

The Optical Appearance of Wood Related to Nanoscale Surface Roughness

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The color and appearance of timber is influenced by a variety of physical and chemical effects. Especially the chemical composition and the surface structure play a major role. In particular, the influence of chromophore extractives and the effect of thermal degradation processes on the color of wood are widely discussed in the literature. The present study deals with the influence of the surface nanostructure on the visual appearance of wood. This new perspective should generally demonstrate various influences on the appearance of natural inhomogeneous surfaces. Therefore, two methods were used to change the nanoroughness of lignocellulose materials. With different oil coatings and UV-irradiation, the surface structures of the samples were changed, and the measured roughness using atomic force microscopy was then correlated with the collected brightness differences. The results show that a clear correlation exists between the nanoroughness altered by oil coatings or UV irradiation and the brightness of the wood surface. Due to various other influences, such as chemical changes and light refractions of the treated wood structure which also influence the color of wood, no quantification can be given at this point.

Keywords: Appearance; Color; Gloss; Nano; Roughness; Wood

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INTRODUCTION

Wood is widely used as a diverse building material, not only because of its mechanical properties, but also for its aesthetic appearance. The color and the contrast of wood surfaces are thus of great interest for wood science in order to explain or influence different color phenomena.

The color of wood is formed by the specific reflection or absorption of light waves of a spectrum. This leads to a variety of relevant optical effects, which all form the resulting appearance. The absorption of light on wood surfaces was studied by Vorreiter (1949) and Norrstrom (1969). They examined the specific absorption of the different macromolecules and extractives in wood. According to their work, lignin is responsible for the greatest part of light absorption. Depending on the wood species, lignin is responsible for 80 to 95% of the overall light absorption in wood. In comparison, carbohydrates are only responsible for 5 to 20% of the absorption and reflect most of the incoming light. Gierlinger *et al.* (2004) showed in their work that variations of extractives and phenolic compounds have an influence on the color of larch wood. This was also found later by Moya *et al.* (2012) for *Acacia mangium* and *Vochysia guatemalensis* wood.

The absorption of light by the irradiated wood defines not only its appearance, but also the specific reflections on its surface. Hence, the variation in the overall appearance of different wood species cannot only be explained due to the variation of the chemical composition. The specific reflections are also influenced by the roughness of the irradiated surface, which is therefore an important factor for the appearance of an object (de la Rie 1987). The gloss, as well as the color saturation, is influenced by the roughness (de la Rie 1987). On a rough surface, light is not only specularly reflected, but also diffused. Scattered light from a rough surface appears whiter (the reflection spectrum in the reflection minimum increases). Consequently, the appearing color will be desaturated (de la Rie 1987). This effect is most noticeable in darker areas because there is a higher proportion of scattered light (de la Rie 1987). In addition, the work of Watanabe *et al.* (2007) showed that roughness values, as obtained with an atomic force microscope (AFM), allowing height differences of less than 1 nm to be measured, have an influence on the apparent color of an object.

For investigations of surface properties on wood, the atomic force microscope is widely used (Tshabalala *et al.* 2003). The roughness (Meincken and Evans 2009) and also the hardness (Gindl *et al.* 2002) and chemical properties of cellular solids (Koljonen *et al.* 2003) can be analyzed.

The influences that lead to color changes on wood are of scientific and also of industrial interest. Studies providing analysis of the relationship between color changes of wood and structural changes at the nano level were not found in the literature. Therefore, the present work examines the influence of the surface roughness on the appearance of wood. Specifically, the brightness difference due to the various rough surfaces was analyzed.

MATERIALS AND METHODS

To study the appearance of wood depending on its roughness, the roughness of lignocellulose materials was altered using two different methods.

Oil Coatings

In the first method, the wood surface was altered by thin oil coatings. The resulting changes in color and brightness were then correlated with the measured structure.

For these thin coatings, commercially available walnut oil, linseed oil, sunflower oil, and castor oil were used. All of the oils were able to crosslink and thereby form a stable structure. Prior to the application, the oils were boiled for one hour to ensure a faster hardening of the coatings. They were then applied on ten 120x60x4 mm oak wood (*Quercus robur*) lamellae, which were ground in advance with a grain size of 220 μm . For each sample, 135 g/m² of oil was applied to the wood surface with a pipette and consequently spread with a fresh paper towel to provide a uniform distribution.

