Performance Testing of Modified Waterborne Polyurethane Coating Applied on Laminated Bamboo

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The effects of different UV absorbents and preservatives on the weatherability of modified waterborne polyurethane (WPU) printed laminated bamboo were investigated. Three types of UV absorbents including 2-hydroxy-4-n-octoxy-benzophenone (UV-531), 2-(2 H-benzotriazol-2-yl)-4-(1,1,3,3 tetramethylbutyl) phenol (UV-329), and nano-TiO_2, and four types of preservatives including boric acid (BA), borax (BX), ammonium polyphosphate (APP), and disodium octaborate tetrahydrate (DOT) were selected to modify WPU coatings. The printed laminated bamboo was tested to evaluate the coating physical and chemical properties and dimensional stability. Thirteen coating types were tested. The results showed that the 0 (20% WPU), 5 (UV-531-BA/BX), 6 (UV-531-BA/BX/APP), 7 (UV-531-BA/BX/DOT), 8 (UV-531-BA/BX/APP/DOT), and 9 (nano-TiO2/BA/BX) samples performed well in adhesion, abrasion resistance, hardness, and temperature denaturation. Fourier transform infrared (FT-IR) spectra analyses and dimensional stability analysis were carried out on the six kinds of coatings screened out. FT-IR spectra analyses showed the successful introduction of UV light absorbers and flame retardants, whereas test results of hygroscopicity showed that the coated test material improved the dimensional stability performance. Test material 8(UV-531-BA/BX/APP/DOT) had the best dimensional stability performance.

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INTRODUCTION

Bamboo has the advantages of excellent mechanical properties, short growth cycle, and strong carbon fixation ability, which meet consumer needs for environmental materials with low-carbon, energy-saving, and emission-reducing characteristics. Bamboo has been widely used in construction, furniture, packaging, and many other fields. However, when bamboo-based materials are used outdoors, they are easily destroyed by severe climate conditions, resulting in serious problems such as bamboo cracking, discoloration, and mold (Cheng *et al*. 2020; Cui *et al*. 2021; Dogan *et al*. 2021; Greiner *et al*. 2021). Thus, bamboo-based materials need to be pretreated or modified before use (Fang *et al*. 2019; Yang *et al*. 2019b). Ultraviolet light radiation is the main factor affecting the photodegradation of the bamboo surface, which accelerates its aging. Boron-based preservatives provide suitable anti-biological deterioration (Yang *et al*. 2016; Wang *et al*. 2018) and flame retardant ability for bamboo (Salmeia *et al*. 2018). Ultraviolet light absorbents such as zinc oxide, nano-titanium dioxide, aluminum oxide, and iron oxide improve the color and dimensional stabilities (Lin *et al*. 2017).

Bamboo does not have radially distributed parenchyma cells and ray cells, but it is rich in the colloidal substances, infiltrating bodies, and other non-cell wall substances (Zhang *et al*. 2020; Kim *et al*. 2021). It is difficult to introduce a modification solution completely into the bamboo-based materials using the traditional impregnation or immersion treatment (Guo *et al*. 2019; Hajj *et al*. 2020; Hamidov *et al*. 2021). The application of printing technology in bamboo-based materials ensures the uniformity and thoroughness of the modification treatments (Yang *et al*. 2019a; Shi *et al*. 2020). Waterborne polyurethane (WPU) coatings use water as the solvent and have excellent mechanical properties; they have become a hot spot in the research on wood/bamboo coatings (Dai *et al*. 2020; Chang *et al*. 2021; Cui *et al*. 2021). However, WPU coating is also easily destroyed by exterior factors (Deng *et al*. 2018; Hormaiztegui *et al*. 2020). UV-531 is a high-efficiency anti-aging additive with excellent performance. It can absorb ultraviolet light in the wavelength range 240 to 340 nm (especially strong absorption at 270 to 330 nm), and has the characteristics of light color, non-toxicity, good compatibility, and easy processing. It helps delay yellowing and retard physical property loss. It is widely used in plastics, plexiglass, and polypropylene fibers. In terms of coatings, it can provide good light stabilization effect for dry phenolic, polyurethane, and other air-drying products. It also has a protective effect on plant fibers such as bamboo and wood. UV-329 is also suitable for plastics, epoxy resins, resin fibers, and ethylene vinyl acetate, *etc*. It mainly absorbs ultraviolet light of 270 to 340 nm, and it can be used for packaging materials such as plastic containers and food packaging boxes, providing them with a good light stability, with almost no visible light absorption, which is especially suitable for colorless, transparent and light-colored products (Liu *et al*. 2020; Montesdeoca-Esponda *et al*. 2021).

