UV-Assisted Multiscale Superhydrophobic Wood Resisting Surface Contamination and Failure

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ABSTRACT: In the modern forestry, the demand for renewable and environmentally friendly wood protection is increasing. This paper reports a green method for preparing stable and self-cleaning superhydrophobic coating for wood protection by dripping polyvinyl alcohol cross-linked hollow silica nanoparticles on the surface of wood in combination with polydimethylsiloxane modification. The coating is based on a laminated structure with layers stacked on the surface of the wood and cured quickly with the assistance of UV. The coatings obtained on wood substrates with appropriate ratios have excellent superhydrophobic properties, with an optimum water contact angle of up to 160.4 ± 0.2°. The coating also exhibits good transparency in the UV−visible spectrum and a maximum transmittance of 91%. With transmission electron microscopy, the microscopic morphology of the self-assembled hollow silica nanoparticles was observed. Scanning electron microscopy, Fourier transform infrared spectroscopy, and X-ray diffraction were also applied to investigate the morphology and chemical composition of the coatings. A water contact angle of 151.5 ± 0.7° was maintained even after the abrasion tests with sandpaper at a distance of 300 cm. Meanwhile, the resultant coatings exhibit good self-cleaning properties apart from mechanical durability and chemical stability, which enables effective resistance to contamination. Evidenced by the abovementioned data, this composite coating is capable of optimizing the surface wettability of wood, offering a new dimension to the extensive and prolonged application of wood and wood-based products. Furthermore, considering the advantages of this method, it could also be used in other areas in the future, such as glass, solar substrates, and optical devices.

INTRODUCTION

Wood plays an essential role in carbon neutral actions and can have a long service life if it is adequately protected to minimize its negative environmental impact. Natural wood is highly hygroscopic due to its porous, layered structure and the presence of a large number of hydrophilic groups, resulting in wood products being susceptible to decay, deformation, and cracking, which severely limits the widespread use of wood as a green and sustainable material.1−3 The affinity of wood for moisture is attributed to the hydroxyl-containing cellulose and hemicellulose components. In the plight of the shortage of high-quality wood resources and the shortage of supply in the wood market, it is imperative to break through the expansion and efficient utilization of wood resources.4 The improvement of dimensional deformation, discoloration, and decay of the wood will not only broaden the areas of its application but also prolong its service life.5−7 The modification of superhydrophobic properties on wood surfaces has attracted widespread research interest in recent years due to the enormous potential of superhydrophobic surfaces in areas such as self-cleaning materials, corrosion protection, and wood protection. The droplets remain almost spherical on biological surfaces with superhydrophobic properties, such as cicada wings, butterfly wings, and lotus leaves, from which they can easily roll off and carry away stains.8−11 The biomimetic
modification of wood not only provides the basis for the development and utilization of wood-based functionalized materials but also enables the construction of new multifunctional wood materials. Based on these previous research studies, it is known that two parameters are essential for designing superhydrophobic surfaces on wood: (1) low surface energy and (2) micro/nanoscale roughness of the substrate surface, which reduces the adhesion between the solid surface and the liquid droplets.\textsuperscript{12−14} In light of this finding, a number of related methods for fabricating superhydrophobic surfaces have been reported, such as sol−gel, mussel-inspired, hydrothermal, and plasma etching methods.\textsuperscript{15−22} The natural microgroove structure of wood, including roof-like raised cell walls and grooved cell cavities, provides the prerequisites for the design of superhydrophobic surfaces. Therefore, the fundamental constructing strategy for superhydrophobic coatings on wood surfaces is to introduce micro/nanoscale rough structures and modify them in combination with low-surface-energy materials.

