# Color and Dimensional Stability of Oil Heat-Treated Radiata Pinewood after Accelerated UV Weathering

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# Abstract

*Pinus radiata* specimens at a moisture content of 10 to 12 percent were heat treated at temperatures of 160°C, 180°C, and 210°C in commercial-grade raw linseed oil. The end-matched treated and untreated wood was exposed to an accelerated UV weathering environment for 2,100 hours using UV radiation and intermittent water spray. Changes in color and dimensional stability properties of the oil heat-treated wood were examined after (1) heat treatment and (2) accelerated UV weathering. The results show that the oil heat-treated wood turned darker and was more dimensionally stable. After oil heat treatment, the color of both the surfaces was uniform. Although color was darker on the surface than at the core, this contrast decreased with increasing treatment temperature. After the accelerated weathering test, the oil heat-treated wood retained its dimensional stability and color better than the untreated wood. No surface checks were observed in weathered treated wood, and volumetric swelling was also less compared with the untreated wood. Similarly, no color fading was observed in wood treated at 160°C, and 180°C, while slight fading was noted for specimens treated at 210°C.

The color of wood is very important for applications related to aesthetics. Color preferences are cultural and depend on end use, location, and change with time. Some markets, such as Scandinavia and Japan, prefer light-colored wood for certain end uses (Wahl et al. 2002, Esteves et al. 2008), whereas in South Asia, people like dark-colored wood, as it is considered to be a symbol of strength, stability, and durability (Dubey 2001). Similarly, in Portugal, darker woods are also preferred, as the dark color is associated with high-quality tropical species (Esteves et al. 2008). In the latter markets just mentioned, species of pine, poplar, and eucalyptus are considered less attractive because of their light or pale color. In addition, homogeneous wood color is considered an advantage for uses with large surfaces, such as flooring. This makes certain species like black locust (Robinia pseudoacacia) less preferable for such applications because of its nonhomogeneous color caused by deposition of extractives (Tolvaj and Molnar 2006). Similarly, after felling, red alder wood (Alnus rubra Bong.) acquires a mottled orange color that is undesirable for decorative applications (Thompson et al. 2005).

Another important aspect of wood utilization is dimensional instability, which is one of the major shortcomings of wood compared with synthetic materials coming from nonrenewable resources. The instability issue is more important for fast-grown plantation species like radiata pine in comparison to indigenous, natural grown species because of the higher proportion of juvenile wood in plantation species. As a popular plantation species in Australasia, South America, and South Africa, radiata pine has a creamy white color, a clear wood pattern, and average mechanical properties; however, its stability and durability are regarded as inferior in comparison to other commercial species, such as ponderosa pine (Maclaren 2002).

Heat treatment in a controlled environment at temperatures ranging from 160°C to 250°C is an effective way of altering wood to achieve desired properties (Hill 2006). The heat treatment of wood can be done in air, vacuum, nitrogen, steam, or vegetable oil. Previous studies have reported darkening and improved dimensional stability of wood after heat treatment (Stamm 1964, Sailer et al. 2000,

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Syrjänen and Kangas 2000, Bekhta and Niemz 2003, González-Pena and Hale 2009). While discoloration of wood during kiln drying is generally undesirable, the uniform darkening of light-colored species during heat treatment can be a tool to achieve a desired darker-colored wood, giving them a tropical flavor. Furthermore, there are increasing environmental concerns in the furniture industries over the use of solvent-borne wood finishes (Pandey 2005). Heat treatment, unlike traditional wood finishing or painting, does not involve any emission of health-hazardous volatile organic compounds, such as toluene and xylene (Mitsui 2004). Therefore, heat treatment could be an effective and environmentally friendly way to darken the wood color and, at the same time, improve wood stability (Johansson and Morén 2006, González-Pena and Hale 2009).

However, studies have also shown that the color of heattreated wood is not stable and fades with time when exposed to UV radiation (Syrjänen and Kangas 2000, Militz 2002, Ahajji et al. 2009). In addition to discoloration, Mayes and Oksanen (2002) reported that surface checking occurred for the heat-treated wood exposed to UV radiation, resulting in poor aesthetics. Several reports are available on the study of color after accelerated weathering of wood heat treated in air, vacuum, nitrogen, or steam. The exposure time to UV radiation in most of the previous studies was not more than 900 hours (Avadi et al. 2003, Letourneau et al. 2005, Temiz et al. 2006, Deka et al. 2008, Ahajji et al. 2009). However, investigations of the dimensional stability of heat-treated wood after accelerated weathering are very limited (Feist and Sell 1987), and there have been no published works on the accelerated weathering of oil heat-treated wood.

