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Article

Fabrication of Green Superhydrophobic/Superoleophilic Wood Flour for Efficient Oil Separation from Water

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Abstract: The removal of oil from waste water is gaining increasing attention. In this study, a novel synthesis method of green superhydrophobic/superoleophilic wood flour is proposed using the deposition of nano-zinc oxide (nZnO) aggregated on the fiber surface and the subsequent hydrophobic modification of octadecanoic acid. The as-prepared wood flour displayed great superhydrophobicity and synchronous superoleophilicity properties with the water contact angle (WCA) of 156° and oil contact angle (OCA) of 0° for diesel oil. Furthermore, the as-prepared wood flour possessed an excellent stability, probably due to the strong adhesion of nZnO, which aggregates to the fiber surface of wood flour with the action of glutinous polystyrene. The maximum adsorption capacity of as-prepared wood flour was 20.81 g/g for engine oil, which showed that the as-prepared wood flour is a potential candidate as an efficient oil adsorbent in the field of water-oil separation. Moreover, it has good chemical steadiness and environmental durability. Taken together, all the information acquired from this research could be valuable in evaluating the potential of as-prepared wood flour as a competitive and sustainable oil-water separation material.

Keywords: wood flour; oil adsorption; superhydrophobic; superoleophilic; oil-water separation; sustainable material

1. Introduction

With the rapid development of modern industry, the growing crisis of global water pollution is severely affecting the environment [1–5]. A representative case is the explosion at British Petroleum's Deepwater Horizon oil rig in 2010, which resulted in the loss of life and property, as well as the spillage of huge amounts of oil into the ocean [6]. To date, various materials and approaches have been adopted to remove spilled oil from water bodies, including physical diffusion [7], activated carbon [8], oil containment booms [9], exfoliated graphite [10], waste barley straw [11,12] and membranes [13,14]. Nevertheless, these traditional techniques have certain deficiencies, such as being time-consuming, economically infeasible, environmentally damaging and non-renewable. Therefore, the exploration and use of novel green materials, which can effectively separate oil contaminants from water, is highly desirable. This is not only important for environmental protection, but also for sustainable urban development.

In recent years, a number of self-cleaning plant surfaces have drawn significant research attention. The superhydrophobic nature of lotus leaves is a good example of this [15–18]. In general, the water contact angle is a critical index used to characterize the wettability of a surface. Due to their remarkable

waterproofing performance, superhydrophobic materials have been prepared through various approaches, involving a sol-gel process [19], vapor phase deposition [20], chemical etching [21,22], surface fluorination [23] and electrospinning [24]. To date, a number of advanced materials with superhydrophobic and superoleophilic properties have been synthesized and applied to the disposal of spilled oil [25–29]. For example, Wang et al. [30] developed a facile electrochemical deposition method to prepare a novel functional micro-nano hierarchical structured copper mesh film with special superhydrophobic and superoleophilic characteristics for the effective removal of oil from water. Zhang and Seeger [31] successfully synthesized superhydrophobic and superoleophilic polyester textiles using silicone nanofilaments using one-step growth, which could be used for oil/water separation. Cortese et al. [32] described a convenient approach to fabricate cotton textiles with superhydrophobic and superoleophilic properties using plasma-enhanced chemical vapor deposition. Yue et al. [33] developed a kind of superhydrophobic cellulose/LDH (layered double hydroxide) membrane and applied it in an open oil/water two-phase system. Li et al. [34] successfully synthesized superhydrophobic/superoleophilic cotton fabrics combined with polyvinylsilsesquioxanes polymer and nano- Al_2O_3 particles.

Wood flour is one of the most common forms of agricultural waste, and when burnt, can cause severe environmental problems, such as dust or air pollution. There is an urgent need to reuse waste wood flour in a sustainable way, which could alleviate these problems. Wood flour has the advantages of having a low density, being bio-degradable, eco-friendly and low-cost [35]. However, the synthesis of superhydrophobic and superoleophilic materials using waste wood flour has received little research attention. In this work, wood flour was evenly coated with a layer of octadecanoic acid-modified nZnO aggregates with the assistance of glutinous polystyrene, which enabled it to acquire an excellent performance with regards to both superhydrophobicity and superoleophilicity. The superhydrophobic/superoleophilic wood flour with a WCA of 156° and OCA of 0° was prepared for an efficient oil removal from contaminated wastewater. Regardless of the oil or organic solvent, the maximum oil adsorption capacity of the as-prepared wood flour was almost three-fold higher than the pristine wood flour. The adsorption efficiency ranged from 98% to 100%. After the adsorbed oils were removed using acetone, the superhydrophobic/superoleophilic wood flour could be reused multiple times with a good chemical stability and environmental durability.

