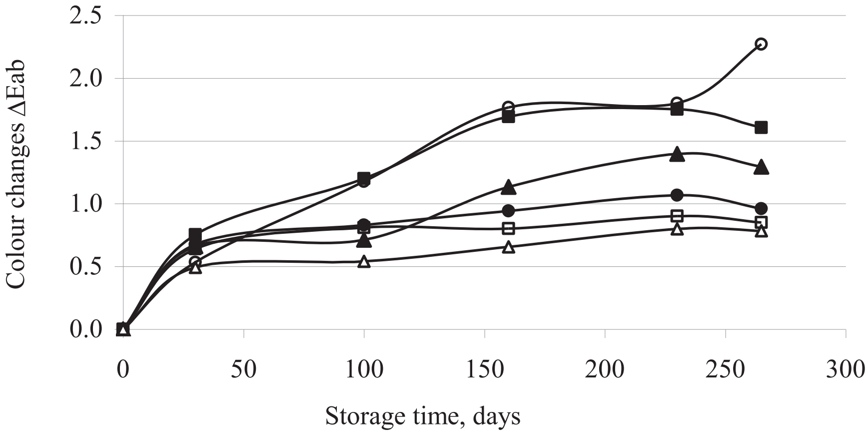

Figure 2. Colour changes ΔEab of thermally modified (170 °C) and untreated aspen, alder and birch wood as a function of storage time in the dark: filled symbols – thermally modified wood; open symbols – untreated wood; ○ (circles) – aspen; □ (squares) – alder; D (triangles) – birch.

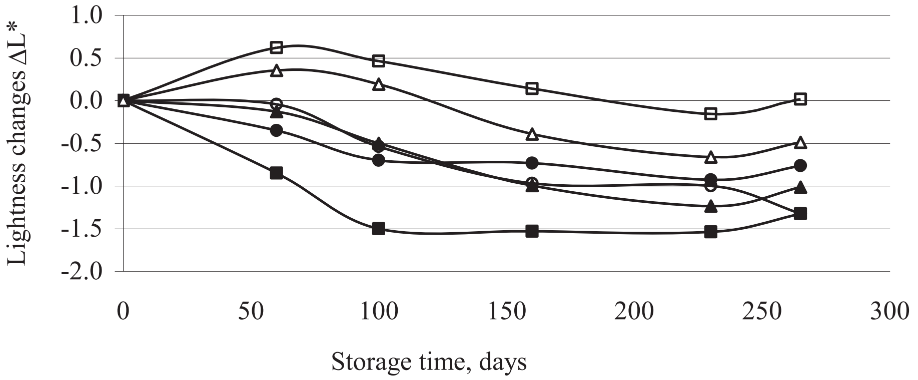

Figure 3. Lightness changes DL* of thermally modified (170 °C) and untreated aspen, alder and birch wood as a function of storage time in the dark: filled symbols – thermally modified wood; open symbols – untreated wood; ○ (circles) – aspen; □ (squares) – alder; D (triangles) – birch.

changes during the first 30 days, the discoloration subsided and wood attained almost a constant colour change DEab values. After storage for 270 days in the dark, untreated alder and birch wood showed the minor colour changes ΔEab, which were even less than one unit. Thermally modified aspen wood also showed colour changes of almost the same value. The rapid initial discolouration after planning can infer that some chemically reactive substances with chromophoric groups that do not need any photoexcitation to be oxidized, were exposed on the wood surface after wood processing. Untreated aspen wood was the most unstable one with respect to discoloration during storage in the dark. At the end of the experiment, the colour change ΔEab of untreated aspen wood had reached value of 2.3 units and there was no evidence that the process of discolouration had stopped. Aspen wood varied from the other untreated woods also by the pattern of colour changes. Untreated aspen wood darkened throughout the entire experiment (Fig. 3) and by this differed from the other untreated specimens, which became lighter at first.

The reason of such a different behaviour of the three studied hardwoods could be the differences in their chemical composition, mainly the difference in content and composition of extractives. It has been established that some extractives have an antioxidant capacity and play important role in colour evolution and change of wood (Ahaji et al., 2009; Varga and Van der Zee, 2008).

The colour changes of all thermally modified wood species (Fig. 2) increased up to approximately 220 days and then a slight decrease can be seen at the end of the experiment. The pattern of discolouration of the thermally modified wood can be related to the lightness changes ΔL* (Fig. 3). The correlation between the trends of colour change ΔEab and lightness change ΔL* for thermally modified wood agrees with the finding that the major contributor to the colour changes, in the case of thermally

