Environmental-benign methods for the color protection of stripe long-shoot bamboo (*Bambusa dolichoclada*) culms

Min-Jay Chung, Sen-Sung Cheng, Shang-Tzen Chang*

School of Forestry and Resource Conservation, National Taiwan University, No. 1, Section 4, Roosevelt Road, Taipei 106, Taiwan

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Abstract

Fresh stripe long-shoot bamboo (*Bambusa dolichoclada* Hayata) culm is attractive for its yellow and green stripes. The stripes, however, discolor quickly after harvest, and currently there is no method to preserve the colors. The objective of this study was to find methods to preserve the colorful yellow–green stripes. Results indicated that excellent color preservation was achieved by treating the bamboo culms with 1% copper nitrate in alcohol at 60 °C for 2 h. Such treatment could be shortened to 30 min by the aid of ultrasonic vibration during the treatment. Immersing bamboo culms in 1% copper nitrate alcohol solution at the ambient temperature for 4 h also achieved the same result. Treated specimens exhibited outstanding colorfastness and mildew resistance after 18 months of indoor exposure.

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1. Introduction

Bamboo is one of the most abundant non-timber forest resources in Asia [1], which has been used as construction materials, handicrafts and other purposes [2]. In Taiwan nearly 58 species and four varieties have been cultivated all over the country [3,4], including such important and special species as ma bamboo (*Dendrocalamus latiflorus* Munro), thorny bamboo (*Bambusa stenotachys* Hackel), green bamboo (*Bambusa oldhamii* Munro), makino bamboo (*Phyllostachys makinoi* Hayata), moso bamboo (*Phyllostachys pubescens* Mazel), stripe common bamboo (*Bambusa vulgaris* Schrad. ex Wendl. var. *striata* Gamble), convex giant bamboo (*Bambusa vulgaris* cv. *Wamin* *Bambusa ventricosa*), square bamboo (*Chimonobambusa quadrangularis* (Fenzi) Makino) and fernleaf hedge bamboo (*Bambusa multiplex* (Lour.) Raeuschel cv. *Fernleaf* Young). Among these species, the stripe common bamboo or stripe long-shoot bamboo (*Bambusa dolichoclada* Hayata) fascinates people with its bright alternating green and yellow vertical stripes. The bright green stripes fade quickly after

being harvested. Therefore, it is desirable to find appropriate methods to preserve the colorful surface color of stripe long-shoot bamboo culms.

Several inorganic salts have been proven to be effective to preserve the green color of bamboo culms [5–9]. But there has been no report on color preservation of stripe long-shoot bamboo culms. It is known that ultrasonic waves can promote chemical reactions, and the technique has been employed in treating bamboo culms to preserve surface green color [7,8]. The objectives of this study were to search for simple methods for preserving and enhancement of the surface color of stripe long-shoot bamboo culms with environmentally friendly reagents and to investigate the effect of ultrasonic technique in such treatments.

2. Materials and methods

2.1. Sample preparation

Three-year-old stripe long-shoot bamboo (*B. dolichoclada* Hayata) culms were harvested from the experimental forest of National Taiwan University in Nan-Tou County.

*Corresponding author. Tel.: +886 2 3366 4626; fax: +886 2 2365 4520.
E-mail address: peter@ntu.edu.tw (S.-T. Chang).

The bamboo culms were cut into 50 mm (longitudinal) $\times$ 15 mm (tangential) $\times$ 4 mm (radial) strips and stored at 4 °C in darkness prior to use.