2. Materials and Methods

2.1. Materials and Chemicals

The wood flour used in this study was obtained from the Larch species (*Larix gmelini*), provided by the Carpentry Laboratory of Northeast Forestry University, China. Hydrogen peroxide, sodium hydroxide, hydrochloric acid, octadecanoic acid and anhydrous ethanol were provided by Tianjin Kaitong Chemical Reagent Co., Ltd., Tianjin, China. Zinc nitrate was purchased from Tianjin Hengxing Chemical Reagent Manufacturing Co., Ltd., Tianjin, China. Polystyrene ($M_w = 101,900$) was purchased from Shanghai xibao Biological Technology Co., Ltd., Shanghai, China. Methylbenzene was obtained from Tianli Chemical Co., Ltd., Guangdong, China. The gasoline, crude oil, diesel, engine oil, chloroform, n-hexane and toluene were purchased from Tianjin Hengxing Chemical Reagent Manufacturing Co., Ltd., Tianjin, China. All chemical reagents were of analytical grade, and used without further purification.

2.2. Pretreatment of Wood Flour

Raw wood flour was first sifted through a filter screen with a uniform grading smaller than $74 \mu\text{m}$. Then, 1.5 g wood flour was dipped into 7 mL hydrogen peroxide (30%) and 200 mL sodium hydroxide aqueous solution (0.5 wt.%) at room temperature under constant stirring for 14 h. After that, hydrochloric acid (6 mol/L) was added to adjust the pH within the range of 6.5–7.0. Finally, the wood

flour was washed several times using ultrapure water and dried at 50 °C for 3 h until the weight became constant.

2.3. Preparation of nZnO Particles

2.4 g sodium hydroxide and 200 mL ultrapure water were added to a 250 mL round-bottom flask in a water bath with constant magnetic stirring at 70 °C for 10 min. Then, 2.0 g of zinc nitrate was added to the sodium hydroxide solution and continuously stirred for 8 h. The solution was placed at room temperature for some time. Next, the non-reacted reagents or by-products were removed using ultrapure water and anhydrous ethanol. The final solution was dried for 5 h in a vacuum oven at 60 °C. Finally, the particles of nZnO were obtained as white powder.

2.4. Fabrication of the Green Superhydrophobic/Superoleophilic Wood Flour

0.1 g wood flour and 0.1 g nZnO particles were mixed and immersed in a beaker containing 10 mL ethanolic octadecanoic acid solution (5%, m/v). The mixture was maintained at 70 °C for 5 h, washed with anhydrous ethanol and dried in an oven at 60 °C for 4 h to obtain octadecanoic acid modified-nZnO nanoparticles coated on the wood flour surface. Afterwards, the modified-nZnO wood flour composites were mixed with 10 mL (2%, m/v) of polystyrene methylbenzene solution. Finally, the superhydrophobic/superoleophilic wood flour was dried at 50 °C for 2 h until the weight became constant.

The mechanism of the interaction of nanoparticles, coated by octadecanoic acid with wood flour fibers, was as follows. Hydroxyl groups on the surfaces of ZnO particles interacted with the hydroxyl groups on the surfaces of wood flour fibers through the formation of hydrogen bonds, thus making the ZnO particles homogeneously cover the wood flour surface. Octadecanoic acid chemically reacted with ZnO to generate modified-ZnO particles, as shown in Figure 1b,c. Polystyrene reagent served as a binder to cover the surfaces of the modified-ZnO particles and wood flour fibers, which resulted in a firm adhesion of nanoparticles onto the fiber surface. Figure 1a shows the generation of ZnO aggregates on the sample surface. In fact, the ZnO nanoparticles on the surface of the final product existed as ZnO aggregates.

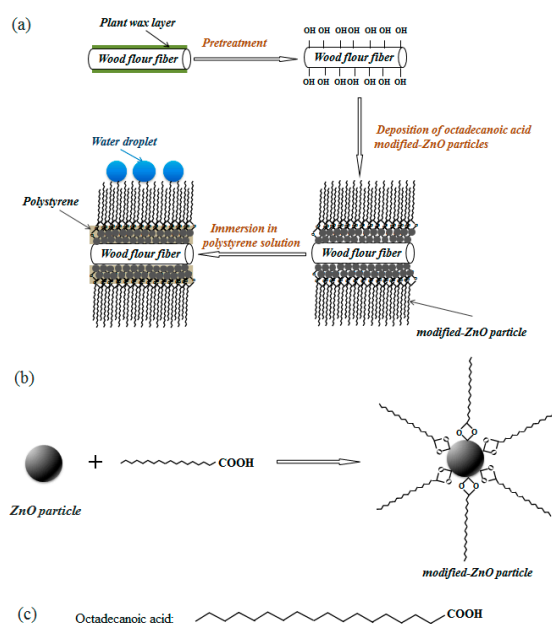


Figure 1. (a) Synthesis route for the preparation of superhydrophobic/superoleophilic wood flour. (b) Graphical representation for the modification of the ZnO particle with octadecanoic acid. (c) Chemical structure of octadecanoic acid.

