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SOME PHYSICAL PROPERTIES OF VARNISH COATED WOOD PREIMPREGNATED WITH COPPER-CHROMATED BORON (CCB) AFTER 3 MONTHS OF WEATHERING EXPOSURE IN SOUTHERN EAGEN SEA REGION

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ABSTRACT

This study evaluates weathering performance of Scots pine (*Pinus sylvestris* L.) and chestnut (*Castanea sativa* Mill.) wood specimens preimpregnated with copper-chromated-boron (CCB) before varnish coating such as polyurethane and an alkyd-based synthetic varnish. Varnishes were applied as coating or some containing paraffin was impregnated into wood as a water repellent. The wood specimens were then exposed weathering for 3 months at during autumn. After 3 months of weathering exposure; some physical properties such as surface hardness, color stability, water repellency, and mass loss of the wood specimens were tested. Wood surfaces preimpregnated with CCB showed enhanced resistance to natural weathering. Weathering performance of coated and treated wood specimens was remarkably increased as compared to that of noncoated and untreated wood specimens. Althouhg, weathering softened both of the untreated wood species, all the treated surfaces were hardened after weathering exposure.

KEY WORDS: weathering performance, CCB, varnish, impregnation, wood, surface hardness, color stability, water absorption, mass loss

INTRODUCTION

Wood has been popularly and favorably used as a decorative material owing to its aesthetic appearance and characteristic properties (Chang and Chang 2001). Because of its chemical composition and composite structure, wood offers a unique combination of properties including high strength, low cost, and ease of processing (Kiguchi and Evans 1998). Wood is a naturally durable material that has been recognized for centuries throughout the world for its versatile and attractive engineering and structural properties (Anderson et al. 1991). However, wood surfaces exposed outdoors are rapidly degraded because lignin strongly absorbs UV light, which leads to radical-induced depolymerisation of lignin and cellulose, the major structural constituents of wood (Evans et al. 2002). The ultraviolet (UV) light portion of the solar radiation and the presence of moisture are the main causes for the weathering degradation of wood (Feist and Rowell 1982, Denes and Young 1999). The rate of degradation is increased by water (rain, dew, and snow), changes in relative humidity, increased temperature, and windblown sand and/or other particulates. Attack by decay fungi is not considered weathering, nor is mildew growth on the wood surface, which usually accompanies weathering. Weathering of wood is primarily a surface phenomenon that results in the slow erosion of wood fibers from the surface (Williams et al. 2001).

Coatings provides wooden materials with the desired aesthetical properties like color and gloss, but are mostly also of vital importance in the protection of wood against environmental influences like moisture, radiation, biological deterioration or damage from mechanical or chemical origin (Meijer 2001). Unfinished wood can be used both outdors and indoors without further protection. However, wood surfaces exposed to the weather any finish change color one roughned by photodegradation and surface checking and erode slowly. Transparent film forming finishes such as spar, urethane, and marine varnishes are not generally recommended for exterior use on wood surfaces, because these varnishes allow transmissions of sunlight and surfaces degradation can take place underneath the coatings (Evans et al. 1992). Protecting wood and wood based materials from degradation caused by severe outdoor conditions has been the focus of several research groups, and a great deal of effort has been invested into the development of efficient protection systems (Hon 1996). Therefore, impregnation of wood with an appropriate water repellent or applying a varnish compatible preservative chemical prior to coating for exterior use under hazardous service conditions has been under taken to make wood more stable primarily against photochemical degradation, dimensional changes, biological decomposition, and fire (Williams et al. 1996, Wilkinson 1976). The most effective method of preventing the photodegradation of wood involves treatment with dilute aqueous solutions of inorganic salts, particularly hexavalent chromium compounds. Application of chromium trioxide to wood surfaces prevents lignin degradation during natural weathering (Kiguchi and Evans 1998, Evans et al. 1992) and predictably, when applied as a wood pretreatment, retards the deterioration of clear finishes and stains during exterior exposure (Feist 1987).

