

**THE EFFECT OF NATURAL WEATHERING ON CHANGE
IN THE COLOR OF HEAT TREATED AND VARNISHED
SCOTS PINE AND ORIENTAL BEECH WOODS**

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ABSTRACT

This study was designed to investigate color changes of heat treated and varnished Scots pine (*Pinus sylvestris* L.) and Oriental beech (*Fagus orientalis* L.) after 3 months of weathering. Heat treatment of Oriental beech and Scots pine wood was carried out by hot air in an oven for 1, 2, and 3 hours at 205, 215, and 225°C. After heat treatment, wood specimens were varnished using a polyurethane varnish (PV) and cellulosic varnish (CV). The results of study showed that heat treatment generally caused decrease of lightness of both wood specimens before weathering. In general, while heated and PV coated Scots pine and Oriental beech wood surfaces turned to reddish and yellowish, heated and CV coated Scots pine and Oriental beech wood surfaces turned to reddish and bluish after weathering. The total color changes of heated and varnished both wood specimens were lower than only varnished both wood specimens after weathering.

KEYWORDS: Scots pine, Oriental beech, color, weathering, varnish, heat treatment.

INTRODUCTION

Wood has been preferred for residential construction since ancient times because of its natural beauty and excellent properties, such as, high specific strength, heat insulation, and ease of handling and processing (Su 1997). However, being biological material, it is susceptible to environmental degradation. So, it is necessary to treat wood to provide long service life and improve specific surface properties on the intended application (Srinivas and Pandey 2012). Wood materials are usually exposed to solar radiation, water, wind, and dust during their useful life, when used outdoor. Wood surfaces exposed outdoors are rapidly degraded because lignin strongly absorbs UV light, which leads to radical-induced depolymerization of lignin and cellulose, the major structural constituents of wood (Evans et al. 2002). The natural weathering process often results in discoloration, a physical deterioration of the wood surface, and loss of paint-retaining properties. The change in color is the first obvious indication of the chemical changes in wood during weathering. Wood also loses its gloss and lightness (Jirous-Rajkovic et al. 2004). The main factor that causes the greatest changes in the surface properties of wood during outdoor exposure is sunlight (Tolvaj et al. 2011). Because lignin is the main chemical component of wood, the aromatic polymer, strongly absorbs sunlight (Jebrane et al. 2009), which causes depolymerization of lignin. The yellowing, browning, and/or graying of wood surfaces indicate the modification of lignin when wood is exposed outdoors (Grelier et al. 2000). The weathering initially causes the discoloration and gloss loss, followed by the occurrence of surface checking and increased roughness of the wood (Denes and Young 1999, Ozgenc et al. 2012). 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 (Evans et al. 1992, Kiguchi and Evans 1998). However, chemical treatment presents a serious threat to the environment. Environmental concerns associated with the use of conventional wood preservatives, however, have increased interest in thermal modification approaches (Hill 2006). Heat treatment is an alternative method for improving these properties with no use of chemical additives (Johansson 2008). Since it doesn't include the use of chemicals and is not toxic it is the most suitable method for wood. In last decade, utilization of heat treatment for wood modification has been increased (Bachle et al. 2010, Khalid et al. 2010). Heat treated wood possesses new

physical properties such as reduced hygroscopy, improved dimensional stability, better resistance to degradation by insects and micro-organisms, and most importantly, attractive darker color. These new versatile and attractive properties help heat treated wood to become popular for outdoor applications (Huang et al. 2012). Heat treated wood has a large application for outdoor use in cladding, decks, garden furniture, and window frames as well as indoor use for kitchen furniture, parquet, decorative panels, and mainly for the interior of saunas (Esteves and Pereira 2009). It is known that various paints, varnishes, and other coatings are also available to enhance or maintain against weathering (Chang and Chou 2000, Kielmann et al. 2016, Kielmann and Mai, 2016). In recent years, there has been a growing customer demand for the use of transparent or semi-transparent coatings that retain the natural appearance of wood, such as color, grain, and texture (Jirous-Rajkovic et al. 2004, George et al. 2005, Scrinzi et al. 2011). The efficiency of exterior wood finishes against weathering factors has been investigated by several researchers (Feist and Ross 1995, Richter et al. 1995, Nejad and Cooper 2001, Gobakken and Westin 2008, Cristea et al. 2010). The combined treatment both heat treatment and varnish application can be expected to improve the performance of wood surface against weathering effects. Therefore, in this study, it was aimed to investigate color stability of heated and polyurethane varnish (PV) and cellulosic varnish (CV) coated Scots pine and Oriental beech wood after 3 months of weathering exposure in Mugla which is in Southern Aegean Region of Turkey.

