# COLOUR CHANGE OF PHOTODEGRADED SPRUCE WOOD BY WATER LEACHING

Antal Kannar University of Sopron, Institute of Applied Mechanics and Structures Sopron, Hungary

Laszlo Tolvaj University of Sopron, Institute of Physics and Electrotechnics Sopron, Hungary

Endre Magoss University of Sopron, Institute of Woodworking Machinery Sopron, Hungary

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# ABSTRACT

Spruce samples (*Picea abies* Karst.) were irradiated by strong ultraviolet (UV) light emitter mercury lamp and another series of specimens were treated with the combination of UV radiation and water leaching. The total duration of UV radiation for both series of specimens was 50 days. The colour parameters (CIE L\*, a\*, b\*) were measured and evaluated after both UV radiation and water leaching. The increase of redness value was two times greater than the yellowness increase based on the initial value at the end of 50 days treatment. The leaching partly removed the yellow and red chromophore molecules generated by the UV radiation. The samples become slightly lighter after water leaching. The leached samples increased slightly more in yellowness and redness during the first 4-6 days of UV radiation than those of the dry series. The change of surface roughness of the specimens was monitored by diffuse reflectance infrared spectroscopy, because the change of roughness alters the light scattering properties of the surface. The roughness of the samples increased during the UV irradiation, and the tendency of roughness change mirrored that of yellowness change. The leaching reduced the roughness value in all examined cases. Good correlation was found between the hue angle and the lightness in both dry UV treatment and combined UV treatment and water leaching.

KEYWORDS: Spruce, colour change, photodegradation, leaching by water.

#### **INTRODUCTION**

The colour of wood is important for users, especially for indoor wood products such as claddings, floorings and furniture. The colour inhomogenity of wood generates a beautiful colour harmony. The machined surface in any anatomical planes features the colour diversity of earlywood and latewood. The colour hue of wood gives us the feeling of warmness. The cellular structure of wood includes microscopic "mirrors" aligned parallel to the grain. Gloss from these micro mirrors gives us a more elegant, soft, natural and beautiful texture than that of plastics and metal (Masuda 2001). The main chemical components determining the colour of wood are the extractives (Umezawa 2001).

This beautiful colour of solid wood is sensitive to light and heat. The main factors that cause the greatest changes in the colour of wood are sunlight and rain during outdoor exposure (Tolvaj and Mitsuki 2005, Tolvaj and Papp 1999). During natural weathering, the surface starts yellowing and turns grey indicative of the rainwater washing out the decomposed lignin and extractives (Tolvaj and Papp 1999). Chemical analyses show that the deterioration is primarily related to the decomposition of lignin (George et al. 2005, Pandey and Vuorinen 2008, Popescu et al. 2011, Timar et al. 2016, Cogulet et al. 2016).

The degradation of lignin creates free phenoxyl radicals. These free radicals react with oxygen to produce carbonyl chromophoric groups (Tolvaj and Faix 1995, Pandey 2005, Xie et al. 2005). The chromophoric groups are partly responsible for the discolouration of wood. The extractives in wood are also sensitive to light irradiation and the chromophoric degradation products also generate colour change (Chang et al. 2009, Fan et al. 2010). Detailed research was carried out using 15 wood species to monitor the photodegradation behaviour of the species (Persze 2011, Csanády et al. 2015). The results showed that the red hue chance was highly species dependent due to the differences in extractive content, but the alteration of the yellow hue was largely independent of species. Among wood species, differences of chemical structures of three major cell wall components, cellulose, hemicellulose, and lignin, are few. However, a great diversity in extractive composition is found throughout wood species (Umezawa 2001).

The colour change of wood is a sensitive indicator for detecting the effects of UV light. The colour alteration of black locust wood is detectable after 2–3 hours of light irradiation by the naked eye (Tolvaj and Varga 2012). Rapid colour change usually occurs in the first 50 hours of irradiation followed by moderate colour alteration. The photodegradation of wood generates intensive and continuous yellowing with moderate shift towards red (Oltean et al. 2008, 2010, Sharrat et al. 2009, Miklecic et al. 2011).

The colour coordinates (L\*, a\*, b\*) are highly complex parameters. The calculation of the colour coordinates use different parts of the reflection spectrum applying complicated equations. Because of the complexity of the colour coordinates, there is no direct correlation between the absorption change of the individual chemical groups and the total colour change. The total colour difference ( $\Delta E$ ) is an even more complex parameter than the colour coordinates. Nevertheless, researchers often try to find correlation between total colour difference and the absorption change of individual chemical group.