The main objective of this work was to examine changes in color and dimensional stability of wood with an oil heattreatment process. The study also examined the effect of accelerated UV weathering on the previously mentioned properties. Radiata pinewood was heat treated in linseed oil over a range of temperatures and times. Treated and untreated specimens were exposed to an accelerating UV weathering environment for 2,100 hours. Changes in color were studied by a spectrophotometer using the Commission Internationale de l'Eclairage (CIE) L\*a\*b\* color coordinate system, and volumetric swelling percentages were determined after a water immersion test. The percent weight changes were also measured to determine the net oil uptake by the specimens after oil heat treatment.

## Experimental

## Oil heat treatment

Specimens were prepared from 3- to 4-m-long boards of *Pinus radiata* obtained from a local sawmill. All boards were kiln-dried, flatsawn sapwood from which clear specimens with dimensions of 300 mm (long) by 90 mm (wide) by 20 mm (thick) were cut. These were oven dried for 48 hours, cooled over silica gel in a desiccator, and weighed before being placed in a conditioning chamber under 65 percent relative humidity (RH) and 20°C for 2 to 3 weeks to attain equilibrium.

The conditioned specimens with equilibrium moisture content of 10 to 12 percent were heated in an oil bath using commercial-grade raw linseed oil as the heating medium at 160°C, 180°C, and 210°C for 1, 3, or 6 hours. The oil was preheated to temperature before immersing the specimens and treating them for the targeted time. The heated oil was recirculated throughout the treatment, and the temperature was recorded at three points in the bath.

After treatment, heating was turned off, the lid of the oil bath was removed, and specimens were kept in the oil bath for 15 minutes; the oil was then drained. The specimens were oven dried at 103°C for 48 hours, cooled over silica gel in a desiccator, and weighed. Finally, the specimens were placed in a conditioning chamber at 65 percent RH and 20°C for 2 to 3 weeks, by which time equilibrium was attained.

# Accelerated weathering test

After the previously mentioned oil heat treatment, control and treated wood specimens were cut into two halves at the midthickness along the specimen length; thus, each half was 300 by 90 by 10 mm. One of each pair of specimens was exposed for accelerated UV weathering, and the other was kept unexposed in a dark room for comparison. The UV weathering tests followed US standard ASTM-G154 (ASTM International 2006). During the UV weathering tests, specimens were exposed to repetitive cycles of UV light and moisturization under controlled environmental conditions in a weathering box. UVA 340 light was used, and moisturization was produced by spraving the specimens with water. Each test lasted 2,100 hours and consisted of 175 cycles, each of which included 8 hours of UV 340 exposure at  $60 \pm 3^{\circ}$ C black panel temperature and 4 hours of condensation at 50  $\pm$  3°C black panel temperature (ASTM International 2006). During the 4-hour condensation period, water was sprayed for 15 seconds every 2 minutes. All the specimens were placed 100 mm beneath the light source.

# **Measurement of properties**

Weight percent change.—The weight percent change (WPC) was determined using Equation 1, which gives an averaged relative difference in ovendry weight of the wood specimens after the oil heat treatment:

$$WPC = \left[\frac{(W_a - W_b)}{W_b}\right] \times 100\% \tag{1}$$

where  $W_b$  is the ovendry weight of specimens before the treatment (g) and  $W_a$  is the ovendry weight of specimens after the treatment (g).

Color.—Variation in wood color after oil heat treatment and accelerated UV weathering was examined by visual inspection and quantified through the CIE L\*a\*b\* coordinate system, which is a well-established procedure for color measurement (St.-Onge et al. 2005, Esteves et al. 2008, Ahajji et al. 2009). This system measures total color variation ( $\Delta E$ ) from changes in three spatial coordinates using the following equation (British Standards 1986):

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

where L\* is the brightness coordinate or luminance ranging from black (0) to white (100), a\* is the color coordinate varying from redness (+) to greenness (-), and b\* is the color coordinate varying for yellowness (+) to blueness (-).

