

Change in Surface Properties of Coating Film on Styrax Wood Treated with Rosin-Copper During UV Irradiation

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Abstract: In this study, Styrax wood (*Styrax tonkinensis* Piere) before coated by Polyurethane (PU), was impregnated with the mixture of 1%, 2%, or 4% rosin sizing agent solution and 3% copper sulfate, then was exposed under UV lights. The effects of rosin-copper on the color change and gloss of coating film on the treated and coated wood surface after UV exposure were also studied. The result showed that the wood samples after being preserved by rosin-copper solutions improved the color stability of paint film on the wood surface under the influence of UV lights, however, it caused a slight decrease in the gloss of the coating film. When the concentration of the rosin sizing agent increases, the gloss of the coating film tends to increase, however, it did not affect the color stability of the paint film. The use of rosin-copper formulations to treat wood could not only reduce the damage of the copper preservative leaching into the environment but also improve the UV resistance of the coating film on the treated wood.

Keywords: UV resistance, color change, gloss, styrax wood, rosin-copper.

Date of Submission: 26-03-2022

Date of Acceptance: 06-04-2022

I. Introduction

Wood is a natural material whose main structural components are cellulose, hemicelluloses, and lignin. Even though wood has had a large number of applications such as construction materials, furniture, and other household items for thousands of years because of its advantages of being easy to process, having a good sound, heat insulation, and being friendly to humans and the environment. However, wood also has limitations such as being easily damaged by fungi and insects, especially wood is very susceptible to warping, cracking, and dimensional instability when used in outdoor conditions. This has reduced the range of use as well as the life of the wood. Therefore, wood needs to be treated and preserved to prolong its life.

Rosin is a natural product, it has very good hydrophobicity and is friendly to humans, so it has been widely used in the paper industry as a sizing agent (Zhang, 2005). In addition, Rosin has also been used alone or in combination with copper/boron to impregnate into the wood and the results have shown that rosin also could improve the wood decay resistance (Nguyen et al., 2012; 2013a, 2017 and 2020). However, to enhance the aesthetic value of wood and contribute to the protection of wood by the effects of organisms, wood products are often painted before being used. Many studies have shown that: preservative compounds such as CCA, Tanalith E, boric acid and Immersol aqua did not affect the scratch resistance but significantly increase the abrasion resistance of the paint film on the surface of pine, oak, and chestnut wood (Ozdemir et al., 2015); The pine wood before varnishing, treated with borate compounds increased the hardness and gloss of the coating film, but reduced the adhesion of coating film (Toker et. al., 2009). However, when used in outdoor conditions, in addition to factors such as moisture, fungi and insects that damage wood, there is also sunlight radiation, especially UV light, which is also a factor possible affects the properties of the wood, causing undesirable effects such as discoloration and reduced mechanical strength as well as the life of the wood.

Yalinkilic et al. (1999) reported that Scotland pine and chestnut wood treated with chromium-copper-boron (CCB), followed by coating with polyurethane varnish or alkyd-based synthetic varnish increased color stability of the wood surface, reducing the weight loss of the wood and can protect the wood in outdoor conditions for a long time. Wood treated with chromium or copper compounds can improve the resistance of wood surfaces to UV irradiation and weathering factors (Temiz et al., 2005). However, there are no reports on the effect of rosin-copper on the quality of coating film on the treated wood surface when used in outdoor conditions or under UV irradiation. Therefore, the main purpose of this study is to evaluate the effect of treatment by rosin-copper formulations on some characteristics such as gloss and discoloration of paint film on the Styrax wood surface after UV irradiation.

II. Materials And Methods

2.1. Material preparations

Styrax tonkinensis (Piere) wood was selected according to the ISO 3129 standard. Wood specimens were cut from untreated *Styrax* wood into blocks with dimensions of 150 × 50 × 15 mm. Deficiency-free specimens were selected for the tests. In addition, wood samples with dimensions of 20 x 20 x 20 mm were also prepared to determine the retention of wood samples.

