Research Article

Coating Performance of Water-Based Polyurethane-Acrylate Coating on Bamboo/Bamboo Scrimber Substrates

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In this paper, a kind of high solid content water-based polyurethane-acrylate (PUA) coating was synthesized in laboratory. Bamboo, bamboo scrimber, and heat-treated bamboo scrimber were selected as substrates and coated with the PUA coating. The coating properties of those materials were investigated. The results showed that PUA coating could well cover the surface of the three kinds of materials. However, the coating performances varied among these substrates due to their different chemical and morphological surfaces. Due to the densified structures, small pores, and improved hydrophobicity of bamboo scrimber, it was difficult for PUA coating to wet and penetrate into the substrate, where the smaller contact angle and penetration depth were obtained compared with bamboo. In contrast, heat-treated one had better hydrophilicity and smaller contact angle with PUA coating than pure bamboo scrimber. Therefore, the coating performance of heat-treated bamboo scrimber was better with lower average roughness value. However, the adhesion classification between the coating and all substrates was 2 (with 5-15% cross-cut area of flaking along the edges).

1. Introduction

Bamboo is considered as an abundantly available, biodegradable, and sustainable natural resource, which is widely used in papermaking, decking, flooring, building, and so on, because of its rapid growth, good flexibility, and short production period [1–5]. The bamboo elements are composed of bamboo fibers, parenchyma cells, and vessels. It is known that the strength and modulus of bamboo fibers are much higher than others. Therefore, when load applies, the weak constitute is easy to break [6]. The performance defects of bamboo had been effectively compensated by Yu et al. [7] with a novel composite, namely, the bamboo scrimber. Compared with other bamboo composites, the bamboo scrimber has comparatively higher raw material utilization rate because the bamboo core and bamboo yellow were all utilized. The bamboo scrimber has a desirable texture, high hardness, and longitudinal strength properties [8–11].

Coating can protect and beautify bamboo and bamboo products. It is generally considered that the whole coating process can be divided into several steps, wetting of substrate, leveling, infiltrating, and drying into film [12–14]. The hydrophilicity and porosity of substrate has great impact on the coating process and coating performance [15–17]. The bamboo scrimber is a kind of high strength bamboo product formed by changing the cell structure of bamboo under the dual action of pressure and resin, which led to the densification of material, less the hydrophilic groups, and the parenchyma and fiber filled by adhesive of bamboo. For heat-treat one, improved hydrophobicity is achieved. Therefore, the changes may affect the coating performance, especially the water-based coating.

Up to present, few research focus on the coating technology of bamboo scrimber, especially the water-based coating. Polyurethane-acrylate resin (PUA) contains acrylic functional groups and urethane bonds. And its coating has high abrasion resistance, adhesion, flexibility, and excellent optical properties and weather resistance. Therefore, in this paper, bamboo, bamboo scrimber, and heat-treated bamboo scrimber were chosen as substrates and coated with a self-made water-based PUA coating. The PUA made in laboratory has more hydrophilic groups in the molecular chains, such
The reactor was heated to 40 °C and blended into a three-neck glass reactor at 150 r/min. 8 g DBTD were dissolved in the prepolyarylate chain. 100 g TDI, 200 g PEG-400, and kept for 4 h. The next step was to graft the prepolyurethane 90 wt% and viscosity of 200 mPa·s into the emulsion at 100 r/min for 10 min. The PUA prepolymer was obtained after cooling and filtration. 2% defoamer agent was added into the emulsion at 150 r/min and blended at 150 °C and kept for 4 h. The next step was to graft the prepolyurethane onto the prepolyarylate chain. 100 g TDI, 200 g PEG-400, and 8 g DBTD were dissolved in N-methyl-2-pyrrolidone solvent and blended into a three-neck glass reactor at 150 r/min. The reactor was heated to 40 °C. During stirring, 66 g HEA was dripped into the reactor within 2 h. The prepolyurethane emulsion was obtained. Afterwards, both the prepolyarylate and prepolyurethane emulsions were mixed in the three-neck glass reactor and blended at 150 r/min. The reaction was heated up to 85 °C and kept for 4 h. The PUA emulsion was obtained after cooling and filtration. 2% defoamer agent was added into the emulsion at 100 r/min for 10 min. The PUA waterborne coating was obtained with the solid content of 90 wt% and viscosity of 200 mPa·s. It was kept in a black bottle and protected against light.

