

COLOR CHANGES OF VARIOUS WOOD SPECIES IN RESPONSE TO MOISTURE

*J. Baar**†

Academic Staff - Assistant
E-mail: jan.baar@mendelu.cz

Z. Paschová

Academic Staff – Assistant
E-mail: zuzana.paschova@mendelu.cz

P. Čermák

Academic Staff - Assistant
Department of Wood Science
Mendel University in Brno
Zemědělská 3
Brno 613 00, Czech Republic
E-mail: xcerma24@mendelu.cz

R. Wimmer†

Professor
Institute for Wood Technology and Renewable Materials
University of Natural Resources and Life Sciences, Vienna
Konrad Lorenz Strasse 24
Tulln 3430, Austria
E-mail: Rupert.Wimmer@boku.ac.at

(Received February 2018)

Abstract. Color including texture and gloss is one of the most important esthetical attributes of wood. For any porous materials in contact with water, the phenomenon of surface darkening arises from a change in the refractive index, as water enters air-filled pores. For the first time, this research investigated to what extent MC differences affect the surface color, below fiber saturation and after the pores are filled with water, with respect to extractive contents. Samples from 13 wood species were exposed to various climates, and as soon as EMC was reached, the color was determined. Color changes were also recorded after water submersion and also after vacuum impregnation in water. Chromaticity parameters a^* and b^* widely increased, along with higher moisture contents. Lightness changes were mostly stable at lower moisture contents. An exception was demonstrated by the heartwood species black locust, plum, walnut, and oak, which showed a brightening effect at lower moisture contents, followed by the darkening phase. The overall darkening of wood at high RH is connected to capillary condensation. Liquid water present in the cell lumen most caused darkening and an intensification of hue (increase in a^* and b^*). Darkening was even more expressed after vacuum impregnation with water. In conclusion, with only limited fluctuations in moisture contents below fiber saturation, the expected color changes are minor to neglectable.

Keywords: Chromaticity, extractives, lightness, moisture content, spectral reflectance, water.

INTRODUCTION

Color is the human eye's perception of an object surface induced by stimulation of the retina, with

a reflected radiation in the visible region of the electromagnetic spectrum (400-700 nm). Color including texture and gloss is one of the most important esthetical attributes of wood, which is distinctively influencing its commercial value (Pandey 2005; Papp et al 2012). Color widely varies across wood species and is primarily

* Corresponding author

† SWST member

determined by the amount and chemical nature of the prevalent extractives. Molecules having chromophore bonds that are responsible for light absorption at specific wavelengths are lignin and phenolic extractives such as tannins, flavonoids, stilbenes, and quinones (beyond 500 nm), and their derivatives (Hon and Minemura 2001; Pandey 2005). Because of the continuous character of the reflectance distribution as a function of wavelength, the actual color of wood is always the result of several components (Csanády et al 2015). Wood color appears inhomogeneous and differs even within given specimens (Nishino et al 1998). Wood discolorations potentially occur along the entire processing chain and may be caused by factors such as drying (Sandoval-Torres et al 2010), steaming (Varga and van der Zee 2008), thermal treatment (Esteves and Pereira 2009), and weathering (Feist and Hon 1984), or various microbial processes (Zabel and Morell 1992).

Practically, all wet surfaces appear darker than when dry. This is also true for unseasoned wood with the lightness being lower and the chromatic parameters being higher, compared with seasoned wood, as shown with *Liriodendron tulipifera* or *Abies sachalinensis* (Sullivan 1966; Hon and Minemura 2001). A common situation is a thin water layer covering a rough solid surface, as classically experienced with a school blackboard or a concrete construction surface (Lekner and Dorf 1988). Here, parts of the light get absorbed by the material, after passing through a superficial water layer, whereas the rest is diffusely reflected and scattered. In general, light passing through a water film gets repeatedly reflected to the surface by the air–water interface, because of total internal reflection, followed by surface absorption (Ångström 1925; Lynch and Livingstone 2001).

For a given porous material, the phenomenon of surface darkening arises from a change in the refractive index (RI), as water enters air-filled regions. The amount of light reflected at any interface depends on the ratio of RIs of the participating materials: the larger the difference in RIs, the higher the light reflection from the media interfaces (Choudhury 2014). Water has an RI higher than air (1.33 vs 1.0), which is close to the

RI of wood (1.54–1.56; Fink 1992; Marschner et al 2005). In any wetted material, the surface entering light gets forward scattered, which leads to a higher chance of surface absorption before leaving the surface (Twomey et al 1986; Jensen et al 1999). RI changes could be best observed with well-defined granulated materials such as sands or soils (Lekner and Dorf 1988).

The mentioned phenomena are valid for materials having surface water layers or pores that are internally water-filled. Water in wood can exist not only as absorbed (free) water in the cell lumens but also as adsorbed (bound) water, which interacts with hydroxyl groups of hydrophilic wood polymers prevalent in cell walls (Walker 2006). The MC at which all cell walls are fully water saturated, with theoretically no absorbed water in the cell cavities, is defined as the FSP (Skaar 1988). The FSP for most wood species is around 30%, but could be also as low as 19%, as in the case of African padauk, Ipe, or Wengé (Rijsdijk and Laming 1994). The wide-spreading FSP values are mainly attributed to existing differences in extractive contents (Wangaard and Granados 1967; Rijsdijk and Laming 1994). FSP is often considered as a value up to that physical and mechanical properties of wood are subject to change, with little changes at moisture contents above the FSP (Simpson and TenWolde 1999).

For low relative humidities, equivalent to an EMC of wood of about 5%, water molecules are superficially adsorbed in monolayers and bonded directly to the adsorbent through accessible hydroxyl groups. With increasing moisture contents (>5%), water starts forming polymolecular layers or clusters, with more water molecules getting entirely bonded to hydrophilic sites of the wood polymers through water–water hydrogen bonds (Hartley et al 1992; Walker 2006). At high RH (>90% RH), water vapor will get transformed to liquid water by capillary condensation, as it enters wood nanopores (Stamm 1964). However, the contribution of capillary condensation to adsorbed moisture is only poorly understood (Wang et al 2014).

