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Effects of Hygrothermal Environment in Cooling Towers on the Chemical Composition of Bamboo Grid Packing

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Abstract: Bamboo grid packing (BGP) is a new kind of cooling packing, used in some Chinese hyperbolic cooling towers, which has excellent potential to complement or replace cooling packing made of polyvinyl chloride (PVC), cement, and glass fiber-reinforced plastic. For bamboo applications, mechanical properties and service life are matters of concern; this is strongly associated with bamboo's chemical composition and mass loss. To better understand the mechanics of mechanical property deterioration and service life reduction, this study investigated the effects of hygrothermal environments in cooling towers on the chemical and elemental composition, mass loss, Fourier-transform infrared (FTIR) spectrum, and color changes of BGP. The results showed that BGP that had been in service for nine years in cooling towers exhibited major decreases in content of hemicellulose and benzene-ethanol extractives, as well as a significant increases in the content of α-cellulose and lignin. Exposure to the hygrothermal environment led to a decrease of oxygen content and around 8% mass loss, as well as an increase in carbon content compared to control samples. The hot water flow in cooling towers not only hydrolyzed hemicellulose, but also degraded some functional groups in cellulose and lignin. The lightness (L^*) and chromaticity (a^* and b^*) parameters of the used BGP all decreased, except for the a^* value of the outer skin. The total color change (ΔE^*) of the inner skin of used BGP exceeded that of the outer skin.

Keywords: bamboo grid packing; cooling packing; cooling tower; chemical composition; elemental composition; FTIR; color

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1. Introduction

A hyperbolic cooling tower is a device wherein hot water from the system is cooled by the ambient air with the assistance of cooling packing [1], and it is widely used in industries due to its high capacity for heat rejection and energy saving [2]. As the core component of a cooling tower, good cooling packing not only increases effective contact between air and water, which promotes heat and mass transfer, but also provides less resistance to the movement of air to reduce pressure drop [3]. In order to improve heat dissipation efficiency and reduce cost, many different materials have been used as cooling packing, such as polyvinyl chloride (PVC), cement, and glass fiber-reinforced plastic. PVC packing with smooth- and cross-ribbing is the most popular kind due to its outstanding cooling performance and lightness in weight. It is used in 96% of cooling towers [4]. However, PVC packing also has lots of disadvantages, such as short service life, poor anti-fouling properties, environmental burdens, etc.

Furthermore, the price of PVC is on the rise. Therefore, many attempts have been made to seek an environmentally-friendly, low cost, and longer-serving alternative to PVC.

Owing to its fast growth speed, short rotation, great mechanical strength, and low energy consumption [5], bamboo, an abundant and sustainable plant resource, has been used as an innovative material for cooling packing. The product, known as bamboo grid packing (BGP), has been used in some hyperbolic cooling towers in China in recent years [6]. BGP has showed good temperature adaptability and anti-fouling properties, and also costs less than its PVC counterpart, which makes it a promising substitute for PVC packing. Mechanical properties and service life are a concern for all bamboo applications; this is strongly associated with its chemical composition and mass loss. The mechanical properties of bamboo were studied in our previous report [2]. However, it is still largely unknown how the hygrothermal environment in cooling towers affects the chemical composition of BGP. The lack of research in this aspect hinders the development of BGP for wider industrial application.

To better understand the reasons of mechanical property deterioration and service life reduction, the effects of hygrothermal environments in cooling towers on the chemical and elemental composition of BGP were investigated. In order to prevent damage to BGP's chemical composition and structure, Fourier-transform infrared (FTIR) spectroscopy was applied in this study. The presence of compounds or functional groups can be determined according to the number, shape, position, and intensity of the spectral bands, thus revealing the structure of the compound and its variation [7,8]. The color changes of BGP were also investigated, as they suggested variation in the chemical composition and structure. This study not only offers a new perspective for understand the changing patterns of the chemical composition, chemical structure, and color of BGP used in cooling towers, but may also contribute to the exploration of new approaches to alleviate hygrothermal aging.