2.2. Chemical treatments

Four water-borne salts (chromated phosphate (CP), copper phosphate (CuP), copper azole (CuAz) and ammoniacal copper quaternary compound-type B (ACQ-B)) [5,8,10], five alcohol-borne salts (copper sulfate (CuSO_4), copper acetate $\text{Cu}(\text{CH}_3\text{COO})_2$, copper nitrate $\text{Cu}(\text{NO}_3)_2$, copper chloride (CuCl_2) and copper hydroxide $\text{Cu}(\text{OH})_2$) and copper naphthenate (CuN) containing 10% copper [7,11] in xylene/ethanol (1:1, v/v) were used in this study. Bamboo specimens were treated with salts of different concentrations (0%, 0.25%, 0.5%, 1%, 2% and 4%) in a water bath at different temperatures (25, 40, 60, 80 and 100 °C) for 0.5, 1, 2, 4 and 6 h. In addition to water bath treatments, specimens also were treated by immersing them in salt solutions at the ambient temperature for 1, 2, 4 and 6 days. Finally, the 1% $\text{Cu}(\text{NO}_3)_2$ solution was selected to treat specimen in an ultrasonic bath at 60 °C for 5, 15, 30, 60, 90 and 120 min. The ultrasonic bath used was Branson PC620 manufactured in USA with 180 W of power and an output frequency of 44 kHz. After treatment, all specimens were dried at 60 °C for 12 h before color measurements.

2.3. Color measurements

Color measurement was performed with a colorimeter (Dr. Lange Co., Germany) with a D₆₅ light source and a test-window diameter of 5 mm. The tristimulus values *X*, *Y* and *Z* of all specimens were obtained directly with the colorimeter. Based on these data, the following color parameters were calculated: *L*^{*} (value on the white/black axis), *a*^{*} (value on the red/green axis), *b*^{*} (value on the yellow/blue axis), ΔL^* (difference in brightness, $\Delta L^* = (L_n^* - L_0^*)$), Δa^* (difference in *a*^{*} component, $\Delta a^* = (a_n^* - a_0^*)$), Δb^* (difference in *b*^{*} component, $\Delta b^* = (b_n^* - b_0^*)$), and ΔE^* (color difference, $\Delta E^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$), where *L*_{*n*}^{*}, *a*_{*n*}^{*} and *b*_{*n*}^{*} were the *L*^{*}, *a*^{*} and *b*^{*} values of treated samples, respectively, and *L*₀^{*}, *a*₀^{*} and *b*₀^{*} were *L*^{*}, *a*^{*} and *b*^{*} values of the untreated reference, respectively. The above parameters were established by the Commission Internationale de l'Eclairage (CIE) in 1976 [6].

2.4. Indoor exposure test

Specimens treated with 1% alcohol-borne $\text{Cu}(\text{NO}_3)_2$ were exposed to indoor conditions to study their color-fastness. The specimens were placed in a room and exposed for 6, 12 and 18 months to natural light ($\lambda > 300$ nm) transmitted through a glass window during daytime. The average temperature and relative humidity during the study was 25 ± 3 °C and $80 \pm 10\%$, respectively. Mold growth on various bamboo specimens during indoor

exposure was evaluated following the procedures described in the ASTM (D3274-82) standard [12].

3. Results and discussion

3.1. Color changes of untreated and treated bamboo culms

Colors of untreated culms faded after oven drying as indicated by increased *a*^{*} value (from −5.1 to 2.0) of green color and varied *b*^{*} and *L*^{*} values of yellow color (from 36.4 and 59.9 to 27.4 and 69.1, respectively) (Table 1). Table 1 also shows results of treating bamboo culms with various 2% salt solutions at 60 or 100 °C for 2 h in preserving colors. Based on *a*^{*} values $\text{Cu}(\text{NO}_3)_2$ and CuP treatments were most effective, treatments with CuSO_4 , and CuCl_2 and CuN were somewhat effective but treatments with CP, CuAz, $\text{Cu}(\text{OH})_2$, $\text{Cu}(\text{CH}_3\text{COO})_2$ and ACQ had no effect. In addition, the $\text{Cu}(\text{NO}_3)_2$ - and CuP-treated specimens exhibited a slightly increased green brightness since the *L*^{*} values increased from initial (fresh sample) 50.1 to 53.3 and 53.7, respectively. In lieu of the fact that ACQ was found effective in preserving the green color of makino bamboo (*P. makinoi* Hayata) [10], the effectiveness of ACQ was further studied.