The rosin sizing agent (R) was an industrial product and was provided by Guangxi Wuzhou Arakawa Chemical Industries Co., Ltd. In this study, it was used to impregnate into the wood at the concentration of 1%, 2%, or 4%. And 3% copper sulfate (CuSO₄·5H₂O) was provided by Tianjin Kermel Chemical Reagent Co., Ltd., was used as a preservative salt, and was also combined with the rosin sizing agent to impregnate wood.

Two-component polyurethane (PU) paints (including 612G primer with 56% dry content and 2099 code gloss paint with dry content 52%) and PU hardener code number OL17 were used to paint the surface of the test sample. All finish systems were obtained from the Oseven company.

2.2. Treating wood blocks

Before the treatment, all wood blocks were oven-dried at to constant weight of 60 ± 2°C, weighed, and recorded as W₁. The blocks were then treated using a full-cell pressure process at 0.1 MPa vacuum for 60 min followed by 0.6 MPa pressure for 60 min. Then the blocks were kept in the treatment solution at ambient conditions for 60 min. The blocks were then individually removed from the solution, wiped lightly to remove the solution from the wood surface, and immediately weighed to determine the mass after impregnation (W₂). The retention of each block was calculated using the following formula:

$$\text{Retention, kg/m}^3 = \frac{GC}{V} \times 10 \quad (1)$$

where G = W₂-W₁ is the weight in grams of the treating solution absorbed by the block, C is the weight (g) of preservative in 100 grams of treating solution, and V is the volume of the block in cubic centimeters.

After treatment, all samples were stored at ambient laboratory temperature for air drying for 4 weeks.

2.3. Application of finishes

All untreated control and treated samples were coated with two-component PU paint, using the pneumatic spray method. The painting process is carried out as follows: First, the wood samples are surface treated, then primed for the first time. Next, the samples are air-dried, sanded, and primed for the second time. When the paint film was dry, the wood sample is further sanded and polished for the last time. After polishing, the wood samples are placed at ambient laboratory temperature for about 1 month to let the paint film dry naturally and stably.

2.4. UV exposure

After coating, all wood specimens were exposed to irradiation source using 40W UV lamps and 350nm wavelength for 960 h at ambient laboratory temperature and air environment. The distance from the lamp to the sample surface is about 50 mm.

Color measurements were performed on the exposed surfaces of the wood samples before and after UV irradiation and were measured using an NF-333 Spectrophotometer (Nippon Denshoku Industries Co. Ltd., Tokyo, Japan) with a standard measuring aperture of 4 mm diameter. The CIELAB system is characterized by three parameters, lightness (L*) and color coordinates (a* and b*) (Fig. 1). The color change, ΔE* after each UV irradiation period, was calculated according to Eqs. 1 through 4,

$$\Delta L^* = L^*_f - L^*_i \quad (1)$$

$$\Delta a^* = a^*_f - a^*_i \quad (2)$$

$$\Delta b^* = b^*_f - b^*_i \quad (3)$$

$$\Delta E^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \quad (4)$$

where ΔL*, Δa*, and Δb* are the changes between the initial (i) and final (f) values. The changes in L*, a*, and b* values contribute to the overall color change, ΔE*.

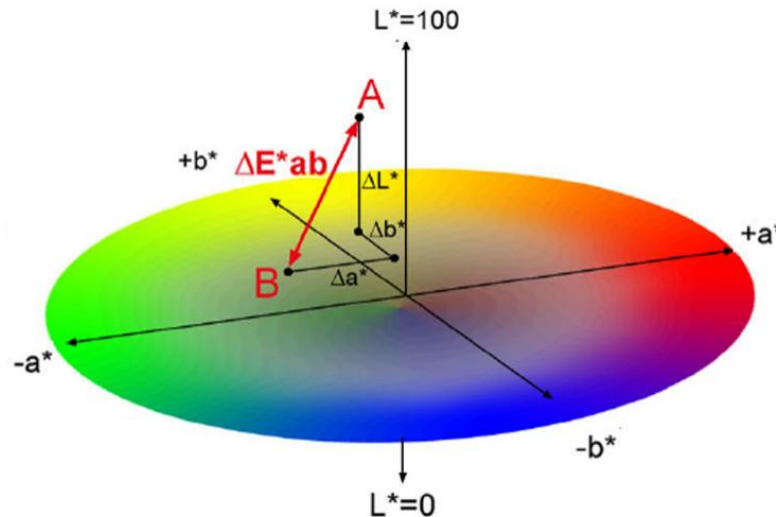


Figure 1: The CIELab color space [10]