Liquid water present on a surface or inside a given material, including wood, may show an intensive change of color lightness and saturation

(Csanády et al 2015). Whereas the above-FSP wood color changes are well known, the color changes below fiber saturation have rarely been examined. The presence of bonded water in porous cell walls, particularly with capillary condensed water, may alter the optical appearance of wood. During artificial weathering, the MC may vary distinctively, especially during long-term exposure (Baar et al 2015). This means that wood samples need to be properly conditioned prior and during exposure in case individual color measurements are carried out to obtain comparable measurement conditions. In this research, the question is raised to what extent unwanted differences in MC may have an effect on the resulting wood surface color. Here, we hypothesize that the FSP, which varies across species, has a significant role in observed moisture-induced moisture changes. We also hypothesize that interactions exist between extractive types, extractive contents, moistening, and color appearance. The research is performed by using several temperate and tropical wood species, which were exposed to various MC changes.

MATERIALS AND METHODS

Wood Species and Samples

Thirteen wood species investigated are listed in Table 1. Each species was represented by a set of five samples. Bulk samples were cut from

randomly selected boards, regardless of growth site and within-tree positions. The wood species were allocated into four groups. Group A contained the softwood species Norway spruce (NS), Scots pine sapwood (SPS), and Scots pine heartwood (SPH). Group B included the diffuse porous species alder (A), poplar (PO), common beech (CB), and European hornbeam (EH). These species have light-colored wood. Group C referred to tropical wood species, with Wengé (WE), African padauk (AP), and rubberwood (R), with Wengé and African padauk showing distinctly colored heartwood. Finally, group D contained ring-porous and semiring-porous species growing in the temperate zone with obligatory heartwood formation, including walnut (WA), black locust (BL), plum (PL), and oak (O). The group C species samples were kiln-dried, whereas the other groups were all air-dried. Samples were stored for a long period at standard climate conditions (65% RH, 20°C), before the experiments. Rift-sawn samples had the dimensions 20 × 7 × 45 mm ($L \times R/T \times R/T$); the exposed surfaces were sanded with 180-grid sanding paper and cleaned with compressed air, and the sample dried in an oven at 103°C. The longitudinal sample size was 20 mm only to fit in the measuring aperture of the used spectrophotometer. The short samples in fiber direction also allowed a fast adjustment to EMC. In addition to

Table 1. Color parameters values ($L^*a^*b^*$; oven-dry) and coefficients of variation (in parentheses) of two softwood and 11 hardwood species.

Common name	Botanical name	L^*	a^*	b^*
NS	<i>Picea abies</i> (L.) Karst.	79.1 (1.8)	5.1 (16.1)	23.5 (7.9)
SPS	<i>Pinus sylvestris</i> L.	80.6 (1.0)	4.5 (8.4)	22.6 (4.2)
SPH	<i>Pinus sylvestris</i> L.	74.6 (1.6)	8.8 (8.8)	35.8 (9.7)
PO	<i>Populus</i> spp.	81.0 (1.8)	3.3 (10.6)	18.5 (7.2)
A	<i>Alnus glutinosa</i> L.	72.2 (5.1)	7.4 (15.9)	20.9 (4.8)
CB	<i>Fagus sylvatica</i> L.	76.0 (1.2)	5.5 (3.9)	16.1 (3.4)
EH	<i>Carpinus betulus</i> L.	76.9 (1.8)	3.6 (13.9)	19.8 (6.8)
O	<i>Quercus</i> spp.	64.6 (4.5)	6.2 (9.0)	20.4 (5.7)
BL	<i>Robinia pseudoacacia</i> L.	63.8 (4.3)	4.8 (12.4)	22.7 (11.7)
PL	<i>Prunus domestica</i> L.	54.5 (3.2)	12.4 (4.0)	18.2 (7.0)
WA	<i>Juglans</i> sp.	57.7 (5.7)	5.5 (10.2)	16.2 (7.6)
R	<i>Hevea brasiliensis</i> Müll.Arg.	74.8 (5.4)	6.1 (21.2)	20.3 (5.7)
AP	<i>Pterocarpus soyauxii</i> Taub.	43.2 (6.2)	27.9 (4.5)	26.9 (8.1)
WE	<i>Millettia laurentii</i> De Wild.	36.6 (5.4)	7.1 (10.1)	11.1 (14.7)

NS, Norway spruce; SPS, Scots pine (sapwood); SPH, Scots pine (heartwood); PO, poplar; A, alder; CB, common beech; EH, European hornbeam; O, oak; BL, black locust; PL, plum; WA, walnut; R, rubberwood; AP, African padauk; WE, Wengé.

the bulk samples, veneers with 0.8 mm in thickness were prepared. The thin veneers had the advantage of fast adaptations to climatic conditions, which has reduced even more the required experimental times.

Samples Moistening

The MC was gradually raised in a conditioning chamber by adjusting the RH to 35%, 65%, 85%, 95%, and 98%, respectively. Temperature was held constant at 20°C throughout. After a period of 10 to 20 d, the color was measured at each conditioning level, along with the actual EMC, which were determined by the Eq 1:

$$\text{EMC} = \frac{m_w - m_0}{m_0} \times 100[\%] \quad (1)$$

where m_0 is the oven-dry weight (g) and m_w is the wet weight (g). In addition, the FSP was reached by storing samples above water in a closed desiccator to equilibrium. Furthermore, surface color changes due to liquid water absorption were measured at two stages: 1) after 20-s distilled water submersion and 2) after distilled water impregnation under vacuum (10 kPa) for 15 min. Excess surface water was removed using paper towels.

Color Change Measurements

Samples were kept in the dark to avoid unwanted photodegradative processes. Only during the actual color measurement, the samples were briefly exposed to daylight. Changes in wood surface color were measured on oblique surfaces, positioned between radial and tangential faces, using the mobile spectrophotometer Konica Minolta CM-2500 (Konica Minolta, Inc., Tokyo, Japan; d/8 measuring geometry, 10° standard observer, D65 standard illuminant, measurement area 8 mm, CIEL* a^*b^*) at a wavelength range between 360 nm and 740 nm, at 10-nm resolution. Two fixed measuring spots were selected on each sample. Surface reflectance at the different wavelengths were determined, along with L^* , a^* , and b^* color coordinates. These parameters are defined in the CIEL* a^*b^* color space as follows:

L^* is the achromatic lightness axis, a^* the chromatic green–red axis (+ a^* red, – a^* green), and b^* the chromatic blue–yellow axis (b^* , + b^* yellow, – b^* blue). The overall discoloration was assessed by the color difference ΔE^* , which was calculated according to Eq 2:

$$\Delta E^* = \sqrt{[(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]}, \quad (2)$$

where ΔL^* , Δa^* , and Δb^* represent the differences between the oven-dried and wet surface color.