It has been pointed out that green color preservation is due to interactions between copper salts and chemical constituents of bamboo surface tissues [13,14]. Interactions between copper salts and cellulose, hemicelluloses, lignin and chlorophyll form stable and water-insoluble complexes during treatment, resulting in development and preservation of green color and excellent colorfastness.

3.2. Effects of salt concentration, treating temperature and treating time

Table 2 shows changes in *a*^{*} and ΔE^* values of bamboo culms after treating with various concentrations of $\text{Cu}(\text{NO}_3)_2$, CuP and ACQ at 60 or 100 °C for 2 h. Specimens treated with 1% $\text{Cu}(\text{NO}_3)_2$ at 60 °C for 2 h not only exhibited the lowest *a*^{*} value (−7.5) but also a small ΔE^* (2.9). Results also show excellent green color preservation when specimens were treated with 0.5% CuP (*a*^{*} = −5.8; ΔE^* = 1.6) and 0.25% ACQ (*a*^{*} = −5.5; ΔE^* = 2.5). Table 3 presents results of the effect of treating temperatures on green color preservation. When treating time was fixed at 2 h the *a*^{*} value of specimens treated with 1% $\text{Cu}(\text{NO}_3)_2$ decreased with increasing treating temperature to a minimum of −7.5 at 60 °C, while the *a*^{*} values of specimens treated with 0.5% CuP and 0.25% ACQ also decreased with increasing treating temperature and reached minimum values of −5.8 and −5.5, respectively, at 100 °C. Because the $\text{Cu}(\text{NO}_3)_2$ treatment resulted in an *a*^{*} value (*a*^{*} = −7.5) lower than that of fresh samples (*a*^{*} = −5.1), its effect was beyond preserving the original green color and the additional green color apparently was contributed to interactions between copper and cell wall constituents.

Table 1

Color changes of stripe long-shoot bamboo after treating with 2% of various copper salts at 60 or 100 °C for 2 h

Solvent system	Treating salt	Color region	CIE LAB		
			L^*	a^*	b^*
Untreated	Fresh	Green	50.1 ± 0.7	−5.1 ± 0.5	18.9 ± 0.8
		Yellow	59.9 ± 0.6	12.1 ± 0.7	36.4 ± 0.6
	Oven dried	Green	46.1 ± 1.2	2.0 ± 1.0	18.5 ± 1.4
		Yellow	69.1 ± 1.1	7.6 ± 0.9	27.4 ± 1.0
Alcohol-borne ^a	CuSO ₄	Green	47.0 ± 2.0	−5.3 ± 2.1	22.4 ± 1.8
		Yellow	80.1 ± 2.1	3.3 ± 1.8	21.6 ± 1.9
	CuCl ₂	Green	53.2 ± 1.6	−6.2 ± 1.7	20.0 ± 1.6
		Yellow	72.5 ± 1.4	1.2 ± 1.3	26.5 ± 1.3
	Cu(CH ₃ COO) ₂	Green	52.5 ± 2.0	−0.6 ± 1.9	27.2 ± 2.1
		Yellow	72.8 ± 2.5	4.6 ± 2.7	24.8 ± 2.4
	Cu(NO ₃) ₂	Green	53.3 ± 1.1	−7.3 ± 1.6	16.4 ± 1.9
		Yellow	64.6 ± 1.3	2.8 ± 1.8	24.4 ± 1.4
	Cu(OH) ₂	Green	57.8 ± 2.2	0.4 ± 2.0	28.6 ± 2.0
		Yellow	77.8 ± 1.9	5.9 ± 2.1	28.4 ± 2.3
Water-borne ^b	CP	Green	51.6 ± 2.4	2.5 ± 2.2	26.9 ± 2.0
	CrO ₃ : H ₃ PO ₄ = 1 : 1 (% , w/w)	Yellow	70.7 ± 2.2	5.5 ± 2.5	28.6 ± 2.3
	CuP	Green	53.7 ± 1.3	−8.7 ± 2.1	20.0 ± 2.0
	CuSO ₄ : H ₃ PO ₄ = 1.2 : 0.8 (% , w/w)	Yellow	71.8 ± 1.7	4.3 ± 1.5	23.6 ± 1.6
	CuAz	Green	52.5 ± 2.1	0.0 ± 2.5	24.3 ± 2.3
		Yellow	62.3 ± 2.4	5.4 ± 2.6	28.2 ± 2.2
	ACQ	Green	29.8 ± 2.1	4.7 ± 2.3	5.5 ± 2.0
	ACQ-B (16%)	Yellow	42.1 ± 2.2	11.3 ± 2.1	23.1 ± 1.9
Solvent-borne ^a	CuN	Green	54.7 ± 1.2	−5.1 ± 1.4	24.9 ± 2.0
	Xylene/ethanol = 1/1 (v/v)	Yellow	62.5 ± 1.0	4.3 ± 1.7	32.6 ± 1.7