In this research, three kinds of ultraviolet light absorbents (UV-531, UV-329, and nano-TiO_2) and four kinds of bamboo preservatives (boric acid (BA), borax (BX), ammonium polyphosphate (APP), and disodium octaborate tetrahydrate (DOT)) were selected to improve the weatherability of WPU-coated laminated bamboo. The coating physical and chemical properties as well as the dimensional stability of the printed laminated bamboo after 3 months outdoor exposure were analyzed to determine the seasonable modification conditions in order to provide useful information for the laminated bamboo industrial production.

EXPERIMENTS

Materials

Moso bamboo (*Phyllostachys edulis* (Carr.) H.de Lehaie) was taken from Jiangxi Province, China. The bamboo strips were hot-pressed into the laminated bamboo with dimensions of 100 mm (tangential) × 100 mm (longitudinal) × 9 mm (radial). The adhesive was phenolic glue, the hot pressing temperature was 130 °C, the hot pressing pressure was 3.9 to 5.9 MPa, and the pressure was segmented, from low to high, and the hot pressing time was 1.5 to 2.0 h, with a radial fit between them. The moisture content of bamboo lumber was maintained at 4% to 6%. SETAQUA 6515 water-based paint was purchased from Allnex Resin Co., Ltd. (Shanghai, China), in which the main film-forming component was polyurethane, and the solid content was 50%.

Table 1. Modifications of WPU Coatings

Specimens	Preservatives	Preservative components ratio	UV absorbents
0	-	-	-
1	BA/BX/W(water)	1:1:8	UV-329
2	BA/BX/APP/W	75:75:7:600	
3	BA/BX/DOT/W	75:75:42:600	
4	BA/BX/APP/DOT/W	75:75:7:42:600	
5	BA/BX/W	1:1:8	UV-531
6	BA/BX/APP/W	75:75:7:600	
7	BA/BX/DOT/W	75:75:42:600	
8	BA/BX/APP/DOT/W	75:75:7:42:600	
9	BA/BX/W	1:1:8	nano-TiO_2
10	BA/BX/APP/W	75:75:7:600	
11	/BA/BX/DOT/W	75:75:42:600	
12	BA/BX/APP/DOT/W	75:75:7:42:600	

-stands for without any modification.

WPU coatings were modified with four types of bamboo preservatives compounded with three types of UV absorbents, as shown in Table 1. WPU coating with a solid content of 20% was mixed with one type of these ultraviolet light absorbents at room temperature for 6 h, and a magnetic stirrer was used at a speed 100 r /min. The added amount of the ultraviolet light absorbent was 1% (mass fraction) of the target coating, and then mixed with one type of these four compound preservatives. The modified WPU coating was stirred completely before painting.

The bamboo laminate was polished with 200-grit sandpaper to get a smooth surface. The modified WPU coating was applied on the surface of the laminated bamboo with a brush, with a balance for weighing, each brushing 2 g, and then dry in natural conditions for 1 hour, and then after reaching the finger dry brushing next time, a total of 4 times. For further experiments, the specimens that had been finished by brushing were subjected to three months of outdoor exposure.

Characterization Experiment of Coating Physical and Chemical Properties

Considering the significant mean change, the test was performed according to the following Chinese national standard description. The following detection method adopts three sets of repeated patterns according to the standard, and the error is calculated as the error of the three sets of data.

Coating adhesion (AH) test

The laminated bamboo was cut to a size of 100 mm × 100 mm × 9 mm, and three replicates in the same treatment condition submitted to the AH test referring to the GB/T 17657 (2013) standard. Three levels of AH grades were taken to evaluate the coating adhesion. Grade 1 meant little shedding at the cutting intersection, and the cross cutting area was not affected by more than 5%; Grade 2 meant that the coating fell off at the cutting edge or at the intersection, and the affected cutting area was 5% to 15%; Grade 3 meant the coating completely fell off, with a large area of peeling at the cutting edge.