Chromated copper arsenate (CCA) has been a major wood preservative for more than 50 years for many applications such as utility poles; children's play grounds, residentical application, etc. (Yildiz et al. 2004). The chromated copper arsenate impregnation can greatly extend the life time and durability of partially UV- light-transparent stain applied to the treated wood surface because the chromium stabilized the wood surface against UV light degradation. In addition to improving the durability of finishes on wood substrates, chromium salts impart other benefical properties to wood surfaces, such as fungal resistance, decreased swelling by water, increased water repellency (Tshabalala and Ganstad 2003). Williams and Feist (1988) found that pretreatment of wood with chromium trioxide or chromium nitrate retarded the deterioration of western red cedar (*Thuja plicata* D. Don) and Douglas fir (*Pseudotsuga menziesii* Mirb.Franco) after 30 months of exposure. Feist (1979) and Feist and Hon (1984) reported that the application of aqueous solutions of chromium trioxide to wood surfaces had an inhibiting effect on the outdoor weathering process and enhanced the life of surface finishes applied over the treated wood. Therefore, treatment of wood with a chromium-containing preservative such copper- chromated-arsenate (CCA) before coating was suggested to extend the life time and durability of the coating system (Williams and Feist 1986, Bardage et al. 1998). Since environmental awareness has forced the use of environmentally safe and arsenic free chemicals for wood and wood based composite protection (Evans 1995, Suzuki 1995), alternatives to arsenic containing preservatives are required. Borates have several advantages as wood preservative in addition to imparting flame retardancy, providing sufficient protection against wood destroying organisms, having a low mammalian toxicity and low volatility. Moreover, they are colorless and odorless (Yalinkilic et al. 1999, Hafizoglu et al. 1994, Chen et al. 1997). As a boron-containing wood preservative which is generally considered to have low mammalian toxicity and providing sufficient protection against all forms of wood destroying organisms (Hafizoglu et al. 1994, Chen et al. 1997, Drysdale 1994), copper-chromated-boron (CCB) was expected to fulfill this requirement. Therefore, chromium in CCB was expected to protect the wood surface against UV-light degradation while copper and boron enhanced the biological resistance of wood. Sell and Feist (1985) reported that high resistance of wood that was preimpregnated with CCB before varnish coating against weathering has been attributed to the protective effect of chromium and copper salt solutions. Peylo and Willeitner (1995) found that water-repellent coatings on borate treated wood increase the wood's service life. In this study, the weathering performance of two commercial varnish types (a polyurethane varnish and an alkyd-based synthetic varnish) applied as coating or WR impregnants and preimpregnated with CCB before varnish coating were studied.

MATERIALS AND METHODS

Preparation of wood specimens

Scots pine (*Pinus sylvestris* L.) and chestnut (*Castanea sativa* Mill.) wood specimens (sapwood) of 10 x100 x150 mm (radial by tangential by longitudinal) were air-dried.

Impregnation and coating

Copper-chromated-boron (CCB) aqueous solutions of 7.5 % concentrations were prepared (composed of 25 % boric acid, 36 % sodium bichromate, 37 % copper sulfate and 2 % additives). An alkyd-based synthetic varnish and a polyurethane varnish of a two component type consisting of an aliphatic isocyanate-terminated component and an active hydrogen-bearing monomer, which when blended cures at room temperature with 4–5 h pot life of the blend. Alkyd-based synthetic and polyurethane varnishes were applied over untreated and CCB impregnated wood. They were also impregnated in to the wood as water repellent (WR) solutions by dissolving in white spirit (20 % v/v) containing 1 % paraffin wax, which reportedly has no effect on proper adhesion of the paint if it is allowed to cure sufficiently after treatment (Williams et al. 1996). Vacuum was applied for 30 min. at 760 mHg^{-1} before supplying the solution into the chamber followed by another 30 min. at 760 $mmHg^{-1}$ diffusion period under vacuum. After impregnation, the wood specimens were conditioned for 3 weeks at 65 % relative humidity and 20°C ± 1°C before coating. Weight percent gain due to chemical load was calculated as follow formula:

$$\text{Weight percent gain} = \frac{W_f - W_i}{W_i} \times 100 \quad (\%)$$

Where W_f is the final conditioned weight of wood specimens and W_i is the initial weight of wood specimens.