The leaching effect of rain is the second most harmful effect during the outdoor weathering of wood. However, the effect of water leaching is a hardly investigated phenomenon. Some paper deal with simultaneous light exposure and water leaching during artificial weathering (Pandey and Khali 1998, Kamdem and Greiler 2002, Hansmann et al. 2006, Futa et al. 2013). The combined effect of the two factors was monitored in these studies. Their individual impact was not separated. The aim of this study was to investigate the separate individual colour alteration effects of UV light exposure and water leaching during artificial weathering of spruce wood.

# MATERIALS AND METHODS

Spruce (*Picea abies* Karst.) specimens having low extractive content were chosen for the colour change test. The specimens were prepared with dimensions of  $150 \times 30 \times 10$  mm (long. x rad. x tang.). Two series of 15 specimens each were created, and 10 points were measured on each specimen. The sun radiation and the leaching effect of the rain are harmful factors effecting mostly the degradation of outdoor wood. These effects were simulated by the following experiments. Samples were irradiated by a mercury lamp, and then plunged into distilled water (wet treatment). A double mercury vapour lamp, as a strong UV light emitter, provided the light irradiation. The total electric power of the applied double mercury lamps was 800 W. The UV radiation was 80% of the total emission of the lamps. Specimens were located at a distance of 64 cm from the lamp. The temperature in the chamber was 50°C during the irradiation. The irradiation time was 24 hours, followed by 6 hours water leaching. These two treatments together constituted one cycle, and this cycle was repeated 50 times. The other series of specimens got light irradiation only (dry treatment). All colour data presented in this study are the average of 150 measured data.

The colour of the wood specimens was measured after each (UV and leaching) treatments. The radial surface was used for colour measurement, generating the average colour data of earlywood and latewood. The colour measurement was carried out after both light irradiation and water leaching during the first 10 cycles. During the next 40 cycles the colour data were measured after UV radiation only. Wet samples were dried at 30°C up to the initial weight after each leaching. This process guaranteed that the colour measurement was done at the same moisture content each time. Measurements were carried out with a Konica-Minolta 2600d colorimeter. The CIE L\* (Lightness), a\* (Redness), b\* (Yellowness) colour space data were calculated based on the D<sub>65</sub> illuminant and 10° standard observer with a the aperture diameter of 8 mm.

The change of surface roughness was determined according to the infrared light scattering process (Csanády et al 2011, Tolvaj et al. 2014). The size of specimens was 30 x 10 x 5 mm (long. x rad. x tang.) for IR measurement. The diffuse reflectance infrared Fourier transform (DRIFT) spectrum of the specimens was measured using a spectrophotometer (JASCO FT/IR 6300), applying a diffuse reflectance unit. The resolution was 4 cm<sup>-1</sup> and 64 scans were obtained and averaged. The background spectrum was determined against an aluminium plate.

Wood has no absorption at 1900 and 3800 cm<sup>-1</sup>. It means that the reflection should be 100% at these wave numbers. But the reflection was less than 100% because of light scattering. The scattering depends on the roughness of the surface. The optical roughness was defined as the quotient of two reflection intensities.

Optical roughness = R(3800)/R(1900)

where: R(3800) is the reflection intensity at 3800 cm<sup>-1</sup> and R(1900) is the same at 1900 cm<sup>-1</sup>.

#### **RESULTS AND DISCUSSION**

The main goal of this study was to determine the colour modification of photodegraded wooden surface by water leaching. These alterations are visible in Figs. 1-3 for the 3 colour coordinates. Fig. 1 represents the change of lightness. The first bar represents the initial lightness of the samples. The second bar shows the effect of UV radiation (24 hours), followed by the bar generated by water leaching (6 hours). These two treatments were repeated 10 times. The UV radiation produced continuous lightness decrease, as reported in earlier papers (Cogulet et al. 2016, Tolvaj and Faix 1995, Oltean et al.2010, Sharrat et al. 2009). The water leaching partly washed out the degradation products of the light irradiation. This phenomenon generated lightness increase in all 10 cycles of treatments. The lightness intensity showed very little decrease after the seventh cycle. The lightness values are calculated from the Y tristimulus value. (The other coordinates are calculated using two tristimulus values.) The Y tristimulus value is determined by the total visible part of the reflection spectrum. That is why the lightness change is not useful in determining the individual chemical changes.