Fig. 1. Adhesion test

Coating abrasion resistance (AR) test

A hole was punched in the center of the coated test material, referring to the GB 23999 (2009) standard, under the gravity of 750 g, with 500 r as the test revolution, 60 r/min rotation speed, and recorded the sample grinding. The mass after pre-grinding and the average weight loss (AWL) were calculated to test the AR of water-based coatings.

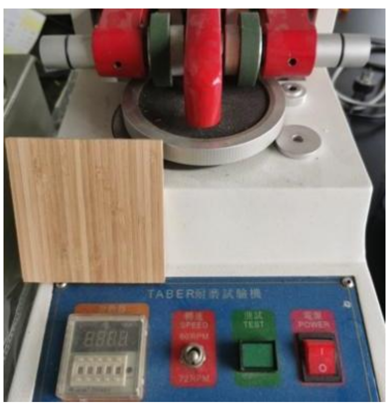

Fig. 2. Abrasion resistance test

Coating hardness (H) test

The hardness of the coating was tested with a scratch tester composed of a series of pencils with the hardness from the hardest 6H to softest 6B according to the GB/T 6739 (2006) standard. The pencils with different hardness were inclined at 45° to the coating and scratched the coating under a load of 1 kg. When the coating began to have scars, the hardness of the pencil was taken as the hardness of the tested coating.

Fig. 3. Hardness test

Temperature change resistance (TD) test

According to GB/T 4893.7 (2013) method, the dried painted samples were treated for 1 h in a constant temperature and humidity box with a temperature of 40 °C and a humidity of 95%. After the treatment, the samples were immediately placed at -20 °C for 1 h. After 3 repeated cycles, the sample was observed by a 4x magnifying glass starting at 20 mm from the edge of the sample to the surface of the coating in the middle of the samples. If any defects such as cracks, bubbling, obvious loss of light, and/or discoloration appeared, the sample was rated as unqualified.

Fig. 4. Temperature change resistance test

Dimensional Stability Test

The dimensional stability measurements were conducted after the samples were painted with different WPU coatings. The dimension of the laminated bamboo was 20 mm × 20 mm × 9 mm (GB/T 1934.2 2009) and conditioned at 20 ℃ and 65% relative humidity. The relevant calculations indices were as follows,

$$S_L = \frac{L_W - L_D}{L_D} \times 100\% \quad (1)$$

$$S_V = \frac{V_W - V_D}{V_D} \times 100\% \quad (2)$$

where S_L is the linear swelling ratio; S_V is the volume swelling ratio; L_W and L_D are the radial or chord-wise dimensions of bamboo after water absorption and absolute drying, respectively; and V_W and V_D are respectively absorbent and dry bamboo volume.

$$ASE_L = \frac{S_{LC} - S_{LT}}{S_{LC}} \times 100\% \quad (3)$$

$$ASE_V = \frac{S_{VC} - S_{VT}}{S_{VC}} \times 100\% \quad (4)$$

where ASE_L is the linear average expansion ratio; ASE_V is the average volumetric expansion ratio; S_{LC} is the linear swelling ratio of untreated specimens, S_{LT} is linear swelling of treated specimens; S_{VC} is the volume swelling ratio of untreated specimens, and S_{VT} is volume swelling of treated specimens.

$$M_A = \frac{m_M - m_D}{m_D} \times 100\% \quad (5)$$

$$M_{EE} = \frac{MA_C - MA_T}{M_{AC}} \times 100\% \tag{6}$$

In Eqs. 5 and 6, M_A and M_{EE} are moisture absorption rate and moisture excluding efficiency; m_M is the mass of bamboo after water absorption, m_D is the mass of dry bamboo, MA_C is the water absorption of untreated bamboo, and MA_T is the dimensionally stabilized bamboo water absorption of treated bamboo. During the process of hygroscoping/water absorption, the chordwise dimension changes were tested every 6 h with 2 to 3 styles. When the difference between the two measurements was less than 0.2 mm, the dimension was considered stable.