A varnish, used as a primer coating to fill voids, was applied twice to untreated and CCB impregnated wood specimens. A topcoat was also applied to reveal the absolute effect of weathering through the clear varnish layers. Sufficient time for coat setting was allowed between succesive application until the target retention of 75 g/m^2 for primer and 100 g/m^2 for topcoat controlled by consecutive weighing. Specimens were left at ambient conditions for 24 h before the topcoating according to the instructions given by the varnish manufacturer. Surfaces were gently sanded with abrasive paper to obtain a smooth surface prior to applying the topcoat. Treatment systems applied to wood specimens prior to weathering exposure are given in Tab. 1.

Treatment group	Impregnation concentration	Varnish type
Untreated (control)	-	-
Untreated (coating alone)	-	Polyurethane
Untreated (coating alone)	-	Synthetic
CCB[a] (Preimpregnation)	7.5 % aqueous solution	Polyurethane
CCB (Preimpregnation)	7.5 % aqueous solution	Synthetic
WR[b] impregnation with polyurethane varnish	20 % dissolved in white spirit includes 1 % paraffin wax	Polyurethane
WR impregnation with synthetic varnish	20 % dissolved in white spirit includes 1 % paraffin wax	Synthetic

[a] Copper-chromated-boron
[b] Water repellent

Tab. 1: Treatment systems applied to wood specimens prior to weathering exposure

Weathering exposure

Each treatment group consisted of 12 individual wood specimens. In total, seven groups of wood specimens for each species were exposed to weathering conditions during autumn (September-October-November) in 2004.

Wood panels were prepared for weathering exposure according to ASTM D 358–55 (1970). A test site was established close to the Regional Meteorological Observation Station of Mugla, which is in Southern Aegean Region, to enable practical assessments.

Surface hardness

The surface hardness of test specimens was measured according to ASTM D 4366–95 (1995).

Color changes

The color of the specimen was measured by a colorimeter. The color difference, (ΔE^*) was determined for each wood as follows (ASTM D 1536–58 T 1984):

$$\Delta a^* = a^*_f - a^*_i$$
$$\Delta b^* = b^*_f - b^*_i$$
$$\Delta L^* = L_f - L^*_i$$

$$(\Delta E^*) = [(\Delta a)^2 + (\Delta b^*)^2 + (\Delta L^*)^2]^{1/2}$$

Where Δa^*, Δb^*, *and* ΔL^* are the changes between the initial and final intervals values.

Mass loss

The mass loss of the wood specimens were calculated based on the initial (W_i) and the final (W_f) conditioned weight.

$$\text{Mass loss} = \frac{W_f - W_i}{W_i} \times 100 \quad (\%)$$

Water droplet test

A water droplet test was applied to determine the water-repellency of coated and treated systems. The diameter of diffused water from five individual droplets (0,047 ± 0,003g/drop) on a single panel at different locations was measured 3 min after application.

RESULTS AND DISCUSSION

Surface hardness

Surface hardness is given in Tab. 2 along with the weight percent gain of the wood specimens due to the chemical load. Higher retentions were achived in Scots pine because of the easy penetrability of these species compared to chestnut (Evans et al. 1992).