^aSpecimens were treated at 60 °C.^bSpecimens were treated at 100 °C.

Table 2

Changes in the a^* and ΔE^* values of stripe long-shoot bamboo after treating with different concentrations of Cu(NO₃)₂, CuP and ACQ at 60 or 100 °C for 2 h

Conc. (%)	Cu(NO ₃) ₂ ^a		CuP ^b		ACQ ^b	
	a^*	ΔE^*	a^*	ΔE^*	a^*	ΔE^*
Untreated	−5.1 ± 0.5 ^C	—	−5.1 ± 0.5	—	−5.1 ± 0.5	—
0	2.1 ± 0.8 ^A	9.2	2.1 ± 0.8	9.2	2.1 ± 0.8	9.2
0.25	−4.7 ± 2.1 ^B	11.1	−5.0 ± 1.2	5.6	−5.5 ± 2.0	2.5
0.5	−5.3 ± 1.7 ^C	7.1	−5.8 ± 1.7	1.6	−4.5 ± 2.3	5.6
1	−7.5 ± 1.4 ^D	2.9	−5.3 ± 1.4	2.1	4.3 ± 1.9	21.8
2	−7.3 ± 1.6 ^D	3.7	−8.7 ± 2.1	4.0	4.7 ± 2.3	27.7
4	−4.4 ± 1.4 ^B	2.8	−3.8 ± 2.3	8.8	2.0 ± 2.7	23.1

 a^* values marked with different letters are significantly different at the level of $p < 0.05$ according to Scheffe's test.^aSpecimens were treated at 60 °C.^bSpecimens were treated at 100 °C.

Fig. 1 shows the effect of treating bamboo culms with 1% Cu(NO₃)₂ at 60 °C and with 0.5% CuP and 0.25% ACQ at 100 °C for various times on color changes. For the 1% Cu(NO₃)₂ treatment green color faded initially when the reaction was carried out for only 30 min but steadily increased beyond 1 h reaction time as indicated by decrease in the a^* values until a maximum green color was reached at 2 and 4 h, followed by a decrease at 6 h. A similar trend

was found for the 0.5% CuP and 0.25% ACQ treatments at 100 °C, in which maximum green color was reached at 2 h reaction time, followed by decreases at 4 and 6 h.

These results clearly demonstrated that the 1% Cu(NO₃)₂ treatment at 60 °C for 2 and 4 h was superior to all other treatments in enhancing the green color, while treatments with 0.5% CuP and 0.25% ACQ at 100 °C for 2 h were only able to preserve the green color at the original

Table 3

Changes in the a^* and ΔE^* values of stripe long-shoot bamboo after treating with 1% $\text{Cu}(\text{NO}_3)_2$, 0.5% CuP and 0.25% ACQ at different temperatures for 2 h

