

THESES OF THE DOCTORAL (Ph.D.) DISSERTATION

**Colour change of the most important wood species in the furniture
industry, caused by the photodegradation**

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1. Actuality and objective of the dissertation

The colour is the conscience deriving from the spectrum of light interacting in the eye. The colour stimulus is generated by the visible light with wavelength between 380 nm and 760 nm. In the human eye, cone cells are the receptors of the colour perception, and have three types, each with different pigment, namely: S-cones, M-cones and L-cones. Each cone is therefore sensitive to visible wavelengths of light that correspond to short-wavelength, medium-wavelength and long-wavelength light.

In terms of use, colour is one of the most important physical features of wood. Natural wood colour is between red and yellow, however the colour of the end product may change continuously, during ageing, depending its finish and the exposure. In case of indoor applications, the colour change is induced by photo-degradation and slow thermal degradation, while for outdoor use the leaching effect of the rain must also be considered.

The change of the wood colour is caused mainly by ultraviolet radiation.

Energy of the photons in the UV region is high enough to free electrons in chemical bonds, thus the structure degrades. Wood in outdoor usages takes a lot of impacts, from these impacts the sun radiation is the most intensive. Beside the sun radiation, moisture has an important role in surface degradation. Degraded wood parts are leached out by rain ensuring the path for the degradation of further layers. The colouring compounds of the wood surface are completely washed out during a one-year outdoor usage and the surface becomes grey. During indoor ageing the wood surface turns to dark and brown. The indoor colour change is the most conspicuous in case of light coloured wooden applications, primarily for pine wood products.

Due to the customer demands the end users must be informed about the changes of the products during its life cycle. Colour change of the furniture is one of these possible changes. Therefore it is important to research and clarify the time dependence of colour change of indoor wooden applications.

Colour change of wood can be described by objective colour measurement. For this purpose, colorimeter devices can be used. Although the demand for objective colour measurement is increasing in the industry, these devices are used mostly in laboratory researches.

The basic criterion of colorimetry is to elaborate a colour stimulus area in which every colour is demonstrated by a single point. For this method CIELAB colour stimulus area is used developed by the CIE. In this system the colour dots are described by the L^* , a^* and b^* colour coordinates.

The subject of my dissertation, as stated above, is the colour change of the 15 most important wood species in the Hungarian furniture industry caused by photo- and thermal degradation.

Based on the result of these investigations, the effect of light radiation and temperature on the studied wood species can be exactly described, thus the colour change of the given wood species due to a given light irradiation at a given temperature can be forecasted as time elapses. In this way, the colour change of the investigated wood species can be projected.

Investigation of the changes on outdoor wood surfaces exposed to sunlight is, however, obstructed by several circumstances. The intensity of the sun radiation changes not only

during the day, but it depends also on the inclination angle of the sunbeams altering with the change of seasons. Furthermore, the calculation of the real sunshine duration gets completely impossible due to the unexpected cloudy periods. The rain may leach out the water-soluble compounds. Air humidity also affects the colour change, but it may change continuously during the day. Considering all these unpredictable circumstances, photo degradation of the wood is usually examined using artificial light sources. Therefore, also in frame of this doctoral research an artificial light source was used. For comparison, colour change of samples placed behind window and irradiated to sunlight was also studied.

2. Applied materials and research methods

In this doctoral work, 15 most important wood species used in furniture industry were involved. Pine wood species: scotch pine (*Pinus Sylvestris* L.), spruce (*Picea abies* Mill.) and red pine (*Larix decidua* L.), frondous wood species: black locust tree (*Robinia pseudoacacia* L.), wild cherry (*Prunus avium* L.), black cherry (*Prunus serotina* Ehrh.), walnut (*Juglans regia* L.), alder (*Alnus glutinosa* L.), linden, (*Tilia cordata* Mill.), sycamore maple (*Acer pseudoplatanus* L.), european ash (*Fraxinus excelsior* L.), pannonia poplar (*P.x euramericana* Pannonia), silver birch (*Betula pendula* Roth) and sessile oak (*Quercus petraea*) were examined.