The color of the wood was measured three times per sample using a Chroma Meter CR-410 (Konica Minolta, Tokyo, Japan), with a measuring diameter of 50 mm and a D65 light source in a 2° observer angle. To calculate the color differences, the CIELAB system was used (DIN 6174 1979). In the CIELAB system, the *L* axis represents the brightness (*L* varies from 100 for white to zero for black), and *a* and *b* are the chromaticity coordinates (+*a* for red, -*a* for green, +*b* for yellow, -*b* for blue) (DIN 6174

1979). Because no major chroma changes due to structural differences are known from the literature, only the brightness change was investigated. The specific brightness differences were calculated as follows,

$$\Delta L = \sqrt{(L_1 - L_2)^2} \quad (1)$$

where the indices stand for the color of the treated sample (1) and untreated sample (2).

An atomic force microscope was used to measure the roughness of the oil-coated samples. Because it was not possible to perform AFM scans on solid wood due to the macroroughness, coated *Typha* fibers (*Typha latifolia*) were used as an alternative. To defibrate the *Typha* plants, three trunks were disintegrated with a razor blade. The individual fibers were then removed using a forceps. The *Typha* fiber provided a relatively large surface without any major cavities, but it is also a lignocellulosic material like wood. Three fibers were dipped into the oil and then placed on a paper towel; therefore the surplus oil could drain off the fiber. After that, the fibers were conditioned (20 °C/65% RH) for a period of two weeks. To select appropriate measurement points for the AFM, electron microscopy (Quanta 250 FEG, FEI, Hillsboro, OR, USA) was used. Spots with a similar surface structure were chosen for every used fiber. The following AFM measurements were performed on a Bruker Dimension Icon (Bruker, Santa Barbara, CA, US) in standard tapping mode. The tip, from the same manufacturer, had a nominal tip radius of < 10 nm. The scanning frequency was 1 Hz for all of the scans. An image analysis was performed using the Gwyddion 2.26 software. For each coating, three positions were scanned. Nanoroughness was measured with 50 points per data record at the same area where the color was measured and represented as a mean. The nanoroughness (R_n) of the different data points was calculated as follows,

$$R_n = \frac{1}{l} \int_0^l |y(x)| dx \quad (2)$$

where y is the deviation from the mean of the profile and l is the reference distance.

UV-Irradiation

In the second method employed to verify the influence of roughness parameters on the appearance of wood, maple wood (*Acer pseudoplatanus*) was irradiated with UV-light. Because much more extractive materials are included in oak and thus potentially more chemical processes occur, the wood species was changed to maple. Two thin sections of maple wood with a thickness of 30 μm were prepared with a microtome. The thin sections were irradiated in four steps, for 1, 2, 4, and 8 h, according to ISO 4892-2 Cycle 6 (2013). In the beginning and after every step, the color and the surface roughness of a specific point on the sample using AFM were measured. The color measurements were performed with a Phyma Codec 400 Vis spectrometer (Gaaden, Austria). The reflection spectrum was acquired from a measuring spot of 12 mm in the 400 to 700 nm region. The color parameters were calculated using the CIELAB system (DIN 6174 1979).

The AFM measurements on the thin sections were measured and analyzed, similar to the process used with the oil-coated fibers. The AFM measurements were performed on three spots after each irradiation step. Care was taken to scan approximately the same position after each step.

Using Fourier transform infrared spectroscopy (FTIR), the thin sections were measured after each step to analyze chemical changes in the tested areas. The samples were measured using an Attenuated Total Reflection (ATR) device (Helios, with a diamond crystal) and a Tensor 27 (Bruker Optics GmbH) spectrometer measuring a spectral range between 4000 cm^{-1} and 400 cm^{-1} with a spectral resolution of 4 cm^{-1} and 8 scans per spectra. Each wood sample was measured four times after each irradiating step, and the averages of the spectra were used for the analysis. A baseline correction with the rubber band method was applied with the software package Opus (Version 6) from Bruker. To determine in more detail the chemical degradation, the ratio between the lignin (1505 cm^{-1}) and the carbohydrates (1375 cm^{-1}), as well as the ratio of the carbonyl (1734 cm^{-1}) to the carbohydrates, was calculated (Pandey 2005). The carbohydrate peak at 1375 cm^{-1} was not affected by irradiation and was therefore used as a reference. Hereinafter, the maximum heights of the specific peaks at 1505 cm^{-1} , 1375 cm^{-1} , and 1734 cm^{-1} were measured and divided.