Oxidation of wood chemical compounds is usually a slow color changing process, and this could be accelerated by exposing wood to light or higher temperatures. To avoid uncontrolled oxidations, tested samples were kept in the dark at stable temperature. Because the entire experiment took months, oxidative products could have potentially contributed to color changes. Therefore, color measurement was also performed on veneer samples, which allowed a faster experimental process (1 mo).

Wood Powder Extraction

Wood materials were cut and milled to a size of about 5 μm , using the mixer mill MM 400 (Retsch®, Germany). Wood powder from different samples of the same wood species were mixed and homogenized. Five grams of wood powder was taken and extracted in 60 mL of demineralized water according to the following procedure: 20 min exposure in an ultrasound bath (GT Sonic®, Shenzhen, China) at 40°C, followed by 20 min in a rocking shaker (Hangzhou MIU Instruments Co., Ltd.®, Hangzhou, China) at 150 rpm and room temperature. The water extract was then decanted and filtrated. An additional 40 mL of demineralized water was added to the already extracted powder, and the described process repeated. Combined filtrates were dried at 103°C to mass constancy, and weighed. The water-extracted powder was dried at room temperature, and 2.5 g was used for follow-up extractions in 30 mL of solvent mixture (acetone/methanol; 1:1). Here, the amount of solvent was

reduced, to maintain the same ratio of powder mass and solvent; likewise, in the second extraction round, the amount of solvent was lower—20 mL. The MC of used powders was determined separately for each wood species and extraction step. Total extractive content was determined as percentage from absolutely dry powder weight.

Bulk Sample Preextractions

The tested bulk samples (AP, BL, O, PL, R, WA, and WE) were extracted in 100 mL of demineralized water for 15 d, with the water changed daily. During this time, the baskets containing the samples were placed four times (2nd, 3rd, 9th, and 14th day) in an ultrasonic bath for 5 h each, at 50°C, for the purpose of intensifying extraction of the water-soluble compounds. After preextraction, the samples were slowly dried to oven-dry weight. The amount of extractives removed during preextraction was expressed as sample mass loss (%). Finally, samples were exposed to the same moistening and color measurement procedure as before.

RESULTS AND DISCUSSION

Surface Lightness Changes and Moistening

The color parameters of the investigated wood species determined at zero MC (oven-dry) are listed in Table 1. Wengé turned out to be the darkest species, with a L^* of 36.6. By contrast, poplar represented the brightest one, with L^* being as high as 81.0. The lightness change data due to different moisture contents are shown in Fig 1. At MC up to 20%, the lightness changes (ΔL^*) of the Fig 1(a)-(c) species remained at a moderate level ($p < 0.05$). The group D species showed and expressed brightening effect. These temperate-zone wood species are characterized by intensively colored heartwood and high extractive content. African padauk (Fig 1(c) – AP) started to darken right from the very beginning. All wood samples showed the darkening when exposed to a RH between 85% and 95%, which corresponded to EMC between 12% and 17%. These results were confirmed also with the veneer

samples (not shown). With the obtained and confirmed data, it can be concluded that the color changes are primarily caused by moistening.

FSPs appeared very different among the individual species (Fig 1) and the FSP averages are listed in Table 2. They are ranging between 16.8% for African padauk and 29.2% for hornbeam. Skaar (1988) stated FSP values between 25% and 30% (d.m.) for most temperate-zone wood species, but FSPs could also be much lower for many tropical wood species because of high extractive contents. Rijdsdijk and Laming (1994) have reported for Wengé and African padauk a FSP of 19.5% and 17.0%, respectively. According to Choong and Achmadi (1991), extractives in cell walls influence only the water adsorbed at higher air RH levels, with the monolayer adsorbed water vapor not being affected. The EMC measured at the higher humidity levels could be connected to the different extractive contents, which have reduced the available sorption sites and their relative potential energy levels for water attraction. Extractives include a wide range of chemical compounds that can be solubilized by polar or nonpolar solvents. In this research, we have performed water and acetone/methanol extractions, with the data shown in Table 2.

Water-soluble extractives range from 0.68% in poplar to 6.27% in oak. Organic solvent-soluble extractives reached higher values in many species than water-soluble ones, especially in Scots pine heartwood and African padauk; it varies between 0.5% in common beech and 13.0% in Scots pine heartwood. The extractive contents (excluding storage materials) are low in the sapwood, whereas the amounts in heartwood may reach high levels (Hillis 1987). In some tree species, the sapwood gradually transforms—with loss of moisture—into a light-colored wood (occasionally called ripewood), which does not contain living cells or reserve material, with low amounts of extractives. A typical example here is common beech, which has extractive contents being among the lowest known and primarily represented by various mono and oligosaccharides, sugar alcohols and acids, and simple phenols