Temp. ($^{\circ}\text{C}$)	$\text{Cu}(\text{NO}_3)_2$		CuP		ACQ	
	a^*	ΔE^*	a^*	ΔE^*	a^*	ΔE^*
Untreated	-5.1 ± 0.5	–	-5.1 ± 0.5	–	-5.1 ± 0.5	–
25	2.6 ± 3.7	12.8	0.6 ± 3.0	6.5	-1.5 ± 2.8	4.1
40	-2.7 ± 3.4	9.3	1.1 ± 4.2	8.5	-1.6 ± 2.4	7.0
60	-7.5 ± 1.4	2.9	1.4 ± 3.7	8.8	-1.8 ± 2.7	5.3
80	–	–	3.5 ± 4.0	11.8	-2.7 ± 2.3	11.4
100	–	–	-5.8 ± 1.7	1.6	-5.5 ± 2.0	2.5

Fig. 1. Changes in a^* value of stripe long-shoot bamboo culms after treating with 1% $\text{Cu}(\text{NO}_3)_2$ at 60°C , and 0.5% CuP and 0.25% ACQ at 100°C for different times (value marked by different letters are significantly different at the level of $p < 0.05$ according to Scheffe's test).

level. Table 4 shows that the 1% $\text{Cu}(\text{NO}_3)_2$ treatment time could be shortened to 30 min if the reaction was carried out in an ultrasonic bath, where the green color reached the same a^* value (-8.0) as that treated at 60°C for 2 or 4 h ($a^* = -7.5$ to -8.0) without the aid of ultrasonic bath. Table 4 also shows the effect of ultrasonic treatment on yellow color. After the ultrasonic treatment at 60°C for 30 min, the yellow stripes had a least decrease in yellow color b^* value ($\Delta b^* = 8.1$) and a least color change ($\Delta E^* = 19.2$) among all treating durations. The brightness (L^*) of the yellow stripes also increased slightly from 59.9 to 62.4. These results suggest that the ultrasonic treatment with 1% $\text{Cu}(\text{NO}_3)_2$ at 60°C for 30 min greatly enhanced the green color with little impact on yellow color.

3.3. Colorfastness

Table 5 presents results of colorfastness and mildew resistance of bamboo culms treated with 1% $\text{Cu}(\text{NO}_3)_2$ at 60°C for 2 h as compared to untreated samples. After 6, 12 and 18 months of indoor exposure the a^* values of

untreated specimens steadily increased from -5.1 to -0.4 , 1.8 and 2.0 , respectively, while the corresponding a^* values of treated specimens only increased from -7.5 to -5.4 , -4.9 and -4.8 . As to changes in yellow color, the b^* value of untreated specimens decreased from 36.4 to 26.6 ($\Delta b^* = 9.8$) after 18 months of indoor exposure and that of treated specimens only decreased from 27.2 to 24.3 ($\Delta b^* = 2.9$). These results indicate the remarkable colorfastness of specimens treated with 1% $\text{Cu}(\text{NO}_3)_2$, in that after 18 months exposure to indoor light the specimens still maintained a green color very close to the fresh green color. Table 5 also shows that untreated specimens sustained some mold growths before drying out during indoor exposure, while the treated specimens totally prohibited mold growth on the surfaces.

3.4. Dipping treatments

Results of dipping treatments at the ambient conditions are shown in Fig. 2. The green color a^* value reached a minimum reading of -11.2 when the specimens were immersed in alcohol solution of 1% $\text{Cu}(\text{NO}_3)_2$ for 4 days. On the contrary, the green color of specimens treated with 1% CuP and ACQ aqueous solutions faded continuously with immersion time. Difference in the effect of green color preservation between the alcoholic and aqueous salt solutions is attributed to much quicker penetration of the alcoholic solution than the aqueous solutions into bamboo tissues for the interactions between copper salt and tissue constituents to occur.