In recent years, many advances have been made in the study of superhydrophobic wood surfaces,\textsuperscript{23−27} where functional coatings are attracting more and more interest, as these properties could be used in more cost-effective and environmentally friendly situations. Yue et al.\textsuperscript{28} fabricated a novel room temperature-vulcanized silicone rubber (RTVSR) superhydrophobic coating on the wood surface with the aid of SiO\textsubscript{2} nanoparticles. This RTVSR/SiO\textsubscript{2} superhydrophobic coating could serve well on hydroxyl-containing substrates, especially wood, in the outdoor environment. Liu et al.\textsuperscript{29} constructed an epoxy resin/SiO\textsubscript{2} composite superhydrophobic coating on the surface of wood. The composite superhydrophobic coating exhibited good mechanical stability, which was mainly attributed to the epoxy resin firmly adhering the SiO\textsubscript{2} nanoparticles to the wood surface. Wu et al.\textsuperscript{6} successfully applied inorganic nanoparticles and vinyltriethoxysilane to construct robust organic/inorganic composite superhydrophobic coatings on the surfaces of various cellulosic materials such as wood, bamboo, filter paper, and cotton, which exhibited good superhydrophobic properties after a series of mechanical abrasion tests such as sandpaper rubbing, blade cutting, and finger scraping. In order to further improve the mechanical robustness of the superhydrophobic coatings, Tu et al.\textsuperscript{7} proposed the construction of SiO\textsubscript{2}/epoxy/FAS composite superhydrophobic films by first prepping the wood substrate with a transparent epoxy resin base layer to cover the natural microgroove structure of the wood surface, thus ensuring a consistent and stable superhydrophobic performance of the coatings. Despite some innovations in the construction of superhydrophobic surfaces on wood substrates, there are challenges in the preparation of these methods, such as the inability of large-scale production, the complexity of the procedure, and the high costs.\textsuperscript{30−32} Thus, one of the important aspects of current research into the functionalization of wood is to design facile ways to fabricate stable and robust superhydrophobic surfaces on wood.

Polymers such as polyvinyl alcohol (PVA), epoxy resins, or polydopamine are often chosen as adhesives in order to strengthen the interfacial interaction between the nanoparticles and the substrate.\textsuperscript{33−37} PVA is a polyhydroxy substance with poor mechanical properties and excellent and inexpensive adhesive properties; however, PVA composites demonstrate good mechanical strength. Polydimethylsiloxane (PDMS) is a reagent commonly used in the fabrication of superhydrophobic surfaces with low variation with temperature, low surface tension, high transparency, good weather resistance, and high hydrophobicity. In this work, a hybrid coating of hollow SiO\textsubscript{2}/PVA was prepared and coated onto the wood substrate, combined with UV-assisted rapid curing, and then modified by PDMS on the wood surface to prepare the superhydrophobic surface. We also expect that this method could be widely used for other wood-based materials or substrates with multifunction integration, in pursuit of the sustainable utilization, longevity, and high performance.

### RESULTS AND DISCUSSION

A detailed process of fabricating a superhydrophobic coating on wood can be seen in Figure 1. Briefly, the mixed coating solution was applied to the wood substrate using a dripping method, cured under UV irradiation, and then stored at room temperature. The dried samples were immersed in PDMS/\textsubscript{n}-hexane solution and finally dried on the wood substrate to obtain a superhydrophobic coating. The group with 0.2 wt % HSNPs was recorded as PVA/HS-PDMS (PHP0.2) wood, the group with 0.5 wt % HSNPs was recorded as PVA/HS-PDMS (PHP0.5) wood, the group with 1 wt % HSNPs was recorded as PVA/HS-PDMS (PHP1) wood, the group with only the PDMS group was recorded as PDMS wood, and the group

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**Figure 1.** Schematic illustration for the fabrication of superhydrophobic coating on the wood substrate.
with 0.5 wt % HSNPs containing no PDMS was recorded as PVA/HS wood.

**Microstructure and Morphology.** Typical SEM images of the wood surface are shown in Figure 2. The scanning electron microscopy (SEM) images of the natural wood samples clearly illustrate that there are many natural micro-grooves and gully structures, which are mainly made up of roof-like raised cell walls and groove-like cell cavities. Since wood specimens were treated with PDMS, it was obvious that the surface of the treated wood became smoother and some of the original groove structures were covered. It is not difficult to find from the figure that we have succeeded in introducing nanoparticles on the surface of the wood samples, which covered some of the original microgrooves and retained the microstructure we were interested in. On this basis, a nanoscale roughness was established, which created conditions for the formation of superhydrophobic coatings.28,38 As illustrated in Figure 2C–F, the surface of the substrate was almost entirely coated with particles. These silica nanoparticles adhered to the surface of the wood, and the cross-linked agglomeration of the nanoparticles presented a porous sponge shape on the surface.

Transmission electron microscopy (TEM, JEM-2100, Japan) was carried out to observe the morphology of hollow silica nanoparticles (HSNPs). The TEM images of the nanoparticles at different magnifications are shown in Figure 2J–M. It is clear from the figures that HSNPs were successfully prepared in this work and the prepared nanoparticles were mono-disperse with an average particle size of around 20–70 nm.