2. Materials and Methods

2.1. Materials

The materials used in this study were identical to those in the previous study [2]. Raw materials were obtained from Moso bamboo (*Phyllostachys edulis* (Carr.) J. Houz), aged for four years, grown in Shaowu, Fujiian Province, China. The bamboo culms were sawn into segments 1200 mm long (Figure 1a) and then split longitudinally into several strips (1200 mm in longitudinal direction and 40 mm in tangential direction) (Figure 1b). Three holes with a diameter of 10 mm were made on the strips with a distance about 400 mm between each hole (Figure 1c). As shown in Figure 1e, round bamboo sticks 600 mm in length were inserted into the holes to connect the bamboo strips. The spacing between the strips was 50 mm. The dimensions of one piece of BGP were around 1200 mm × 600 mm × 40 mm. BGP units were stacked to a height of around 1.5 m in hyperbolic cooling towers [2]. BGP that has been in use for nine years was collected from two hyperbolic cooling towers located in Fujian and Shandong Provinces, respectively. The BGP collected from Fujian Province (FJBGP) and Shandong Province (SDBGP) were used in hyperbolic cooling towers of a thermal power plant and a steel mill, respectively. The bamboo materials used to fabricate the BGPs in these two towers were obtained from the same bamboo species and the same place as mentioned above. In both cooling towers, the inlet water temperature was around 45 to 50 °C, and the water mass flux was around 6500 $kg/(h \cdot m^2)$. Control samples were collected from unused BGP units. Prior to testing, all specimens were kept in a conditioned room at 21 ± 2 °C and 65 ± 3% relative humidity until their weights stabilized.

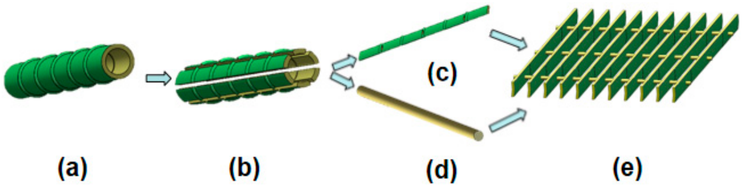

Figure 1. Preparation process of bamboo grid packing (BGP): (**a**) Raw bamboo; (**b**) radial splitting; (**c**) bamboo strip; (**d**) round bamboo stick; (**e**) bamboo grid packing.

2.2. Chemical Composition Measurements

Chemical composition of the used BGP and control samples were both measured. Specimen from each experiment was ground to pass a 40-mesh screen. The following contents in the ground meal was measured in accordance with relative standards: Acid-insoluble lignin using GB/T 2677.8-1994 [9], holocellulose content using GB/T 2677.10-1995 [10], α-cellulose content using GB/T 744-2004 [11], and benzene-ethanol extractives using GB/T 2677.6-1994 [12]. The content of hemicellulose is the difference between the content of holocellulose and that of α-cellulose.

2.3. Elemental Composition and Mass Loss

The used BGP and control samples were ground to fine powder and passed through different sieves to obtain a particle size between 0.2 and 0.5 mm. The powder was then conditioned at 103 °C for 24 h and stored in closed bottles before analysis. Elemental analyses were performed using a Thermofinnigam Flash EA1112 micro-analyzer (Sundy Co., Hunan, China). The content of carbon, oxygen, and hydrogen were measured. At least three replicates were tested for each sample. Mass loss due to chemical degradation caused by the hygrothermal environment in cooling towers was calculated according to the following equation:

$$ML(\%) = \frac{m_0 - m_1}{m_0} \times 100\% \tag{1}$$

where m_0 is the initial anhydrous mass of the sample before being put to use in cooling towers, and m_1 is the anhydrous mass of the same sample after nine years of service.

2.4. FTIR Spectroscopy Analysis

Before being ground, the used BGP was gently brushed to remove impurities from the surface. Then all specimens for the FTIR analysis were prepared by being ground in a mill with a 100-mesh screen (FW100, TAISITE Co., Tianjin, China). The FTIR spectra of the samples were obtained in a Nexus 670 spectrometer (Nicolet, WI, USA) within the range of 500–4000 cm^{-1}, with a resolution of 4 cm^{-1} and 64 scans. The KBr pellet, consisting of KBr and randomly-selected bamboo powder with a weight ratio of 100:1, was prepared prior to the measurement. The FTIR analysis for each sample was performed in quintuplicate. For evaluation of the spectra, only the area between wavenumbers 800 and 1800 cm^{-1} will be discussed; this includes the most important values of lignocellulose materials [13].