The surfaces of the pine wood species contain both light early wood and dark late wood, therefore radial sections were measured. Steamed beach samples were also examined (steaming temperature: 95°C, steaming time: 24 h).

Four different tests were performed: 800 W mercury-vapour lamp irradiation at 30 and 80°C, thermal treatment at 80°C in dense darkness, and natural sunlight irradiation behind window.

Light irradiation tests were performed in a climate chamber with adjustable temperature using two mercury-vapour lamps as light source. The joint power of the two lamps was 800 W and the distance between the samples and the light sources was 64 cm. 80% of the emission of the mercury vapour lamp was in the ultraviolet (UV) region. 31% of the emitted UV light was in UV-A (380-315 nm) region, while 24 % of that was in UV-B (315-280 nm) and 25 % in UV-C (<280 nm) region. The temperature within the chamber was stabilized at 30°C and at 80°C. 20 samples were irradiated for each species.

Colour changes were monitored with a Konica-Minolta 2600d spectrophotometer equipped with a D65 illuminant and a 10° standard observer. The sensor head was 8 mm in diameter. White etalon was used for calibration.

The colour was measured with interruptions after 0, 8, 20, 40, 90 and 200 hours of treatment for all treatments. The surface colour was measured on 10 points on each sample, thus the results are the average of 200 measurements. Data were then recorded in the CIE L*a*b* system.

3. Summary of the research results

The colour change caused by 80 C° mercury vapour lamp radiation and natural sun light radiation behind window were studied, and the two effects compared. Depending on the changes, the examined wood species can be classified into three distinct groups. Pine woods and ash belong to group 1, black locust, linden, alder, walnut and cherry wood belong to group 2, while steamed beech, maple, poplar, birch, and oak wood belong to the third group. The research proved the followings:

- On a long term, wood species in the second group have the most stable colour against photo degradation although these species showed the most intensive changes at the beginning of the treatments. The colour of this group changed intensively in the first 8 hours, but the modified colour was proved to be stable, even in case of the strong UV emitter mercury vapour lamp.
- The birch showed the smallest colour change.
- Pine and ash has the most unstable colour. These wood species showed continuous colour change during the studied period.
- The complete colour change (E*) could not give as detailed information about the colour change as the detailed analysis of single colour coordinates.
- Results also show that the colour change is a complex process. Light radiation degrades the chromophore chemical groups in wood, but at the same time new chromophore groups arises due to the oxidation which follows the degradation of the extractives. The scale of the two changes varies depending on the species, however it significantly depends on the distribution of the light radiation wavelength.
- In case of sunlight, during the first 8 hours of the treatment, for most of the wood species the degradation of the existing chromophore groups was more intensive than the development of the new chromophore groups.
- Results confirm former observation that the mercury vapour lamp is not suitable to imitate short-time sunlight radiation.

During the research the colour change caused by mercury vapour lamp radiation at 30 and 80°C with the results of the treatment in dense darkness at 80°C temperature was compared and the followings were found:

- Temperature has a strong effect on wood photodegradation.
- The same light irradiation caused much higher red colour coordinate growth at 80°C than at 30 C°.
- Pine wood samples suffered 57 % higher red colour change at 80°C than at 30 C°.
- Wood species with less extractives showed less red colour change. In accordance with the literature, it can be stated that the extractives have a decisive role on red colour change.
- The yellow colour showed two alterations. Photodegradation caused yellow colour increase, while the thermal effect resulted in its decrease. The effect of photodegradation was stronger on yellow colour change than that of the thermal degradation.

Based on the statistical analysis of the measured data the followings can be stated:

- In case of the majority of the examined wood species the light irradiation increased the colour inhomogeneity of the samples.
- Black locust and black cherry showed the highest increase in terms of standard deviation. In case of these species the deviation of the lightness was the highest with about 5 units, while the deviation for the other two coordinates was about 2 units.
- Slight, but obvious colour homogenization was found only in case of pine wood.
- For the other studied wood species only a slight light irradiation induced colour homogeneity change was observed.