MATERIAL AND METHODS

Preparation of test specimens

Specimens 10 x 100 x 150 mm (radial by tangential by longitudinal) were machined from air-dried sapwood of Scots pine (*Pinus sylvestris* L.) and Oriental beech (*Fagus orientalis* L.) lumbers. All specimens were conditioned at 20°C and 65% relative moisture content for two weeks before tests.

Heat treatment

Three different temperatures (205, 215, and 225°C) and three treatment durations (1, 2, and 3 h) were applied to wood specimens under atmospheric pressure and in the presence of air. Heat treatment was performed using a temperature-controlled laboratory oven. Heat treatment is performed in three stages. The stages are heating stage, drying stage, cooling and acclimatization stage. During the application heat treatment oven temperature is increased gradually. When the internal heat of the wood and temperature of the oven was equalized desired heat treatment is commenced. This process is named as 1st stage heating stage. In the 2nd stage drying was performed. In the cooling and acclimatization stage as 3rd stage gradual cooling was performed. During the cooling and acclimatization of the wood specimens, in order to balance the moisture and help cooling water is sprayed to wood specimens.

Varnish application

Cellulosic and polyurethane varnishes were used. PV and CV varnishes were applied over heat treated Oriental beech and Scots pine wood. The varnishes were applied to all surfaces and sides of the heated and unheated Scots pine and Oriental beech specimens with a spray gun according to the ASTM D3023-98 (2017) standard. Filler was used as the first coating applied to the wood surface was for filling the voids, and the second and third coatings were applied for top coating. Sufficient time for layer settling was allowed between successive applications until the target retention of

100 g·m⁻² for the primer and 100 g·m⁻² for the top coating were reached, controlled by consecutive weighting. Specimens were left in ambient conditions for 24 h according to the manufacturer's recommendations after the first coating, and then surfaces were gently sanded using a fine-grit sandpaper (220 grit) to obtain a smooth surface before the top coating.

Color test

The color parameters L*, a*, and b* were determined by the CIEL*a*b* method. The L* axis represents the lightness, whereas a* and b* are the chromaticity coordinates. The +a* and -a* parameters represent the colors red and green, respectively. The +b* parameter represents yellow, whereas -b* represents blue. The L* value can vary from 100 (white) to zero (black) (Zhang 2003). The colors of the specimens were measured by a colorimeter (X-Rite SP Series Spectrophotometer, X-rite Pantone, MI, USA) before and after weathering. The measuring spot was adjusted to be equal or not more than one-third of the distance from the center of this area to the receptor field stops. The color difference, (ΔE*) was determined for each wood according to ASTM D1536-58T (1964). The color changes were calculated using Eqs. 1 to 4;

$$\Delta a^* = a_f^* - a_i^* \tag{1}$$

$$\Delta b^* = b_f^* - b_i^* \tag{2}$$

$$\Delta L^* = L_f^* - L_i^* \tag{3}$$

$$(\Delta E^*) = [(\Delta a^*)^2 + (\Delta b^*)^2 + (\Delta L^*)^2]^{1/2} \tag{4}$$

Where Δa*, Δb*, and ΔL* represent the changes between the initial and final interval values.

Natural weathering test

Each groups consisted of 5 individual wood specimens. In total, 20 groups of wood specimens for each species were exposed to weathering conditions during April-May-June in 2015. Wood panels were prepared for weathering exposure according to ASTM D 358-55 (1970). Weathering test apparatus is demonstrated in Fig. 1.



Fig. 1: Weathering test apparatus.

A test site was established close to the Regional Meteorological Observation Station of Mugla which is in Southern Aegean Region to enable practical assessments.

Tab. 1: Details of the climate condition of Mugla city during April-May-June in 2015.

