Wood Sponge Reinforced with Polyvinyl Alcohol for Sustainable Oil–Water Separation
Yijing Cai, Yan Wu,* Feng Yang, Jian Gan, Yajing Wang, and Jilei Zhang

ABSTRACT: The excellent oil absorption capacity and sustainability advantages of adsorbent-type oil-absorbing products have become the primary method to deal with marine oil spills and organic pollution at this stage, especially aerogel products. However, this type of material also has some problems, such as secondary pollution during nanocellulose preparation. Lignin and hemicellulose were removed from the natural wood, and followed by the action of freeze drying, the wood sponge was prepared. Then, followed by immersing the wood sponge into polyvinyl alcohol solution (PVA) and dipping it in polydimethylsiloxane solution, the target PVA-reinforced wood sponge with better mechanical compressibility and hydrophobic properties was obtained. The new wood sponge showed high mechanical compressibility (reversible compression rate of 40%) and elastic recovery rate (the height retention rate was about 100% after 200 cycles of 30% strain). It also showed excellent hydrophobic and oleophilic properties, and the water contact angle was up to 138°, and the oil absorption capacity was 25 g·g⁻¹. The ability of oil absorption can be recovered by compression, and the high absorption rate was maintained after 50 cycles. The wood sponge has great potential in reusable oil–water separation due to low cost, high efficiency, high performance, biodegradability, environmental friendliness, and other advantages.

1. INTRODUCTION
With the development of exploiting and transporting petroleum in the sea and the surge in demand for industrial raw materials, the discharge of industrial wastewater and the frequent occurrence of marine oil spills have seriously threatened the marine ecological environment and species.⁷⁻⁹ Therefore, it is necessary to find an effective method to solve the problem, especially for the marine oil spills, which includes in situ combustion, gravity separation, bioremediation, and so on at this stage, but these methods are expensive and less efficient, causing secondary damage to the environment.⁶⁻⁹ Developing the easy-to-operate and eco-friendly adsorbent is an effective method to deal with the problem of marine wastewater.¹⁰ In recent years, adsorbents with a three-dimensional porous structure which has the advantages of a large surface area, high porosity, high absorption capacity, and special wettability have received widespread attention.¹¹⁻¹³ However, these materials still have the disadvantages of the complicated manufacturing process, poor recyclability, and low economic efficiency.¹⁴⁻¹⁶ Moreover, the detailed process and feasible mechanism of high-viscosity crude oil adsorption are not yet understood, which actually limits the design and application of high-efficiency adsorbents. In view of these shortcomings, it is extremely important to find alternatives that are environmentally safe, cost-effective, more effective, and sustainable.¹⁷

Considering the natural abundance, low cost, biodegradability, and environmental friendliness, biomass materials as raw materials for adsorbent production have a promising future and attract more and more researchers, such as the 3D-printed biomimetic super-hydrophobic structure;¹⁸ graphene nanoribbon/polyurethane sponge;¹⁹ and nanofibrous cellulosic membrane.²⁰ As a naturally renewable resource, wood is composed of a cellulose framework and structural supports of hemicellulose and lignin.²¹⁻²³ At this stage, biomass energy was widely used. Researchers reported a lignocellulosic porous carbon which had high-performance deionization (CDI), high salt adsorption capacity, and good cycle stability.²⁴ In addition, cellulosic materials were also potentially used as bio-absorbents to remove dyes from contaminated water.²⁵ With the advantages of light weight, high porosity, low density, and a larger specific surface area, as absorbents, aerogels extracted from plant cell walls as raw materials and prepared using freeze drying and supercritical drying methods have strong absorption capability and can be used to effectively absorb
impurities and pollutants in water, but the complicated process and high manufacturing cost limit their applications. By selectively removing lignin and cellulose, a flexible cellulose skeleton structure can be obtained, which can be transformed into an ideal porous adsorbent with a 3D structure. It not only can absorb a certain amount of crude oil but also recover the oil in it through simple mechanical extrusion. As an oil-absorbing agent, the wood sponge has simpler production methods and lower manufacturing costs than other biomass materials, such as aerogel materials based on bacterial cellulose, nanocellulose, cotton, and lignin. However, for the wood sponge, there are still some problems, such as the material being limited to light wood such as balsa wood and the relatively low of reuse rate. Although the balsa wood has the characteristics of low density and high porosity, its high production cost and low mechanical strength are not conducive to repeated extrusion for reuse such as separating oil from oil–water mixtures. Furthermore, the wood with almost complete removal of lignin and hemicellulose has the lightest quality, which reasonably brings the largest oil absorption mass ratio but also leads to poor mechanical properties, especially for compressibility. Therefore, if the honeycomb structure of the original wood is not damaged, the mechanical properties could be retained, which makes the wood sponge after the removal of lignin and hemicellulose have enough strength to perform oil/water separation after repeated extrusion.