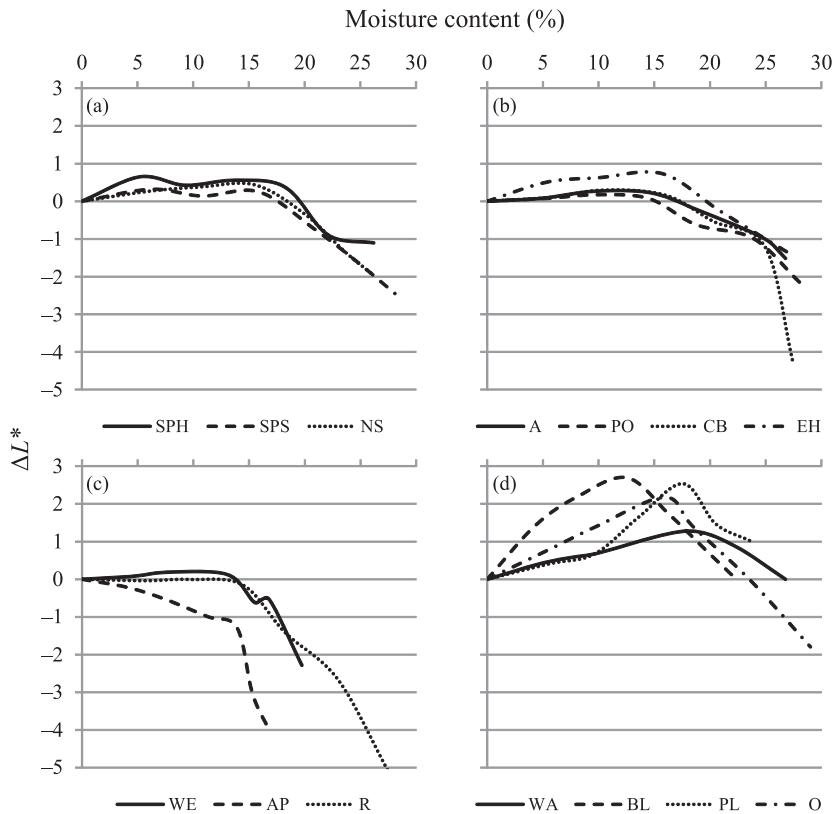


Figure 1. Lightness change (ΔL^*) with increasing wood MC; (a) softwoods, (b) diffuse-porous temperate-zone wood species, (c) tropical-zone wood species, and (d) ring- and semiring-porous temperate-zone wood species; see Table 1 for abbreviations.

(Vek et al 2016). By contrast, the most prevalent extractives in the heartwood of Scots pine are the hydrophobic stilbenes pinosylvin and pinosylvin monomethylether, monoterpenoids, and resin acids (Ekeberg et al 2006). African padauk is known for its very low water solubility and high extractive content (Brémaud et al 2011), with the most abundant compounds being biflavanoids, isoflavanoids, or isoflavanes (Jansen and Cardon 2005). Generally, higher extractive contents were found in the obligatory heartwoods of pine, oak, black locust, African padauk, and others. Walnut heartwood showed relatively low extractive amounts. The highest ratio between water and organic solvent-soluble extractives was found for oak heartwood, in which hydrolyzable tannins represent dominant group of extractives (Scalbert et al 1988). Water leaching of selected entire samples leads to an increased yield of extractives,

which can be related to higher water temperature or distinctively longer process time.

As shown, the lightness changes (Fig 1) were constant at lower EMC, where water is absorbed by cell wall as vapor, with the RI of water vapor being close to the one of air. According to Kulasinski (2016), it can be assumed that bonded water molecules in cell walls are closer and more stringently organized than bulk water because of exerted compressive pressure in the wood polymers, which can further influence its other properties including optical. The darkening of wood at higher moisture contents could be associated with capillary condensation, ie the adsorption of vapor in small capillaries forming droplets and consequently liquid surfaces at small radii of curvature. Kohonen and Christenson (2000) showed that water condensating between

Table 2. Extractive contents from wood powder (water-extracted, acetone/methanol [ac/me], total), bulk samples (water extraction), along with FSPs (coefficient of variation in parentheses).

Species	Wood powder			Bulk sample	
	Water (%)	Ac/me (%)	Total (%)	Water (%)	FSP (%)
NS	0.81	0.97	1.78	—	26.5 (3.4)
SPS	0.97	2.11	3.08	—	28.2 (3.7)
SPH	1.53	12.96	14.49	—	26.2 (2.1)
PO	0.68	1.74	2.42	—	28.8 (1.7)
A	3.56	2.15	5.71	—	26.8 (1.1)
CB	1.46	0.50	1.96	—	28.5 (1.4)
EH	4.77	0.82	5.59	—	29.2 (2.8)
O	6.27	3.36	9.64	13.30	29.0 (2.5)
BL	4.02	6.50	10.52	4.46	22.5 (7.8)
PL	2.14	6.07	8.21	4.95	23.6 (2.8)
WA	1.96	1.68	3.64	5.70	26.8 (3.4)
R	3.03	1.18	4.21	—	27.3 (7.3)
AP	0.73	12.66	13.39	3.34	16.8 (3.5)
WE	3.96	5.32	9.28	4.68	19.7 (10.4)

NS, Norway spruce; SPS, Scots pine (sapwood); SPH, Scots pine (hardwood); PO, poplar; A, alder; CB, common beech; EH, European hornbeam; O, oak; BL, black locust; PL, plum; WA, walnut; R, rubberwood; AP, African padauk; WE, Wengé.

sheet silicate mica surfaces had RIs equal to bulk water (RI = 1.33). This means that the presence of capillary condensed water may in part alter wood optical properties, similarly to free water in lumens. The water present in nanocapillaries enables multiple absorption of light because of changes in the RIs, which is resulting in darker surfaces because of a decreased amount of reflected light. Similar observation was published by Senthilkumar et al (2011) for dyed cotton fabric, where the depth of color drastically change at higher RH level (85%) because of capillary condensation.

For the group D species, the lower EMC ranges showed a brightening phase, instead of relatively stable lightness as seen in the other groups (A, B, and C). This is most likely linked to the nature of the acting extractives. In most temperate-zone hardwood species, hydrolyzable gallotannins and ellagitannins are predominantly present components (Rowe and Conner 1979). Aromatic components derived from glucose, such as flavonoids and condensed tannins, are having free hydroxyl groups that are water soluble, making them highly reactive with water molecules during moistening (Nzokou and Kamdem 2006). Furthermore, heartwood extractives of Wengé and

African padauk are known to be hydrophobic, with a low water-soluble content determined when extracted (Richter and Dallwitz 2000; Kilic and Niemz 2012; Saha et al 2013). To further confirm the extractive-induced brightening effect, the wood samples were extracted in warm water. The sample mass-loss analyses reassured that the group C (Fig 1) species all contain high amounts of water-soluble extractives. The same was confirmed for the tropical species Wengé.