MUGLA	April	May	June
The highest temperature per month (°C)	24.7	31.8	32.0
The lowest temperature per month (°C)	1.3	6.4	10.6
Average temperature per month (°C)	11.4	18.2	20.8
Humidity per month (%)	60.5	61.9	61.7

The details of the climate condition of Mugla city in Tab. 1 are informed by Turkish State Meteorological Service database.

RESULTS AND DISCUSSION

Color changes of Scots pine after weathering

Color changes of heat treated and varnish coated Scots pine before and after 3 months of weathering are given in Tab. 2.

Tab. 2: Color changes of Scots pine before and after weathering.

Temperature (°C)	Duration (Hour)	Varnishes	Before weathering			After 3 months of weathering			Total color changes				
			L_i^*	a_i^*	b_i^*	L_f^*	a_f^*	b_f^*	ΔL_f^*	Δa_f^*	Δb_f^*	ΔE_f^*	
Only PV coated		PV	62.71	13.03	37.65	54.67	16.68	34.88	-8.04	3.65	-2.77	9.25	
205	1		65.28	9.91	41.10	58.67	12.31	44.75	-6.61	3.40	3.65	8.28	
	2		62.59	11.48	42.85	56.53	13.91	45.30	-6.06	2.43	2.45	6.97	
	3		21.04	16.43	22.44	26.20	17.62	25.79	5.16	1.19	3.35	6.62	
215	1		44.85	16.52	38.99	51.30	19.10	41.70	6.45	2.58	3.71	7.87	
	2		28.67	13.87	16.49	32.97	15.49	24.50	4.30	1.61	8.01	7.56	
	3		19.52	15.25	17.82	24.22	16.73	21.54	4.70	1.51	3.72	6.18	
225	1		50.44	16.03	43.20	56.00	18.80	46.44	5.56	2.77	3.24	6.99	
	2		15.44	12.30	12.54	20.18	14.75	15.65	4.74	2.45	3.11	6.17	
	3		12.82	7.19	6.03	17.08	9.50	8.65	4.26	2.31	2.62	5.50	
Only CV coated			CV	68.88	8.73	37.84	32.52	23.61	32.61	-36.35	14.87	-5.23	39.63
205	1			66.63	9.91	39.72	34.03	24.00	32.60	-32.60	14.08	-7.12	36.22
	2	57.50		17.42	33.43	37.08	21.73	31.30	-20.42	4.31	-2.13	20.98	
	3	14.73		12.05	13.24	18.39	11.65	7.82	3.66	-0.40	-5.42	6.55	
215	1	54.96		13.52	40.57	27.25	23.02	23.63	-27.70	9.50	-16.93	33.83	
	2	27.58		17.46	26.56	21.78	17.17	13.38	-5.79	-0.28	-13.17	14.39	
	3	20.21		15.40	19.15	22.87	16.53	14.00	2.66	1.12	-5.15	5.90	
225	1	28.08		17.71	30.83	20.71	17.69	13.48	-7.36	-0.02	-17.35	18.84	
	2	14.38		13.91	17.35	14.99	10.58	6.89	0.61	-3.32	-10.46	10.99	
	3	9.49		6.97	4.50	14.22	6.60	2.04	4.72	-0.36	-2.46	4.96	

Note: Five replicates were made for each group.

Color is one of the quality attributes that influence the customers' perceptions of wood products (Barcik et al. 2015). Higher duration and temperature resulted in lower L^* values of Scots pine before weathering. Especially, heat treated and varnish coated Scots pine for 3 h heat treatment groups. For heated and PV coated Scots pine, except for 1h treatment at 205°C, L^* values of all heat treated and varnish coated Scots pine were decreased before weathering. The lower L^* values shows that the wood become darker on the effect of heat treatment. Previous studies have shown that the treatment temperature and the duration make darker the wood specimens (Militz 2002, Akgul and Korkut 2012, Mitsui et al. 2003). Bourgios et al. (1991) reported that wood darkening after heating might be related to a reduction in holocellulose content as the temperature increases. The degradation of lignin and other non-cellulosic polysaccharides may increase the darkening of heat treated Scots pine wood (Grelrier et al. 2000, Hon and Chang 1985, Petric et al. 2004). For heat treated and PV coated Scots pine, except for only PV coated and heated at 205°C for 1 and 2 h and PV coated Scots pine, all heat treated and PV coated Scots pine gave positive ΔL^* values after 3 months of weathering. For heated and