2.5. Color Measurements

The changes in color of BGP surface due to hygrothermal treatment were measured using a Technibrite Brightmeter micro S-5 colorimeter (Technidyne Corporation, New Albany, IN, USA) with an aperture size of 1 cm^2. CIELAB (CIE$L^*a^*b^*$) is a color space that describes all the colors visible to the human eye and it is created to serve as a device-independent model used as a reference [14]. In the CIELAB system, the L^* axis represents the lightness (L^* varies from 100 (for white) to zero (for black)), a^* and b^* describe the chromatic coordinates on the green–red and blue–yellow axis, respectively (+a^* is for red, $-a^*$ for green, +b^* for yellow, $-b^*$ for blue) [15]. ΔE^* was calculated using the following equation:

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

where ΔL^*, Δa^*, and Δb^* are the difference of initial (control samples) and final (used BGP) values of L^*, a^*, and b^*, respectively.

2.6. Statistical Analysis

The effects of hygrothermal environment on the chemical composition and color changes in comparison with control samples were analyzed by analysis of variance (ANOVA) using SPSS 19.0 (IBM SPSS Corporation, Chicago, IL, USA). The significance ($p < 0.05$) between average values of control samples and used BGP specimens was compared using Duncan's test. Different letters given along with the average values of tested parameters indicated a significant difference by Duncan's test.

3. Results and Discussion

3.1. Chemical Composition Analysis

The structure of bamboo is stable because the natural tissues of the cell walls are composed of three bio-based chemicals—cellulose, hemicellulose, and lignin. Cellulose provides structural support in the cell walls. Hemicellulose and lignin are the binding and packing materials for the skeleton structure. The three components are interwoven into thin layers that together form bamboo cell walls [16]. In addition to lignocellulosic structures, bamboo also contains a variety of low molecular weight organic compounds known as extractives—such as terpenes, resins, fatty acids, waxes, and phenols—which can be extracted using solvents [17,18]. The temperature of inlet water in both cooling towers was around 45 to 50 °C, and the effects of hot water flow on the chemical composition of the used BGP are presented in Table 1.

Table 1. Analysis of main chemical composition of the used BGP and control samples.

Sample	Holocellulose (%)	α-Cellulose (%)	Hemicellulose (%)	Lignin (%)	Benzene-Ethanol Extractives (%)
Control	64.08a (1.25)	41.03a (1.94)	23.05c (0.09)	24.37a (2.23)	5.77c (0.86)
FJBGP	66.27ab (2.01)	45.43b (4.08)	20.84b (2.75)	27.68b (2.44)	1.62b (1.33)
SDBGP	67.81b (2.35)	48.35b (2.24)	19.46a (2.65)	26.63b (4.06)	1.15a (1.23)

Note: FJBGP: bamboo grid packing collected from Fujian; SDBGP: bamboo grid packing collected from Shandong. Values in parentheses are coefficient of variation. The different letters in the same column indicate a significant difference at the 0.05 level.

The hemicellulose content of FJBGP and SDBGP were 20.84% and 19.46%, respectively. Both data were significantly lower than those of control samples, which indicates that hot water flow in the cooling towers partially degraded hemicellulose. Hemicellulose is a heterogeneous low molecular weight polysaccharide composed of acetyl groups, aldonic acid groups, and different glycosyl groups, most of which are thermally labile, especially the acetyl groups. Furthermore, because of its branched structure and amorphous tissues, hemicellulose is considerably more susceptible to thermal degradation than other chemical components [19].

The benzene-ethanol extractive contents of FJBGP and SDBGP were 1.62% and 1.15%, respectively, which were also significantly lower than those of control samples. The changes in benzene-ethanol extractive content of the used BGP were not consistent with the results of heat-treated bamboo and wood [20,21]. Changes in extraction content are related to the degradation of hemicellulose and cellulose. Volatiles, extractives, and water are products that result from hemicellulose and cellulose degradation [22]; the phenolic compounds, which can be very soluble in benzene-ethanol mixture, can also be formed in depolymerization reactions [23]. The decrease in the benzene-ethanol extractive content of the used BGP may be attributed to the dissolution of extractives in hot water flow and the degradation of hemicellulose.