The gloss test of wood specimens was determined using a Gloss checker (HORIBA IG-320, HORIBA, Ltd., Japan) according to ASTM D523-14. The chosen geometry was from an incidence angle of 60° . Results were based on a specular gloss value of 91, which related to the perfect condition under identical illumination and viewing conditions of a highly polished, plane, and black glass surface.

2.5. Statistical Analysis

To determine the effects of wood preservatives on the UV resistance of coating film on the treated wood surface, the mean comparison of different formulations used in the color change, gloss surface study has been analyzed by one-way ANOVA and homogeneous groups by using SPSS 25.0 statistical software package.

III. Results And Discussion

3.1. Gloss test results

The gloss values of the paint film on the StyraX wood treated with rosin-copper at different UV irradiation intervals are shown in Figure 2 and Table 1. We can see that before UV irradiation, the gloss of the paint film on the treated wood surface was slightly lower compared to the untreated control. The average gloss value of the paint film on the surface of the untreated controls was 85.34, while the treated wood samples ranged from 78.09 to 84.48. This is due to the wood being treated with the solutions before painting might increase the surface porosity, thereby reducing the gloss value of the paint film (Ozdemir et al., 2015).

After UV irradiation, the glossiness of the paint film on both treated and untreated wood surfaces was increased after the first 48 hours of UV irradiation. However, the gloss of the paint film then tended to decrease, and when the irradiation time was up to 744 hours, the gloss of the paint film did not continue to decrease but was almost unchanged. After 960 hours of UV irradiation, the gloss of the paint film on the control wood surface was 88.21, an increase of 3.4% compared to before UV irradiation. The gloss of the paint film on the wood surface treated with rosin sizing agent alone or in combination with copper was about 76.23 – 86.84, decreased from 0.05 % - 6.76 %. This result agreed with the results reported by Baysal et al. (2014) that there is a slight decrease in glossiness on Scots pine treated with copper-based preservatives, followed by a coating of synthetic and polyurethane varnishes after 500 h accelerated weathering. When the concentration of the rosin sizing agent increases from 1% to 4%, the gloss of the paint film tends to increase, notably wood samples treated with 4% rosin sizing agent increased by nearly 3.4%. However, the results of ANOVA analysis (Table 1) showed that there was a significant difference in the gloss of paint film on treated and untreated wood surfaces after UV irradiation. There was also a significant difference in the gloss of paint film on rosin-copper solution treated wood surfaces when the concentration of rosin sizing agent increased from 1% - 4%. These results have shown that UV rays caused a slight decrease in the glossiness of the paint film on the treated wood surface. The lowest percentage decrease in glossiness values after UV irradiation showed that impregnation with 2% or 4% rosin sizing agent plus 3% copper sulfate before PU coating protected the wood surface against gloss losses.