Effects of Water Preextraction on Moistening-Color Changes

The sample preextraction led to less distinct lightness changes at the beginning of moistening process compared with unextracted, but the color lightening was still visible at the later phase of exposure (Fig 2). The removal of water-soluble extractives in Wengé before light exposure has caused darkening from the beginning of moistening. An even more intensive darkening was observed for African padauk (Fig 2). The influence of water preextraction on the chromaticity (a^* and b^*) was not observed; all samples exhibited similar behavior, not different from the unextracted samples. It was not possible to prove that the presence of water-soluble extractives explain completely the brightening effect of the group D species. All six preextracted wood species differed in ratio of water and organic

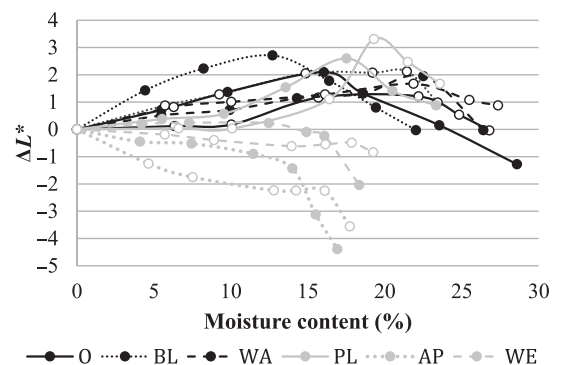


Figure 2. Change in lightness (ΔL^*) with increasing wood MC of selected woods before (full circle) and after water extraction (empty circles); see Table 1 for species abbreviations.

solvent-soluble extractives: even the extractive types of these species differ significantly, eg oak heartwood is typically rich in hydrolyzable ellagitannins and phenolic acids (Zhang et al 2015), whereas black locust has various groups of flavonoids and stilbens (Sergent et al 2014). The heartwood of walnut is known to have high contents of gallic acid (Gupta et al 1972), as well as various ellagic acid derivatives or naphthoquinones (Burtin et al 1998). African padauk is known for an “insoluble” red dye, which was identified mainly as biflavonoids and isoflavanoids (Jansen and Cardon 2005). The common feature for all temperate species differing in discoloration behavior is lightness (dry state) in range between 55 and 65. All other species were distinctively lighter or darker (AP and W). Rubberwood is known to have high content of hydrophilic water-soluble compounds (mainly amino acids) in the heartwood (Simatupang et al 1994), a fact that was also confirmed by own data (Table 2). The rubberwood heartwood has a light blonde color, indistinct from sapwood, with color parameters similar to the group B species. The color change shown by rubberwood was equivalent to those observed for the light-colored temperate species of group B. Therefore, the combination of specific colored compounds of individual woods and wood surface lightness can be responsible for such a behavior, which would need more detailed investigation.

Chromaticity and Overall Color Changes

Results of chromatic parameters a^* and b^* are listed in Table 1. Redness a^* of oven-dried wood was about the same across the species, ranging between 3 and 7 (Table 1). Higher values were observed for Scots pine heartwood, for plum, and particularly for African padauk. The a^* value of African padauk is multiple times higher than those for the other species, and it fits to the color appearance which is from vivid orange to deep red color. Distinct differences between species in the parameter b^* (yellowness) were found with Wengé (less yellowish) and with Scots pine heartwood and African padauk (more yellowish) (Table 1).

The chromatic parameter a^* increased uniformly with moistening till an EMC of 15% was reached, with Δa^* ranging between 0.5 and 1.5 (Fig 3). Beyond that MC, the color trends continued inconsistently, reflecting the existing variability among the species. For all species, redness (a^*) appeared more intensive around the FSP than at oven-dried state. Here, the most significant color changes were observed not only for the group D species, especially for plum wood, but also for the group B species beech, with Δa^* being greater than 2 (Fig 3).

Parameter b^* (yellowness) behaved similar to a^* for the group D species and rubberwood, which showed a uniform increase during the entire process of moistening. For softwoods and the group B species, almost no b^* change was observed at lower wood moisture contents. More yellow color (higher b^*) occurred only when wood was exposed to a higher air humidity (85-95%). The Δb^* has reached values twice as high as Δa^* or having a similar value (not shown). Parallel to the lightness progress, African padauk differed from the remaining species in b^* by changing from the beginning of moistening to lower b^* values, with loss in yellowness.

The intensification of chromaticity of selected species, particularly at higher moisture contents, can be explained by the presence of capillary condensed water. If pores are filled with water instead of air, more light gets absorbed and is subject to more intense internal reflections.

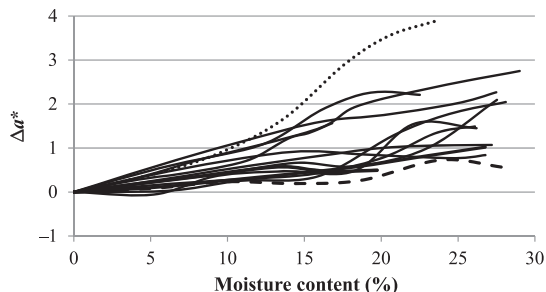


Figure 3. Change in parameter a^* with increasing wood MC (dotted line = plum, dashed line = poplar; solid lines = all remaining species without distinction, as only general trends are shown).

Internal reflection accentuates the effect of spectral variations and results in hue as well as color saturation changes (Dorsey et al 2008). Meints et al (2017) stated that for central European wood species, those with darker color and high chroma or hue are highly susceptible to color intensification due to wetting. Likewise, Senthilkumar et al (2011) stated that the increase in chroma of dyed cotton fabric was paralleled with higher RH, and a change in intensity was dependent on dye color and its concentration. Under moist condition, more light has the chance to get absorbed by the wood substances or directly by the embedded extractives, both resulting in a darker and also more saturated color.

Figure 4 shows reflectance spectra for selected species. Beech wood (Fig 4(c)) is a typical representative for all light-colored temperate hardwood species (H, A, PO) and for rubberwood. The reflectance of the wood surface declined across the entire wavelength range, meaning the surface was reflecting less light, which intensified a darker appearance. In oak and black locust,

which showed intensive reddening and yellowing at the FSP, a convergence of both spectral reflectance curves (oven-dried and FSP state) beyond 560 nm (yellow color) was observed, and even a crossing of curves at longer wavelengths (red color, Fig 4(c)). A distinctive change in reflectance spectra was seen for plum wood: wood samples with MC at FSP reflected more intensively light with wavelength longer than 600 nm than dry wood. This corresponds to the highest positive change of parameters a^* and b^* at the end of moistening cycles for plum wood. Dorsey et al (2008) stated that the occurrence of liquids of higher RI in pores causes more internal reflections, which results in changes of reflected distributions of light. This means that if a wood absorbs light in short wavelengths range more than in long wavelengths in dry state, the reflected light distribution after wetting, due to more internal reflections, includes even greater percentage of energy with longer wavelengths rather than shorter ones. The consequence of changed ratio between short and long wavelength of reflected light is a hue shift and more saturated color.