Color variation of wood specimens was recorded by a Minolta model cm2500d spectrophotometer. The diameter of the color-measuring circle on the specimen surface was 10 mm with the corresponding area of  $0.78 \text{ cm}^2$ . The light

source was the Standard Illuminant D 65, which represents the average daylight, including UV region and 10-degree standard observer. For the color measurement, the specimens were first stabilized at 65 percent RH and  $20 \pm 2^{\circ}$ C before the measurements. For each specimen, six measurements were taken at different marked locations. The values of coordinates L\*a\*b\* of the specimens before the oil heat treatments, after the oil heat treatment, and after the accelerated UV weathering were used to determine the color changes.

Dimensional stability.—Dimensional stability was determined by measuring volumetric swelling percentage of the specimens, which were immersed in a water bath at a controlled temperature of 20°C. During the tests, the water in the water bath was recirculated continuously to maintain the required temperature (20°C). The specimens were weighed every 24 hours, and once the weight changes of the test specimens were less than 0.1%, it was assumed that the specimens had reached equilibrium, which took 2 weeks. Then the dimensions in longitudinal, width (tangential), and thickness (radial) directions were measured to an accuracy of  $\pm 0.01$  mm. The volumetric swelling percentages were determined by

$$S = \left(\frac{V_2 - V_1}{V_1}\right) \times 100\% \tag{3}$$

where  $V_1$  is the ovendry volume of the specimens before water immersion (cm<sup>3</sup>) and  $V_2$  is the volume of the specimens after the water immersion (cm<sup>3</sup>).

## Results

# Weight percent change

Figure 1 shows the effect of treatment temperature and time on the mean WPC of the treated specimens. The weight gain decreased with increasing treatment temperature and duration. For the treatment duration of 3 hours, the WPCs at 160°C, 180°C, and 210°C were approximately 11, 7.5, and 1 percent, respectively. At a given treatment temperature, the WPC values were maxima for the 1-hour treatments and minima for the 6-hour treatments. For the specimens treated at 210°C for 6 hours, a net weight loss of 1.8 percent was recorded.

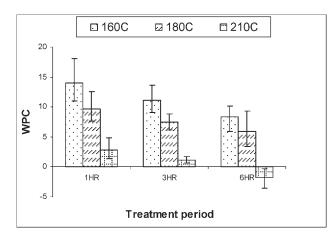


Figure 1.—Effect of treatment temperature and duration on mean WPC of wood specimens (n = 8).

# Color

The heat treatment affected the wood color as expected, with the wood turning darker. However, no surface cracking, warping, or twisting was observed in treated wood. The surface appeared shiny near the rays, which may be due to high uptake of oil by parenchyma cells compared with other wood cells. The color change was identical on both flat surfaces (Table 1), the difference between the two surfaces becoming reduced with increasing the treatment temperature. When the wood was treated at 180°C and 210°C, the total color variation ( $\Delta E^*$ ) and brightness variation ( $\Delta L^*$ ) between the two surfaces were not more than 1.5 units, although the wood surface became darker. The human eyes can distinguish color variation only if  $\Delta L^*$ is more than 3 units (Phelps et al. 1994) or the value of  $\Delta E^*$ is greater than 2 (Sundqvist 2002).

Color change was noted throughout the cross section of the treated specimens, the intensity of the color being greater on the surfaces than in the core. Table 2 gives the values of CIE L\*a\*b\* coordinates for the surface and core of treated wood. The average lightness values (L\*) for the surface are 22 to 28 percent less than the corresponding values at the core. This difference in color between the surface and the core decreased with increasing treatment temperature, i.e., for specimens treated at 210°C, the total color difference between the surface and the core was 15 compared with the corresponding values of 15.9 and 16.7 for the specimens treated at 180°C and 160°C, respectively.

Table 1.—CIE (L\*a\*b\*) values at the two flat surfaces of treated and untreated specimens.