2. Materials and Methods

2.1. Materials. Moso bamboo (Phyllostachys pubescens), age 3–4 years, was taken from Jian’ou Forest Reserve, Fujian province, southeast of China. The reagents for water-based polyurethane-acrylate resin (PUA) coating are listed in Table 1.

2.2. Preparation of Water-Based PUA Coating. The monomers of 24 g AA, 4 g ST, and 7 g EA with 44 g distilled water were blended in a three-neck glass reactor at 150 r/min. Afterwards, the initiator of 8 g APS was added at 10 wt%. In the next 70–90 min, the rest of 96 g AA, 16 g ST, and 133 g EA were dripped into the reactor. Afterwards, the rest of 32 g APS and 36 g KH550 was dripped into the reactor within 90–110 min. The reactor was heated up to 85 °C and kept for 4 h. The next step was to graft the prepolyurethane onto the prepolyarylate chain. 100 g TDI, 200 g PEG-400, and 8 g DBTD were dissolved in N-methyl-2-pyrrolidone solvent and blended into a three-neck glass reactor at 150 r/min. The reactor was heated to 40 °C. During stirring, 66 g HEA was dripped into the reactor within 2 h. The prepolyurethane emulsion was obtained. Afterwards, both the prepolyarylate and prepolyurethane emulsions were mixed in the three-neck glass reactor and blended at 150 r/min. The reaction was heated up to 85 °C and kept for 2 h. The PUA emulsion was obtained after cooling and filtration. 2% defoamer agent was added into the emulsion at 100 r/min for 10 min. The PUA waterborne coating was obtained with the solid content of 90 wt% and viscosity of 200 mPa·s. It was kept in a black bottle and protected against light.

2.3. Heat Treatment of Bamboo. According to actual industrial production and previous studies [18–20], the hot dry air treatment was employed on bamboo slivers. The details could be found in previous study [11], namely, the green bamboo was sawn into a bamboo tube with a length of 2600 mm and then split longitudinally into two semicircular tubes. The inner nodes were removed and the semicircular bamboo tubes were pushed into the fluffer as well as removed the wax siliceous surfaces parallel to the grain direction with a series of dotted and linear shaped cracks, thereby forming the net-structured bamboo bundles, where the diameter of bamboo fiber bundle was lower than 0.2 mm. The bamboo bundles were oven-dried at 105 °C and weighed before the thermal treatment. The dried samples were placed in an iron bucket in an oven and heated at 180 °C for 8 h. For fear of flame, the samples in the bucket were covered by sand as protecting medium. Afterwards, the samples were taken out and washed with water and oven-dried again.

2.4. Fabrication of Bamboo Scrimber. An impregnation process was employed to load phenol-formaldehyde (PF) resin. The target PF loading was 13 wt%. The bamboo bundles were immersed into the PF resin for 6 min at room temperature at PF concentration of 22 wt%. Afterwards, the bundles were taken out and leaked for about 6 min to remove excessive resin and weighed. As stated, the amounts of resin loading can be controlled by the solid content of resin and weight gains. The immersed bamboo bundles were dried in an oven at 70 °C to moisture content of 12% for manufacturing. The manufacturing of bamboo scrimber was carried out by a cold-in and cold-out process. The bamboo bundles were weighed at desired density of 1.30 g/cm³. They were assembled along the grain direction in the hot-pressing mold. When the temperature of the hot plate heated up to 60 °C, the bamboo mat was sent into the press. The pressure was kept at 3.5–70 MPa according to relative density at 140 °C for 30 min to cure the mat. Afterwards, cold water was introduced into the hot plate and cooled down to 60 °C. The pressure was released and the cured mat was removed from the press. The nominal dimension of the bamboo scrimber was 2600 × 1300 × 16 mm³. After then, all the mats were cut into required dimensions and conditioned in a room at 20 °C and 65% RH for 2 weeks for further tests.

Table 1: Reagents for water-based PUA coating.