Tab. 2: Surface hardness of wood specimens before and after 3 months of weathering exposure

Treatment chemical	Weight percent gain (% w/w)	Varnish type	Surface hardness	
			Before exposure Mean ± SD[c]	After exposure Mean ± SD
Scots pine				
Untreated	-	-	27.85 ± 3.75	19.18 ± 4.51
Untreated	-	Polyurethane	54.00 ± 8.84	66.85 ± 7.48
Untreated	-	Synthetic	20.89 ± 5.70	28.43 ± 6.35
CCB[a]	25.3	Polyurethane	51.45 ± 6.41	72.80 ± 7.72
CCB	25.3	Synthetic	19.24 ± 6.35	37.75 ± 9.69
Polyurethane-WR[b]	23.6	Polyurethane	40.17 ± 8.57	60.66 ± 7.65
Synthetic-WR	31.9	Synthetic	19.96 ± 6.30	26.14 ± 5.05
Chestnut				
Untreated	-	-	38.65 ± 3.54	33.54 ± 3.66
Untreated	-	Polyurethane	61.58 ± 6.48	70.09 ± 9.57
Untreated	-	Synthetic	24.89 ± 5.87	30.27 ± 7.40
CCB[a]	4.2	Polyurethane	53.70 ± 4.69	79.40 ± 8.81
CCB	4.2	Synthetic	21.13 ± 5.59	40.39 ± 6.65
Polyurethane-WR[b]	5.8	Polyurethane	52.66 ± 4.55	63.08 ± 9.45
Synthetic-WR	7.4	Synthetic	23.28 ± 7.08	31.12 ± 4.77

[a] Copper-chromated-boron
[b] Water repellent
[c] Standard deviation

Outdoor conditions softened both of the untreated wood species. The combined action of the moisture, sunlight, and temperature could destroy the lignocellulosic network, limiting performance of unprotected wood in outdoor applications (Müller et al. 2003). In contrast, all the coated and treated wood surfaces were hardened after 3 months of exposure. Copper-chromated-boron (CCB) pretreatment reduced the degradation of wood specimens during exterior exposure. Feist and Williams (1991) observed that small amounts of chromium salts on the wood surface greatly decreased wood weathering. In addition to improving the durability of finishes on wood substates, such as decreased swelling by water, increase water repellency, enhanced resistance to natural outdoor weathering, and extractive bleed. Surface hardness of wood specimens coated with polyurethane varnish was higher than synthetic varnish-coated wood surfaces before and after 3 months of exposure. Decker et al. (1991) reported that organic coatings obtained by photopolymerization of aliphatic urethane-acrylate telecholic oligomers have been shown to be quite resistance to accelerated weathering. Remarkable increases of surface hardness of wood specimens coated with synthetic varnish were recorded after 3 months of exposure. It may be due to the progressive cross-linking of alkyd molecules on exposure, followed by degradation reactions (Majumdar et al. 1998).

The hardness of alkyd coatings is a function of their formulations and can be improved by additives. Generally speaking, the types of binder resin and pigments will mainly affect the resistance ability against light irradiation and water action (Motohashi 1996).

Color changes

Color changes of wood specimens after 3 months of weathering exposure are given in Tab. 3. The results showed that the color of the untreated wood specimens remarkably changed after 3 months of weathering exposure conditions. The natural colors of wood vary rapidly changed when exposed to weathering. First it becomes dark, intensifying the yellow or brown colors and after a period of a silver grey color is predominant. The first part of the process is due to the formation of brown products resulting mainly from lignin degradation. These products are leached by the rain, leaving a grey surface composed of partially degraded cellulose with low lignin content (Feist and Hon 1984, Pastore et al. 2004). In general, light colored woods tend to turn yellow or brown, and some dark woods tend to bleach (Feist and Hon 1984, Feist 1983). Copper-chromated-boron (CCB) pretreatment of the wood before synthetic varnish coating gave the best color stabilization of the both wood species. Surface coating with polyurethane varnish resulted in higher color changes as compared to synthetic varnish coated for both wood species after 3 months of exposure during autumn. The high ΔE of polyurethane alone coated wood surfaces may be due to the oxidation of the terminal aromatic amine. So, pretreatment of wood with commercially available UV absorbers and hindered amine light stabilisers can improve the longevity of clear coatings applied to wood (Evans et al. 2002). Feist (1987) found that given good construction practice, any pretreatment such as water repellent, a water repellent preservative, or similar material would help protect painted wood from decay and improve the overall performance of wood. If impregnated, varnish acts as a multilayer coating that may reduce sunlight diffusion into wood. The present study, synthetic varnish coated wood specimens showed better color stability as compared to polyurethane coated wood specimens after 3 months of exposure. These results are consistent with earlier assessment on alkyd-based coatings (Kraft et al. 1967).