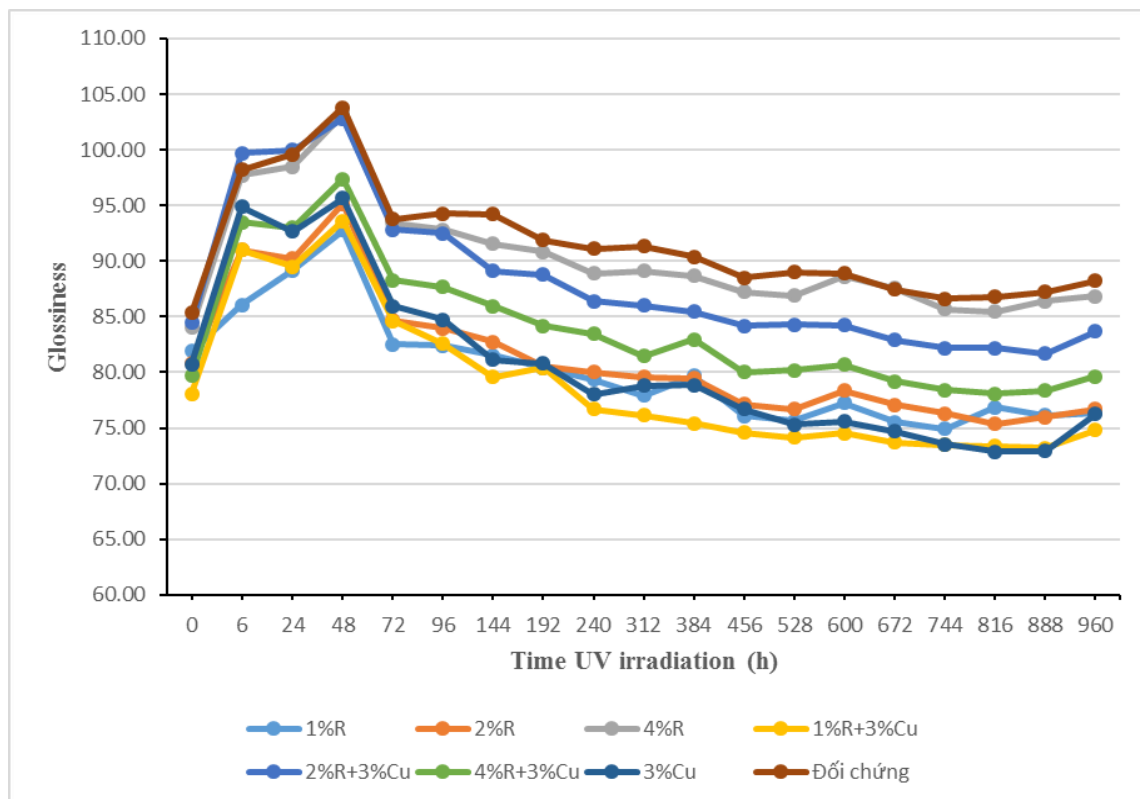


Figure 2: Gloss change of coating film on StyraX wood treated with rosin-copper at different UV irradiation intervals

Table 1: The gloss values of coating film on StyraX wood treated with rosin-copper after 960h exposed to UV light

Solutions Concentrations	and Retention (kg/m ³)	Before UV irradiation	After UV irradiation	Change (%)
1%R	6.70 (0.55)	81.90 (2.37)	76.37 (1.57) ^{cd}	-6.76
2%R	12.88 (0.88)	80.77 (5.95)	76.67 (6.77) ^{cd}	-5.08
4%R	25.49 (1.40)	84.00 (1.49)	86.84 (2.21) ^b	3.37
1%R+ 3% Cu	26.44 (1.53)	78.09 (3.29)	74.80 (6.84) ^d	-4.20
2%R+ 3% Cu	34.29 (2.23)	84.47 (2.62)	83.67 (8.62) ^b	-0.95
4%R+ 3% Cu	44.26 (3.64)	79.69 (2.31)	79.65 (2.45) ^c	-0.05
3% Cu	20.90 (1.60)	80.73 (4.70)	76.23 (5.95) ^{cd}	-5.57
Control	-	85.34 (2.89)	88.21 (5.54) ^a	3.37

Note: Standard deviations are in brackets; Cu: anhydrous copper sulfate and R: rosin sizing agent; Means within a column followed by the same letter are not significantly different at 5% level of significance using the one-way ANOVA test.