<table>
<thead>
<tr>
<th>Reagent</th>
<th>Type</th>
<th>Abbreviation</th>
<th>Manufacturer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ammonium persulfate</td>
<td>Initiator</td>
<td>APS</td>
<td>Xilong Chemical Co. Ltd. (Guangzhou, China)</td>
</tr>
<tr>
<td>Acrylate acid</td>
<td>Monomer</td>
<td>AA</td>
<td>As above</td>
</tr>
<tr>
<td>Styrene</td>
<td>Monomer</td>
<td>ST</td>
<td>As above</td>
</tr>
<tr>
<td>Ethyl acrylate</td>
<td>Monomer</td>
<td>EA</td>
<td>As above</td>
</tr>
<tr>
<td>Hydroxyethyl acrylate</td>
<td>Monomer</td>
<td>HEA</td>
<td>As above</td>
</tr>
<tr>
<td>Toluene-2,4-diisocyanate</td>
<td>Monomer</td>
<td>TDI</td>
<td>As above</td>
</tr>
<tr>
<td>Polyethylene glycol (Mn=400)</td>
<td>Monomer</td>
<td>PEG-400</td>
<td>As above</td>
</tr>
<tr>
<td>Dibutyltin dilaurate</td>
<td>Catalyst</td>
<td>DBTD</td>
<td>As above</td>
</tr>
<tr>
<td>γ-aminopropytriethoxysilane</td>
<td>Silane</td>
<td>KH550</td>
<td>Qufu Chengguang Chemical Co., Ltd (Shandong, China)</td>
</tr>
<tr>
<td>Polyether siloxane copolymer composition</td>
<td>Defoamer agent</td>
<td>/</td>
<td>Tianjin Jinke Fine Chemical Institute (Tianjin, China).</td>
</tr>
<tr>
<td>N-methyl-2-pyrrolidone</td>
<td>Solvent</td>
<td>–</td>
<td>As above</td>
</tr>
<tr>
<td>2-hydroxy-methylphenypropan-1-one</td>
<td>Photoinitiator</td>
<td>/</td>
<td>As above</td>
</tr>
</tbody>
</table>

as carboxyl, and the PUA resin can dissolve well in water and different solid content PUA coatings can be obtained. The coating properties and the different mechanism among those materials were investigated, which aimed to get effective method for better process conditions of water-based coating for bamboo scrimber.

Table 1: Reagents for water-based PUA coating.
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2.5. Coating of Bamboo and Bamboo Scrimber. The moso bamboo, bamboo scrimber (BS), and heat-treated bamboo scrimber (HBS) were coated by semiautomatic roll coater (BGD 218, BIUGED instruments, China) according to ISO standard 15528: 2011 and the relative film thickness requirement. In theory, the dry film thickness is 18 μm. The film sample was then dried in a cool, dry, dark environment for 1 week before use [21].

2.6. Characterizations and Tests. The characterizations of the samples were carried out by scanning electron microscope coupled with energy dispersed X-ray analysis, atomic force microscopy (AFM), Fourier transform infrared spectroscopy equipped with an attenuated total reflection (ATR-FTIR), and X-ray photoelectron spectra (XPS) analyses [21].

The microstructures of the above samples were observed by scanning electron microscope (JEOL JSM-6301F, Japan) with an acceleration voltage of 20 kV. The samples were sputter-coated with gold. Energy dispersed X-ray analysis (EDXA, Horiab 7021-H, Japan) was carried out in mapping mode. The images and distributions of elements N which were mainly from PUA resin were captured digitally to allow for further analysis of impregnation of PUA in each substrate.

The surface morphologies were captured by AFM and collected by a scanning microscope in contact mode.

The chemical groups of the samples were examined by ATR-FTIR (BRUKER Vertex 70v, Germany).

XPS analysis of the specimens were performed on Escalab 250Xi (Thermo Scientific, USA) to determine the number of functional groups present on the surface, with a pass energy of 10 eV and nonmonochromatic MgKα and AlKα X-radiations (hv=1253.7 eV and 1486.7 eV, respectively). A current of 10 mA and a voltage of 13 kV were used. The survey scans were collected from the binding energy range of 0 to 1350 eV. The layers were mounted onto a holder with double-sided adhesive tape and placed in vacuum ranging from 1.33 × 10⁻⁶ to 1.33 × 10⁻⁵ Pa. The test sample area was 10 × 10 mm² and the atomic percentages of the elements present were derived from the spectra in this area.