Fourier Transform Infrared (FT-IR) Spectra Text

After stabilization, 6 kinds of coatings were poured on a square glass plate and dried naturally for a week. After drying to form a film, a sample was made, and the sample was tested with an infrared tester for analysis.

RESULTS AND DISCUSSION

Analysis of Physical and Chemical Properties of Coatings

The physical and chemical performance tests were conducted on 13 kinds of test materials that had been exposed to the outdoors for three months. The effects of different modifications on the WPU coating properties including AH, AR, H and TD were recorded in Table 2. Specimens in the modifications of 0, 5, 6, 7, 8, and 9 had relatively better adhesion between the coating and the bamboo surface, which may be due to the good compatibility of the added UV absorbent UV-531 with WPU. With the increase of the viscosity of the added preservatives, the cross-linking and curing of the coating during the drying process was affected, and the AH of the coatings in the specimens of 10, 11, and 12 were all decreased. For UV-531 and WPU, the emulsion still performed well after several months of standing, demonstrating that there was no mutual reaction and good binding. The AH of coatings in the specimens of 3, 4, 11, and 12 was poor, and the reason may be attributed to the addition of DOT and APP, which has increased the viscosity of the coating, as the result it was difficult for the coating to penetrate into the interior of the laminated bamboo during the drying process.

As shown in Table 2, the mass loss of coatings 1 to 4 (with UV-329) and coatings 5 to 8 (with UV-531) were more than those of coatings 9 to 12, which may be due to the addition of nano-TiO_2 in the WPU coatings. This was attributed to the fact that after WPU has been painted, a dense network structure was formed on the surface of the laminated bamboo, which has a strengthening effect on the coating. In this study, the coating losses of the samples with different the modifications were not obvious.

The hardness of coating in the samples without any modification (0 group) reached 4H. After adding different UV absorbents, the hardness of the coatings decreased in different degrees (1H-3H). UV absorbents destroy the stability of the WPU emulsion (Patel and Kapatel 2019; Qian *et al*. 2021; Schirp and Schwarz 2021) The most obvious decrease of coating hardness was observed in the samples of group 6, which dropped to H. Thus, the mixture of APP and UV-531 was not suitable to modify WPU coating.

In the TD experiment, according to the national standard, the gloss was observed with eyes, and there was no obvious change, no obvious defects including cracks, bubbling, obvious loss of gloss and discoloration on the surface of all painted laminated bamboo was observed compared with the samples before the TD test. Combination of AH, AR, H and TD test results, the coatings of the painted laminated bamboo with the modifications of 0 (20% WPU), 5 (UV-531-BA/BX), 6 (UV-531-BA/BX/APP), 7 (UV-531-BA/BX/DOT), 8 (UV-531-BA/BX/APP/DOT), and 9 (nano-TiO_2/BA/BX) had better physical and chemical performance and these modifications were selected for the dimensional stability analysis.

Table 2. Coating Physical and Chemical Properties

Specimen	0	1	2	3	4	5	6	7	8	9	10	11	12
AH grade	2	3	3	2	3	1	1	2	2	1	2	3	3
AR (g)	0.05	0.05	0.05	0.05	0.13	0.09	0.05	0.09	0.05	0.01	0.02	0.01	0.02
H	4H	3H	3H	2H	3H	3H	H	3H	3H	3H	3H	3H	3H

Analysis of Moisture Absorption Experiments

The hygroscopicity of the specimens subjected to different painting treatments is shown in Table 3. The data in the table above were calculated after taking the average of three measurements. The specimens treated with different finishes showed better hygroscopic properties than the untreated specimens. At 24 h and 48 h, the S_L of the laminated bamboo without painting was 0.29% and 0.44%, the S_v of the laminated bamboo without painting was 0.67% and 0.94%; and the MA of the laminated bamboo without painting was 1.37% and 2.28%. Correspondingly, the specimens treated with different finishes showed better hygroscopic properties than the untreated specimens. The reason may be due to the large number and amount of compound flame retardants added to the coating material, the strong binding performance, and the good combination with the WPU. The coating film formed on the surface of the substrate was relatively dense, which improved its ability to resist the penetration of water vapour. In tests at 24 h and 48 h, the ASE_L of the specimens in group 8 was the best, reaching 82.76% and 77.27%, respectively. The reason may be that the APP in the flame retardant was insoluble in water, and a hydrophobic film was formed on the surface of the substrate, which also made it resistant to water intrusion (Wang *et al*. 2021; Wu *et al*. 2021). The specimens in group 8 performed was best at 24 h and 48 h, the MA rate was good, and the MEE was the best among the test materials. Among the test materials, only the water-blocking properties of specimen 0 were not significantly improved. The reason may be that only the WPU without modifier was coated on the surface of the substrate, and the formed film was not block moisture.