Tab. 3: Color changes of wood specimens after 3 months of weathering exposure

Treatment chemical	Varnish type	Color changes (ΔE) After exposure Mean ± SD[c]
Scots pine		
Untreated	-	16.33 ± 2.09
Untreated	Polyurethane	12.44 ± 2.18
Untreated	Synthetic	6.77 ± 1.66
CCB[a]	Polyurethane	8.11 ± 2.33
CCB	Synthetic	6.04 ± 1.90
Polyurethane-WR[b]	Polyurethane	9.07 ± 1.60
Synthetic-WR	Synthetic	7.18 ± 1.87
Chestnut		
Untreated	-	14.43 ± 3.03
Untreated	Polyurethane	11.87 ± 3.56
Untreated	Synthetic	5.54 ± 1.39
CCB[a]	Polyurethane	8.19 ± 2.07
CCB	Synthetic	4.58 ± 0.77
Polyurethane-WR[b]	Polyurethane	7.15 ± 1.47
Synthetic-WR	Synthetic	5.12 ± 1.66

[a] Copper-chromated-boron
[b] Water repellent
[c] Standard deviation

Water repellency

Water diffusibility levels of wood specimens are given in Tab. 4. Chesnut wood specimens were more water repellent than Scots pine before and after 3 months of exposure owing to its higher density and structural difference. The reduction in water droplet spread on untreated Scots pine was an indicator of early degradation of hemicelluloses and cellulose (the primary companents of the cell wall responsible for water absorption) due to erosion effects of weathering. Varnishs reduced the water diffusibility of wood within a given time. Because, coatings reduce the rate of uptake of water into the wood by its barrier function against liquid water and water vapour diffusion. It also reduces the drying rates of wood by same water vapor diffusion barrier (Meijer 2001). Water diffusibility was similar for both varnish types along with exposure. Pretreatment of wood specimens with CCB before varnish coating cause decrease in droplet spread compared with the only varnish coated surfaces within the 3 months period of exposure. The application of aqueous solutions of hexavalent chromium (e.g. chromium trioxide) to wood surfaces had an inhibiting effect on the outdoor weathering process and enhanced the water repellency surface finishes applied over the treated wood (Feist and Hon 1984, Feist 1979). Owing to the interaction of the chromium trioxide with the wood components, the majority of the chromium on the treated wood was reduced from the ($^{+}$6) to the ($^{+}$3) state. In addition, the surface concentration of hydroxyl groups decreased while the hydrocarbon level increased. Those observations are consistent with a mechanism involving an oxidation of the surface wood components by chromium ($^{+}$6) and subsequent fixation of the reduced chromium to wood carbohydrates and lignin. In a chromate-cellulose interaction, the resulting wood surface would be expected to have an increased hydrophobic character. This was borne out by the observation of surface water repellency on wood freshly treated with aqueous chromata solution (Ross and Feist 1991).