In this study, the poplar wood (Populus trichocarpa) was used as the raw material to prepare the wood sponge with better oil/water separation ability. First, the lignin and hemicellulose in poplar wood were removed through sodium chlorite (NaClO2) and sodium hydroxide (NaOH) to retain a cellulose skeleton with a three-dimensional hierarchical structure. Then, the cellulose skeleton was immersed in polyvinyl alcohol (PVA) solution to reinforce the three-dimensional structure. Finally, the PVA-reinforced wood sponge (PWS) was immersed in the polydimethylsiloxane (PDMS) solution with n-hexane as the solvent to successfully prepare a new wood sponge with better hydrophobic and oleophilic properties (PPWS). The prepared PPWS has strong oil absorption and high oil–water separation efficiency. In addition, the microstructure, chemical composition, mechanical strength, hydrophobic properties, oil absorption performance, and reusable oil/water separation functions were also studied.

### 2. RESULTS AND DISCUSSION

#### 2.1. Structure Characteristic and Chemical Analysis

Poplar wood as the raw material for preparing wood sponge has the advantages of low density (0.47 ± 0.007 g cm⁻³), high porosity (84.9 ± 0.01%), and thinner pore walls (5 μm) compared with other natural wood (Table 1). As shown in Figure 2a, the poplar wood cells with a honeycomb shape and high porosity were neatly arranged from the cross-section, and the tube pores containing infill and horizontally arranged intercellular pathways which transfer the nutrients trees need during the growth of the poplar and help the modified drugs to enter the interior of the poplar block can be seen intuitively. The wood cell wall is mainly composed of three parts, including cellulose, hemicellulose, and lignin. Among them, the cellulose, hemicellulose, and lignin provided natural wood with powerful mechanical properties, but hemicellulose and lignin reduce the low specific surface area of wood. Therefore, this cellulose skeleton with a 3D structure has great potential for further functionalization.

After selectively removing lignin from NW with NaClO2 aqueous solution, white wood blocks were obtained (Figure 2b), which indicated that the dark lignin had been removed, leaving colorless polysaccharides. In addition to the change in wood color, the removal of lignin also reduced the density (0.25 ± 0.01 g cm⁻³) and increased the porosity (90.2% ± 0.1) of wood, as shown in Table 1. It was confirmed by FTIR that the characteristic peaks of lignin in the wood after chemical treatment disappeared at 1593, 1506, and 1459 cm⁻¹ (Figure 2e), corresponding to the vibration of the benzene ring carbon skeleton in lignin. However, the peaks at 1733 and 1235 cm⁻¹ were retained, which were related to hemicellulose, indicating that the NaClO2 solution largely removed lignin while retaining hemicellulose. The characteristic peaks of lignin in WS were weakened at 1593, 1506, and 1459 cm⁻¹ when compared with the ones in NW. Meanwhile, the C=O stretching vibration (xylan acetyl) of DW blocks near the peak of 1733 cm⁻¹ disappeared, and the peak at 792 cm⁻¹ related to the CO–OR bond in hemicellulose and Ph–O–R vibration in lignin was also weakened. These results indicated that lignin and hemicellulose in WS were largely removed. This could also be known from the chemical composition analysis; compared with NW, the lignin in DW was significantly reduced (Figure 2f). The microscopic and structural properties of NW have also changed by delignification treatment. As shown in Figure 1b, while the impurities in the cells and tracheid were removed, the neat honeycomb structure was scattered and replaced with a layered stacked structure in which the cell-to-cell connections were favorable, and the cell wall shrank to varying degrees. Parenchyma cells appeared to shrink or even break after the chemical modification of removing lignin, which was conducive to the expansion of the specific surface area and the increase in porosity. It was interesting that the shape of the lignin-removed wood has also changed from the original rectangular shape to a parallelogram-like structure, which produced recoverable deformation without damaging the cell wall when it encountered compression, which provided it with excellent compression performance.