	L*	a*	b*	$\Delta L^*$	∆a*	$\Delta b^*$	ΔΕ
Untreated							
Surface 1	64.3	12.2	31.8				
Surface 2	68.1	11.5	33.8	3.8	-0.8	2.0	4.3
OHT at 160°C							
Surface 1	65.9	12.3	38.2				
Surface 2	62.5	13.2	37.0	-3.3	0.9	-1.2	3.6
OHT at 180°C							
Surface 1	61.2	13.7	36.8				
Surface 2	59.8	14.1	36.6	-1.4	0.4	-0.3	1.5
OHT at 210°C							
Surface 1	47.1	13.8	26.3				
Surface 2	46.1	13.3	25.6	-0.9	-0.5	-0.7	1.2

Table 2.—CIE (L*a*b*)	values	at	the	surface	and	core	of	the
treated specimens.								

	L*	a*	b*	$\Delta L^*$	$\Delta a^*$	$\Delta b^*$	$\Delta E$
OHT at 160°C							
Surface Core	55 70	13.4 7.3	31.8 27.5	15	-6.1	-4.2	16.7
OHT at 180°C							
Surface Core	61.2 74.7	12 6.9	33.5 27	13.6	-5.1	-6.6	15.9
OHT at 210°C							
Surface Core	36.9 47.3	11.3 11.8	15.9 26.7	10.4	0.6	10.8	15

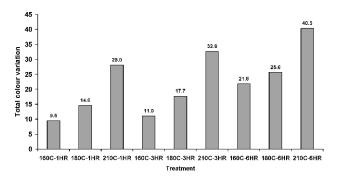


Figure 2.—Total color variation ( $\Delta E L^*a^*b^*$ ) for different temperatures and times.

The color of treated wood varied with the treatment temperature such that at 160°C, 180°C, and 210°C, the wood was, respectively, dark yellow, reddish brown, and dark chocolate. Figure 2 shows that the total color variation ( $\Delta E^*$ ) increased with treatment temperature and duration. For a given treatment time, the total color variation was greater for specimens treated at 210°C than for specimens treated at 180°C, which in turn, was greater than the specimens treated at 160°C. Similarly, specimens treated at a given temperature for 6 hours were darker than those treated at the same temperature for 1 hour. However, the treatment temperature has a more profound influence than the treatment time.

Representative color changes after the accelerated UV weathering test are shown in Figure 3 for both heat-treated and untreated samples. Figure 4 shows the results of total color variation ( $\Delta E^*$ ) during the accelerated UV weathering as a function of exposure time. The untreated specimens displayed the greatest total color variation throughout the exposure period. Among the treated wood, values of  $\Delta E^*$  for specimens treated at 160°C were generally greater than those treated at 180°C and 210°C. The  $\Delta E^*$  values of specimens treated at 180°C and 210°C were very close.

Figure 5 compares changes in chromatic coordinates  $(\Delta a^*, \Delta b^*)$  and the lightness coordinate  $(\Delta L^*)$  after 2,100 hours of exposure to the accelerated UV weathering. The untreated wood clearly showed greater total color variation  $(\Delta E^* = 32)$  than the treated wood  $(\Delta E^* = 4 \text{ to } 10)$ . The darkness and blueness of the untreated wood increased significantly with weathering  $(\Delta L^* = -27 \text{ and } \Delta b^* = -16)$ , but after oil heat treatment, the color of radiata pinewood

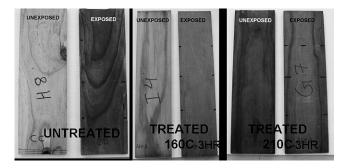


Figure 3.—Comparison of color in unexposed and exposed specimens of after 2,100 hours of accelerated UV weathering. The unexposed and exposed samples belong to the same piece cut from the middle of the thickness of the same specimen.

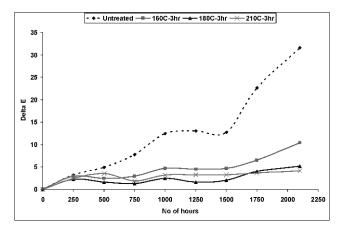


Figure 4.—Total color variation for untreated and heat-treated wood with accelerated UV weathering tests for different exposure times.

