Assessment of Color and Contact Angle Change of Weathered Wood in Relation to Wood Species and Different Coating Types

Seungmin Yang,[a] Junho Kim,[b] and Seoggoo Kang [b,]*

Color change was compared through artificial and outdoor weathering tests according to wood species and stain type. In the artificial weathering tests, the color change Δ*E* was the highest for the initial 200 hour, and teak solvent-based stain was the most effective in preventing color change. Outdoor weathering tests also showed a rapid color change until the initial 60 days, and the uncoated larch specimens exhibited graying after 120 days. Teak solvent-based stain had the highest preventing color effect, whereas water-based white semi-transparent stain had the highest contact angle. It is difficult to check the color change of wood due to the addition of pigment in teak, as its resistance to moisture is rapidly reduced and its surface protection effect is poor. Water-based white semi-transparent stain prevented color change and maintained a contact angle of 57.1° for up to 150 days, confirming the effect of moisture resistance. This study aimed to provide basic data on weather resistance by wood species and to suggest that the development direction of outdoor exposed wood is a water-based semi-transparent stain.

DOI: 10.15376/biores.18.4.8296-8310

Keywords: Artificial weathering; Outdoor weathering; Contact angle; Solvent-based stain; Water-based stain; Transparent stain; Semi-transparent stain; Wood species

Contact information: a: Institute of Agricultural Science, Chungnam National University, P.O. 34134, 99 Daehak-ro, Yuseong-gu, Daejeon, Korea; b: Department of Biobased materials, Chungnam National University, P.O. 3414, 99 Daehak-ro, Yuseong-gu, Daejeon, Korea;
** Corresponding author: lachesis@cnu.ac.kr*

INTRODUCTION

The world is declaring a goal of 2050 carbon neutrality to prevent global warming (Wang *et al.* 2021). To achieve carbon neutrality, attention is being paid to forests that absorb carbon dioxide and wood materials that store carbon (Kazulis *et al.* 2017). Wood is an environmentally friendly renewable resource and building material that has been in use for thousands of years (Keržič *et al.* 2021). In addition to building materials, wood is used for various purposes such as indoor interior materials, furniture, outdoor benches, street lamps, and tables (Nguyen *et al.* 2020). In 2023, Stockholm, Sweden, announced that it would create the "world's largest wooden city". The project used more wood than any other project and designed the world's largest mass timber city. In addition, by 2022, the Korea Forest Service is promoting a project to create urban wood utilization to promote the use of domestic timber (Lim *et al.* 2021). Urban wood utilization will create a street where the city's landscaping facilities are made of wood and wood experience centers that run wood experience programs. This is expected to change consumer's awareness of the use of wood through natural contact with it, increase demand for domestic wood products through the use of wood in the region, and create jobs using wood in the region.

The primary objective of the wood street development, which is a representative project in the realm of urban wood utilization, is to develop outdoor facilities in the city, including benches, streetlamps, bus stops, and shelters, all crafted from locally sourced wood. However, when wood with an unprotected surface is used outdoors, it loses its original properties and rapidly degrades, causing the wood to crack or turn gray (Oberhofnerovă *et al.* 2018). This phenomenon is called weathering, and in general, wood surface deterioration by climate is initiated by UV radiation among solar radiation and other environmental factors (Žlahtič and Humar 2017; Kropat *et al.* 2020). Shorter wavelengths of the solar spectrum with higher photon energies are absorbed by materials such as wood. The absorbed energy has direct effects on the chemical transformation of the wood cell walls (Teacă *et al.* 2019). Weathering initially changes the wood surface color, and then surface cracking and roughness increase (Arpaci *et al.* 2021). Weathered wood has a more pronounced yellow color than wood that has not been exposed to weather because of the transformation of lignin and hemicellulose (Kubovský *et al.* 2018). It is mainly affected by the decomposition and photochemical reaction of lignin, and most of the decomposed lignin is washed away during rainfall (Sudiyani *et al.* 1999). However, cellulose, which is highly resistant to UV degradation, is preserved without deformation, and the color of wood turns gray (Jirouš-Rajković and Miklečić 2021).

To change the color of wood and increase its lifespan, wood products used outdoors are treated with various types of chemicals or are finished with paint. Wood species that are resistant to weather factors, as well as oil stain coatings are used to improve weather resistance. Pánek *et al.* (2018) reported that oak wood (*Quercus petraea* L.) resists biological damage and can be used outdoors as a species with higher weather resistance. However, even for wood species with high weather resistance, when exposed outdoors without surface protection, the color changes rapidly, and the extractive compounds are leached. Heshmati *et al.* (2018) manufactured a wooden deck using Douglas fir, western hemlock, and white spruce, all which are commercial species used in Canada, and studied the checking and cupping of the wooden deck according to outdoor exposure. Kim *et al.* (2017) compared the durability of wooden decks by species with tropical hardwoods such as lpe (*Tabebuia ipe Standle),* malas (*Homalium foetidum Bth*), and massaranduba (*Manilkara huberi (Ducke.) Standl*), which distributed as wooden deck materials, and Douglas fir (*Pseudotsuga menziesii*) and Japanese larch (*Larix leptolepis*), which are softwoods in Republic of Korea. Investigation on wood color change under exposure to environmental outdoor conditions was conducted for commercial wood species (Heshmati *et al.* 2018; Kim *et al.* 2017). Various stain options include transparent, semi-transparent, and opaque stains. As contemporary architectural trends evolve and consumer preference shift, transparent stains have gained prominence for preserving the innate beauty of natural wood. Nonetheless, it is known that transparent stains typically fall short in terms of durability compared to opaque stains (Cogulet *et al.* 2018). The potential issues surrounding the longevity of transparent stains could potentially hinder the increasing use of wood as a construction material. Consequently, there is a need for research delving into the color retention and durability of different stain types.