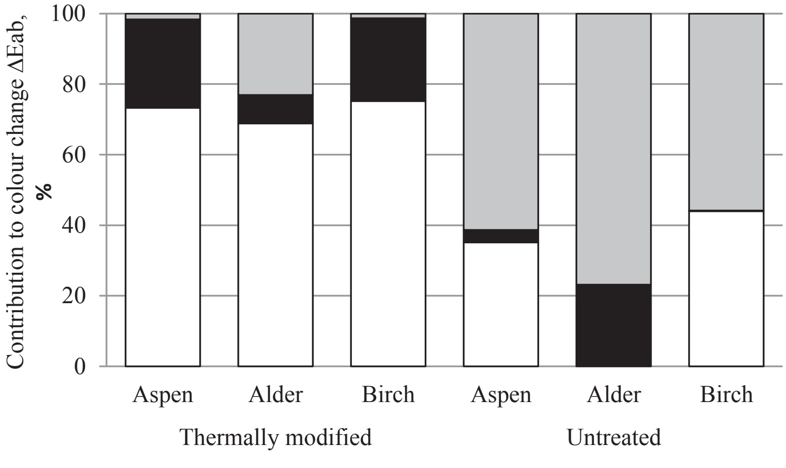

Figure 4. Contribution of each color parameter changes (DL*, Da*, Db*) into total color changes DEab after storage in dark for 270 days for untreated and thermally modified (170 °C) aspen, alder and birch wood: white - ΔL*; black - Δa*; grey - Δb*.

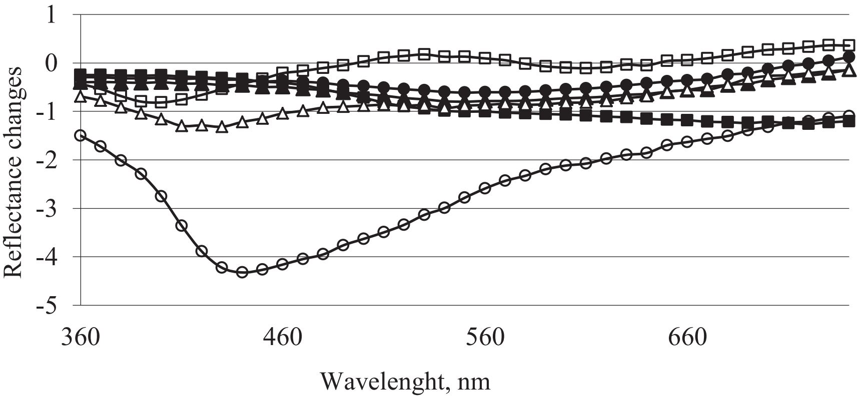

Figure 5. Reflectance difference spectra of untreated and thermally modified (170 °C) aspen, alder and birch wood after storage in the dark for 270 days: filled symbols – thermally modified wood; open symbols – untreated wood; ○ (circles) – aspen; □ (squares) – alder; D (triangles) – birch.

modified wood, were the lightness change ΔL* values (Fig. 4).

For thermally modified wood, the lightness change DL* values contributed more than 70% to the total colour changes DEab. In the case of untreated wood, the bigger contributors were chromaticity parameters.

Alternating directions of the lightness changes ΔL* were observed for all the specimens during the experiment. As it was mentioned above, untreated woods showed different patterns concerning the lightness changes. Unlike it, common trend for thermally modified specimens was observed. Colour of all the thermally modified specimens became darker throughout the entire experiment. It was indicated by decreasing values of lightness changes DL* (Fig. 3). The results of final measurements showed that the darkening had stopped and even a tendency of slight lightening can be noticed at the end of the experiment.

The results showed that the pattern of colour changes due to oxidative aging differed between the untreated and thermally modified specimens and also among untreated wood species under study. It was also supported by reflectance difference spectra for the reflectance changes at the end of the experiment (Fig. 5).

It can be seen that at the end of the experiment, reflectance of untreated aspen wood had considerably decreased in comparison with other examined specimens. Pronounced band of reflectance difference had appeared at a wavelength region 380 to 560 nm. It indicates that chromophoric groups were formed with characteristic absorbance in the shorter wavelength region of the visible light. Reflectance reduction in the same wavelength region, although on a noticeably smaller scale, can be observed also for untreated alder and birch wood. The increase in absorption of violet, blue and green light (400 – 520 nm) agrees with the finding that the yellowness parameter changes Δb* contributed more than 50% into the total colour change ΔEab of untreated wood (Fig. 4).

At the end of the 270 days experiment, the final colour changes were greater for the thermally modified wood than for the untreated wood with the exception of untreated aspen wood. This is in contrast to findings that the colour of thermally modified wood is more stable than that of untreated wood when exposed to ultraviolet light (Rosu et al., 2010; Miklecic et al., 2011). Obviously, thermally modified wood contains relatively more components with chromophoric groups that can react with oxygen without any photoexcitation.

After storage in the dark for 270 days, the colour changes ΔEab were in the range between one and two units for all the modified wood species. Colour differences of the magnitude one to two units are absolutely common and accepted for wood products (Buchelt and Wagenfuhr, 2012). Such differences are considered as an observable colour difference, which is barely perceptible. This indicates that thermally modified wood inhere stable colour when not exposed to light.

## Conclusions

1. Freshly planed surfaces of both untreated and thermally modified aspen, alder and birch wood discolour during storage in the dark. The colour changes DEab of thermally modified aspen, birch and alder wood do not exceed two units, which is common and acceptable for wood products.
2. Untreated and thermally modified wood shows different trends of discoloration during storage in the dark. For thermally treated wood, the dominant changes occur with the lightness parameter L* values while the yellowness parameter b* is most changed in untreated wood.

3. The colour changes after storage for 270 days in the dark were greater for the thermally treated wood. It indicates that thermally modified wood contains relatively more components with chromophoric groups that can react with oxygen without any photoexcitation.

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