CV coated Scots pine, all heated 205, 215, and 225°C for 3 h treatment and coated CV showed positive ΔL^* values after 3 months of weathering. Also, heated 225°C for 2 h and CV coated Scots pine gave positive ΔL^* values after 3 months of weathering. Olărescu et al. (2014) studied ΔL^* of heat treated wood panels after three months of natural weathering. They found that while untreated samples become dark (negative ΔL^* measurements), the heat treated panels have a disposition to become light (positive ΔL^* measurements). Kucuktuvek et al. (2017) investigated color changes of heat treated Scots pine (*Pinus sylvestris* L.) after weathering. Heat treatment of Scots pine wood was carried out by hot air in an oven for 1, 2, and 3 hours at 210, 220, and 230°C. They found that while heat treated Scots pine wood become lighter after weathering, un-heated wood become darken after weathering. The results are in good agreement with data Olărescu et al. (2014) and Kucuktuvek et al. (2017). While the positive values of Δa^* indicate a tendency of wood surface to become reddish, the negative values of Δa^* indicate a tendency of wood surface to become greenish. For heat treated and PV coated Scots pine, the results showed that only PV coated and all heat treatments and PV coated Scots pine gave positive Δa^* values after 3 months of weathering exposure. This positive values of Δa^* after heat treatment show a tendency of wood surface to become reddish. The increase of the chromaticity coordinate (Δa^*) may be explained by the modification of some chromophoric groups of lignin (Grelier et al. 2000). For heat treated and CV coated Scots pine, all heat treatment at 225°C gave negative Δa^* values after 3 months of weathering exposure. Also, heat treatments at 205°C for 3 h and 215°C for 2 h gave negative Δa^* values. This negative values of Δa^* indicate a tendency of wood surface to become greenish after 3 months of weathering. While positive Δb^* measurements show wood surface's disposition to appear yellowish, negative Δb^* measurements shows wood surfaces disposition to appear as bluish after weathering. For heat treated and PV coated Scots pine, except for only PV coated Scots pine, all other treatments gave positive Δb^* values indicate a disposition of wood surface to become yellowish after 3 months of weathering exposure. For heat treated and CV coated Scots pine, all treatments gave negative Δb^* values shows wood surfaces disposition to appear as bluish after 3 months of weathering exposure. Total color changes (ΔE^*) at the end of the 3 months of weathering for the only PV varnished wood were higher than heated and PV varnished wood. In our study, total color change was 9.25 for only PV coated Scots pine, this value between 5.50 and 8.28 for heat treated and PV coated Scots pine. Also, while ΔE^* value was 39.63, for only CV coated Scots pine, it was between 4.96 and 36.22 for heat treated and CV coated Scots pine. ΔE^* value of heat treated and varnish coated Scots pine wood decreased with increasing duration and temperature of the treatment. The experiment results showed that total color changes of heat treated and PV coated Scots pine were lower than heat treated and CV coated Scots pine after 3 months of weathering exposure. It may be due to among of the wood coatings polyurethane is hard, abrasion-resistant and durable, and is known to act as a good moisture barrier for wood products (Poaty et al. 2013).

Color changes of Oriental beech after weathering

Color changes of heat treated and varnish coated Oriental beech before and after 3 months of weathering exposure are given in Tab. 3.

Tab. 3: Color changes of Oriental beech before and after weathering.