Fig. 1: Lightness change caused by UV radiation and water leaching (w).

Fig. 2 shows the redness change during the cyclic UV radiation and water leaching. The UV radiation generated continuous redness value increase during the treatments. This increase was diminished after the seventh cycle. The water leaching partly washed out the water-soluble chromophore degradation products. The greatest leaching effect can be observed during the third cycle. After the seventh cycle, the leaching effect was negligible.



Fig. 2: Redness change caused by UV radiation and water leaching(w).

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Fig. 3 represents the yellowness change during the cyclic UV irradiation and water leaching. The UV radiation produced a substantial yellowness value increase during the first day of treatment, as reported in earlier papers (Cogulet et al.2016, Tolvaj and Faix 1995, Sharrat et al. 2009, Wang and Ren 2008, Persze and Tolvaj 2012). This rapid change was followed by moderate increase up to the fifth cycle. After that, the yellowness value decreased slightly. The water leaching reduced the yellowness intensity in all cases.



Fig. 3: Yellowness change caused by UV radiation and water leaching(w).

Fig. 4 shows the roughness change during the cyclic UV radiation and water leaching. The optically measured roughness increased during the first five cycles of treatment and decreased slowly after it. The greatest change was generated by the first 24 hours of UV treatment. The tendency of roughness change was similar to the change of yellowness. This is because the coloured degradation products of lignin generate most of the yellowness increase. Lignin is the most sensitive macromolecule of wood during photodegradation (Cogulet et al. 2016, Tolvaj and Faix 1995, Pandeya and Vuorinen 2005, Sharrat et al. 2009, Wang, Ren 2008, Persze and Tolvaj 2012, 2008, Prekle tet al. 2018). Water leaching reduced the value of roughness in all cases. The roughness values measured after water leaching do not exactly follow the change of yellowness. The reason is that lignin degradation followed by oxidation of free radicals generates not only coloured chemical compounds, unconjugated carbonyl groups are also generated. One example is  $\gamma$  lactone (Tolvaj and Faix 1995, Csanády et al. 2015, Varga et al. 2017). The leaching of unconjugated carbonyls affects the roughness but do not change the yellowness.



Fig. 4: Roughness change caused by UV radiation and water leaching (w).

#### WOOD RESEARCH

Earlier figures showed the detailed effects of UV radiation and water leaching during ten cycles. The experiments were continued up to 50 cycles, but the colour measurement was performed only after 12, 16, 21, 25, 30, 42 and 50 cycles. Figs 1 through 3 showed that the leaching effect was small right before the tenth cycle. This is why the cumulative effect of leaching was determined compared to the effect of pure UV treatment (dry treatment) in the second part of investigation. The changes are presented based on UV irradiation time as independent variable.

The greatest difference between the effects of dry and wet treatments was found for the yellow colour coordinate (Fig. 5). The irradiation time in Fig. 5 include the duration of UV irradiation only, for the correct comparison of the two types of treatments (leaching time is not included). The yellowness value increased rapidly up to the fourth cycle. The curve of leached samples ran slightly above the curve of dry UV treatment during the first four cycles. After this point the two curves separated and the curve of leached samples was under the curve of dry UV treatment. The distance between the two lines increased with irradiation time. This fact shows that yellow chromophores were leached out continuously by water. The exact determination of the types of leached molecules requires further chemical investigation. The yellowness intensity of dry samples increased up to 12 days of UV radiation, and remained almost constant during further treatment. The yellowness value of leached samples increased during the first four days of treatment only, and slightly decreased after that. The 50 days UV radiation multiplied the initial yellowness value 2.13 times, and the leaching reduced this factor up to 1.71.



Fig. 5: Yellowness change caused by UV radiation (Dry) andby UV radiation and water leaching (Leached).

Three phenomena contributed the yellowness intensity change. The UV light degraded the chromophore compounds of wood reducing the yellowness value. The chromophore products of lignin degradation intensified this value but the leaching mitigated it. During the first four cycles the effect of lignin degradation was dominant. Between the fourth and the twelfth cycles, the effect of leaching compensated for that of the yellow chromophore production and the intensity of yellowness remained constant.