In order to further optimize the structure, NaOH solution was used to perform additional chemical treatment on the delignified wood to remove the residual hemicellulose. Inferred from the FTIR, the disappearance of the peaks at 1733 and 1235 cm⁻¹ can infer that hemicellulose had been largely removed, and the chemical composition analysis also clearly showed that the hemicellulose in WS has been largely removed (Figure 2f). The XRD pattern showed that after chemical treatment, the arrangement of cellulose nanofibers in the cell wall was well preserved (Figure 2g), and the crystal structure of NW did not change during the treatment of cellulose Iβ. After the removal of hemicellulose and lignin, the cellulose skeleton was retained, forming a WS with a high porosity and very low density (0.17 ± 0.002 g cm⁻³) of about 60% lower than those

Table 1. Density and Porosity of NW, DW, WS, and PWS

| wood sample | density (g cm⁻³) | porosity (%) |
|-------------|-----------------|--------------|
| NW          | 0.47 ± 0.01     | 84.9 ± 0.1   |
| DW          | 0.25 ± 0.01     | 90.2 ± 0.1   |
| WS          | 0.17 ± 0.01     | 92.3 ± 0.1   |
| PWS         | 0.19 ± 0.02     | 92.4 ± 0.1   |
| PPWS        | 0.19 ± 0.01     | 92.3 ± 0.2   |
of NW. In the SEM image, the original honeycomb structure disappeared further and turned into a spring-like structure by stacking multiple wave-shaped layered structures (Figure 2c). The reason for this phenomenon was that the thicker wood ray part was preserved, but the parenchyma cells were completely broken in the process of chemical treatment, which were frozen into a wave shape under the action of freezing and broken during the drying process. Parenchyma cells were often attached or coupled to the area of uninterrupted wood rays, thus forming a unique layered structure consisting of multiple stacked, arched layers.28,33

According to the report in 2018, WS could be compressed to 60% of its original thickness and completely recovered after releasing the force; however, it could be damaged if it is manually compressed about 10 times after absorbing water.33,34 The reason was that when the WS was saturated with water, the cell wall was swollen, and the connection between the hydrogen bonds was broken by several instances of squeezing, which causes damage to the cell wall.27 In order to further increase the water saturation strength of WS, a vacuum-impregnated PVA-modified WS (PWS) was adopted. Although from the perspective of density, the PWS had not changed much (0.19 ± 0.004 g·cm⁻³), it could be clearly seen from the microstructure that the special layered structure was connected by filamentous substances which had a support force and distributed between layers, including cells (Figure 2d). It could be further seen from the XPS spectrum that the relative intensity of the peak in Figure 2g (2) increased compared with the O–C–O group in Figure 2g (1), and the peak at C–C/C–H also changed relatively.35 These results indicated that a large number of PVA chains have been attached to the WS and became the filamentous substance in the layered structure of the WS, which played a role in stretching and supporting.36,37

2.2. Mechanical Behavior. According to the reports,33 wood sponges made from balsa wood have excellent compressive properties in the stacking direction, which can withstand up to 60% compressive strain and completely restore their original height after the stress was released. Compared with balsa wood, poplar wood has lower porosity. The compression and resilience of wood sponge made from poplar wood are not as good as those of balsa wood sponge, but benefited from the lower porosity, poplar wood sponge has good mechanical stability. Compared with NW, the compression deformation of WS under the same pressure reached 50% of the length in the compression direction. After the pressure was released, it quickly returned to the original state (Figure 3a), while the natural wood remains as the original wood.38