Temperature (°C)	Duration (Hour)	Varnishes	Before weathering			After 3 months of weathering			Total color changes			
			L_i^*	a_i^*	b_i^*	L_f^*	a_f^*	b_f^*	ΔL_f^*	Δa_f^*	Δb_f^*	ΔE_f^*
Only PV coated			52.37	17.29	33.37	45.15	13.68	28.19	-10.22	-3.61	-5.18	12.01
205	1	PV	45.18	18.90	32.33	50.54	17.51	39.39	5.27	-1.38	7.06	8.91
	2		45.14	18.77	32.15	52.02	17.33	37.22	6.88	-1.42	5.06	8.65
	3		8.40	4.23	1.86	14.66	7.34	6.43	6.26	3.11	4.57	7.78
215	1		27.74	16.60	22.78	34.09	19.34	28.01	6.35	2.73	5.23	8.66
	2		11.18	4.44	2.96	18.26	7.95	6.08	7.08	3.51	3.12	8.49
	3		11.08	4.55	2.50	18.04	5.72	5.60	6.96	1.17	3.10	8.40
225	1		21.94	15.96	19.63	26.52	17.34	25.89	6.58	1.36	6.25	7.86
	2		9.26	3.78	1.95	17.52	6.72	5.68	5.25	2.93	3.73	7.07
	3		9.99	1.59	6.98	15.45	3.90	8.82	5.46	2.31	1.84	6.20
Only CV coated			55.81	15.90	31.38	32.48	22.41	28.18	-23.33	6.51	-3.20	24.43
205	1	CV	48.95	18.38	32.94	36.71	22.51	34.29	-12.24	4.13	1.35	12.98
	2		43.57	18.68	30.29	35.16	22.14	32.56	-8.40	3.46	2.26	9.36
	3		8.93	1.40	1.07	14.20	1.57	0.08	5.27	0.16	-0.98	5.36
215	1		34.60	17.83	28.76	29.05	21.84	25.45	-5.54	4.01	-3.31	7.60
	2		15.59	10.72	9.92	16.54	10.01	5.65	0.95	-0.71	-4.26	4.43
	3		10.10	6.10	4.72	15.41	7.67	3.34	5.31	1.57	-1.37	5.71
225	1		15.13	9.05	9.72	16.57	11.36	7.78	1.44	2.31	-1.94	3.35
	2		8.12	3.29	2.47	13.86	2.29	0.54	5.74	-1.00	-1.93	6.13
	3		10.07	0.82	0.24	13.65	1.17	0.19	3.58	0.50	-0.04	3.59

Note: Five replicates were made for each group.

L^* values of all heat treated Oriental beech specimens before varnish coating were decreased. The decrease in L^* indicates that the specimens become darker after heat treatment. Generally, darkening with heat treatment increased with treatment temperature and duration and this is consistent with earlier findings (Militz 2002, Mitsui et al. 2003, Akgul and Korkut 2012, Guller 2012, Cademartori et al. 2013a, b, Baysal et al. 2014, Yildiz et al. 2011, Aksoy et al. 2011). The darkening of Oriental beech might be due to degradation of lignin and other non-cellulosic polysaccharides (Hon and Chang 1985, Grelier et al. 2000, Petric et al. 2004). As wood is heated, acetic acid is formed from acetylated hemicelluloses by hydrolysis (Forsman 2008). The released acid serves as catalyst in the hydrolysis of hemicelluloses to the soluble sugars (Thermowood Association 2003). The heat caramelizes the sugar to a brown color which affects the color of wood. As the degradation of hemicelluloses accelerates with temperature, the color will become darker with increased treatment temperature (Forsman 2008). For heat treated and PV coated Oriental beech, except for only PV coated Oriental beech wood, all heat treated and PV coated Scots pine gave positive ΔL^* values after 3 months of weathering. For heated and CV coated Oriental beech, except for only CV coated and heat treatments at 205°C 1 and 2 h, and at 215°C 1 h, other treatment groups showed positive ΔL^* values after 3 months of weathering. Turkoglu et al. (2017) determined ΔL^* values of heat treated Oriental beech above 200°C and then coated with PV after 500 h accelerated weathering. They noted that while only PV coated Oriental beech gave negative ΔL^* values after 500 h accelerated weathering, heat treated and PV coated Oriental beech gave positive ΔL^* values after 500 h accelerated weathering. Baysal et al. (2014) determined heat treated Oriental beech wood samples become lighter after exposure to natural weathering and on the other hand non-heat treated Oriental beech wood samples become darker after exposure to natural weathering. Turkoglu et al. (2015) studied color characteristics of heat treated Oriental beech wood specimens after natural weathering. In their study, heat treatment