This study compared the changes in wood color due to outdoor exposure and artificial deterioration for malas, tropical hardwood, hemlock, and Japanese larch, which are softwoods used for outdoor landscaping facilities. With an increasing number of landscaping facility materials, the basic data on the initial weatherability of each wood species is reported.

EXPERIMENTAL

Sample Preparation

The wood species malas (*Homalium foetidum*), hemlock (*Tsuga heterophylla* Sargent), and Japanese larch (*Larix kaempferi*), which are used for outdoor landscaping facilities, were selected, and kiln-dried lumber was purchased. The supplied lumber was cut into 10 mm thick, 70 mm wide, and 150 mm long sections. The surface of lumber was finished according to ISO 16474-1 (2013). The prepared specimens were equilibrated for 7 days before coating at 20 ± 2 ℃ and a relative humidity of 65 ± 2%.

The stain was a commercial product (Aju Chemical Co., Ltd., Gimchen City, Republic of Korea), and five types of outdoor oil- and water-based stains were selected as transparent and semi-transparent (Table 1). The selected stain was applied twice at an application amount of 0.03 mL/cm^2 and was re-applied after drying for 24 h after application. The coated specimens were stored for 7 days in a room not exposed to UV radiation and subjected to artificial deterioration and outdoor exposure tests. At this time, a Plustuk OpticBook 4800 (Plustek Inc., China) flatbed scanner with a resolution of 1200 dpi was used to observe the color change of the specimen over time.

Table 1. Description of Selected Coatings

Coating	Base	Type	Color	Repetitions		
				Malas	Hemlock	Japanese larch
Control	N/A	N/A	N/A	5	5	5
SC	Solvent	Transparent	Clear	5	5	5
ST	Solvent	Opaque	Teak	5	5	5
WC	Water	Transparent	Clear	5	5	5
WT	Water	Semi-transparent	Teak	5	5	5
WW	Water	Semi-transparent	White	5	5	5

Contact Angle Test

The contact angle was measured to compare the water absorption of wood species and stains and compared according to the outdoor exposure time. Contact angle analyzer (Phoenix 150, SEO Co., Ltd. Korea; contact angle: 0 to 180° (±0.1°), frame: 70 frames/s, CCD camera resolution: 640 × 480 pixels) was used (Kim *et al.* 2022). The wood was installed in a contact angle measuring device, and the instantaneous surface contact angle was measured by dropping 0.1 mL of distilled water and stains on the sample surface from a height of approximately 1 cm from the wood surface. The contact angle was measured 10 times per specimen, and the average contact angles were compared.

Artificial Weathering Test

For the artificial deterioration test, a xenon-arc lamp weatherability test Ci3000+Weather-Ometer (ATLAS, USA) was used according to ISO 16474-2 (2013). The specimen was exposed for 600 h and was configured to repeat UV irradiation and water spray at 120 min intervals. One cycle of 120 min consisted of 102 min of UV irradiation and 18 min of UV irradiation and water spray. A total of 600 h was exposed, and the color of the specimen was measured every 200 h.

Outdoor Weathering Test

Among wood species, Japanese larch, a domestic species in Republic Korea, was tested for outdoor exposure. The coated specimens were exposed to the natural environment at the College of Agriculture and Life Science, Chungnam National University. The specimens were randomly placed in racks, and one wide face was exposed at a 45° surface oriented at a 180° azimuth. The specimens began exposure on June 16, 2020, and ended testing on November 16, 2020. The daily maximum and minimum temperatures and monthly rainfall during exposure is shown in Fig. 2. The Republic of Korea has four seasons, and in particular, during the long rainy season in summer, wood deterioration due to UV radiation and moisture exposure began during the earliest period. At 60, 120, and 150 d after exposure, samples were taken, and image scans, color and contact angles were measured.