On closer inspection of the PHP0.5 wood surface in Figure 2G–I, it is easy to observe the beautiful nanoparticles deposited on the wood surface. The sample surface was completely blanketed with particles and formed a microbump structure with a large surface roughness. Meanwhile, it is worth noting that in these TEM images at high magnification, we have successfully prepared silica nanoparticles of approximately 63 nm in size. These nanoparticles exhibited the status of cross-linking and aggregation on the wood surface, which could be explained by the fact that PVA acted as an effective adhesion agent. On the one hand, it helps the bonding between the nanoparticles and the wood substrate, and on the...
FTIR spectroscopy confirms the successful attachment of PDMS and silica to the surface of the wood. The peak at 3325 cm\(^{-1}\) corresponds to the antisymmetric and symmetric stretching vibrations of the hydroxyl groups on the cell walls in the wood. A stretching vibration peak of methyl at 2958 cm\(^{-1}\) and a bending vibration peak of Si–C at 1251 cm\(^{-1}\) indicated that PDMS has been effectively coated onto the wood substrate. The Si–O–Si antisymmetric stretching vibration near 1010 cm\(^{-1}\) and the Si–O–Si symmetric stretching vibration at 802 cm\(^{-1}\) show absorption peaks with higher intensities, indicating the presence of a large number of Si–O–Si functional groups in the coated wood. The characteristic peak intensity of Si–O–Si in the infrared spectrum of PHP0.5 wood is significantly higher than that of PDMS wood and natural wood. This phenomenon is mainly due to the large amount of Si–O–Si contained in SiO\(_2\) nanoparticles. In conclusion, it can be known that PDMS and SiO\(_2\) nanoparticles have been successfully coated on the wood surface through the stretching vibration of the functional group in the infrared spectrum.

Table 1. Weight Percentage of Surface Elements before and after Treatment

| groups    | elements | weight percentage/% | atomic percentage/% |
|-----------|----------|---------------------|---------------------|
| natural wood | C        | 51.92               | 58.99               |
|           | O        | 48.08               | 41.01               |
| PDMS wood | C        | 47.99               | 55.81               |
|           | O        | 48.78               | 42.58               |
|           | Si       | 3.24                | 1.61                |
| PHP0.5 wood | C        | 24.58               | 35.05               |
|           | O        | 41.15               | 44.05               |
|           | Si       | 34.26               | 20.89               |

This table uniformly illustrates that PDMS and SiO\(_2\) have been coated on the wood surface and that the modified coating has not significantly affected the crystalline properties of the cellulose on the wood surface.

Wettability. The wettability of the natural wood and the coated wood is shown in Figure 5A–F. It can be clearly seen in the graph that the water contact angle (WCA) of the composite coating varies from 148.9 ± 0.3° to 160.4 ± 0.2° and 150.6 ± 0.6°, respectively, as the content of HSNPs changes from 0.2 to 0.5 and 1 wt %. The improvement in...
hydrophobicity can be attributed mainly to the increased surface roughness, the microgrooves formed by more silica nanoparticles cross-linked together to trap more air, and the lower surface energy created by the PDMS overlay. These two factors succeeded in minimizing the contact area between the water droplets and the composite coating. The hydrophobicity of the coated sample surface can be effectively improved by introducing different levels of nanoscale hollow silica particles. The maximum contact angle of the fabricated composite coating is 160.4 ± 0.2°, when the content of HSNPs reaches 0.5 wt %, as shown in the figure. While the WCA of the PHP wood is higher, the superhydrophobic effect is superior to natural wood, PDMS wood, and PVA/HS wood. This result can be explained by the fact that the appropriate ratio of PVA and HSNPs helps the nanoparticles to adhere to the wood surface in a homogeneous dispersion forming a suitable layered structure and microgroove structure in order to trap more air.42 This structure is also preferable to a single arrangement, which results in a smaller contact area between the water droplets and the surface. The figure also reveals that the WCA of the composite coating is decreased when the nanoparticle fill content is either less or more, and this result indicates that the appropriate ratio can effectively improve the hydrophobic properties of the coating. The addition of HSNPs can increase the roughness of the wood surface, which in turn displays good hydrophobicity. However, the agglomeration of HSNPs affects the material properties when the roughness reaches a certain level, which explains why the additional addition of HSNPs can no longer improve the WCA. Therefore, the coated wood mentioned in the next characterization tests in this paper refers only to coated wood with an HSNP content of 0.5 wt %.

The adhesion behavior of the composite coating is investigated by separating 5 μL water droplets from the surface of the coating. The dynamics of the water droplets approaching, contacting, deforming, and leaving the coating were recorded, as shown in Figure 5G–L. It is observed that the water droplets easily depart from the coating and remain spherical even when they are compressed onto the superhydrophobic surface, indicating that the composite coating has significantly low water adhesion.

