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Novel color stabilization concepts for decorative surfaces of native dark wood and thermally modified timber



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ABSTRACT

UV and visible light exposure of dark colored wood and thermally modified timber (TMT) is strongly connected with discoloration and photobleaching of the respective wood surfaces. Conventional and nanoscale UV absorbers as well as radical scavengers (HALS), mainly effective for the protection of softwood and bright hardwood, were found to be improper for the light stabilization of dark wood surfaces. In the current work discoloration mechanisms are discussed considering the role of wood extractives and the sensitivity of dark colored wood types to visible light and results are presented which were obtained by applying novel protecting systems for the light stabilization of transparently coated surfaces from dark colored wood types and TMT.

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1. Introduction

High-value wood surfaces for exclusive wood-based floorings, furniture and interior outfitting of residences, luxury yachts, aircrafts, and railway vehicles are preferentially made of dark colored tropical wood species. Decreasing availability, high material costs, and environmental issues led to efforts to substitute those wood species by other materials, e.g., thermally modified timber (TMT), possessing comparable surface properties. TMT can be made from European wood species such as Norway spruce, Scots pine, European beech and European ash which in the unmodified form have bright colored surfaces. By varying the thermal modification processes different medium to dark color tones are generated in a reproducible manner [1].

However, exposure to daylight often causes brightening or graying of transparently coated dark wood or TMT surfaces [2–5]. The thereby induced intensive discoloration of solid dark wood or veneer surfaces cannot be sufficiently stabilized by conventional light protecting systems such as UV absorbers and radical scavengers (e.g., hindered amine light stabilizers – HALS) mainly effective for the protection of softwood and bright hardwood

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http://dx.doi.org/10.1016/j.porgcoat.2015.06.017 0300-9440/© 2015 Elsevier B.V. All rights reserved. surfaces [6–8]. In some cases (e.g., spruce TMT) their addition was found to intensify discoloration in comparison to counterparts without any stabilizing agent [9]. Similarly, nanoscale zinc, titanium, or cerium oxide UV absorbers which could be successfully applied on softwood and bright hardwood [10–13] were proven to be unsuitable for the light stabilization of dark wood surfaces [14,15]. A limited success was achieved with modified UV absorbers which cover a part of the visible light spectrum [7,16].

Furthermore, it was found that discoloration of dark wood species is not only caused by UV-induced bond cleavage and oxidation of the lignin macromolecule causing the formation of colored low-molecular fragments [17]. It is also a result of (1) photoinduced darkening due to the oxidation of colorless precursors (leucochromophores) to chromophores, e.g., the formation of stilbene ortho-quinones from monohydroxystilbenes [18] and (2) photobleaching of chromophores by visible light leading to the formation of colorless polymers and degradation products [19–21]. Nevertheless, better understanding of the discoloration behavior of TMT and dark tropical wood is necessary. Generally, these wood species contain relatively high concentrations of chromophores such as guinones and phenolic or furfuryl derivatives [22-24]. Particularly quinones form dark colored charge-transfer complexes with hydroquinones may contribute to the color of the respective wood species [20,21] and are prone to photobleaching [19]. In particular, the role of wood extractives and the sensitivity of dark wood species to visible light require more investigations. In the present paper, discoloration mechanisms for dark wood species are discussed considering these aspects and testing results are presented which were obtained by the application of novel protection systems developed for the light stabilization of transparently coated surfaces from dark colored wood and TMT.

2. Materials and methods

Wood species. For the investigations, furniture panels veneered with Brazilian rosewood (*Dalbergia nigra* Fr. All), East Indian rosewood (*Dalbergia latifolia* Roxb.), Macassar ebony (*Dalbergia melanoxylon* Guill. et Perr.), Wengè (*Millettia laurentii* De Wild.), African Padouk (*Pterocarpus soyauxii* Taub.), Eastern Black walnut (*Juglans nigra* L.), teak (*Tectonia grandis* L.F.), American black cherry (*Prunus serotina* Ehrh.), African mahogany (*Khaya* ssp.), *European oak* (*Quercus* ssp.), and European beech (*Fagus sylvatica* L.) as well as thermally modified softwood (Norway spruce – *Picea abies* L.) obtained by different technologies and temperature profiles were used.

Extractives. Wood extractives were obtained by a 4 h Soxhlet extraction of 10g of milled wood material with 250 ml of acetone. The extracts were used as obtained. Filter paper samples were impregnated through drop-by-drop addition of the extract and subsequently air-dried at 23 °C and 50% relative humidity. This procedure was repeated until all filter sheets subsequently used for irradiation experiments contained the same mass of solid extract.