3.2. Color test results

The result of the change in value for all three color parameters (ΔL^* , Δa^* , and Δb^*) and the total color change (ΔE^*) of coating film on StyraX wood treated with rosin-copper at different UV irradiation intervals are shown in Figure 3 and Table 2. Before UV irradiation, the values L^* , a^* , and b^* of the paint film on the surface of untreated samples (control) are 75.88, 8.89, and 26.54, respectively. For the wood samples only impregnated to rosin sizing agents, L^* values ranged from 73.29 to 76.01; a^* values ranged from 8.99 to 10.36 and b^* values from 27.75–29.23. However, after combination with copper sulfate, L^* and a^* values slightly decreased only ranging from 71.04 to 72.22 and from 6.48 to 7.63, respectively. This result showed that the color of the paint film on the surface of StyraX wood after being treated with the rosin sizing agents only did not be changed much compared to the untreated sample but after being combined with copper sulfate, the treated wood surface changed greener and slightly darker, thus the color of the paint film on the surface of treated wood also changed

darker. This result was due to the presence of copper in the impregnation solution (Figure 4).

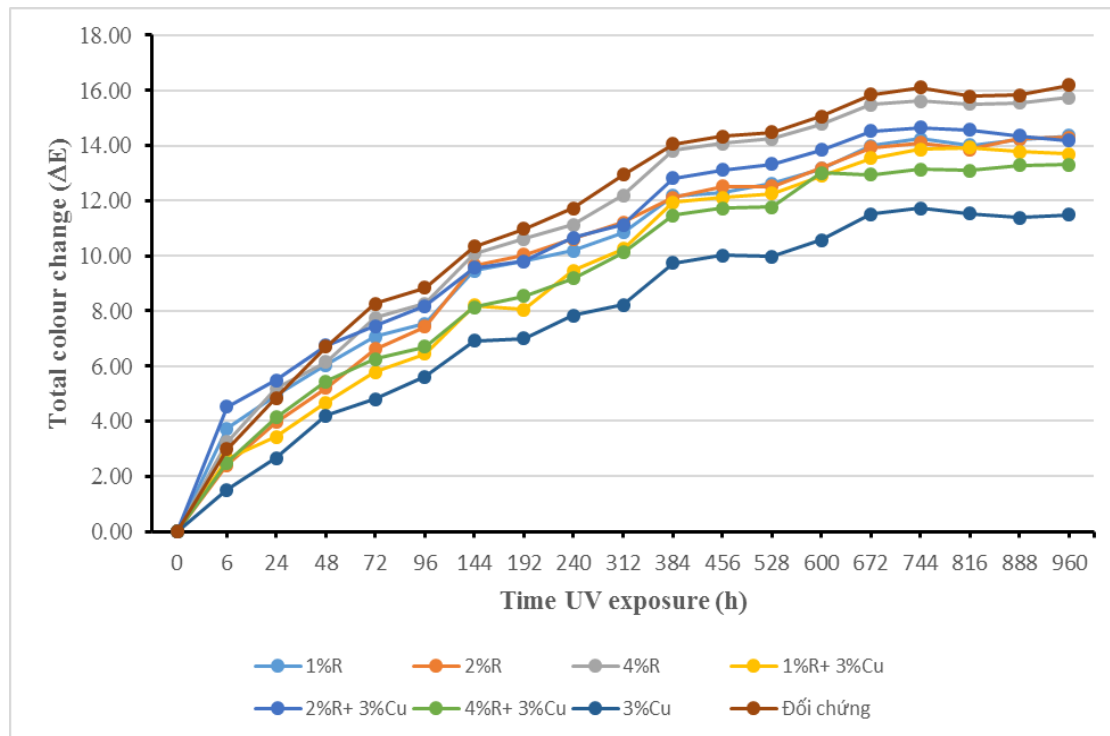


Figure 3: Total color change of coating film on StyraX wood treated with rosin-copper at different UV irradiation intervals

Table 2: Color change and gloss value of coating film on StyraX wood treated with rosin-copper after 960h exposed to UV light