Self-Cleaning and Antifouling Properties. The self-cleaning surface refers to the use of air, sunlight, rain, and other automatic means of removing surface stains, such as the surface of a lotus leaf.43 The principle is that when the water droplets...
slide or roll freely on the surface of the material, the water droplets can adsorb contaminants on the substrate and remove them during the rolling process, achieving a self-cleaning effect on the surface of the material.

Under outdoor exposure in real applications, super-hydrophobic surfaces are often contaminated by dust or particles. As shown in Figure 6A, the coated wood samples were placed obliquely, and the self-cleaning performance was investigated by dripping water onto the surface of the wood sprinkled with blue chalk dust. Moreover, as expected, the water droplets rolled off the coated wood surface quickly and took away the chalk dust in the rolling path, effectively demonstrating the coating’s self-cleaning ability and antifouling properties. On the surface of natural wood, it can be clearly observed that chalk dust is dissolved by water and attached to the wood, resulting in the accumulation of contaminants and further pollution of the surface. As the composite coating provides a multilevel micro/nanosurface structure, there is a rich air layer between the coating and the water droplets. Thus, the contact surface between the dust and the coating can be reduced, and the water droplets can easily carry away the dust from the coating surface.

To evaluate the antifouling properties, different drops of liquid are placed on top of the composite coating and the natural wood. Figure 6B,C demonstrates that after the different drops have been applied to the surface, the surface of the composite coating remains clean, while the natural wood is saturated with droplets, indicating that the coating has good antifouling properties. Once immersed in water, as shown in Figure 6D, a mirror-like surface is observed on the coating due to the strong light reflection from the air cushions trapped in the holes on the superhydrophobic surface. According to the Cassie–Baxter theory, the micro/nanostructure of the coating can well create a stable air layer which has the ability to resist the muddy water mixture and remain clean. This series of optical pictures jointly confirmed the excellent superhydrophobic and self-cleaning properties of the composite coating, which can facilitate a durable application in harsh environments.

Transparency. The well-balanced optical transparency can be explained by the high transmission of light in the visible spectrum, which is verified by UV–vis transmission spectroscopy. The transparency of the PHP0.5 coating was studied with UV–vis spectroscopy. It is observed that in the UV–vis spectrogram at 400–800 nm, the visible spectrum transmission of the coating is approximately 51–91% and the absorption of the visible spectrum is roughly 0.27–0.03 (Figure 7). Noted, where the transmission could reach a maximum of 91% and the average transmission was 71%. The conclusion can be drawn that the coatings prepared in this study exhibit good visible light transmission. This result can be interpreted together with the SEM images to be that the surface roughness below 100 nm reduces the refraction at the interface between the air and the surface of the coatings, thus effectively improving the optical transparency of the coating.

Mechanical Durability and Chemical Stability. Stability and durability are critical factors to be considered in the practical application of superhydrophobic coatings. The micro/nanostructure of bionic superhydrophobic surfaces is prone to failure under external damage and chemical attack. Here, abrasion and chemical solution corrosion tests have been carried out. The mechanical durability of the composite coating was assessed by the sandpaper abrasion test. As illustrated in Figure 8A, a weight (50 g) is placed on the surface of the wood dragging it at a constant speed on 1500 grit metallographic sandpaper and recording the change in contact angle against the increase in abrasion length.
Figure 8B presents the trend of the WCA as the abrasion length increases. It is clear from the results that the wood sample remains superhydrophobic with a WCA of 151.5 ± 0.7° after a distance of 300 cm. This is primarily attributable to the facts that the layered structure formed by the nanoparticles cross-linked and adhesion to the wood surface is stronger than a single arrangement.39 Thus, during the sandpaper abrasion experiment, some of the less firmly bonded nanoparticles were dislodged during the abrasion but still retained the surface microstructure. However, the superhydrophobicity of the composite coating decreases after 300 cm distance yet remains superhydrophobic, which can be explained by the fact that the low-surface-energy material wrapped around the composite coating is destroyed and part of the nanoparticles are shed under prolonged abrasion.

Chemical stability is evaluated by exposure to acidic, alkaline, and salt solution. The stability of the coating in different chemical environments is investigated by dropping 10 μL of 1 M acidic (HCl), alkaline (NaOH), and salt (NaCl) solution onto the composite coating surface. As can be seen from Figure 8C, the droplets remain well hydrophobic on the surface of the coated sample after 1 h, suggesting that the composite coating exhibits excellent resistance to acids, alkalis, and salts.

The abovementioned results reveal that the superhydrophobic coating in this study has outstanding mechanical durability and chemical stability, which can be applied in many fields.