4. Conclusions

Treating stripe long-shoot bamboo culms with 1% $\text{Cu}(\text{NO}_3)_2$ in alcohol at 60°C for 2 h was able to enhance the green color and to preserve the yellow color in the beautiful alternating green and yellow stripes. Treating time could be shortened to 30 min when the treatment was carried out in an ultrasonic bath. The same result was obtained when the bamboo culms were treated by immersing them in 1% $\text{Cu}(\text{NO}_3)_2$ alcohol solution at the ambient temperature for 4 days. Bamboo culms treated as such showed excellent color fastness after 18 months of

Table 4

Changes in color parameters of stripe long-shoot bamboo culms after treatment with 1% Cu(NO₃)₂ at 60 °C for different durations in ultrasonic bath

Treatment time (min)	Color region	CIE LAB			Δa^*	Δb^*	ΔE^*
		L^*	a^*	b^*			
Fresh bamboo	Green	50.1 ± 0.7	−5.1 ± 0.5	18.9 ± 0.8	—	—	—
	Yellow	59.9 ± 0.6	12.1 ± 0.7	36.4 ± 0.6	—	—	—
5	Green	47.6 ± 1.6	−0.2 ± 2.4	16.6 ± 2.7	4.9	2.3	13.3
	Yellow	70.2 ± 1.4	6.7 ± 2.2	24.8 ± 3.0	5.4	11.6	24.4
15	Green	50.4 ± 1.5	−4.3 ± 2.8	26.4 ± 2.9	0.8	7.5	8.1
	Yellow	69.2 ± 1.8	4.3 ± 2.1	23.7 ± 2.4	7.8	12.7	21.3
30	Green	48.9 ± 1.7	−8.0 ± 2.4	27.9 ± 3.1	−2.9	9.0	3.1
	Yellow	62.4 ± 1.4	5.8 ± 2.5	28.3 ± 2.6	6.3	8.1	19.2
60	Green	49.6 ± 1.8	−7.1 ± 2.2	22.8 ± 3.0	−2.0	3.9	4.3
	Yellow	68.4 ± 1.1	1.7 ± 2.6	26.2 ± 2.8	10.4	10.2	19.7
90	Green	50.1 ± 1.3	−6.1 ± 2.0	14.3 ± 2.4	−1.0	4.6	7.8
	Yellow	69.7 ± 1.2	4.3 ± 2.3	23.1 ± 2.7	9.4	13.3	20.5
120	Green	54.9 ± 1.2	−5.5 ± 1.9	25.6 ± 2.3	−0.4	6.7	10.0
	Yellow	72.4 ± 1.0	2.3 ± 2.2	23.4 ± 2.9	7.4	13.0	23.2

Table 5

Color changes and mold growth of stripe long-shoot bamboo culms treated with 1% Cu(NO₃)₂ after indoor exposure for 18 months

Exposure time (months)	Region of culm	Untreated bamboo			Treated bamboo ^a		
		a^*	b^*	M.R. ^b	a^*	b^*	M.R. ^b
0	Green	−5.1 ± 0.5	18.9 ± 0.8	10	−7.5 ± 1.4	27.0 ± 1.7	10
	Yellow	12.1 ± 0.7	36.4 ± 0.6		6.0 ± 1.1	27.2 ± 1.3	
6	Green	−0.4 ± 2.1	21.3 ± 2.4	8	−5.4 ± 1.7	20.7 ± 1.9	10
	Yellow	8.4 ± 1.9	28.2 ± 0.6		5.1 ± 2.0	25.6 ± 1.6	
12	Green	1.8 ± 2.3	17.9 ± 2.0	8	−4.9 ± 1.4	17.7 ± 2.1	10
	Yellow	7.3 ± 1.5	27.1 ± 0.9		4.5 ± 1.6	25.4 ± 1.4	
18	Green	2.0 ± 2.2	17.3 ± 2.3	8	−4.8 ± 1.2	14.9 ± 1.6	10
	Yellow	6.9 ± 1.8	26.6 ± 1.6		4.3 ± 1.1	24.3 ± 1.5	

^aGreen color protector dissolved in menthol and treated in 60 °C for 2 h.^bMildew resistance rating. Fig. 2. Changes in a^* value of stripe long-shoot bamboo culms after treating with 1% Cu(NO₃)₂ in alcohol and 1% CuP and ACQ in water at ambient temperature for different times.

indoor exposure. Treating bamboo culms with 0.5% copper phosphate and 0.25% ACQ aqueous solutions also were effective to preserve the colors at the fresh color levels.

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