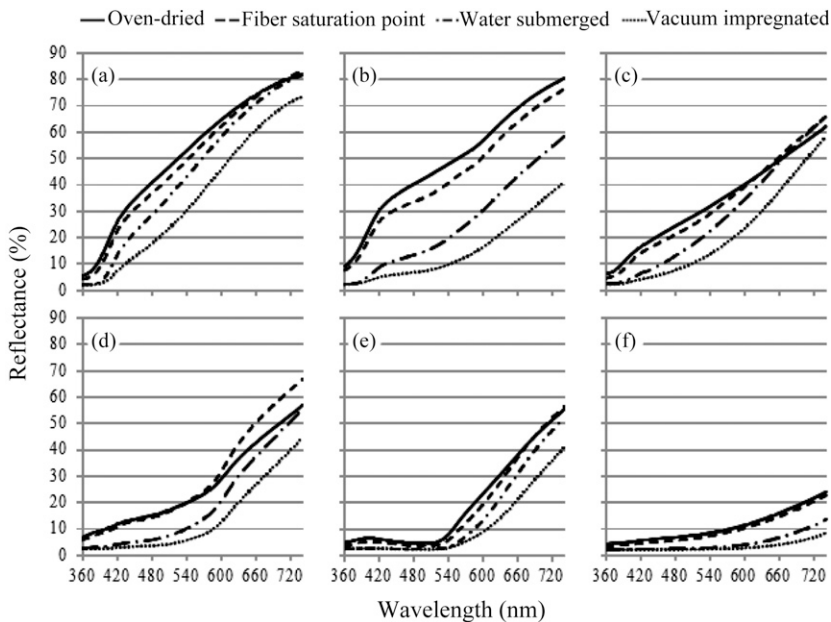


Figure 4. Spectral reflectance curves of selected species having different moisture contents (a-Norway spruce, b-common beech, c-oak, d-plum, e-African padouk, f-Wengé; solid line = oven-dried, dashed line = at FSP, dot-dash line = water submersion, dotted line = water impregnation under vacuum).

Overall color changes ΔE , which refer to oven-dry samples, and samples at FSP, are shown in Table 3. About in half of the species, the color change exceeded the value of 3, which is considered to be the limit for the detection by the human eye (Hon and Minemura 2001). Mokrzycki and Tatol (2011) stated even lower values, based on verified color differences, with 1-2 still being detectable to a trained eye, whereas an untrained eye may detect differences between 2 and 3.5. However, the human eye is even more sensitive in some areas of color (ie gray axis) and less in others (such as highly saturated area). The found changes in color caused by moisture contents below FSP are minor because the MC of wooden products usually fluctuates only in rather narrow ranges (about 10%).

Color of Wetted Wood

Sample surfaces were either wetted with liquid water for a short time to simulate a color intensification through transparent coatings or through a superficial oil film layer. A subsequent vacuum impregnation was performed to transport water deeper into the wood structure. Surfaces of all tested species darkened after water submersion and it appeared that darker wood species were

also more prone to distinct darkening ($R = 0.70$). Lightness and chromaticity parameters after short water submersion are shown in Table 3, with the color shifts depicted in Fig 5.

It was obvious that the surface lightness declined with increasing numbers of pores filled with water because less light is reflected from the material. Capillary condensation is limited to nanopores only, and the final effect of water on darkening is rather insignificant and undetectable by the human eye. Liquid water in cell lumens after water submersion caused more visible darkening. This was even doubled when the samples were vacuum impregnated with water. A change in lightness between water submersion and vacuum impregnation was more intense in permeable or more porous wood species such as Norway spruce, hornbeam, or beech. Higher permeability creates a thicker layer of water-saturated wood, causing the most forward light scattering, which then leads to deeper light penetration and therefore multiple light interactions with the material and thus higher probability of absorption before reflection (Jensen et al 1999).

Chromaticity is intensified by the presence of liquid water, oil, or transparent lacquer present in cell lumens. Whitish or yellowish wood species

Table 3. Color parameters change for individual species after water submersion and overall color change ΔE^* of three stage of wetting.

	ΔL^*	Δa^*	Δb^*	ΔE^*		
				FSP	Water submersion	Vacuum impregnation
NS	-4.9	3.2	11.9	3.2	13.2	22.2
SPS	-7.3	3.1	12.9	3.8	15.1	23.5
SPH	-2.5	2.1	8.5	2.0	9.1	13.3
PO	-4.0	1.9	9.5	2.9	10.5	19.8
A	-10.9	7.2	11.2	1.8	17.2	27.0
CB	-13.3	6.6	9.9	4.9	17.9	28.3
EH	-9.1	3.3	10.6	2.2	14.4	27.8
O	-10.1	5.9	10.9	4.3	16.0	23.2
BL	-7.5	4.6	10.8	6.6	13.9	20.1
PL	-9.5	6.7	6.9	5.6	13.5	19.8
WA	-13.6	4.0	4.8	2.3	15.0	24.9
R	-8.3	5.2	11.3	6.5	14.9	19.3
AP	-11.2	2.2	-1.2	4.7	11.4	18.7
WE	-14.4	-0.5	-4.6	2.6	15.1	19.7

NS, Norway spruce; SPS, Scots pine (sapwood); SPH, Scots pine (hardwood); PO, poplar; A, alder; CB, common beech; EH, European hornbeam; O, oak; BL, black locust; PL, plum; WA, walnut; R, rubberwood; AP, African padauk; WE, Wengé.