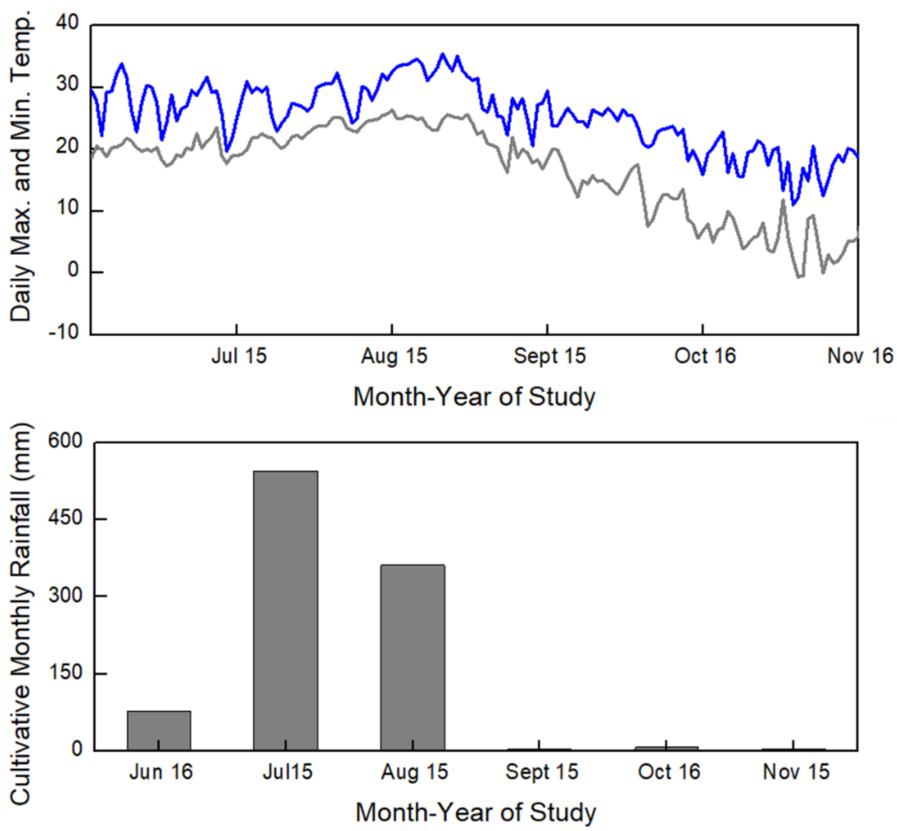

Fig. 1. Maximum and minimum temperature and monthly rainfall for the Chungnam National University (Daejeon, Republic of Korea) during the study

Color Measurement

*L**, *a**, and *b** parameters were measured for the color change of wood using a chroma meter CR-410 (Konica Minolta, Inc., USA) with a diameter of 5 cm. *L** is the brightness factor, and *a** and *b** are chroma coordinates (*a**: redness factor, *b**: yellowness factor) (Bekhta and Niemz 2003; Rosu *et al.* 2010). The *L** represents the gray value from 0 (black) and 100 (white). Positive and negative values of Δ*a** are shown in red and green, respectively. The positive and negative values of Δ*b** are shown in yellow and blue, respectively.

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \quad (1)$$

where ΔL^*, Δa^*, and Δb^* are the changes between initial (i) and final (f) (artificial weathering test: 200, 400, and 600 h; outdoor weathering test: 60, 120, 150 d), the color change is calculated as ΔL^*, Δa^*, and Δb^*. Depending on the exposure time, the overall color changes observed in artificial weathering tests are indicated as ΔE_{200}, ΔE_{400}, and ΔE_{600}, and outdoor weathering tests are indicated as ΔE_{60}, ΔE_{120}, and ΔE_{150}.

RESULTS AND DISCUSSION

Contact Angle Values of Wood and Stains

The wetting results of the wood and stain through the contact angle values are shown in Table 2. The contact angle value between the water and wood specimen was 62° to 69°. In the case of wood species, malas exhibited the highest contact angle values and high resistance to moisture. The contact angle values of the solvent-based stain were smaller than those observed for the water-based stain. According to Nejad and Cooper (2011), the lower the contact angle values, the better the wettability, and the higher the bonding strength and coating performance. Accordingly, solvent-based stains are expected to have better wettability with wood than water-based stains and to improve the bonding strength and coating performance. SC (15.8 to 19.9°) was clear, ST (18.8 to 22.2°) was a teak color stain, and the contact angle values increased by approximately 3° because of the additives added to the pigment. The water-based stains showed high contact angle values in the order of WC, WW, and WT. Similar to the contact angle values of the solvent-based stains, WC indicates clear, WT indicates teak, and WW indicates white. The contact angle increased due to the additives to the pigment in the stain.

Table 2. Comparison of Contact Angle Values according to Wood Species and Stains

Species	Control (water)	SC	ST	WC	WT	WW
Japanese larch	62.2°	15.8°	18.8°	30.4°	59.2°	33.9°
Malas	68.4°	19.9°	22.2°	53.8°	62.9°	40.4°
Hemlock	63.5°	15.9°	19.9°	46.6°	64.0°	37.5°

Color Changes Due to Artificial Weathering Test

The results for ΔE are shown in Fig. 2, and visual change results for specimens are shown in Fig. 3. In general, rapid color changes were observed up to 200 h in the initial stage and showed a high ΔE in the order of hemlock, malas, and larch. Rapid color change in the initial 200 h of the artificial weathering test was reported by Deka *et al.* (2008) and Kržišnik *et al.* (2018). The water-based stain showed a higher color change than the oil-based stain. The color change of the wood decreased in the order of transparent, semi-transparent, and opaque stains.

Larch showed the highest ΔE in the order of ST, WW, WT, SC, and WC depending on the stain type. ST-, WT-, and WW-colored stains were judged to be suitable for preventing color changes in larch.