Solutions and Concentrations	Before UV irradiation				After UV irradiation		
	L*	a*	b*	ΔL*	Δa*	Δb*	ΔE*
1%R	74.90 (6.93)	9.27 (1.02)	28.51 (1.52)	-9.49 (2.98)	7.52 (1.49)	7.47 (3.84)	14.35 (4.60) ^{abc}
2%R	73.29 (3.42)	10.36 (1.44)	27.75 (2.32)	-8.67 (2.27)	7.47 (1.44)	8.29 (2.25)	14.29 (2.53) ^{abc}
4%R	76.01 (1.29)	8.99 (1.39)	29.23 (1.84)	-11.25 (0.84)	8.42 (0.69)	6.99 (1.34)	15.74 (0.96) ^{ab}
1%R+ 3% Cu	71.04 (3.26)	6.64 (1.51)	28.77 (1.93)	-10.63 (1.77)	8.02 (1.77)	2.6 (2.16)	13.69 (2.59) ^c
2%R+ 3% Cu	72.22 (3.28)	6.48 (0.68)	27.06 (2.76)	-11.29 (2.52)	7.54 (1.18)	3.38 (1.38)	14.17 (1.70) ^{bc}
4%R+ 3% Cu	71.04 (1.71)	7.63 (0.28)	29.10 (0.96)	-10.95 (0.82)	7.24 (0.35)	1.96 (0.55)	13.29 (0.67) ^{cd}
3% Cu	67.97 (2.85)	7.88 (0.76)	28.30 (2.51)	-9.41 (2.02)	6.32 (0.76)	1.16 (1.38)	11.49 (1.92) ^d
Control	75.88 (1.65)	8.89 (1.77)	26.54 (2.41)	-8.87 (1.41)	8.21 (1.29)	10.67 (2.4)	16.19 (2.63) ^a

Note: Standard deviations are in brackets; Cu: anhydrous copper sulfate and R: rosin sizing agent; Means within a column followed by the same letter are not significantly different at 5% level of significance using the one-way ANOVA test.

Under the influence of UV rays, the color of the paint film was changed a lot during the first 744 hours of UV irradiation, then the color of the paint film changed little or almost no change. After 960h of UV irradiation, the values of Δa* and Δb* were positive in all untreated samples (control) and samples treated with rosin sizing agent solution and copper sulfate alone or in combination. This showed that the paint film on the surface of StyraX wood becomes more red-pink under the influence of UV lights. The ΔL* value is considered to be the most sensitive parameter of wood surface quality (Baysal, 2012). The luminance stability (ΔL*) was found to be negative for all treated and untreated samples, indicating that the wood surface became darker after UV irradiation. The total color change (ΔE*) of the untreated samples was 16.19 and the samples treated with rosin sizing agents alone ranged from 14.29 to 15.74. However, the samples treated with copper sulfate alone or in combination with rosin sizing agents had a smaller total color change than the control samples and the samples treated with rosin sizing agents alone, only between 11.49 – 14.17. This may be due to the presence of

copper in the impregnation solution. In the process of the wood surface being affected by UV rays, the composition of the wood affected first is lignin and causes discoloration of the wood (Sudiyani 1999; Sudiyani et al. 2003; Evans 2008). When wood is treated with a mixture containing copper, it slows down the photodegradation of the wood by inhibiting the formation of carbonyl groups (Zhang, 2003; Pandey 2005), which reduced wood discoloration compared to untreated controls and the samples treated with rosin sizing agents alone. The results of the Anova variance analysis (Table 2) also showed that there was a significant difference in the color change of wood treated with copper sulfate alone or combined with rosin sizing agent compared with the untreated controls or samples treated with rosin sizing agent alone. However, there was no significant difference when the concentration of the rosin sizing agent increased from 1% to 4%. This result showed that using the rosin sizing agent in combination with copper sulfate to impregnate styraX wood could improve the color stability of the paint film on the wood surface under the influence of UV rays.