In order to further explore the relationship between the mechanical properties of PWS and the concentration of PVA, the mechanical strength of WS impregnated with different concentrations of PVA was measured, ranging from 1 to 5 wt %. It could be seen from the stress–strain curve (Figure 3b) that when the PVA concentration was 4 and 5 wt %, the elastic phase of wood sponges was significantly shorter than that of 1–3 wt %, and the deformations of PWS were 20 and 40%, respectively. PWS entered the elastic phase where it could return to its original shape when the pressure was removed. However, after entering the yield phase, PWS could only partially recover, while the elastic phase of PWS could last to about 50%. When the PVA concentration was 1, 2, and 3 wt %, its elastic stages lasted to 60, 55, and 50% respectively, which was better than that of WS. The stress–strain curve showed that when the PVA concentration was low, the curve tended to be smooth. As the concentration increased, the curve becomes steep (the slope increases). Compared with the pressure required for 60% strain, PWS4 and PWS5 required the largest pressure. As the amount of PVA decreased, the required pressure also decreased. It was worth noting that when the strain was 60%, the pressure required by PWS1 was greater than that of PWS2. Although there was no obvious linear relationship between the compressibility of PWS and the amount of PVA, when the PVA concentration was as high as 5 and 4 wt %, the compression rate of PWS was less than that of WS, and when the concentration was 2, and 3 wt %, the compression rate of the WS was less than that of PWS, and the best concentration was 2 wt %. Adding an appropriate amount of PVA could enhance the compressibility of WS, but when it exceeded a certain amount, it would reduce the compressibility.
of WS. Due to the hierarchical structure of WS and the strong interaction between cellulose, hemicellulose, and lignin, its breaking strength was as high as 500 KPa, while the breaking strength of PWS was only 200 KPa under the same conditions.

When the concentration of PVA reached 2 wt % and the strain was less than 60%, the stress–strain curve presented a linear stage, and the reaction area deformed elastically, which had good mechanical strength without the partial structural

Figure 2. Morphology and structure of different wood samples. (a) Photograph of NW and its cross-sectional SEM image, showing a honeycomb porous structure and compact cell walls. (b) Photo of WS and its cross-sectional SEM image, showing the delamination of the internal structure of the cellulose nanofibers exposed in the cell wall. (c) Photograph of WS and SEM image of its cross-section, showing a layered butt structure. (d) Photo of the PWS and its cross-sectional SEM image, showing the filamentary connection between the layered stacked structure. (e) FT-IR spectra of different wood samples. (f) Relative content of cellulose, hemicellulose, and lignin in different wood samples obtained by chemical composition analysis. (g) XPS spectra of WS and PWS samples. (h) XRD patterns between different samples.
collapsing, while the plastic deformation appeared to be between 60 and 70%. Compared with other samples, PWS2 had good mechanical compression. It was attributed to the special spring-like layered structure and the filiform PVA between the curved layers, which could increase strength.

The compressive stress–strain curve was used to further evaluate the mechanical properties of PWS2, as shown in Figure 3. Figure 3c shows that when the deformation was 30%, both WS and PWS2 could rebound to their original state, but WS required more force than PWS2. After reaching a high strain of 40%, PWS2 could good return to its original state, showing better mechanical compressibility. Meanwhile different strain (20, 30, and 40%) curves of PWS2 were further recorded, as shown in Figure 3d. Due to the elastic deformation of PWS2, the linear area of the curve of 20% was less than 30 and 40% (Figure 3d). Then, although the stress increased to 40%, the stress caused by the collision of the adjacent-stacked layers also increased, resulting in the restoration of the PWS2 shape. The compression test was repeated 200 times, and a constant strain of 30% was controlled to evaluate the fatigue resistance of PWS, as shown in Figure 3e. The results showed that after compression–release cycles 200 times, the plastic deformation was very slight, and the height retention rate was above 94%, showing its excellent structural stability. Compared with other wood sponges based on balsa wood in the references, PWS made from poplar wood in this study had better mechanical properties and fatigue resistance. This was due to a special structure, which resembled a spring. In the layered structure, there were many voids, which allowed greater deformation without local structural collapse. However, the difference in wood (early wood and late wood, chord and radial) will influence the experimental results to some extent.