The contact angle of those specimens was measured according to ISO 15989: 2004 by contact angle meter (ISO, 2004; Krüss K11MK4, Germany), when the test solutions (polyurethane-acrylate coating) were diluted to 70 wt%.

The adhesion classification of coating films was measured according to ISO 2409: 2013 standard methods by a cross-cut test. Classification 0: the edges of the cuts are completely smooth; none of the squares of the lattice is detached. Classification 1: detachment of small flakes of the coating at the intersections of the cuts. A cross-cut area not greater than 5% is affected. Classification 2: the coating has flaked along the edges and/or at the intersections of the cuts. A cross-cut area greater than 5%, but not greater than 15%, is affected. Classification 3: the coating has flaked along the edges of the cuts partly or wholly in large ribbons, and/or it has flaked partly or wholly on different parts of the squares. A cross-cut area greater than 15%, but not greater than 35%, is affected. Classification 4: the coating has flaked along the edges of the cuts in large ribbons and/or some squares have detached partly or wholly. A cross-cut area greater than 35% but not greater than 65% is affected. Classification 5: any degree of flaking that cannot even be classified by classification 4.

3. Results and Discussion

3.1. Coating Morphologies. The surface morphologies of each sample before and after coating are shown in Figure 1. The figures showed that the microstructure of each substrate was clearly visible before coating, including dense fibrous tissue and looser parenchyma cells. After the recombinant processing, the parenchyma cells of bamboo were destroyed and the cell structure became dense. The surface of the HBS became flatter than BS, which indicated that the permeability and distribution uniformity of the phenolic resin in bamboo were improved by the heat treatment.
The surface of bamboo and HBS, both coated by polyurethane-acrylate, were well covered by the coatings and the flat coating film achieved, though there were a small amount of micronanoparticles. It might be the coating particles and bubbles, which were generated during the coating penetration into the interior of the substrate during the curing process of the coating film. In case of HS, some bamboo fibers were exposed on the surface, which led to some defects such as bottom leakage. Due to the high density of the BS, weak penetration into the interior of substrate occurred. On the other hand, the free energy of the BS surface was high, which made the coating not evenly spread out. Hence, an agglomeration of the coating film was got after curing.

In order to further analyze the permeability of the coating in bamboo, BS, and HBS, the coated samples were investigated by N-element spectrum scanning by EDXA. The results are shown in Figure 2. The figure showed that most PUA distributed on the surface of the substrate, and just a small amount penetrated into the substrate. On the surface of bamboo, the penetration depth of PUA was about 4 μm, while on the surface of BS, the penetration depth decreased to 2 μm, which indicated that it was difficult for the PUA resin penetrating into the substrate. It was because that the cells tissue was compacted after the substrate was disintegrated and reorganized. Interestingly, the most penetration depth of PUA resin in HBS increased to 4-5 μm, with partly of 7 μm, which might because of the enlarged pores of HBS during the heat treatment.

The AFM 3D images of surface topography of coated samples are shown in Figure 3. It could be seen that the bamboo, BS, and HBS were well covered by the PUA coating. Smooth surface was obtained, with little defects. Very tiny flow marks, formed on bamboo during curing, were observed. The surface roughness value (Rq:Ra) of coated bamboo was 1.35. The coating film on the BS surface was rough and the average roughness value increased to 1.41. Compared with the others, the surface of the HBS was the smoothest with the lowest roughness value of 1.24, which indicated more uniform distribution of coating on the surface of the HBS was obtained.

3.2. Chemical Characterizations. The ATR-FTIR results of the specimens, including coated and uncoated bamboo, coated and uncoated BS, and coated and uncoated HBS are shown in Figure 4. In Figure 4(a), changes of chemical group peaks were found between raw substrates and coated samples, indicating that there were chemical reactions. However, the results between BS and HBS were not significant. Compared with bamboo, the absorption peaks of –OH at 3420 cm\(^{-1}\) of BS and HBS were both obviously weakened, which indicated that the hydrophilicity of BS and HBS decreased. The reason should be explained by the fact that the fiber surface of BS and HBS was well covered by phenolic resin (PF), which was hydrophobic than bamboo fibers.[22]