After the twelfth cycle the leaching was slightly more dominant than the photodegradation and the yellowness value decreased. The reason, for the yellowness values of leached samples being slightly above the values of dry UV treatment in the first four cycles might be that the leaching opened new surface for the photodegradation.

Fig. 6 shows the redness change during the 50 cycles UV radiation and water leaching. Pure UV radiation (dry treatment) generated intensive redness value increase during the first twelfth days of irradiation. After this period, the redness value had a plateau for up to 25 days of exposure. Further exposure generated slight redness value increase. The curve of leached samples ran slightly above the curve of dry UV treatment in the first six cycles. The reason, could be that the leaching opened new surfaces for the photodegradation. The redness value of leached samples increased during the first twelfth days of treatments, then slightly decreased and later increased again. The distance between the two lines (dry and leached) increased slightly with irradiation time. This fact shows that redcolorants were leached out continuously by water. The 50 days UV radiation multiplied the initial redness value 4.48 times, and the leaching reduced this factor up to 3.28. The exact determination of the types of leached molecules requires further chemical investigation.



Fig. 6: Redness change caused by UV radiation (Dry) and by UV radiation and water leaching (Leached).

The increase of redness value was generated partly by the degradation products of extractives (Timar et al. 2016). The same thing occurs during the steaming of wood (Tolvaj et al. 2010, 2012). Spruce wood has a low extractive content, which is why the degradation products of lignin play an important role in redness change. During light irradiation phenolic groups react with photons and form phenolic radicals that transform into 0 and pquinonoid structures Cogulet et al. 2016, Pandey 2005, Leary 1968). These newly generated quinones increased the value of redness. The majority of red dyestuffs produced by plants are quinones (Melo 2009, Mills, White 2011).

The tendency of lightness decrease (not presented here) is similar as the tendency of yellowness and redness increase. The two curves (dry and leached) diverge after four days of UV irradiation and run almost parallel with one another. The distance between the two curves was small, approximately three units. It is 4% of the initial lightness value.

Earlier studies showed a good linear correlation between lightness (L\*) and hue angle (h\*) for 17 wood species (Tolvaj 2013). Experiments demonstrated that this correlation holds throughout the colour change caused by steaming (Tolvaj 2008), and photodegradation (Tolvaj et al. 2010) as well. The colour data in this study offers an opportunity to investigate the validity of this correlation for combined UV radiation and water leaching as well. Fig. 7 presents the correlation between lightness values and hue values. The dots representing the first cycle are located in the top-right corner followed by the colour dots of the irradiated samples towards left with growing irradiation time. The two trend lines are close to one another. The high values of the coefficients of determination show good correlation between the hue angle and the lightness in both cases. The slope of two trend lines differs slightly, showing that the hue of leached samples decreased somewhat less intensively than the hue of dry samples, as compared to the lightness change.



Fig. 7: Correlation between lightness and hue angle values of spruce wood during UV radiation (Dry) and during UV radiation and water leaching (Leached).

# **CONCLUSIONS**

One series of spruce specimens was irradiated by strong UV light emitter mercury lamp (dry series) and another series of specimens was treated with a combination of UV radiation and water leaching (leached series). The leaching partly removed the yellow and red chromophore molecules generated by the UV radiation. The specimens become slightly lightened after water leaching. The leached specimens become slightly more yellow and redder during the first 4-6 days of UV irradiation than those in the dry series. The 50 days UV radiation multiplied the initial redness value 4.48 times, and the leaching reduced this factor up to 3.28. These values for yellowness were 2.13 and 1.71, respectively. The roughness of the samples increased during UV irradiation, and the tendency of roughness change followed that of yellowness change. Good correlation was found between the hue angle and the lightness in both types of treatments. The determination of the exact types if leached chromophore molecules requires further chemical investigations.

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Antal Kannar University of Sopron Institute of Applied Mechanics and Structures Bajcsy Zs. U. 4 Hu-9400 Sopron Hungary

\*Laszlo Tolvaj University of Sopron Institute of Physics and Electrotechnics Bajcsy Zs. U. 4 Hu-9400 Sopron

Hungary \*Corresponding author: tolvaj.laszlo@uni-sopron.hu PHONE: +36 99 518140

Endre Magoss University of Sopron Institute of Woodworking Machinery Bajcsy Zs. U. 4 Hu-9400 Sopron Hungary

> Bajcsy Zs. U. 4 Hungary

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