The α-cellulose contents of FJBGP and SDBGP were 45.43% and 48.35%, respectively, and the lignin contents were 27.68% and 26.63%. These numbers were all significantly higher than those of control samples, which indicates that the degradation of α-cellulose and lignin by hot water flow was less significant than that of hemicellulose. α-Cellulose is a homogeneous polysaccharide consisting of the same type of glucosyl groups. It is also a straight-chain structural macromolecule without branched chain, therefore its thermal stability is relatively good. Lignin is a complex phenolic polymer synthesized from three alcohol monomers (namely *p*-coumaryl alcohol, coniferyl alcohol, and sinapyl alcohol) [24]. It has large molecular weight, few lyophilic groups, and is insoluble in water and general solvents. Lignin is the most stable compound for which thermal degradation is observed above 100 °C, although high temperatures can result in cleaved bonds within the lignin [25]. Because cellulose and lignin have fewer lyophilic groups and more stable chemical structures, they are more difficult to hydrolyze by hot water flow than hemicellulose. The increase of α-cellulose and lignin contents in SDBGP and FJBDP may largely be the result of the hydrolysis of benzene-ethanol extractive content and the degradation of hemicellulose.

The effects of hygrothermal environments on the chemical composition of BGP may be less conspicuous in the short term because the highest water temperature is usually below 65 °C. The actual extent of change in the chemical composition of used BGP was due to years of accumulation.

Our previous experiment studied the mechanical properties of used BGP that had been in service for nine years in cooling towers [2]; the reduction in the mechanical properties of SDBGP and FJBGP was mainly caused by the depolymerization reactions of polymers, which led to the reduction in hemicellulose, the most thermochemically sensitive component of bamboo. The degradation of hemicellulose is primarily responsible for initial strength loss [26,27].

3.2. Elemental Composition and Mass Loss

Elemental composition and mass loss of the samples are presented in Table 2. Results indicate that the hygrothermal environment led to a drop in oxygen content and an increase of carbon content of used BGP compared to control samples, which was in line with the results of heat-treated wood [21,28,29]. The decrease of the O/C ratio of used BGP manifested severe dehydration due to higher degradation of amorphous polysaccharides and/or higher amounts of carbonaceous materials within the bamboo structure, as reported for heat-treated wood [30–32]. The degradation of polysaccharides involves depolymerization by transglycosylation and dehydration reactions. As a result, the production of anhydro monosaccharides that could participate in subsequent reactions led to the formation of carbonaceous materials [17,33].

Table 2. Elemental composition and mass loss of used BGP and control samples.

Sample	Carbon (%)	Oxygen (%)	Hydrogen (%)	O/C ^{a}	Mass Loss ^{b} (%)
Control	48.68	45.11	6.14	0.695	-
FJBGP	48.95	44.95	6.06	0.689	7.94
SDBGP	49.15	44.73	6.06	0.683	8.85

Note: ^{a} = atomic ratio; ^{b} = mass loss due to hygrothermal treatment.

Elaieb et al. reported that the O/C ratio was negatively correlated with mass loss and inferred that the O/C ratio is a good indicator for mass loss of wood after thermo-degradation [34]. Moreover, the O/C ratio is related to the content of hemicellulose [28]. The O/C ratio and hemicellulose contents of FJBGP were 0.689 and 20.84%, respectively, exceeding those of SDBGP, while the mass loss of FJBGP was 7.94%, lower than that of SDBGP. These indicate that hygrothermal degradation of SDBGP was more significant than of FJBGP.

3.3. FTIR Analysis

To investigate the changes of chemical structure that took place in used BGP, FTIR spectroscopy was applied in this study. Spectra of specimens are shown in Figure 2. Bamboo bears basic structural similarities to wood because of the similar chemical constituents. Therefore, characterization and assignment of IR peaks in bamboo was done by reference to wood, as previously described by Wang and Ren [35,36]. The assignments of the peaks to structural components were as follows: 1730 cm^{-1} for unconjugated C=O in xylan (hemicellulose), 1603 cm^{-1} and 1510 cm^{-1} for aromatic skeleton in lignin, 1460 cm^{-1} for CH^3 deformation in lignin and CH^2 bending in xylan, 1425 cm^{-1} for CH^2 scissor vibration in cellulose, 1370 cm^{-1} for C-H deformation in cellulose and hemicellulose, 1330 cm^{-1} for C-H vibration in cellulose and C$_1$-O vibration in syringyl derivatives, 1240 cm^{-1} for syringyl ring and C-O stretch in lignin and xylan, 1160 cm^{-1} for C-O-C vibration in hemicellulose and cellulose, 1122 cm^{-1} for aromatic skeletal and C-O stretch in lignin and cellulose, 1048 cm^{-1} for C-O stretch in cellulose and hemicellulose, 897 cm^{-1} for C-H deformation in cellulose [37–39].