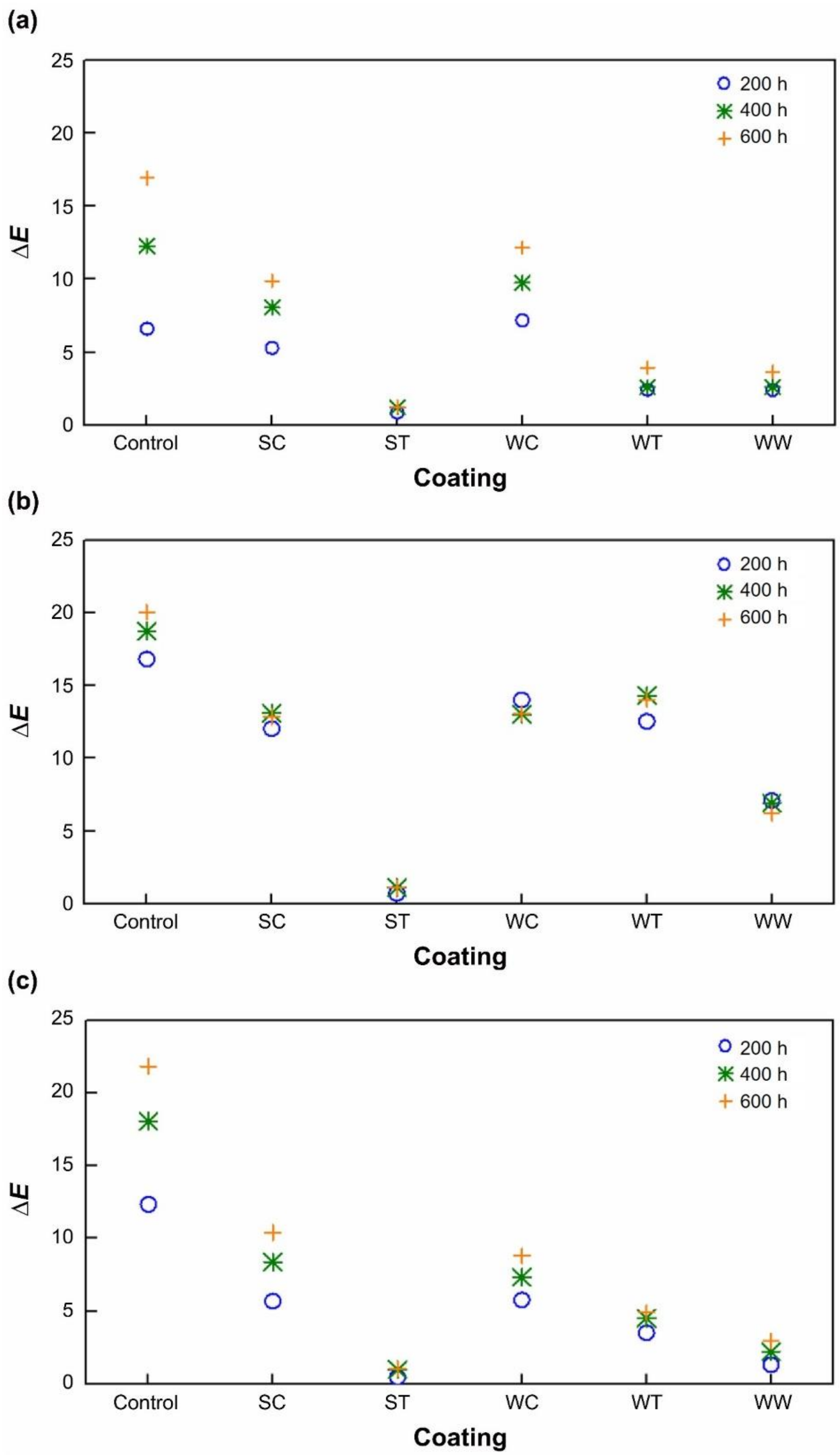

Fig. 2. Result of ΔE according to wood species, stain type, and artificial weathering time ((a) larch, (b) malas, and (c) hemlock)

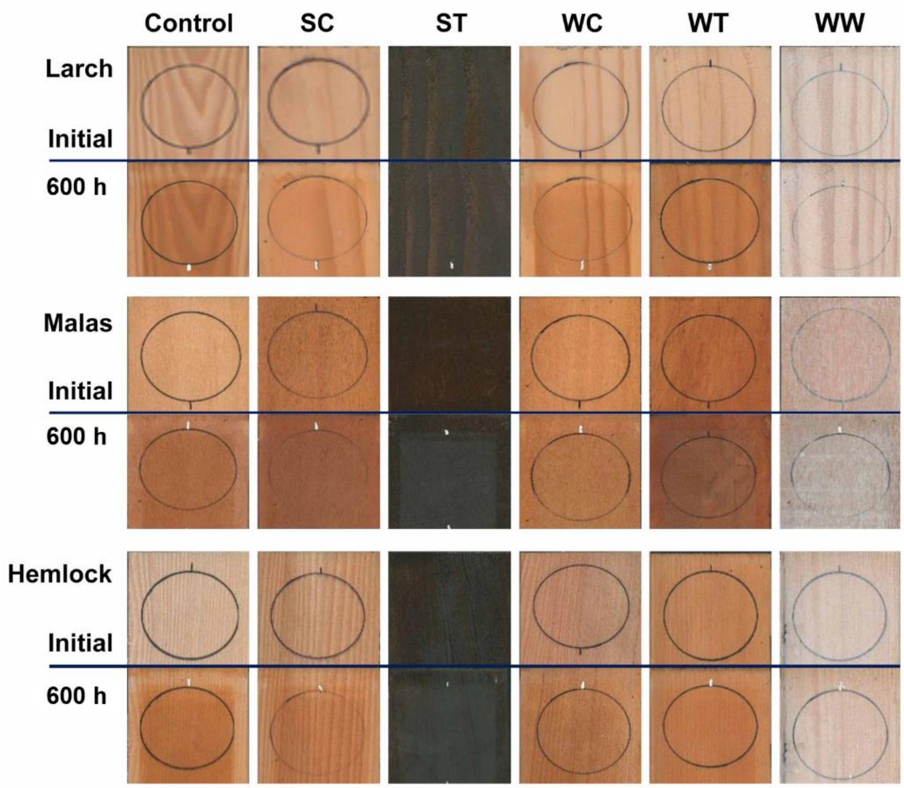

Fig. 3. Visual change of specimens before and after artificial weathering test according to wood species and stains

Malas showed the highest color change among the three wood species. Malas is known to have excellent decay resistant against fungi due to oil contents. However, when exposed outdoors, the extractive component contained in malas are oxidized by UV radiation, promoting color change. Malas showed a difference between WT and WW, which are water-based semi-transparent stains. WT showed a similar color change as WC, but WW showed a decrease in color change. Δ*E* was decreased by reducing the amount of energy absorbed by the light reflected by added white pigment (TiO_2). TiO_2, an inorganic pigment, has received much attention in recent years due to its UV absorption ability and effect on color stability of composites (Wang *et al.* 2017).