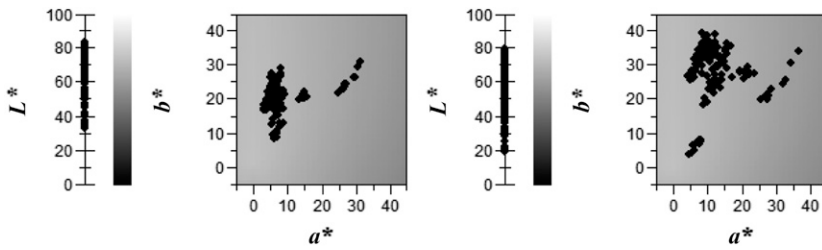


Figure 5. Color parameters range for dry (left) and wet (right, after submersion) wood surfaces.

such as hornbeam or spruce have shown more yellowness (b^*), rather than higher redness (a^*). For both strongly colored tropical species of group C an intensification of hue—an increase in a^* and b^* —was not observed. The chromaticity decline was even more pronounced after vacuum impregnation, with Δa^* and Δb^* reaching in African padauk -5.0 , and -8.9 , and for Wengé -3.8 and -8.3 , respectively. A decrease in yellowness was also found for walnut after vacuum impregnation. After wood wetting, the absorption of shorter wavelengths was more pronounced, so the larger part of the reflected light shifted into the yellow and the reddish range. The tropical species African padauk and Wengé have a low lightness value, which means that the small part of light is reflected at the surface. Wetting of the surface supports additional absorption, and from the reflectance spectra (Fig 4(e) and (f)), it is evident that the yellow portion of the visible light (577-597 nm) is widely absorbed by the wood. Vacuum-impregnated African padauk absorbed more light at longer red wavelengths, with the consequence that the intensity of redness has declined. From the obtained results, it is obvious that the moistening effect depends on the depth in which pores are filled by the medium having a RI close to the one of wood.

CONCLUSIONS

Lightness and chromaticity of surfaces of selected wood species exposed to different air humidity conditions were evaluated. In contrast to oven-dry wood, moist wood turned darker when the RH was greater than 85%. At lower EMC, no changes were observed for most of the species, with exception of the colored temperate hardwood species black locust, plum, walnut, and oak, which showed a brightening effect. All these

wood species have high water-soluble extractives content, but it was not entirely proved that they are cause of brightening. Redness (a^*) and yellowness (b^*) of wood were usually intensified after moistening, and all studied wood species showed their higher value at the FSP state compared with the oven-dried one, with the exception of African padauk and Scots pine heartwood, where the b^* value slightly decreased. Color hue shift, caused by a MC increase, corresponded to spectral reflectance curve change. The wood discoloration due to moisture content change occurs even in the range below FSP, in most species achieved values observable by human eye. Nevertheless, it can be neglected if the narrower range of MC fluctuation is considered or the wood MC is lower than 15%. Surface wetting by liquid water led to more intensive darkening and intensification of chromaticity parameters compared with changes below FSP. The rate of discoloration was influenced by the depth of water penetration beyond the surface.

ACKNOWLEDGMENTS

This work was carried out at Research Center Josef Ressel in Brno-Útěchov and financially supported by the Internal Grant Agency (IGA) of the Faculty of Forestry and Wood Technology, Mendel University in Brno (LDF_PSV_2016015).

REFERENCES

- Ångström A (1925) The albedo of various surfaces of ground. *Geogr Ann* 7:323-342.
- Baar J, Ondroušková J, Rademacher P (2015) Change of wood moisture content during artificial light irradiation and its influence on colour measurement. *Drvna Ind* 66: 189-193.