### CONCLUSIONS

In this paper, we designed and synthesized HSNPs, which were bonded to wood substrates by PVA and were modified with PDMS to fabricate superhydrophobic coatings on the wood surface. The prepared superhydrophobic coating is based on a sandwich structure stacked layer by layer on the wood substrate, which can be constructed by facile UV-assisted curing, dripping, or spraying methods. The coatings obtained on the wood substrates with appropriate ratios have excellent superhydrophobic properties, with an optimum WCA of up to 160.4 ± 0.2°. The coating retains excellent wettability under sandpaper abrasion, acidic solution (1 M HCl), alkaline solution (1 M NaOH), and salt solution (1 M NaCl). The superhydrophobic coating shows outstanding transparency in the UV–visible spectrum with a maximum transmission of up to 91%. In addition to its mechanical durability, chemical stability, and good transparency, the coating also exhibits good self-cleaning properties, which are attributable to the special micro–nanostructure that reduces the contact between chalk dust and the surface. On the one hand, the use of this superhydrophobic coating on the wood surface improves the wettability of wood, expands the application scope of wood, and prolongs the service life of it. On the other hand, beyond the use on wood, this simple and effective method is very promising for various practical applications, such as optical devices, glass, and solar substrates.

### EXPERIMENTAL SECTION

#### Materials.
Balsa wood, purchased from Shanghai, China, was cut into 20 mm (longitudinal) × 20 mm (tangential) × 10 mm (radial). All wood samples were selected without knots, cracks, or other obvious defects. Tetraethyl orthosilicate (TEOS) (98%), ammonium hydroxide solution (25 wt %), poly(acrylic acid) (PAA), ethanol anhydrous (99.7%), and deionized water were purchased from Harbin. PDMS (SYLGARD 184A) and the curing agent (SYLGARD 184B) were purchased from Dow Corning Corporation. N,N′-Methylenebis(acrylamide) (photo-cross-linker), PVA (M_w ~ 27,000), n-hexane (97%), and 2-hydroxy-4′-(2-hydroxyethoxy)-2-methylpropionophenone (2959, photoinitiator) were purchased from MacLean. All chemicals are analytically pure without further purification.

#### Synthesis of HSNPs.
Ethanol anhydrous, ammonium hydroxide solution, TEOS, and PAA were used to prepare HSNPs via a method reported by Wang and Yu. First, 0.7 mL of PAA dissolved in 9 mL of ammonium hydroxide solution was mixed with 180 mL of ethanol anhydrous in a 500 mL Erlenmeyer flask, followed by 4.5 mL of TEOS being dropped within 2 h under vigorous magnetic stirring. The solution was placed at room temperature for 12 h and then centrifuged at 10,000 rpm at 22 °C for 12 min. The supernatant was discarded and retained the solid product. The solid particles were washed with ethanol several times,
then cleaned with deionized water, and dried in a vacuum drying oven at 80 °C for 6 h to obtain HSNPs.

Preparation of Superhydrophobic Coatings on the Wood Substrate. The wood veneer samples were ultrasonically rinsed in ethanol for 30 min and dried at 80 °C in a vacuum drying oven. First, 0.2, 0.5, and 1 wt % nanoparticles, 4 g of PVA, and 0.2 g of a photo-cross-linker were added to 25 mL of deionized water and magnetically stirred at 90 °C for 30 min, and then, 0.1 g of the photoinitiator was added and ultrasonically dispersed at ambient temperature for 30 min. The mixed coating was applied to the wood substrate using a dripping method, cured under UV irradiation for 10 min, and then stored at room temperature. The dried samples were immersed in 1 wt % PDMS n-hexane solution. After 10 min, the wood sample was removed and cured in 80 °C for 3–4 h in vacuum. Finally, the superhydrophobic coating was obtained on the wood substrate.

Characterization. A cold field emission SEM (FE-SEM, JSM-7500F, Japan) instrument was used to observe the surface morphology of the sample coated with a Pt film under a vacuum environment with an accelerating voltage of 5.0 kV. Moreover, an Oxford X-Max cooling energy spectrometer attached to the FE-SEM instrument was applied to check the chemical elements on the wood surface. An FTIR (Nicolet iS50) spectrometer was used to analyze the state of chemical functional groups. The transmittance and absorbance of the coating were studied by XRD (X’Pert Powder, Netherlands). In the wettability test, WCs were measured using a droplet shape analyzer (DSA100, Krüss, Germany) with about 10 µL of water droplets. The final result of the WCA is the average of five measurements made at different locations on the wood surface. A digital camera (DB80, Nikon, Japan) was used to record static optical images in the experiment.

■ ASSOCIATED CONTENT

* Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.1c04207.

Presentation video of the self-cleaning test (AVI)

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Notes
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