of Oriental beech wood was carried out by hot air in an oven for 1, 4, and 8 h at 140, 170, and 200°C. They determined that while positive ΔL^* indicates that heat treated Oriental beech wood specimens become lighter after natural weathering, negative ΔL^* of non-heated Oriental beech wood specimens become darker after natural weathering. Our results are in good agreement with data Turkoglu et al. (2017) and Baysal et al. (2014). While the positive values of Δa^* indicate a tendency of wood surface to become reddish, the negative values of Δa^* indicate a tendency of wood surface to become greenish. For heat treated and PV coated Oriental beech, except for only PV coated and heat treated 205°C 1 h and 2 h and PV coated Oriental beech, other treatment groups showed positive Δa^* values after 3 months of weathering. This positive values of Δa^* values after heat treatment show a tendency of wood surface to become reddish. The increase of the chromaticity coordinate (Δa^*) may be explained by the modification of some chromophoric groups of lignin (Grelier et al. 2000). For heat treated and CV coated Oriental beech, except for heat treatments at 215°C and 225°C for 2 h, other treatment groups gave positive Δa^* values after 3 months of weathering exposure. While positive Δb^* measurements show wood surface's disposition to appear yellowish, negative Δb^* measurements shows wood surfaces disposition to appear as bluish after weathering. For heat treated and PV coated Oriental beech, except for only PV coated Oriental beech, all other treatments gave positive Δb^* values indicate a disposition of wood surface to become reddish after 3 months of weathering exposure. For heat treated and CV coated Oriental beech, except for heat treatments at 205°C for 1 and 2 h, other treatment groups gave negative Δb^* values, it shows wood surfaces disposition to appear as yellowish after 3 months of weathering exposure. Turkoglu et al. (2017) investigated color changes of heated and varnished Oriental beech after accelerated weathering. Heat treatment of Oriental beech wood was carried out by hot air in an oven for 0.5, 1, and 1.5 h at 210 and 220°C. After heat treatment, Oriental beech wood specimens were varnished using a polyurethane varnish. Results showed that Δa^* and Δb^* values of heat treated and PV varnished Oriental beech wood gave positive Δa^* and Δb^* values, they indicated that wood surface turned reddish and yellowish, respectively after 500 h accelerated weathering. In our study, while heat treated and PV coated Oriental beech wood surface turned to reddish and yellowish after 3 months of weathering exposure, heat treated and CV coated oriental beech generally turned reddish and bluish after 3 months of weathering exposure. The results are in this study in good agreement with data Turkoglu et al. (2017). Total color changes (ΔE^*) at the end of the 3 months of weathering were higher for the only PV varnished wood than heated and PV varnished wood. Turkoglu et al. (2017) determined total color changes of heat treated and PV coated Oriental beech after accelerated weathering. They found that total color changes of only PV coated Oriental beech were higher than heat treated and PV coated Oriental beech after 500 h accelerated weathering. In our study, total color change was 12.01 for only PV coated Oriental beech, this value between 6.20 and 8.91 for heat treated and PV coated Oriental beech. Also, while ΔE^* value was 24.43, for only CV coated Oriental beech, it was between 3.35 and 12.98 for heat treated and CV coated Oriental beech. Generally, ΔE^* value of heat treated and CV coated Oriental beech wood decreased with increasing duration and temperature of the treatment.

CONCLUSIONS

This study presented that effects of natural weathering on color changes of heated and polyurethane and cellulosic varnish coated Scots pine and Oriental beech wood. The results of the study showed generally L^* (lightness) values of heated and Scots pine and Oriental beech decreased before natural weathering. Heat treatments cause a strong darkening of wood surface

especially higher durations and temperature. The results showed that ΔL^* values of only varnished both wood specimens were lower compared to heated and varnished both wood specimens.

In general, while heated and PV coated Scots pine and Oriental beech wood surfaces gave positive Δa^* and Δb^* values indicate a tendency of wood surface to become reddish and yellowish, respectively, heated and CV coated Scots pine and Oriental beech wood surfaces showed positive Δa^* and negative Δb^* values show a tendency of wood surface to become reddish and bluish, respectively. Total color changes were higher only varnish coated both wood specimens than heated and varnished both wood specimens. Generally, higher duration and temperature resulted in lower ΔE^* values of both wood specimens after weathering.

In conclusion, heat treatments before varnish coating improved color characteristics of both wood specimens after 3 months of weathering. It enhanced lightness stability and total color changes of both wood specimens. Generally, higher durations and temperatures resulted in better color characteristics of Scots pine and Oriental beech wood specimens after 3 months of weathering exposure.

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