Mass loss

Mass loss of wood specimens during weathering occurs due to photodegradation of lignin and leaching lignin fragments from the exposed wood surfaces (Dahlgren 1973). Untreated and noncoated wood specimens lost up to 1.91 % and 2.61 % of their weight for chesnut and Scots pine respectively, after 3 months of weathering exposure (Tab. 4). Chestnut was more durable to weathering exposure because of its polyphenolic extractive content and high density (Wilkinson 1979, Scheffer 1973). Mass losses levels remained less than 0.4 % for all treated panels. The mass loss of preimpregnated CCB or WR impreganeted wood specimens was lower than only varnish coated panels. But, coating alone imparts to wood only superficial protection against some deteriorating agents for a limited time, often less than 2 years (Evans et al. 1996). Thus, it is concluded that especially, long-term exterior wood protection could have been achieved by a successful combination of appropriate preservative treatment by a compatible surface coating process (Ross and Feist 1991).

Tab. 4: Mass loss and droplet spread of wood specimens before and after 3 months of weathering exposure

Treatment chemical	Varnish type	Mass loss (%)		Droplet spread (cm)	
		Before exposure Mean ± SD[c]	After exposure Mean ± SD	Before exposure Mean ± SD	After exposure Mean ± SD
Scots pine					
Untreated	-	-	2.61 ± 0.27	3.27 ± 0.18	4.66 ± 0.37
Untreated	Polyurethane	-	0.30 ± 0.10	0.70 ± 0.16	1.05 ± 0.20
Untreated	Synthetic	-	0.38 ± 0.13	0.91 ± 0.18	1.22 ± 0.28
CCB[a]	Polyurethane	-	0.12 ± 0.04	0.78 ± 0.22	0.93 ± 0.17
CCB	Synthetic	-	0.15 ± 0.05	0.90 ± 0.10	1.02 ± 0.10
Polyurethane-WR[b]	Polyurethane	-	0.22 ± 0.04	0.63 ± 0.09	1.16 ± 0.19
Synthetic-WR	Synthetic	-	0.25 ± 0.04	0.74 ± 0.08	1.35 ± 0.25
Chestnut					
Untreated	-		1.91± 0.24	1.65 ± 0.23	2.97 ± 0.66
Untreated	Polyurethane	-	0.27 ± 0.04	0.51 ± 0.08	0.98 ± 0.22
Untreated	Synthetic	-	0.33 ± 0.11	0.60 ± 0.07	1.23 ± 0.16
CCB[a]	Polyurethane	-	0.12 ± 0.03	0.45 ± 0.07	0.68 ± 0.21
CCB	Synthetic	-	0.14 ± 0.07	0.49 ± 0.12	0.70 ± 0.13
Polyurethane-WR[b]	Polyurethane	-	0.18 ± 0.04	0.58 ± 0.10	1.05 ± 0.25
Synthetic-WR	Synthetic	-	0.20 ± 0.08	0.69 ± 0.17	1.27 ± 0.18

[a] Copper-chromated-boron
[b] Water repellent
[c] Standard deviation

CONCLUSIONS

A polyurethane varnish and an alkyd-based synthetic varnish produced commercially and applied over untreated or CCB-impregnated Scots pine and chesnut specimens were tested for 3 months of weathering exposure. Varnishes were applied as coating or containing paraffin wax impregnated into wood for WR.

The results of this study indicated that Scots pine and chesnut preimpregnated with CCB have enhanced natural outdoor weathering characteristics (ultraviolet degradation, subsequent erosion). While untreated and noncoated both wood specimens very rapidly softened in weathering conditions, treated and coated wood specimens were hardened after 3 months of weathering exposure. Color stability of polyurethane coated wood specimens was lower than synthetic alkyd-based varnish coated wood specimens. The high color changes of polyurethane-coated wood surfaces may be attributed oxidation of some terminal aromatic amine. In this respect, polyurethane varnish requires additional UV inhibitor in weathering exposure. Mass losses of noncoated and untreated wood specimens were higher as compared that of coated and treated wood specimens. Some macroscopically visible failure occured on coated film after 3 months of weathering exposure, while noncoated and untreated wood specimens tended to discolor, flake, and crack on the wood surfaces.

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