RESULTS AND DISCUSSION

The color measurements of the oil-coated lamella showed clear differences between the different surface treatments (Fig. 1). Also, a variation in the nanoroughness of the fibers measured with the AFM was found for the different oil treatments. Figure 1 shows a clear relation between the nanoroughness of the fibers and the brightness change of the coated wood ($R^2= 0.93$).

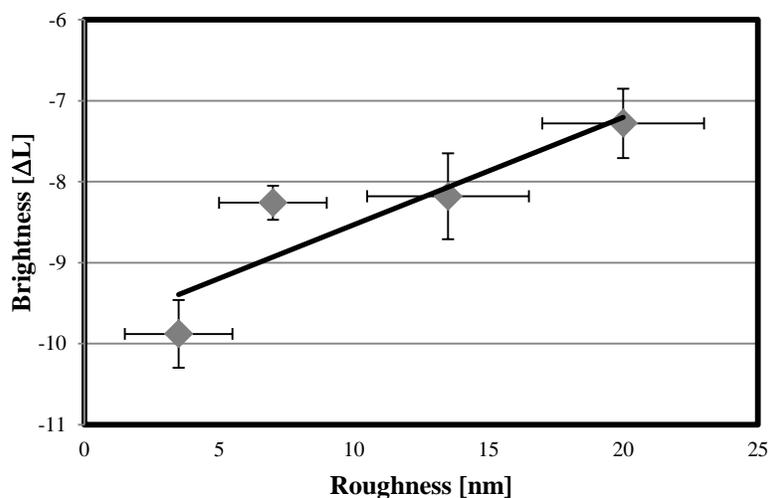


Fig. 1. Relationship between the roughness (nm) of the coated *Typha* fibers measured with the AFM (Atomic force microscope) and the brightness difference in the coated oak wood (measured before and after the oil treatment)

The reason for this effect seems to be the relatively stronger diffuse reflection associated with an increased nanometer-scale roughness. Parts of the incoming light reflect specularly on a smoother surface, but not all of it does. A portion of the incident light gets reflected diffusely to a varying degree. The greater the dispersion of light is, the brighter the sample appears. This effect can also be found in several other studies (Klein 2004; Meichsner *et al.* 2011). For example, rougher pigments appear much brighter than

smooth pigments (Klein 2004). The correlation of nanoroughness with brightness (Fig. 1) is non-trivial, as the roughness is well below the visible wavelength. Although the nanoroughness is below the wavelength of light, minor local surface irregularities may still lead to diffused light reflections.

Non-homogeneities in the distribution of the oils or different interactions with the wood can be alternatively explained by a similar physical effect. Thereby, to varying degrees, diffuse light from the entire reflected radiation will be filtered. This means that thicker layers and more homogeneous coatings filter more diffuse light due to back reflection; consequently, the surface appears darker (Meichsner *et al.* 2011). The differences between the homogeneous and smooth linseed oil and the rather rough and inhomogeneous sunflower oil can be seen in Fig. 2.

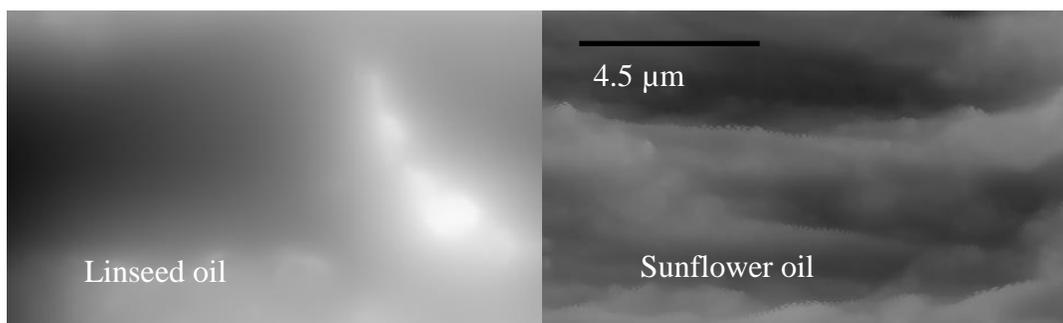


Fig. 2. Two AFM images (topography images) of linseed oil- and sunflower oil-coated Typha fibers