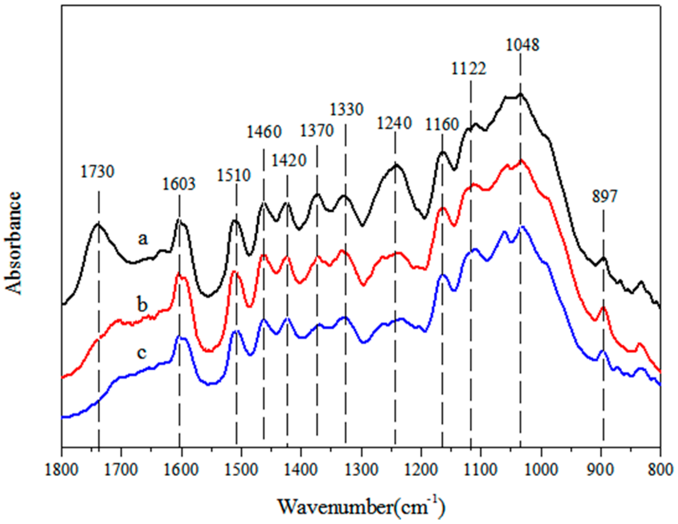

Figure 2. Fourier-transform infrared (FTIR) spectra of used BGP and control samples. (**a**) Control samples; (**b**) BGP collected from Fujian Province (FJBGP); (**c**) BGP collected from Shandong Province (SDBGP).

Hygrothermal treatment seemed to have a significant impact on the chemical structure of used BGP (Figure 2). Peaks at 1730, 1370, 1240, and 1048 cm^{-1} for used BGP decreased compared to control samples. The intensity of the unconjugated C=O peak at 1730 cm^{-1}, often used as an important baseline for the degradation of hemicellulose [25], experienced a dramatic decrease, indicating that hemicellulose was degraded most severely. The degradation of unconjugated C=O was caused by deacetylation that occurred during hygrothermal treatment, which was a result of the cleavage of acetyl groups linked as ester groups to the hemicellulose [40]. The hygrothermal treatment caused a marked reduction in the 1370 cm^{-1} peak in used BGP due to damage of C-H bonds in hemicellulose and cellulose. The peak intensity of 1240 cm^{-1} of used BGP was lower than that of control samples owing to the degradation of syringyl ring and C-O bonds in lignin and xylan. The decrease in intensities of absorption bands at 1048 cm^{-1} indicates the loss of C-O in cellulose and hemicellulose. The difference in FTIR spectra between FJBGP and SDBGP was rather slight, except for the absorption bands at 1048 cm^{-1}. The intensities of absorption bands at 1048 cm^{-1} in SDBGP was lower than that of FJBGP, and the absorption peak at 1048 cm^{-1} in SDBGP was split into two small peaks, which showed the degradation of C-O bonds in SDBGP was more noticeable than that of FJBGP. Although the content of cellulose and lignin increased, the C-H and C-O bonds in cellulose and syringyl ring in lignin were both partially degraded.

The intensity of the remaining bands for used BGP were almost the same with control samples, which indicates that functional groups corresponding to these peaks were relatively unaffected by hot water flow.

3.4. Color Changes

The color of lignocellulosic material is related to its chemical composition. Since the main chemical composition and structure of bamboo is similar to wood, the theory and method of analyzing color changes of wood can be applied to analyze bamboo. The effects of hygrothermal environment on color changes of the outer and inner skin of all samples are shown in Table 3. Control samples were light-colored. L^* values of the outer and inner skin were 66.27 and 74.03, respectively. The surface color grew significantly darker after nine years of use. In terms of the outer and inner skin, the L^* value of FJBGP decreased from 66.27 to 40.95 and from 74.03 to 36.30, respectively, and that of SDBGP decreased from 66.27 to 50.85 and from 74.03 to 41.95. Changes in color reflected alteration in the chemical composition and structure of BGP during hygrothermal treatment. Some studies reported that that a decrease in L^* caused by a loss of lightness was possibly due to a degradation of hemicellulose and an increase in lignin content [41,42], as well as the degradation of oxygen-containing groups such as carboxyl groups and acetyl groups [19]. The darkening of used BGP may mainly be attributable to the degradation of acetyl groups in hemicellulose and oxidation of lignin.