Table 3. Hygroscopic Expansion Parameters of Each Sheet

		Raw	0	5	6	7	8	9
S_L (%)	24h	0.29	0.28	0.26	0.24	0.06	0.05	0.15
	48h	0.44	0.39	0.26	0.42	0.35	0.10	0.39
S_V (%)	24h	0.67	0.52	0.29	0.20	0.05	0.27	0.57
	48h	0.94	0.84	0.58	0.49	0.64	0.28	0.66
ASE_L (%)	24h	-	3.45	10.34	17.24	79.31	82.76	46.94
	48h	-	11.36	40.91	4.55	20.45	77.27	11.36
ASE_V (%)	24h	-	22.39	56.72	70.14	92.54	59.70	14.93
	48h	-	10.64	38.30	47.87	31.91	70.21	29.79
MA (%)	24h	1.37	0.94	0.84	0.79	0.90	0.42	0.86
	48h	2.28	1.88	1.69	1.59	1.79	1.55	1.72
MEE (%)	24h	-	31.39	38.40	42.07	34.54	69.34	37.35
	48h	-	17.54	25.98	30.38	21.33	31.84	24.70

Water Absorption Test Analysis

Results for the specimens painted with different coatings after 24 h and 48 h water absorption are shown in Table 4. The S_L and S_V values of the specimens were smaller than those of the untreated bamboo, and the ASE_L and ASE_V also showed a similar change rule, indicating that the coating had a positive effect on the dimensional stability of the laminated bamboo. The MA and the MEE also showed a similar change rule, indicating that the coating had a positive effect on the dimensional stability of the test material. Specimen 8 had the best dimensional stability at 24 h and 48 h, and the ASE_L and the ASE_V at 48h were 35.55% and 35.68% higher than the material board, respectively. The reason may be that the UV-531 ultraviolet light absorber and boron-based flame retardant component contained in the coating can play a certain role in cross-linking and curing during the drying process of the coating, which improves its hydrophobicity. Specimens 7 and 8 also showed better linear and volumetric resistance to expansion compared with unmodified specimen 0. Surface finishing had a certain positive effect on improving the dimensional stability of the test material. The poor performance of the MA and the MEE of test material No. 0 may be due to the poor binding ability of single WPU to the surface of the test material.

In contrast to the moisture absorption test, the long-term soaking of the water absorption test easily caused the surface coating to fall off, and the water was more likely to penetrate into the interior of the test material. Based on the relevant parameters of water absorption, the introduction of flame retardant can effectively improve the waterproof performance of the test material, and the performance of test piece 8 was the best.

Table 4. Water Absorption Expansion Parameters of Each Plate

		Raw	0	5	6	7	8	9
S_L (%)	24h	2.56	1.68	1.95	2.05	1.94	1.36	2.09
	48h	3.01	2.47	2.14	0.42	2.38	1.97	2.64
S_V (%)	24h	6.71	5.07	5.58	5.64	5.05	4.64	5.76
	48h	7.54	6.44	5.90	6.47	5.55	4.85	6.65
ASE_L (%)	24h	-	34.38	23.89	20.05	24.22	46.88	18.42
	48h	-	17.93	28.80	9.33	21.05	35.55	12.44
ASE_V (%)	24h	-	24.47	16.86	15.98	24.72	30.85	14.16
	48h	-	14.59	21.77	14.17	26.38	35.68	11.74
MA (%)	24h	39.53	43.28	31.51	36.86	39.11	30.52	35.20
	48h	49.30	54.20	41.55	48.73	48.89	39.34	47.20
MEE (%)	24h	-	-9.48	20.30	6.74	1.06	22.79	10.95
	48h	-	-9.44	15.71	1.16	0.83	20.21	4.26