The uncoated hemlock had the highest ΔE_{600} values among the wood species, but the coated hemlock showed a rapid decrease in the color change. The SC of hemlock showed the same results as larch SC. The ST of hemlock showed the same very low results as larch and malas. WC showed a lower color change than ST, and the color change in WT was higher than that in WW. Δ*E* was compared according to the type of stains and wood species. The ST showed the lowest color change among all the species. WT and WW differed according to species, but WW was more effective than WT in preventing color change. In contrast, SC and WC showed similar color change, and showed a rapid color change up to ΔE_{200}, similar to the uncoated specimen.

The *L**, *a**, and *b** results are shown in Fig. 4. *L**, *a**, and *b** values for larch specimens before exposure were 62.61, 7.63, and 18.49, respectively. As time passed, Δ*L** decreased and the color shifted to dark, Δ*a** shifted to red, and Δ*b** shifted to the initial yellow and was maintained. SC, ST, WC, and WW all showed similar color changes, although the values of Δ*L**, Δ*a**, and Δ*b** were smaller than those of the uncoated larch.

Moreover, Δ*b** of the WT changed slightly to blue. This is because the pigment in the stain absorbs light and changes color to yellow, rather than the color change of the wood.

Before exposure of malas, its *L**, *a**, and *b** values were 47.45, 12.03, and 18.23, respectively. *L** was lower than that of larch, indicating a dark color, and *a** was high, indicating a red color. As time passed, Δ*L** decreased and the color shifted to dark, Δ*a** changed to redder color, and Δ*b** changed to yellow color. In the SC, WC, and WT of malas, Δ*L** shifted to dark, Δ*a** shifted to green, and Δ*b** turned yellow. In contrast, Δ*L** of WW showed the lowest change among the stain types. This was consistent with the Δ*E* result, in which the white pigment reflected UV radiation to prevent graying by preventing absorption into the wood. The *L**, *a**, and *b** values of uncoated hemlock before exposure were 60.58, 9.04, and 24.11, respectively. The color change differed according to the exposure time. Δ*L** and Δ*a** turned dark and red, similar to larch and malas, respectively. Similar results were obtained for SC, ST, and WT. Δ*L** of the WT showed that the color shifted toward white until the initial 200 h and then toward dark. It was decided to be due to the white pigment added to the stain, such as the color change of malas.

The parameter exhibiting the most significant color change in response to the artificial weathering test is represented by *L**. UV radiation plays a pivotal role in influencing changes in the brightness of wood. This was consistent with previous studies showing that wood initially exhibits a rapid color change when exposed to UV radiation, and graying occurs as the color changes to dark (Deka *et al.* 2008).