Table 3. Color changes of used BGP and control samples.

Samples		**CIELAB**			
		L^*	a^*	b^*	ΔE^*
Outer skin	Control	66.27c (5.08)	1.70a (3.23)	14.34c (12.48)	—
	FJBGP	40.95a (4.91)	3.40b (9.12)	11.69a (4.19)	25.51
	SDBGP	50.85b (5.68)	4.44c (7.21)	13.61b (3.38)	15.68
Inner skin	Control	74.03c (2.34)	5.16c (8.91)	19.91c (4.82)	—
	FJBGP	36.30a (3.5)	2.87a (7.77)	9.99a (8.51)	39.08
	SDBGP	41.95b (12.35)	3.84b (6.14)	13.32b (9.67)	32.78

Note: Values in parentheses are coefficient of variation. The different letters in the same column indicate a significant difference at the 0.05 level.

The a^* value of FJBGP and SDBGP outer skin increased from 1.7 to 3.4 and from 1.7 to 4.44, respectively. However, the changes in the inner skin demonstrated an opposite trend, decreasing from 5.16 to 2.87 (FJBGP) and from 5.16 to 3.84 (SDBGP), respectively. The original brightly-colored lignocellulosic material turned dark and reddish under the hygrothermal environment, as a result of the volatilization of a large amount of colored extracts. However, the color indices of lignocellulosic material (near neutral color) only increase slightly due to the "darkening" effect caused by the rapid oxidation of the chemical composition [43]. Naturally, the original color of bamboo outer skin is green, while the inner skin is mostly red-brownish. After most of the extractives and chromophores in used BGP were hydrolyzed by hot water flow and some chemical composition were oxidized, the color of the outer and inner skin of used BGP become somewhat alike. In other words, the outer skin reddened while the inner skin grew comparatively green.

For the b^* value, FJBGP decreased from 14.34 to 11.69 and from 19.91 to 9.99 for the outer and inner skin, respectively, and SDBGP dropped from 14.34 to 13.61 and from 19.91 to 13.32. The lower b^* values of used BGP indicates that it became bluer compared to control samples. This specific change was induced by the modification of hemicellulose following hygrothermal treatment [15]. The total color difference (ΔE^*) of used BGP's inner skin was greater compared to the outer skin because the inner skin underwent a greater change in terms of lightness. The ΔE^* was caused by the volatilization of color extracts and the oxidation, degradation and polymerization of the chemical composition, which also had a significant correlation with the content of holocellulose, α-cellulose, and acid insoluble lignin [44].

4. Conclusions

The results of this study confirmed that the hygrothermal environment in cooling towers influenced the chemical and elemental composition, mass loss, chemical structure, and color changes of BGP. Statistically significant differences were observed in the content of hemicellulose, benzene-ethanol extractives, α-cellulose, and lignin between used BGP and control samples. Specifically, there was a decrease of hemicellulose and benzene-ethanol extractive content in used BGP and an increase in α-cellulose and lignin content. The O/C ratio of used BGP decreased in general, but it was higher in FJBGP than in SDBGP with less mass loss. The FTIR spectra showed the unconjugated C=O, C-H, and C-O bonds in hemicellulose; C-H and C-O bonds in cellulose; and the syringyl ring in lignin were partially degraded. However, the peak intensities and positions of the rest of the functional groups remained mostly unchanged. Color parameters such as lightness (L^*) and chromatic coordinates a^* and b^* of used BGP surface all decreased due to the hygrothermal environment's influence, with the exception of the a^* value of the outer skin. The total color difference (ΔE^*) of used BGP's inner skin was more substantial than that of the outer skin.

This study provides primary data for BGP used in industrial cooling towers. Still more work needs to be done in order to promote the application of BGP, such as optimizing its design, evaluating its service life, devising appropriate methods to alleviate the effects of hygrothermal aging, etc.

Author Contributions: L.-S.C., X.-X.M., and B.-H.F. conceived and designed the experiments; L.-S.C. performed the experiments; C.-H.F. analyzed the data; J.-P.L. contributed reagents/materials/analysis tools; L.-S.C. wrote the paper, and C.-H.F. revised it.

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