However, with the data available at present, it is very difficult to determine which of these effects is crucial for the altered appearance of wood. Therefore, a second method was used to change the roughness of the wood in the nanometer range without applying any substance. Due to UV-irradiation, the nanoroughness of the wood was altered. With a longer exposure to UV radiation, the roughness of the measured surface decreased (Fig. 3[a]). This effect was also detected in the work of Meincken and Evans (2009), in which the samples were exposed to sunlight. From an initial peak, the roughness subsequently decreased to a minimum level, only to increase again after a long exposure time. In the present work, not only has the nanoroughness changed due to the UV-irradiation, but there was also a change in color (Fig. 3[b]).

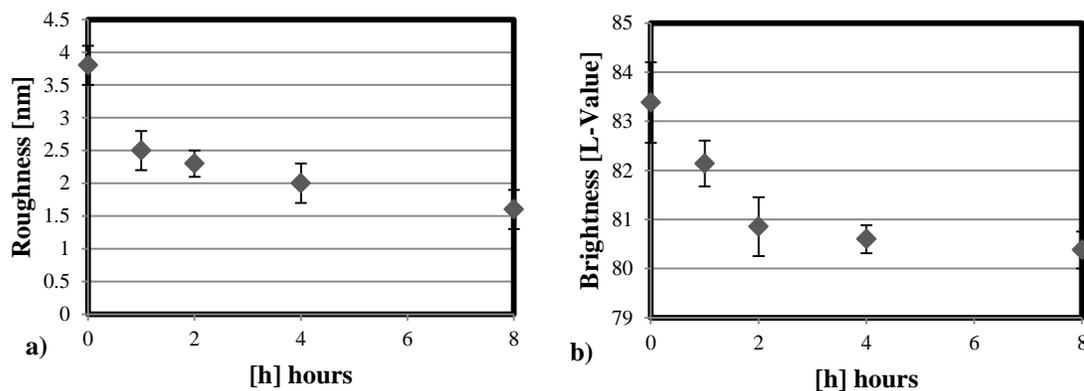


Fig. 3. The measured roughness [a] and brightness [b] after each UV-irradiation step

The maple thin sections got darker after every step of the UV-irradiation. Similar to the oil-coated samples, a significant amount of variability in brightness could be explained by the measured nanoroughness ($R^2 = 0.90$).

To analyze chemical changes due to the UV-irradiation, FTIR-ATR measurements were carried out. The measured spectra from FTIR-ATR showed minor changes; after eight hours of UV-irradiation, a clear increase in carbonyl groups (1734 cm^{-1}) and a decrease at the aromatic lignin C=C band (1505 cm^{-1}) was found (Fig. 4).