The absorption peak of C=O at 1737 cm\(^{-1}\) was the characteristic peak of hemicellulose. And in case of BS and HBS, this peak almost disappeared, which indicated that the hemicellulose and polysaccharide of bamboo degraded during curing. In addition, the C-O absorption peak at 1043 cm\(^{-1}\) increased, and the C-O vibration peak of ester bond at 1265 cm\(^{-1}\) shifted to 1240 cm\(^{-1}\), which indicated that aromatic ester bonds and ether bonds formed between phenolic resin and cellulose. The skeleton stretching vibration
peak of lignin aromatic benzene ring C=C at 1630 cm\(^{-1}\) of BS and HBS shifted to 1607 cm\(^{-1}\), which was due to the side chain substitution on the lignin benzene ring during heating. All these results suggested new chemical bond was formed in BS and HBS compared with neat bamboo.

Figure 4(b) shows the results of the PUA coated bamboo and BS/HBS samples. Compared with the pure polyurethane-acrylate sample directly dried on the glass plate, the spectrum was basically the same except for the stretching vibration peak of -OH at 3375 cm\(^{-1}\), which was shifted to 3414 cm\(^{-1}\). It was mainly due to the fact that the hydrogen bond formed between the PUA and the -OH of bamboo or BS/HBS, indicating that the PUA coating was well covered on the surface of bamboo, BS, and HBS.

To further analyze the surface chemical changes of these materials, the XPS analysis was carried out. Table 2 showed the percentage results of XPS elements of each sample before and after coated. The atomic percentages of C, O, and N in bamboo were 70.35%, 26.63%, and 3.02%, respectively. After fabricated into bamboo scrimber, the C element increased to 78.37%, while the O element decreased and the N content remained basically. The reason may be associated with the PF resin containing higher C which was added into the bamboo scrimber. The C content of the HBS further increased compared with the BS, and the O element further decreased. It was because heat treatment could degrade the -OH of bamboo, which led to a decrease in the bamboo hydrophilicity. Compared with uncoated bamboo, the quantity of C, O, and N element of coated samples changed, with the percentages of 73.47%, 20.43%, and 4.81%, respectively. Due to PUA coating containing much C and N elements, the content of C and N increased, and the content of O decreased, compared with uncoated sample. The C element of coated BS changed slightly while the N content significantly increased, compared to those of coated bamboo. It was because the PF resin in the reconstituted bamboo could react with the ester groups of the urethane-acrylate, so more amino groups were exposed on the surface of the sample.

In contrast, the C content of the coated HBS increased, while the O content decreased, and the N content changed little. It was because the heat-treatment process reduced the surface content of hydroxyl groups on the bamboo scrimber, simultaneously expanding the substrate porosity. Thus the PUA penetrated inside. Therefore, the PUA coating was well distributed on the substrate surface and was more likely to permeate into the substrate interior. Therefore, differences among each substrate were not significant. In general, HBS had better coating performance than that of BS.

In order to further analyze the binding of C\(_{1s}\) in each material, the four peaks were observed, including C1(C-C/C-H), C2(C-O/C-OH), C3(O-C-O/C=O), and C4(O-C=O) [23]. The results are shown in Figure 5 and Table 3. It could be seen from Table 2 that there was no C4 peak except the bamboo scrimber coated by PUA. Although PUA contained some ester bonds, it penetrated well into bamboo and HBS and evenly covered the surface of the substrate. Thus, C4 peak was not detected. On the contrary, it was difficult for PUA to penetrate into bamboo scrimber, which, aggregated on the surface, so C4 peak was detected. The proportions of C1, C2, and C3 of bamboo were 54.82%, 34.78% and 10.40%, respectively. In contrast, the proportion of C1 of BS increased.

Table 2: Elements percentages of bamboo, BS, HBS, and their coated samples.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Percentage (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>C</td>
</tr>
<tr>
<td>Bamboo</td>
<td>70.35%</td>
</tr>
<tr>
<td>BS</td>
<td>78.37%</td>
</tr>
<tr>
<td>HBS</td>
<td>81.91%</td>
</tr>
<tr>
<td>PUA coated bamboo</td>
<td>73.47%</td>
</tr>
<tr>
<td>PUA coated BS</td>
<td>74.21%</td>
</tr>
<tr>
<td>PUA coated HBS</td>
<td>81.44%</td>
</tr>
</tbody>
</table>

Figure 4: ATR-FTIR spectra of bamboo, BS, HBS (a), and their coated samples (b).
Counts (s)

Binding energy (eV)

Counts (s)

Binding energy (eV)

Counts (s)

Binding energy (eV)

Counts (s)

Binding energy (eV)

Figure 5: C\textsubscript{1s} peak spectra of bamboo, BS, HBS, and their coated samples. (a) Bamboo; (b) BS; (c) HBS; (d) PUA coated bamboo; (e) PUA coated BS; (f) PUA coated HBS.