- Brémaud I, Nadine A, Minato K, Grill J, Thibaut B (2011) Effect of extractives on vibrational properties of African padauk (*Pterocarpus soyauxii* Taub.). *Wood Sci Technol* 45:461-472.
- Burtin P, Jay-Allemand C, Charpentier JP, Janin G (1998) Natural wood colouring process in *Juglans* sp. (*J. nigra*, *J. regia* and hybrid *J. nigra* × *J. regia*) depends on native phenolic compounds accumulated in the transition zone between sapwood and heartwood. *Trees* 10:258-264.
- Choong ET, Achmadi SS (1991) Effect of extractives on moisture sorption and shrinkage in tropical woods. *Wood Fiber Sci* 23:185-196.
- Csanády E, Magoss E, Tolvaj L (2015) *Quality of machined wood surfaces*. Springer International Publishing, Cham, Switzerland.
- Dorsey J, Rushmeier H, Sillion F (2008) *Digital modeling of material appearance*. Morgan Kaufmann/Elsevier, Amsterdam, The Netherlands.
- Esteves MB, Pereira HM (2009) Wood modification by heat treatment. A review. *BioResources* 4:370-404.
- Ekeberg D, Flåte P, Eikenes M, Naess-Andresen C (2006) Qualitative and quantitative determination of extractives in heartwood of Scots pine (*Pinus sylvestris* L.) by gas chromatography. *J Chromatogr A* 11:267-272.
- Feist WC, Hon DNS (1984) Chemistry of weathering and protection. Pages 401-451 in R Rowell, ed. *The chemistry of solid wood*. American Chemical Society, Washington, D.C.
- Fink S (1992) Transparent wood—A new approach in the functional study of wood structure. *Holzforschung* 46: 403-408.
- Gupta SR, Ravindranah B, Seshadri TR (1972) Juglandaceae: Polyphenols of *Juglans nigra*. *Phytochemistry* 11: 2634-2636.
- Hartley ID, Kamke FA, Peemoeller H (1992) Cluster theory for water sorption in wood. *Wood Sci Technol* 26:83-99.
- Hillis WE (1987) *Heartwood and tree exudates*. Springer-Verlag, Berlin, Germany.
- Hon DNS, Minemura N (2001) Color and discoloration. Pages 385-442 in DNS Hon and N Shiraishi, eds. *Wood and cellulosic chemistry*. Marcel Dekker, New York, NY.
- Choudhury AKR (2014) *Principles of colour and appearance measurement: Object appearance, colour perception and instrumental measurement*. Woodhead Publishing, Cambridge, U.K.
- Jansen PCM, Cardon D (2005) *Plant resources of tropical Africa 3: Dyes and tannins*. PROTA Foundation/Backhuys Publisher/CTA, Wageningen, The Netherlands.
- Jensen HW, Legakis J, Dorsey J (1999) Rendering of wet materials. Pages 273-282 in Proc. 10th Eurographics Conference on Rendering, June 21-23, Eurographics Association, Granada, Spain.
- Kilic A, Niemz P (2012) Extractives in some tropical woods. *Eur J Wood Wood Prod* 70:79-83.
- Kohonen MM, Christenson HK (2000) Capillary condensation of water between rinsed mica surfaces. *Langmuir* 16:7285-7288.
- Kulasinski K (2016) Effects of water adsorption in hydrophilic polymers. Pages 217-223 in A Mendéz-Vilas and A Solano-Martín, eds. *Polymer science: Research advances, practical applications and educational aspects*. Formatex Research Center, Badajoz, Spain.
- Lekner J, Dorf MC (1988) Why some things are darker when wet. *Appl Opt* 27:1278-1280.
- Lynch DK, Livingstone W (2001) *Color and light in nature*. Cambridge University Press, Cambridge, U.K.
- Marschner SR, Westin SH, Arbree A, Moon JT (2005) Measuring and modeling the appearance of finished wood. Pages 727-734 in Proc. SIGGRAPH, 2005 July 31-August 4, Los Angeles, CA.
- Meints T, Teischinger A, Stingl R, Hansmann C (2017) Wood colour of central European wood species: CIELAB characterisation and colour intensification. *Eur J Wood Wood Prod* 75:499-509.
- Mokrzycki W, Tatol M (2011) Color difference delta E-A survey. *Mach Graph Vis* 20:383-411.
- Nishino Y, Janin G, Chanson B, Détienne P, Gril J, Thibaut B (1998) Colorimetry of wood specimens from French Guiana. *J Wood Sci* 44:3-8.
- Nzokou P, Kamdem DP (2006) The influence of wood extractives on the photodiscoloration of wood surfaces exposed to artificial weathering. *Color Res Appl* 31:425-434.
- Pandey KK (2005) A note on the influence of extractives on the photo-discoloration and photo-degradation of wood. *Polym Degrad Stabil* 87:375-379.
- Papp EA, Csiha C, Valent J (2012) Colour change of some wood species during artificial xenon radiation. *Óbuda Univ e-Bull* 3:89-97.
- Richter HG, Dallwitz MJ (2000) *Commercial timbers: Descriptions, illustrations, identification, and information retrieval*. <http://delta-intkey.com> (25 June 2009) [In English, French, German, Portuguese, and Spanish].
- Rijsdijk JF, Laming PB (1994) *Physical and related properties of 145 timbers. Information for practice*. Kluwer Academic Publishers, Dordrecht, The Netherlands.
- Rowe WJ, Conner AH (1979) *Extractives in eastern hardwoods: A review*. General Technical Report FPL 18. U.S. Department of Agriculture, Forest Service, Forest Products Laboratory, Madison, WI. 67 pp.
- Scalbert A, Monties B, Favre JL (1988) Polyphenols of *Quercus robur*: Adult tree and in vitro growth calli and shoots. *Phytochemistry* 27:3483-3488.
- Saha JBT, Abia D, Dumarçay S, Ndikontar MK, Gérardin P, Noah JN, Perrin D (2013) Antioxidant activities, total phenolic contents and chemical compositions of extracts from four Cameroonian woods: Padouk (*Pterocarpus soyauxii* Taub.), tali (*Erythrophleum suaveolens*), moabi (*Baillonella toxisperma*), and movingui (*Distemonanthus benthamianus*). *Ind Crops Prod* 41:71-77.
- Sandoval-Torres S, Jomaa W, Marc F, Puiggali JR (2010) Causes of color changes in wood during drying. *For Stud China* 12:167-175.
- Senthilkumar M, Selvakumar N, Shamey R (2011) The effect of humidity, fabric surface geometry and dye type on the

- colour of cotton fabrics dyed with a select range of anionic dyes. *Dyes Pigment* 90:225-232.
- Sergent T, Kohnen S, Jourez B, Beauve C, Schneider YJ, Vincke C (2014) Characterization of black locust (*Robinia pseudoacacia* L.) heartwood extractives: Identification of resveratrol and piceatannol. *Wood Sci Technol* 48: 1005-1017.
- Simatupang MH, Schmitt U, Kasim A (1994) Wood extractives of rubberwood (*Hevea brasiliensis*) and their influences on the setting of the inorganic binder in gypsum-bonded particleboards. *J Trop For Sci* 6:269-285.
- Simpson W, TenWolde A (1999) Physical properties and moisture relations of wood. Pages 3.1-3.24 in *Wood handbook: Wood as an engineering material*. Forest Products Laboratory, Madison, WI.
- Skaar C (1988) *Wood-water relations*. Springer-Verlag, Berlin, Germany.
- Stamm AJ (1964) *Wood and cellulose science*. The Ronald Press Company, New York, NY.
- Sullivan JD (1966) Color characterization of wood: Color parameters of individual species. *For Prod J* 17:25-29.
- Twomey SA, Bohren CF, Mergenthaler JL (1986) Reflectance and albedo differences between wet and dry surfaces. *Appl Opt* 25:431-437.
- Varga D, van der Zee ME (2008) Influence of steaming on selected wood properties of four hardwood species. *Holz Roh-Werkst* 66:11-18.
- Vek V, Oven P, Poljanšek I (2016) Review on lipophilic and hydrophilic extractives in tissues of common beech. *Drvna Ind* 67:85-96.
- Walker JCF (2006) *Primary wood processing—Principles and practice*. Springer, Dordrecht, The Netherlands.
- Wang J, Mukhopadhyaya P, Morris PI (2014) Sorption and capillary condensation in wood and the moisture content of red pine. *J Build Phys* 37:327-347.
- Wangaard FF, Granados LA (1967) The effect of extractives on water-vapor sorption by wood. *Wood Sci Technol* 1:253-277.
- Zabel RA, Morell JJ (1992) *Wood microbiology: Decay and its prevention*. Academic Press, San Diego, CA.
- Zhang B, Cai J, Duan CQ, Reeves MJ, He F (2015) A review of polyphenolics in oak woods. *Int J Mol Sci* 16: 6978-7014.