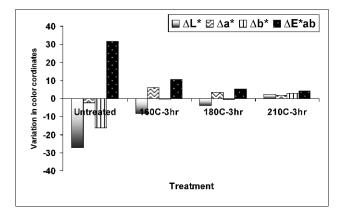


Figure 5.—Changes in CIELAB coordinates in weathered specimens after 2,100 hours.

was more stable, and color stability generally increased with treatment temperature. For example, the total color change ( $\Delta E^*$ ) was 10 for wood treated at 160°C compared with corresponding values of 5 for wood treated at 180°C and 4 treated at 210°C. All specimens were darker without any fading ( $\Delta L^* = -4$  to -8) except for those treated at 210°C, which faded slightly ( $\Delta L^* = 2$ ) during the weathering. Interestingly, specimens treated at 160°C became redder, as evidenced by the increase in the value of the red/yellow chromatic coordinate ( $\Delta a^* = 6$ ) in Figure 5. In all the observations of wood color change through the CIE L\*a\*b\* system, the lightness coordinate (L\*) was the most affected and sensitive parameter during the wood oil heat treatment and accelerated UV weathering.

# **Dimensional stability**

Figure 6 shows weathered surfaces of untreated and oil heat-treated wood. The surfaces of treated wood were smooth without or with only minor checks on visual inspection. However, clearly visible cracks and checks appeared on the untreated specimens.

Table 3 presents the results of volumetric swelling in water immersion tests. The results include the influences of oil heat treatment (unexposed to weathering) and accelerated UV weathering (exposed to weathering). After

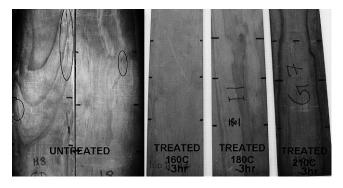


Figure 6.—Surfaces of treated and untreated wood after accelerated UV weathering. The cracks and checks appeared on untreated wood are marked.

weathering, the volumetric swelling in untreated wood was 32 to 55 percent more than that of the treated wood. With the treated wood, the volumetric swelling decreased with increasing treatment temperature, which was true for wood both unexposed and exposed to UV weathering. However, with the UV weathering, the untreated wood gave higher increases in the volumetric swelling than the treated wood. For example, the volumetric swellings of the untreated wood increased from 13.8 to 17.5 percent with UV exposure, while the specimens treated at 210°C increased from 7 to 7.9 percent. The difference in the volumetric swelling between exposed and unexposed specimens was about 21 percent for untreated wood, while for the treated wood this difference was about 20, 13, and 10 percent for the specimens treated at 160°C, 180°C, and 210°C, respectively (Fig. 7).

### Discussion

Both ovendry weight percentage and wood color changed after oil heat treatment at 160°C, 180°C, and 210°C for 1, 3, or 6 hours. This may be the result of the combination of a number of factors: (1) degradation of chemical constituents of wood during heat treatment (Tjeerdsma and Militz 2005, Windeisen et al. 2007); (2) removal or migration of extractives and other nutritive compounds, such as lowmolecular-weight sugars and amino acids (Theander et al. 1993); and (3) oil uptake by wood and the formation of an oil layer on the wood surface as a result of heat treatment.

Some products formed after thermal degradation of hemicelluloses and lignin are colored and lead to color changes in wood (Bekhta and Niemz 2003). The removal or migration of extractives and other nutritive compounds also affects the weight and color change in the treated wood (Theander et al. 1993, Sundqvist and Morén 2002). The

Table 3.—Volumetric swelling of wood specimens in water immersion tests.  $^{\rm a}$ 

	Volumetric swelling	ig, mean (SD) (%)		
Treatment	Unexposed to weathering	Exposed to weathering		
Untreated	13.9 (1)	17.5 (0.9)		
OHT at 160°C	9.5 (0.4)	11.9 (1.2)		
OHT at 180°C	9.5 (0.7)	11 (0.7)		
OHT at 210°C	7 (0.6)	7.9 (1.1)		

a Values are the averages of six replicates.

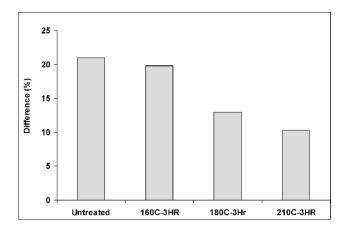


Figure 7.—Difference in volumetric swelling percentage (after 2 wk of water immersion) between exposed and unexposed specimens to UV weathering.

extractives and other colored products tend to migrate toward the surface (Sundqvist and Morén 2002), resulting in a darker surface than the core.