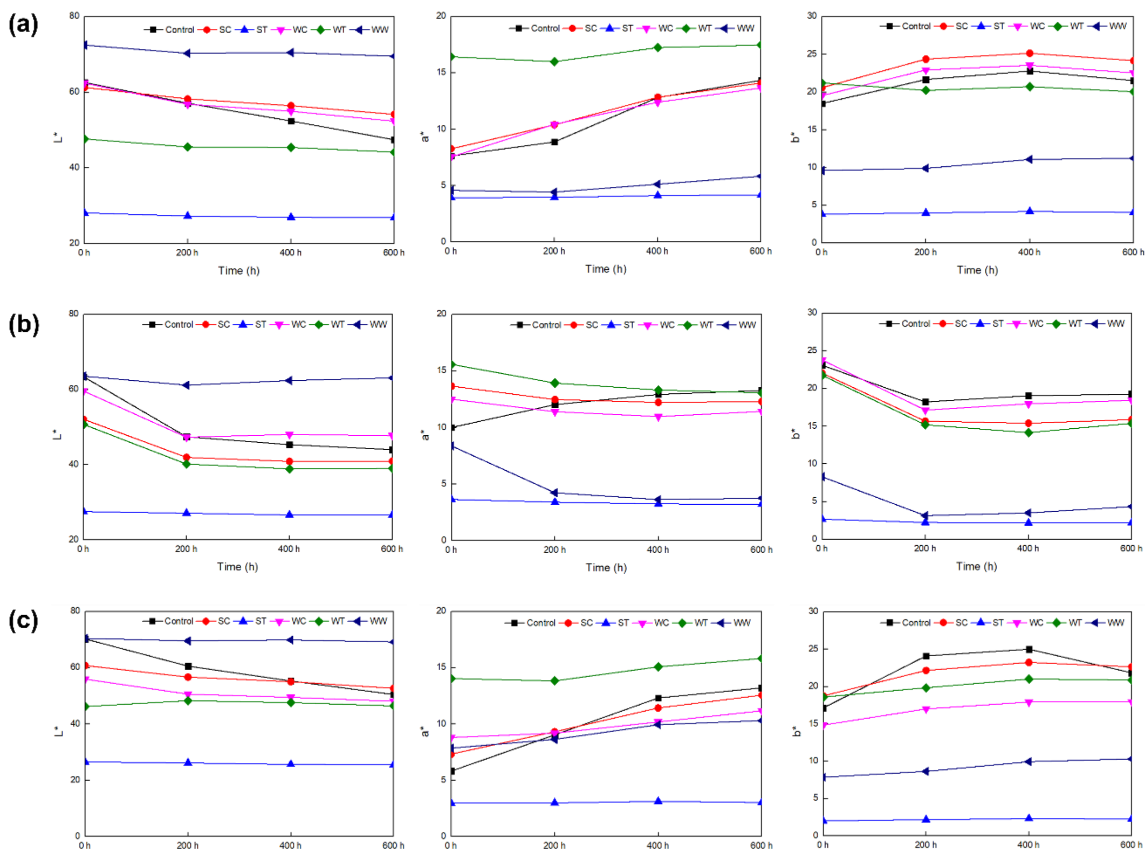

Fig. 4. L*, a*, and b* changes according to artificial weathering time and stain ((a): larch, (b) malals, (c) hemlock)

Color and Contact Angle Values Change due to Outdoor Weathering Test

Following the artificial weathering test, larch emerged as the chosen representative wood species due to its comparable color alteration, between WT and WW conditions. Figure 5 illustrates the Δ*E* in larch specimens, while Fig. 6 shows the visual change results.

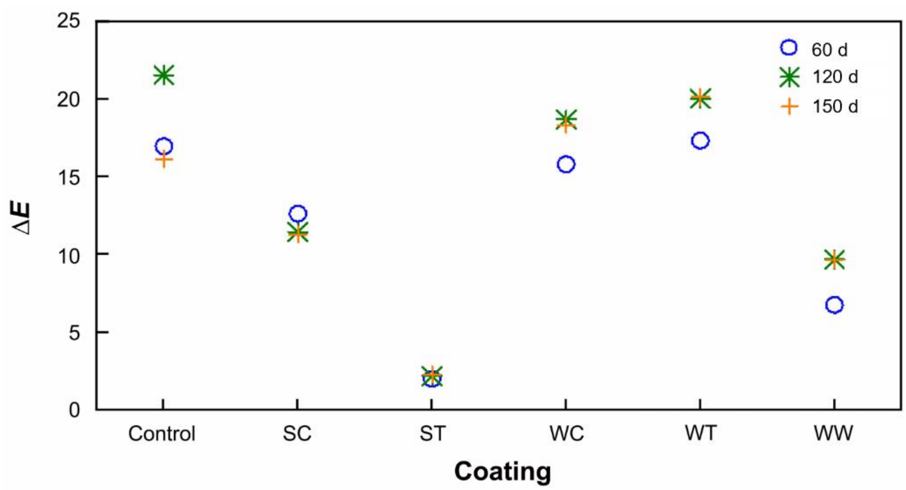

Fig. 5. Δ*E* change according to the stain types and outdoor exposure time

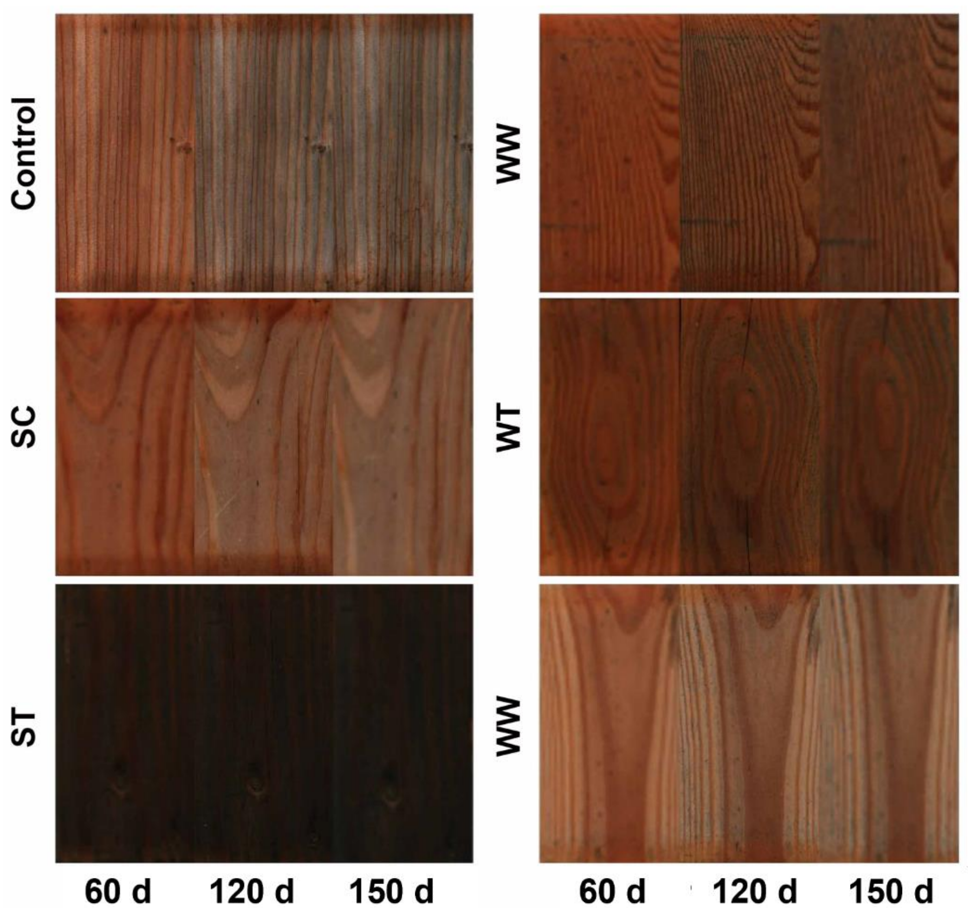

Fig. 6. Visual changes of uncoated and coated larch specimens according to the time and stains