2.3. Hydrophobic and Oleophilic Performance. The PWS was made of a wood cellulose skeleton and reinforced with PVA materials added between the cell layer structures to increase mechanical properties. Both of these materials have strong hydrophilic and lipophilic properties (Figure 4a), so the PDMS was used to make PWS possess hydrophobic properties. The presence of Si–O and Si–C in the XRD spectrum proved that the cellulose skeleton has been successfully silanized, and the SEM image of the PPWS also clearly showed the growth of the oxane coating of the granular polysilicon in the direction of the accumulation layer (Figure 4f). The PPWS showed excellent hydrophobicity, with the water contact angle on the surface reaching 138°, and the hydrophobic angle decreased slightly within 2 min with the hydrophobic angle greater than 135°. Meanwhile, the internal hydrophobicity of the PPWS was also a manifestation of the functionalization of oil–water separation. The internal hydrophobic angle of the PPWS has
also reached 120°, which had good hydrophobic properties. Benefitting from the highly porous structure, the PDMS could be effectively dispersed and penetrated in PWS. The PPWS not only had good hydrophobicity on the surface but also achieved a certain hydrophobic effect inside the PWS, indicating that the silylation in the whole structure was quite uniform.

In order to further observe the absorption state of samples, the capillary absorption of water along the longitudinal direction was measured (Figure 4b). The water was dyed with MB, so the trajectory of water movement in the wood could be clearly observed. Both WS and PWS showed excellent absorption capacity along the capillary, and the dye could reach 40% of the entire length of the samples within 1 s. The PPWS also showed excellent hydrophobic properties. It can be seen that although it absorbed a certain amount of water at the beginning, about 3 mm, it still maintained a water absorption length of 3 mm after 2 min in the water. However, the absorption of water by natural wood was not obvious. The reason for the phenomenon was that the nanopores produced in the cell wall of the WS enhanced capillary action and promoted liquid absorption. The oil absorption performance of PWS was slightly better than that of PPWS (Figure 4c). At 0 s, PWS showed oil absorption performance comparable to NW, while PPWS oil absorption performance was slightly lower than that of PWS. At 60 s, the oil absorbed by PWS reached 75% of the length of the wood stripe, higher than that of PPWS (63%). The PWS showed water/oil absorption capacity, while silanized PWS (PPWS) showed excellent oil absorption and hydrophobicity.

Figure 4. Water and oil absorption capacity of PPWS. (a) WS, PPWS water droplet, and oil drop photos and contact angle, (b) water absorption diagram of NW, WS, PWS, and PPWS along fiber directions, (c) oil absorption diagram of NW, WS, PWS, and PPWS along fiber directions, (d) XPS image of PPWS, and (e) energy-dispersive X-ray spectroscopic maps showing the elemental distribution of Si on the PPWS cross-section.
The PPWS showed high porosity, lightweight, and hydrophobicity, so oil and organic solvent absorption properties have been further explored in this study, as shown in Figure 5. From Figure 5f, it could be seen that the PPWS had inherited the excellent oil absorption performance of WS, but different concentrations of PVA also have a certain impact on the oil absorption capacity of PPWS2.
absorption capacity of PPWS. Silicone oil, soybean oil, and isopropanol solution were used to explore and prove the oil absorption capacity of PPWS. This PPWS with different concentrations of PVA could selectively absorb oil on the water surface (Figure 5b) or underwater (Figure 5c), but its absorption capacity was significantly different (Figure 5f). As far as silicone oil was concerned, the oil absorption capacity of PPWS with a PVA concentration of 2% (PPWS2) was the best, reaching 25%, followed by that of the WS. It was worth noting that they have a good absorption capacity for silicone oil. For soybean oil, the PPWS2 still showed excellent absorption performance, and the absorption energy reached 20 g·g⁻¹. After that, the absorption capacity for soybean oil decreased as the concentration increased. For organic substances such as isopropanol, their absorption capacity was equivalent, around 8 g·g⁻¹. The oil absorption capacity of the PPWS2 was further explored. The content of various oils and organic solvents in the composite material was 5–20 g·g⁻¹, which was obviously better than that of most of the aerogel materials for oil–water separation treatment (Table 2).