The greatest mean weight increase (14%) was found for wood treated at 160°C for 1 hour. This can be attributed to the oil uptake (Fig. 1). However, the actual oil uptake by specimens was greater than the measured weight increase, because indicative weight losses due to wood degradation and the removal of extractives at high temperatures must be taken into account. The weight gain tended to be less at higher temperature treatment, as the wood degradation was more severe with an increasing treatment temperature, which was confirmed by the observation that the treated wood actually lost about 2 percent of its weight in heat treatment at 210°C for 6 hours.

The color of two flat surfaces of the oil heat-treated specimens was found to be identical without resin patches. This contrasted with the findings of Johansson and Morén (2006), who observed a nonhomogeneous distribution of color in thermally treated wood at 175°C to 200°C for 1 to 10 hours in steam. The similarity of the wood color at the treated specimen surfaces is due mainly to uniform heat transfer with an oil medium compared with that in the air or other heating medium (Sailer et al. 2000). Another possible explanation might be that during the heat treatment in air or steam, migration of extractives toward the surface leads to the formation of unsightly resin spots in wood (Mayes and Oksanen 2002), whereas these extractives and other degraded products might have transferred into the heating oil.

No fading of treated wood was observed after weathering except for wood treated at 210°C. This was in line with the findings of Deka et al. (2008), who noted improved color stability of heat-treated wood (in vacuum at 210°C for 2 h) during UV light irradiation for 400 hours. In contrast, Syrjänen and Kangas (2000) reported that heat-treated wood starts fading if not protected from UV light. However, Ahajji et al. (2009) reported mixed results from artificial weathering of different species of wood that had been heat treated at 210°C to 250°C for 1 hour. They observed that the color of heat-treated spruce is more stable to UV radiations than the untreated controls, whereas in the case of beechwood, the contrary results were found. Thus, oil heat-treated wood demonstrated better color stability than untreated wood after the accelerated UV weathering test.

Similarly, after accelerated UV weathering, volumetric swelling of the treated wood increased less than that of the untreated wood, indicating that treated wood is more stable. The oil heat-treated wood did not show noticeable checks with the UV weathering. This differs from Mayes and Oksanen (2002), who reported that surface checks after weathering were observed for heat-treated wood at 70°C to 240°C for 3 hours in a steam environment.

The superior stability of color and dimensional changes of the oil heat-treated wood than for untreated wood during artificial weathering might be due to an increase in lignin stability due to its undergoing condensation reactions during heat treatment (Ayadi et al. 2003, Deka et al. 2008). Another possible reason for these improvements with the oil heat treatment can be related to the lower moisture absorption by the oil heat-treated wood. Apart from UV radiation, moisture is a critical factor in photoinduced degradation of wood, as it facilitates light penetration into the wood surface (Ayadi et al. 2003). Because of the high linolenic acid  $(C_{18,3})$  content, linseed oil has a tendency to dry quickly and harden on the wood surface after oil heat treatment (Gunstone 2002). This process forms a protective layer of oil on the surfaces of the treated specimens. Therefore, the oil layer acts as a barrier for UV ray penetration and for water absorption and thus improves the water repellency in the treated wood.

# Conclusions

This study examined color change and dimensional stability of oil heat-treated radiata pinewood before and after accelerated UV weathering. The results of this study have shown that the oil heat-treated wood turned darker and was more dimensionally stable, depending on treatment time and temperature. The color changes made the two flat surfaces of the specimens more uniform without any color patches being observed, although the surfaces were darker than the core. Color inhomogeneity between flat surfaces and the difference between core and surface color decreased with the increase in treatment temperature.

After an accelerated UV weathering test, treated wood exhibited better color stability, and no color fading was observed in the specimens treated for 3 hours at 160°C and for 3 hours at 180°C; those treated at 210°C faded slightly. In general, no surface checks were observed on the weathered surfaces of treated specimens. The percent swelling after weathering in treated specimens was less than for untreated wood.

The results of this study indicate that oil heat treatment can improve the wood stability and durability, and this can be explained by the changes in the chemical constituents of wood and the formation of a protective oil layer on the wood surfaces. Based on the findings of this study, the oil heat treatment of plantation wood could be an effective way to achieve uniform and stable dark color for some markets and improve stability and durability.

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