During the outdoor weathering test, the uncoated larch specimen increased ΔE_{120} to 21.57 and decreased ΔE_{150} to 16.15. The ΔE of solvent-based stains was lower than that of water-based stains, and colored stains (ST and WW) prevented color change more effectively than transparent stains. This result is consistent with the findings of Cogulet *et al.* (2018). The cited authors reported that clear protection does not provide the same durability as opaque protection, and that it is difficult to provide long-term durability along with surface protection. WC and WT were equal to the ΔE_{60} of the uncoated larch, and ΔE_{120} showed a difference of 1 to 3. ΔE_{150} had a value similar to that of ΔE_{120}. ST showed the lowest ΔE among the stain types, similar the artificially weathered color change, with results of 2.01, 2.18, and 2.28 according to ΔE_{60}, ΔE_{120}, and ΔE_{150}. ΔE_{60}, ΔE_{120}, and ΔE_{150} of WW were 6.76, 9.69, and 9.66, showing rapid color change until the initial 60 d, and then maintaining the color.

The ΔL^*, Δa^*, and Δb^* values according to the exposure time and stain type are shown in Table 3. The ΔE of uncoated larch increased up to 120 days and rapidly decreased at 150 days. This was most affected by ΔL^* and showed graying that moved to the dark by 120 days and then brightened toward white. This was consistent with the results of Feist and Hon (1984). Among the stain types, the solvent-based stain showed lower color change than the water-based stain, and the colored stain showed lower color change. ΔL^*60, ΔL^*120, and ΔL^*150 of WW were -5.43, -6.41, and -6.26, respectively, showing low color change except for ST, but Δb^*60, Δb^*120, and Δb^*150 were -3.45, -5.94, and -6.02, respectively, and the color of the wood was changed to yellow. This is due to the decrease in ΔL^* by the pigment added to WW, which can slow down the graying rate; however, the decrease in Δb^* changes the wood color from yellow to brown.

Table 3. ΔL^*, Δa^*, and Δb^* Changes for Larch Wood Species According to Exposure Time and Stain

	60 d			120 d			150 d		
	ΔL^*	Δa^*	Δb^*	ΔL^*	Δa^*	Δb^*	ΔL^*	Δa^*	Δb^*
Control	-13.0	-5.78	-9.19	-15.92	-8.09	-12.04	-11.46	-6.26	-8.99
SC	-12.08	1.01	-2.73	-9.16	-2.8	-4.55	-8.18	-3.52	-5.15
ST	-1.74	-0.1	-0.92	-1.61	-0.69	-1.21	-1.52	-0.83	-1.35
WC	-15.55	1.5	-4.06	-17.79	-1.69	-5.43	-17.35	-1.91	-5.52
WT	-17.12	0.93	-2.41	-19.22	-2.18	-5.2	-19.26	-2.37	-5.4
WW	-5.43	-1.62	-3.45	-6.41	-3.68	-5.94	-6.26	-3.67	-6.02

The results of the contact angle value according to the stain and exposure time are shown in Fig. 7. The contact angle values before exposure were in the range of 60° to 70°, with uncoated larch at 62.2°, SC 66.5°, ST 64.1°, WC 70.2°, WT 61.2°, and WW 68.6°. After 60 days, the contact angle values showed a rapid decrease, and the uncoated larch showed a rapid decrease to 28.5°. After 120 and 150 days, the contact angle values changed within 10°. As water contributes to wood aging, the contact angle values with water on the exposed wood surface is an important indicator of the weathering rate (Van den Bulcke *et al.* 2011; Petrič and Oven 2015; Žlahtič and Humar 2016). The water-based stain showed lower contact angle values changes than solvent-based paint, and WC and WW showed high contact angle values of 48.5° and 57.1°, respectively. The results of contact angle value according to wood species and stains during coating (Table 2) show that the contact angle value of the solvent-based stain was higher than that of the water-based stain, resulting in high wettability and bonding strength; however, the coating layer was thin, resulting in a high rate of weathering when exposed to moisture.

The difference in contact angle according to pigment color was also shown, and ST and WT with teak color pigments showed lower contact angles than SC and WC. Teak color staining can show the effect of preventing color change in wood, but the dimensional stability will decrease due to the decrease in water resistance due to the decrease in contact angle, and cracking will occur later. WW prevented the color change of wood and the contact angle was maintained high for up to 150 days, so the white pigment prevented wood weathering from UV radiation.