### Table 2. Oil Absorption Capacity of Other Materials

| the adsorbent                                  | absorption capacity (g·g⁻¹) | REFs. |
|-----------------------------------------------|---------------------------|-------|
| hydrophobic silica aerogels                   | up to 15.1                | 41    |
| elastic superhydrophobic silica aerogels      | 9.83–10.64                | 42    |
| 3D macroscopic superhydrophobic magnetic porous carbon aerogel | 10.02–10.83               | 43    |
| poly-g-poly styrene                           | 4–21                      | 44    |
| hydrophobic and nanoporous chitosan–silica composite aerogels | 13–3                      | 45    |
| spongy graphene                               | 20–86                     | 46    |
| chitin sponges                                | 20–40                     | 47    |
| silylated wood sponge                         | 16–41                     | 33    |

The reusability of the PPWS2 was essential for practical application as an oil/chemical absorbent. The expanded PPWS2 impregnated with silicone oil was directly squeezed to discharge the oil in the sponge, and thanks to its excellent compressibility and elasticity, the squeezed sample quickly recovered its original shape without any deformation after the pressure was released (Figure 5a). The absorptive capacity of PPWS2 subjected to periodic extrusion and the absorption of oil were evaluated (Figure 5f). After 50 times of compression and absorption cycles, the adsorption capacity only decreased to 20.2 g·g⁻¹ (Figure 5g), indicating that PPWS had stable absorption performance and good reusability, which ensured that it could still be used after multiple compressions. At this stage, many aerogel materials have weak oil absorption capacity and poor repeatability. The oil absorption capacity of these aerogel materials was usually reduced after multiple compression and absorption cycles because they lacked sufficient mechanical elasticity as absorbents. Meanwhile, different strain (20, 30, and 40%) curves and cyclic compression of PPWS were further recorded, as shown in Figure 3d, which showed superior recyclability.

### 3. CONCLUSIONS

The special honeycomb structure of natural wood gave the characteristics of high porosity. The oil/water separation wood sponge with excellent mechanical properties and reusability was prepared by chemical methods. The lignin and hemicellulose were innovatively removed from poplar wood, and the PVA-reinforced WS was prepared by dipping it in PDMS solution. The PWS had a special spring-layered structure with the PVA wire embedded. Such a structure provided excellent mechanical compression properties and the elasticity of WS, which can be repeatedly squeezed 200 times in the stacking direction without damaging the structure. Meanwhile, the PPWS obtained by silane modification had a higher oil absorption of up to 25 g·g⁻¹, and the oil can be recovered by simple mechanical extrusion. A sustainable WS with mechanical elasticity had great potential in hydrophobic and oil absorption, which can be used as an adsorbent to deal with marine oil spills.

### 4. EXPERIMENTAL SECTION

#### 4.1. Materials

Poplar wood (Populusmentosa) was obtained from Yihua Co. Ltd., China with a density of 0.47 ± 0.007 g·cm⁻³. The wood samples were cut into the sizes of 15 mm × 15 mm × 15 mm (radial × tangential × longitudinal). Anhydrous ethanol of analytical grade was purchased from Sinopharm Chemical Reagent Co., Ltd. NaClO₂ was supplied by Shanghai Macklin Biochemical Co., Ltd. NaOH of analytical grade was purchased from Nanjing Chemical Reagent Co., Ltd. PVA was purchased from Sinopharm Chemical Reagent Co., Ltd. PDMS was obtained from Yuanye Co., Ltd. Methylene blue was purchased from Tianjin Shi Chemical Reagent Research Institute Co., Ltd. Glacial acetic acid (CH₃COOH), toluene, dichloromethane, trichloromethane, n-hexane, n-dodecane, and n-hexadecane were purchased from Nanjing Chemical Works. The soybean and engine oils were purchased from a local market. All chemicals were used as received without further purification.

#### 4.2. Preparation of Wood Sponge

The absolutely dry poplar wood samples were immersed in a mixed solution of 5 wt % NaClO₂ and CH₃COOH at 85 °C for 6 h. The pH value of the solution was adjusted to 4.6. The NaClO₂ solution was changed every 8 h until the color of wood became pure white. The delignified wood samples (DW) were washed with ultrapure water to remove chemical residues. Then, 8 wt % NaOH solution was used to selectively remove hemicellulose from wood again at 85 °C for 8 h. Finally, the treated samples were washed with ultrapure water again. After freeze drying, the wood sponge (WS) samples were obtained.