Fourier Transform Infrared (FT-IR) Spectra Analyses

Figure 5 shows the infrared spectrum of 6 kinds of modified water-based coatings. The peak at 3391 cm^{-1} is the N-H stretching vibration absorption peak of the amide bond in the urea group, 2954 cm^{-1} is the C-H absorption peak, 1725.0 cm^{-1} is the stretching peak of carbonyl C=O, and 1144 cm^{-1} is the stretching vibration peak of polyester C-O bond, which indicates successful introduction of WPU in the modified coating. Compared with the curve of test material 0, the other 5 coatings based on borax borate appeared CH_3 and CH_2 stretching vibration peaks at 2850 cm^{-1} and 2920 cm^{-1}, respectively, indicating the successful addition of boron substances. The peak of coating 9 at 3653 cm^{-1} is the stretching vibration of -OH, which is mainly caused by adsorption of H-O-H on the surface of TiO_2. The structure of WPU was not changed after adding flame retardant and UV light absorber.

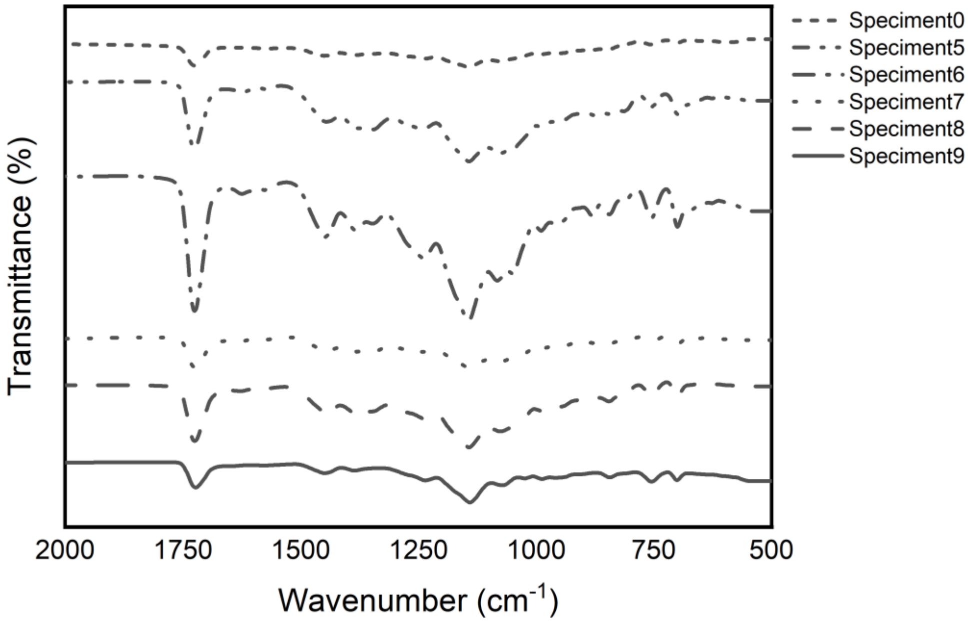

Fig. 5. Infrared spectrum of paint

CONCLUSIONS

1. The study involved the performance of adhesion tests, abrasion resistance tests, hardness tests, and temperature denaturation tests on 13 kinds of composite paint-coated specimens exposed for 3 months outdoors. The results demonstrated that the physicochemical properties of samples 0, 5, 6, 7, 8 and 9 were better.
2. The dimensional stability analysis found that each modified coating showed good moisture resistance and water resistance compared with the unmodified coating, and the dimensional stability of test material 8 was the best. The characteristic peaks displayed by the infrared test also proved the successful introduction of boron species and UV absorbers.
3. The coating composition with the best physical and chemical properties and dimensional stability was paint 8:20% WPU/BA/BX/APP/DOT/water/UV-531, BA/BX/APP/DOT/water quality ratio was 75:75:7:42:600, and the amount of UV-531 added was 1% of the coating mass.

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