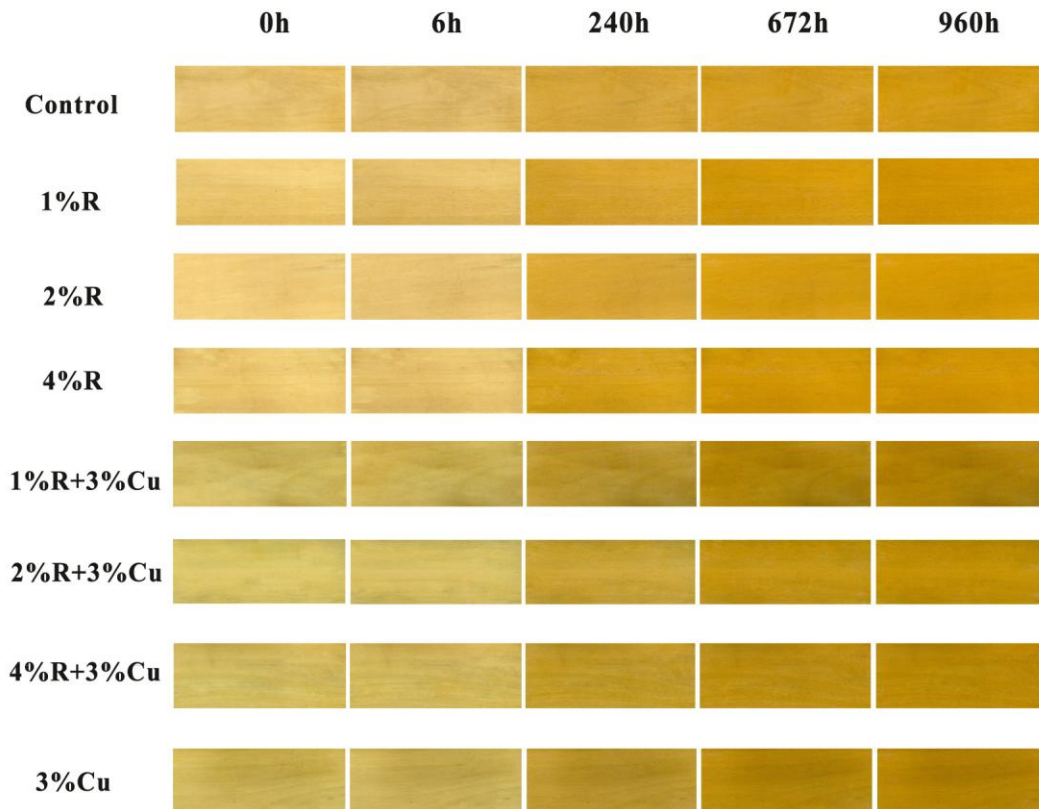


Figure 4: The appearance of coating film on untreated control and treated samples before and after UV exposure (Cu: anhydrous copper sulfate and R: rosin sizing agent).

IV. Conclusions

The glossiness and color stability of wood are very important properties for its aesthetic and decorative appearance. This study evaluated the effect of rosin-copper formulations on the color change and gloss of coating film on the treated StyraX wood surface. The results showed UV lights caused a slight decrease in the glossiness of the paint film on the wood surface treated with the mixture of rosin sizing agent and copper sulfate. The lowest percentage decrease in glossiness values was detected in the specimens treated with 2% or 4% rosin sizing agent plus 3% copper sulfate. Wood surfaces became greener and slightly darker after being treated with rosin-copper formulations in comparison with the initial surfaces of wood specimens, thus the color of the paint film on the surface of treated wood also changed slightly darker before UV irradiation. However, the paint film on the surface of all untreated and treated StyraX wood samples became more red-pink after UV irradiation. All samples treated with rosin sizing agents alone or in combination with copper sulfate had a smaller total color change than the untreated control samples. The concentration of rosin sizing agents used in this study did not significantly affect the color discoloration of the paint film. The result showed using the rosin sizing agents in combination with copper sulfate to impregnate styraX wood could reduce the damage of the copper preservative leaching into the environment, while also improving the color stability of the paint film on the wood surface under the influence of UV rays.

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Thi Thanh Hien Nguyen. "Change in Surface Properties of Coating Film on Styra Wood Treated with Rosin-Copper During UV Irradiation." *IOSR Journal of Polymer and Textile Engineering (IOSR-JPTE)*, 09(02), 2022, pp. 01-07.