#### 4.3. Preparation of Reinforced Wood Sponge

The WS samples were immersed in PVA aqueous solution using a vacuum chamber (0.1 MPa, 2 h). Then, the samples penetrated with PVA were washed with ultrapure water to remove excess residue. The washed samples were freeze-dried in vacuum for 24 h to obtain the PWS. The PWS samples with PVA concentrations of 1, 2, 3, 4, and 5 wt % were also prepared in the same way and coded as PW51, PW52, PW53, PW54, and PW55, respectively.

#### 4.4. Preparation of Hydrophobic and Lipophilic Wood Sponge

The PWS samples were immersed in 1 wt % PDMS/n-hexane solution using a vacuum chamber (0.1 MPa, 30 min). Then, the samples were washed with n-hexane to remove excess PDMS. After placing in an oven at 60 °C for 2 h, the final materials of PPWS were obtained and coded as PPWS1, PPWS2, PPWS3, PPWS4, and PPWS5, respectively. The specific experimental steps are shown in Figure 1.

#### 4.5. Characterization

The surface morphology and structure of nature poplar wood (NW), WS, PWS, and PPWS samples were observed by field emission scanning electron microscopy (FESEM, FEI Co., Ltd., USA). The
elemental maps were obtained by energy dispersive spectroscopy (EDS) with FESEM. The elemental and chemical analysis was performed by X-ray photoelectron spectroscopy (XPS, Thermo Fisher Scientific Co., Ltd) and Fourier transform infrared spectroscopy (FTIR, Bruker Spectroscopy, USA), respectively.

The porosity was measured by the weighing method. Specifically, the weight (g), length (cm), width (cm), and height (cm) of the wood block were measured. Then, the wood block was immersed in isopropanol, and the weight of the wood block was measured after sufficient infiltration. The porosity was calculated by the following formula 1. Multiple samples are measured to calculate the average

$$\text{Porosity} = \frac{(m_2 - m_1)/\rho}{l \times w \times h} \times 100\%$$  \hspace{1cm} (1)

where $m_1$ is the weight of the sample, $m_2$ is the weight of the sample after measuring in isopropanol, and $\rho$ (g cm$^{-3}$) is the density of isopropanol. The $l$, $w$, and $h$ represent the length, width, and height of the sample, respectively.

The mechanical properties and resilience of WS and PWS were tested using universal testing equipment (Shimadzu Corporation, Kyoto, Japan). The water contact angles (CAs) were measured with an OCA20 contact angle system (CAs, Krusch Scientific Instruments Co., Ltd., China). To observe the NW, PWS, and PPWS absorption capacity of water and oil, a water (stained with methylene blue) and oil (stained with Sudan Red) mixture was prepared at first. Then, the bottoms of the samples with the size of 40 mm (radial) × 5 mm (tangential) × 1 mm (longitudinal) were immersed in the mixture in a Petri dish for 60 s. In order to test the absorption capacity of oils and organic solvents, the samples were fully immersed in the oils (silicone oil, engine oil, and soybean oil) and organic solvents (isopropanol, ethanol, methanol, xylene, n-hexane, and dichloromethane) until the weight did not change. The absorption capacity of various oils and the organic solvent was measured as follows

$$\text{Absorption capability} = \frac{(m_4 - m_3)/m_3}{m_2 - m_1}$$  \hspace{1cm} (2)

where $m_1$ and $m_4$ are the weights of the sample before and after absorption, respectively.

To test the oil absorption of WS and PPWS, they were immersed in silicone oil, and the mass before and after weighing was calculated using formula 1. In order to test the circulating oil absorption capacity of PPWS, the sample was completely immersed in oil until no liquid flows out, and then, the PPWS was took out from the oil and weighed. Formula 1 is used to calculate the oil absorption of the tested PPWS. The next oil immersion squeeze cycle starts by putting each PPWS back into the water again until it reaches a saturated state; then, the PPWS was removed and the remaining steps were repeated until 50 cycles are completed.

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**Notes**

The authors declare no competing financial interest.

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