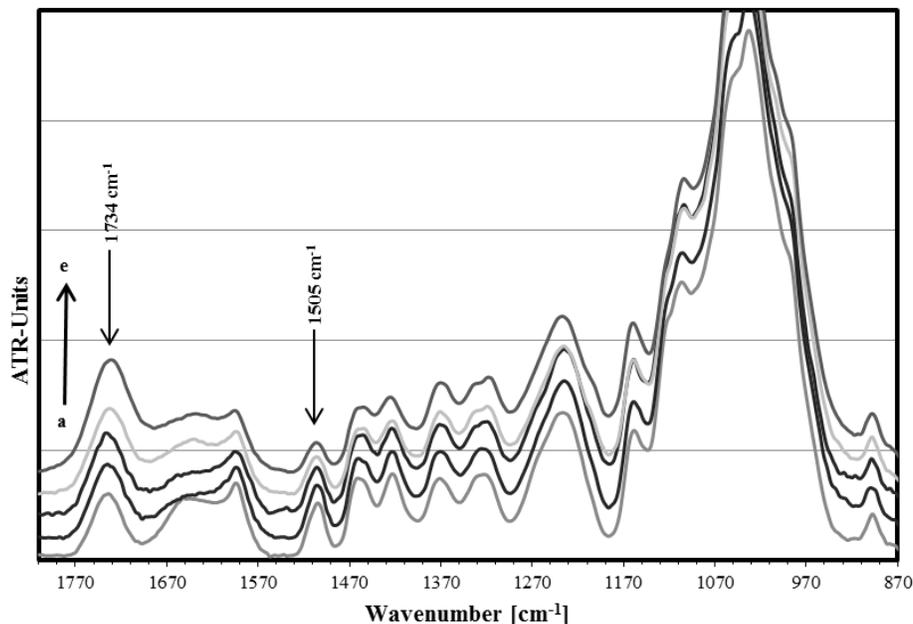


Fig. 4. Vector normalized (at 1375 cm^{-1}) FTIR spectra of maple wood irradiated for (a) 0 h, (b) 1 h, (c) 2 h, (d) 4 h, and (e) 8 h.

Furthermore, it can be seen that the ratio between the carbonyl and carbohydrates (Fig. 5[a]) and the ratio between the lignin and carbohydrates (Fig. 5[b]) both correlated with the brightness of the measured maple thin sections.

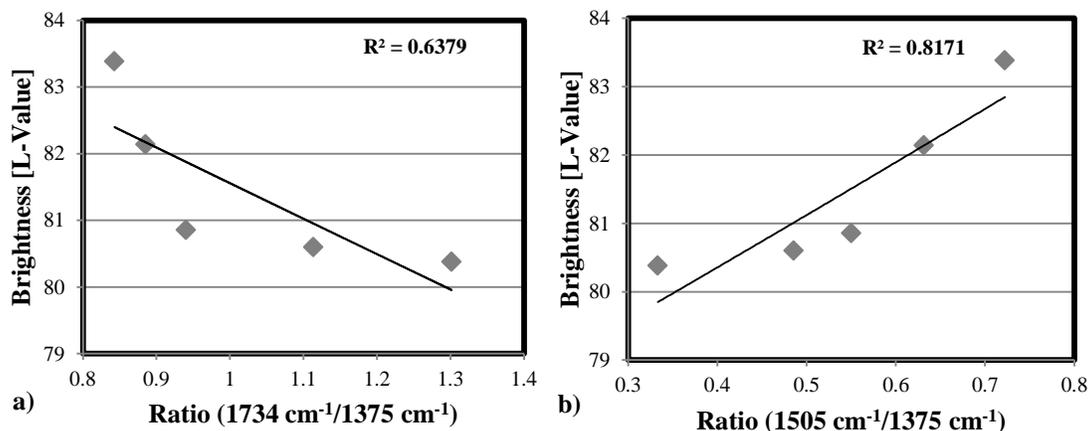


Fig. 5. Correlation of the brightness change (L-Value) and the increase in carboxyl groups [a] as well as the decrease in lignin [b]

In the literature, only strong chemical alterations wherein a degradation of lignin can be clearly seen have been associated with changes in color (Pandey 2005). Due to the measured spectra, chemical changes must be considered to explain the color changes. Both the measured chemical and structural changes correlate with the brightness of the prepared sample. This shows that both have an effect on the resulting appearance of wood. Because it is very difficult to change the nano-scale surface structure of wood without changing the surface chemistry, a quantification of the structural effects remains challenging.

The experiments presented show that the influence of the surface structure could be a partial factor for the appearance of wood. The identified correlations and the evidence from the literature should be reason for further investigation. However, a new approach should be found to characterize the influence of the surface structure. Further studies on this issue should clarify the influence of these effects on the general appearance of wood, independent of the processing or treatment.

CONCLUSIONS

1. This investigation has been carried out to test the surface roughness of wood and its influence on the appearance. The results show that a clear correlation exists between the nanoroughness, which was altered by oil coatings and UV irradiation, and the brightness of the wood surface. This indicates a hitherto unconsidered effect of the surface roughness on the appearance of wood.
2. Due to a similar trend found for surface chemistry changes, no quantification of the strength of this effect can be given at this point.

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