Figure 6: Contact angle of bamboo, BS, and HBS with 70% PUA drop.

significantly and the proportion of C\textsubscript{2} decreased. The reason should be that the PF resin, which contained a large amount of C-H, was added into the BS, resulting in the increase of C\textsubscript{1} ratio. After heat treatment, the C\textsubscript{1} ratio of BS continued to increase, and the C\textsubscript{2} ratio decreased, which was attributed to the degradation of the hydroxyl groups by heat treatment [24].

3.3. Contact Angle between PUA and the Substrates. It was known that the contact angle size depends on the hydrophilicity of the substrate and the distribution of the phenolic resin adhesive on the surface of the substrate when coated by the same PUA [25]. Figure 6 showed the contact angle of the PUA droplets (70 wt% solid) on the surface of the three kinds of materials, including bamboo, BS, and HBS. From the figures, the contact angle of PUA on the surface of the BS was the largest of 31.38°, which showed that the compatibility between PUA and BS was poor, and the droplets were difficult to spread on the BS surface. Therefore, the coating film, with poor surface quality, was obtained, due to the resin particle agglomeration and protrusions during the drying process. There was little difference of the contact

Table 3: Elements percentages of bamboo, BS, HBS, and their coated samples.

<table>
<thead>
<tr>
<th>Sample</th>
<th>C\textsubscript{1} (%)</th>
<th>C\textsubscript{2} (%)</th>
<th>C\textsubscript{3} (%)</th>
<th>C\textsubscript{4} (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bamboo</td>
<td>54.82</td>
<td>34.78</td>
<td>10.40</td>
<td>0</td>
</tr>
<tr>
<td>BS</td>
<td>72.51</td>
<td>19.35</td>
<td>8.14</td>
<td>0</td>
</tr>
<tr>
<td>HBS</td>
<td>80.04</td>
<td>14.63</td>
<td>5.33</td>
<td>0</td>
</tr>
<tr>
<td>PUA coated bamboo</td>
<td>68.81</td>
<td>18.96</td>
<td>12.23</td>
<td>0</td>
</tr>
<tr>
<td>PUA coated BS</td>
<td>66.09</td>
<td>22.42</td>
<td>7.58</td>
<td>3.91</td>
</tr>
<tr>
<td>PUA coated HBS</td>
<td>74.69</td>
<td>15.35</td>
<td>9.96</td>
<td>0</td>
</tr>
</tbody>
</table>
angles of PUA droplets on the surface of bamboo and HBS. Therefore, the wetting effect of them was fairly good [26].

3.4. Adhesion of Coating Films. The images of PUA coated bamboo, BS, and HBS after cross-cut tests are shown in Figure 7. It can be seen that the adhesion classification of the PUA coating film of the three types of substrate was all 2. For each coating film, the coating had flaked along the edges and at the intersections of the cuts. A cross-cut area greater than 5%, but not greater than 15%, was affected. Thus, the PUA adhesion of the three types of materials needs to be improved. In the further research, the substrate surface needs to be modified to increase the adhesion of coating film.

4. Conclusions

The coating substrate affected the resulting coating performance of PUA resin due to the different physical and chemical surface properties of the substrate. Bamboo had the fairly good coating performance. However, because of the densified structures and impregnation of phenolic resin, the coating performance of bamboo scrimber by water-based PUA coating was poor. Heat treatment can improve the performance to a certain extent owing to the decreased hydrophilicity and enlarged micropores. The adhesion classifications of the PUA coating film on the three types of substrate were all classification 2. Therefore, the next step was to modify the surface of the substrate or develop a new kind of water-based coating to further improve the coating performance of bamboo scrimber.

Data Availability

The data used to support the findings of this study have not been made available because it is part of a research program of the National Key Research and Development Program of China, and it will be further studied.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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