Based on the study of the untreated samples the followings can be stated:

- There is a correlation between the hue angle and the lightness.
- The colour dots of beech, pine, alder, linden, maple, sour cherry, ash, spruce, poplar and birch fit well on a positive rising line with a correlation coefficient of 0,91.

Colour dots of heartwood with high extractive content like oak, walnut, locust, and turkey oak are far from the line.

4. The thesis's of the dissertation

1. The colour change effect of the photodegradation is more intensive in case of coniferous wood species than in case of deciduous wood species. This difference was observed both in the first intensive colour change period and in the subsequent period with moderate changes. The behaviour of the ash was similar to that of the coniferous wood species.

2. It was proved that based on the photodegradation induced colour change the conifers can be divided into two groups. Species in the first group (Acacia, late cherry, wild cherry, alder, linden and walnut) suffered intensive colour change in the first period followed by a stagnant colour change period. In case of the second group (beech, maple, poplar, birch and oak) the initial period was less intensive than that for the first group, but it was followed by a gentle colour change.

This prolonged colour change process was much less intensive than that of the coniferous wood species.

3. Measurements proved that the photodegradation increases the inhomogeneity of wood surface colour. The only exception was the pine where a minor homogenization effect was observed.

Acacia and American cherry showed the most intense inhomogeneity increase.

4. It was found that the colour change effect of sunlight behind glass is significantly different from that of the mercury vapour lamp irradiation in the first 20 hours of the treatment. In this period, the mercury vapour lamp irradiation induced an intense and unidirectional change.

Contrarily, in case of the sunlight irradiation behind glass, mainly a constant colour was observed with both some decrease and increase.

5. It was demonstrated that the same light irradiation causes significantly higher red colour increase at 80°C than at 30°C. Pine samples suffered 57% higher red colour change at 80°C than at 30°C. Wood species with the least extractive content (for example the poplar) suffered the smallest red colour change. In line with the results found in the literature, it was proved that extractives have a decisive role in terms of red colour change. This effect must be considered if wooden applications are exposed to direct sunlight.

6. Measurements verified that there is a linear correlation between the lightness and the hue angle of wood in case of Hungarian untreated wood species, except those with high extractive content.

In case of species with high extractive content, such as oak, beech and walnut, the deflection from linearity depends on the extractive content.

Publications related to the topic of the Dissertation

Tolvaj L; Persze L; Albert L (2011)

Thermal degradation of wood during photodegradation

Journal of Photochemistry and Photobiology B: Biology 105: 90–93.

Persze L; Tolvaj L (2012)

Photodegradation of wood at elevated temperature: Colour change

Journal of Photochemistry and Photobiology B: Biology 108: 44-47.

Tolvaj L; Persze L; Lang E (2013)

Correlation between hue angle and lightness of wood species grown in Hungary

Wood Research 58:(1) 141-145.

Tolvaj L; Persze L (2010)

Photodegradation of black locust

The 4th Conference on Hardwood Research and Utilisation in Europe. Sopron, (May 17-18.) 186-194.

Persze L; Tolvaj L (2012)

Superposition of thermal- and photodegradation for wood monitored by colour measurement.

The 5th Conference on Hardwood Research and Utilisation in Europe. Sopron, (2012. sept. 10-11.) 33-40.

Tolvaj L; Takáts P; Persze L (2010)

The role of the steam in the color changing of wood material by thermal treatment on 90°C

Faipar 58: (1) 5-10.

Tolvaj L; Persze L (2011)

Problem of the imitation of sun radiation by artificial light source

Faipar 59: (2-3) 19-26.

Persze L (2011)

Comparison of the photodegradation of the hungarian wood species. Color changing.

Faipar 59: (2-3) 35-49.

Persze L (2012)

Effect of the temepartue to the photodegaradtion of wood. Color changing.

Faipar 60: (2) 5-13.