Coating systems. Commercially available, solvent- and waterborne two-component polyurethane (2K PUR) lacquers (hydroxvlpolyacrylate resin, binder/hardener ratio 10:1 (w/w), solid content after mixing: 39.0-41.0%, applied quantity: 100-120 g m⁻², twofold application) were used. They were applied either with (1.0-3.0%, w/w, calculated on solid content of the coating system) or without conventional light protecting agents: (1) nanoscale cerium dioxide (Nanobyk 3812/3810, Byk-Chemie, Germany), (2) organic UV absorbers (hydroxyphenylbenzotriazine type: SunCare UV concentrate, ISP, USA), (3) HALS: 4-hydroxy-2,2,6,6-tetrametyhlpiperidinoxyl type (Lignostab 1198, BASF, Switzerland). In addition, aqueous alkyd resin impregnating basecoats (binder content: 10.0–13.0%, v/v, applied quantity: $50-120 \text{ gm}^{-2}$, one- or twofold application) were applied with (1.0–3.0%, w/w, calculated on wet coating system) or without novel color stabilizing agents based on water soluble carboxylates and inorganic salts. The effect of the stabilizers is assumed as a result of complexation and salt formation of the central ions of the active components with phenolic substructures of the wood extractives and lignin.

Irradiation experiments. The spectral sensitivity of uncoated wood species (equilibrium moisture content at 23 °C and 50% relative humidity) as well as their extractives (impregnated filter papers) was examined by means of a 150W laboratory xenon high pressure arc lamp at 50% relative humidity and 23 °C (Fig. 1; LOT Quantum Design, Germany; wavelength range 200–900 nm) with exchangeable filters. For the experiments 320, 400, 500, 600, and 700 nm cutting-edge filters were used. The sample specimens were fixed in a 0.5 m distance from the filter. Repeating color measurements were carried out after each hour of irradiation up to a complete irradiation time of 20 h.

Native and coated wood specimens were irradiated according to [25,26], respectively, in a Ci3000 Xenotest weathering device (Atlas Material Testing Technology, USA; relative humidity: $50 \pm 10\%$, irradiation intensity: $50 \pm 2 \text{ W m}^{-2}$ at 300–400 nm, external sodalime filter type C, internal borosilicate filter type S). The total irradiation time was 400 h. Outdoor irradiation was performed according to [27] (total irradiation time 1008 h).

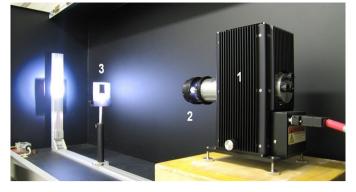


Fig. 1. Laboratory high pressure xenon arc lamp (1: UV/vis radiation source, 2: filter holder, 3: sample holder).

Colorimetry. The measurement of the color changes of uncoated and coated test specimens was carried out with a CM 3610d spectrophotometer (Minolta, Japan; D65 standard light, 10° standard observer, SCI method; two measurements on each sample, measuring spot: 24.5 mm). The color changes were determined in terms of the CIELab color system [28].

3. Results and discussion

3.1. Light-induced discoloration of dark wood species and TMT

Conventional light protecting formulations used for transparent wood coatings are mixtures of benzophenone, benzotriazole or triazine UV absorbers and HALS radical scavengers [6-8,16]. Recently, nanoscale zinc oxide, titanium dioxide or cerium dioxide UV absorbers have been completing the light stabilizer family. They are successfully applied on softwood or bright hardwood surfaces particularly for interior applications [10-13].

On native dark wood species such as Brazilian rosewood (Fig. 2) and TMT (not shown) those UV stabilizers do not lead to color stabilization.

In most cases, their addition even accelerates the discoloration (color difference $\Delta E^* = 23.75/18.9$ after 400 h of irradiation) in comparison to samples without any stabilizing additives ($\Delta E^* = 12.3$). The initial dark color of the respective wood species is quickly

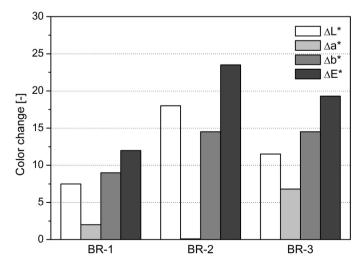


Fig. 2. Effect of conventional UV absorbers (benzotriazine type, 2%, w/w) and HALS (amine-N-oxyl type, 2%, w/w) on light induced CIELab color changes of 2K polyurethane coated Brazilian rosewood. BR-1: no stabilizer added; BR-2: HALS/UV absorber combination (1%, w/w); BR-3: HALS lignin stabilizer (1%, w/w) in impregnating basecoat; 400 h Xenotest irradiation according to [25,26].

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