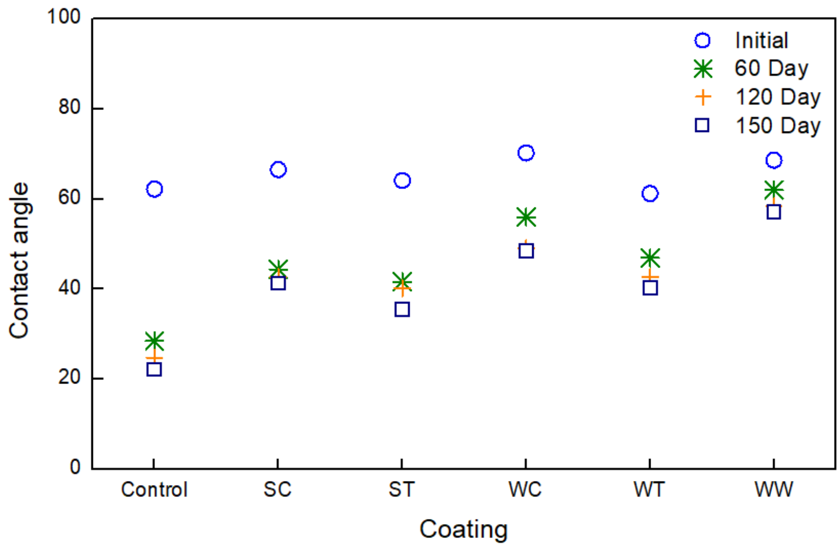

Fig. 7. Change in contact angle according to stains and exposure time

The color changes of larch specimens due to the artificial and outdoor weathering test were compared. ΔE showed similar values after 600 h of artificial weathering and 150 d of outdoor weathering. In contrast, outdoor weathering turned gray after 120 d. Feist and Hon (1984) reported that initial wood surface colors tended to be darker, and darker colored surfaces lightened and turned gray during weathering. This is because exposure to the UV component of sunlight primarily affects the color change of lignin and extractives from the wood surface (Arpaci *et al.* 2021). In particular, UV radiation from sunlight is absorbed inside the wood, starting a photochemical reaction that decomposes lignin, hemicellulose, and extractives. When the decomposition products are leached into water, the surface degradation process starts (Chang *et al.* 2010; Tolvaj and Mitsui 2010; Laskowska *et al.* 2016). This usually causes the wood surface to turn yellow or brown, eventually graying the wood surface (Davis *et al.* 2021; Jirouš-Rajković and Miklečić 2021). Davis *et al.* (2021) studied exterior wood coatings performance. It has been reported that most coatings lose their wood protection effect within one year due to biological and UV-induced deterioration. UV radiation affects the decomposition and photochemical reactions of lignin in wood, leading to its breakdown (Cogulet *et al.* 2018). This process subsequently generates secondary chromophores that shift the color of the wood to yellow hues. Additionally, moisture-induced surface leaching can result in the formation of fractures, splits, and checks on the surface of the wood, which can compromise the dimensional stability of the wood. Prolonged exposure to moisture fosters the growth of fungi on the surface of the wood, causing biological degradation that presents as alterations in color and depth within the wood.

The outdoor weathering test encompasses climatic conditions involving 3 months of heavy rainfall followed by 3 months of nearly no precipitation. Previous research by Williams *et al.* (2001) revealed water to be a potent solvent capable of eroding wood surfaces at an average rate of 50 μm per year. The substantial rainfall impacts wood erosion and contributes to weight loss by transporting products resulting from the photolysis and hydrolysis induced by UV radiation. This study serves as valuable foundational data for assessing the suitability of using wood outdoors in regions characterized by fluctuating climate, marked by exposure to UV radiation and heavy rainfall.

As the climate varies depending on latitude and longitude, research on weathering according to the place of use is necessary. In the Republic of Korea's climate, resistance to moisture and graying occurs within 120 d of exposure due to heavy rainfall in summer; therefore, it is necessary to develop a stain with a UV-blocking effect.

CONCLUSIONS

1. The contact angle values according to the solvent type and wood species showed the highest contact angle values in the order of larch, hemlock, and malas. Among the wood species, malas showed high resistance to water. The solvent-based stain showed a fast absorption rate of less than 30°, and the water-based stain showed 30° to 60°.
2. The color change during artificial weathering initially rapidly changed up to 200 h, and ΔE decreased at 400 and 600 h. The change in ΔE was closely related to the initial color of the wood, and malas, which has a dark color, low color change with ΔE of 5 or less until 400 and 600 h after a rapid color change until 200 h. Depending on the type of stain, solvent-based staining had a higher color-change prevention effect than water-based staining, and among solvent-based stains, opaque stain with teak pigment added had a higher color change prevention effect than transparent staining. For water-based stains, the color change prevention effect of transparent stains with added pigments was high, and white pigments had greater UV-blocking effects than teak pigments.
3. The color change due to outdoor weathering of larch at 60, 120, and 150 d was similar to the result of artificial weathering. Depending on the type of stain, the solvent-based stain had a higher prevented color change effect than the water-based stain, and solvent stain (ST) with teak pigment added had the highest preventing color change effect. Among the water-based stains, WW with a white pigment was the most effective. However, the contact angle value with water over the exposure time was the highest in WW and lowest in ST among the coating specimens. It is difficult to check the color change of wood by adding pigment, but water-based stain is advantageous for the long-term use of wood because its resistance to moisture is rapidly lowered.
4. Through this study, it was confirmed that WW showed the best effect, and that water-based stains with UV-blocking pigments were suitable for the Republic of Korea climate. Research on the development of water-based semi-transparent stains with UV-blocking effect immediately after exposure and high moisture resistance that can respond to concentrated rainfall is necessary.

ACKNOWLEDGMENTS

This study was carried out with the support of "R&D Program for Forest Science Technology (Project No. 2023495A00-2325-AA02)" provided by the Korea Forest Service (Korea Forestry Promotion Institute).

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Article submitted: August 27, 2023; Peer review completed: September 23, 2023; Revised version received and accepted: October 5, 2023; Published: October 20, 2023.
DOI: 10.15376/biores.18.4.8296-8310