2.5. Separation of Oil/Water Mixtures

0.5 g of as-prepared wood flour was added to a beaker containing a 150 mL mixture of water and diesel oil, which was dyed red using Sudan III in order to easily and clearly observe the phenomenon. After the adsorption, the red wood flour was recovered on the surface of the water.

The oil adsorption capability was defined using Equation (1):

$$Q = (m_2 - m_1) / m_1 \quad (1)$$

where Q is the oil adsorption capability (g/g); m_2 is the weight of the wood flour after adsorption (g), and m_1 is the initial weight of the wood flour before adsorption (g).

2.6. Characterization of the Green Superhydrophobic/Superoleophilic Wood Flour

2.6.1. The Surface Morphologies of Wood Flour Sample

Scanning electron microscopy (SEM, FEI QUANTA200, Hillsborough, Oregon, USA) was used to examine the surface morphologies of the pristine and superhydrophobic/superoleophilic wood flours under the condition that all specimens were pre-coated with a layer of gold.

2.6.2. The Chemical Compositions of the Wood Flour Samples

The chemical compositions of the wood flour samples were investigated using Fourier transform infrared spectroscopy (FT-IR, Magna-IR 560, Nicolet, Madison, Wisconsin, USA), X-ray photoelectron spectrometry (XPS, PHI Thermo Fisher Scientific, Waltham, MA, USA) and energy-dispersive X-ray analysis (EDX, Quantax 70, Billerica, MA, USA). The FT-IR spectra of the wood flour samples were obtained by direct transmittance using the potassium bromide (KBr) pellet technique. For each sample, the wavenumber was measured within the range of 500–3400 cm^{-1} , and the spectrum was accumulated from a total of 32 co-added scans at a spectral resolution of 4 cm^{-1} . The preparation of the samples for the FT-IR measurement was performed by fully grinding the mixture of 2 mg wood flour and 200 mg spectroscopic grade KBr powder, which were pressed into a pellet with a diameter of 15 mm. Before analysis, the background spectrum of pure KBr was recorded. For the infrared spectroscopic analysis, the pellets were analyzed directly.

2.6.3. The Water/Oil Contact Angle

A contact angle instrument (CA-A, Hitachi, Tokyo, Japan) was used to measure the water contact angle (WCA) and oil contact angle (OCA). For these measurements, 5 μL of ultrapure water or an oil droplet was dropped at five different locations under ambient conditions. The values of WCA and OCA were obtained as averages of five repeated measurements.

3. Results and Discussion

3.1. Morphology of the Green Superhydrophobic/Superoleophilic Wood Flour

It has been widely reported that the superhydrophobic property of certain materials is primarily due to their dualistic micro/nano surface structures [36]. As a consequence, it is vital to characterize the surface morphologies of samples before and after the treatments. The morphologies of pristine and superhydrophobic/superoleophilic wood flours were characterized using SEM at different magnifications, as shown in Figure 2. According to Figure 2a–d, after the treatment with octadecanoic acid modified-nZnO particles and polystyrene, the integrity of the fiber-like structure of the wood flour survived, which means that the structures of fibers were the same for both the pristine and as-prepared wood flours. In addition, compared with the pristine wood flour, the as-prepared superhydrophobic/superoleophilic wood flour possessed a rougher surface because of the deposition of nZnO aggregates, which had an average diameter of approximately 80 nm on

the surface of the fibers (Figure 2b,d). A hierarchical structure is thus presented on the surface of the superhydrophobic/superoleophilic wood flour, which included the micron fibers and abundant nanoparticles on each fiber. Due to this structure, the as-prepared wood flour achieved enough roughness to bring about the co-existing features of superhydrophobicity and superoleophilicity. The results showed that the presence of nZnO aggregates played a prominent role in the preparation of the superhydrophobic/superoleophilic wood flour. On this basis, and in combination with the modification of an octadecanoic acid (low-surface energy material), a large amount of air could be trapped in the cavities and interspaces in the as-obtained wood flour surface. Then, as soon as a water droplet was dripped onto the surface of a sample, it would come into contact with the trapped air and bounce off without any residue. The superhydrophobicity of wood flour was due to the combination of trapped air and low-surface energy material, which prevented the adsorption of water at the interface.

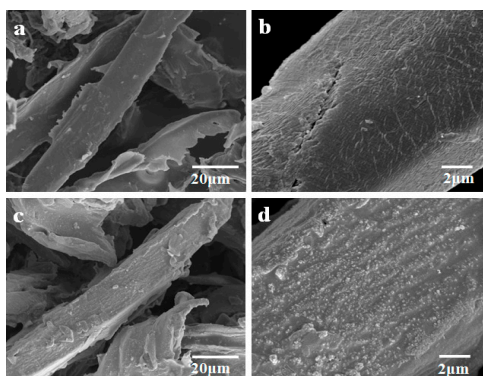


Figure 2. SEM images of pristine wood flour. (a,b) without nZnO aggregates and superhydrophobic/superoleophilic wood flour, and (c,d) with nZnO aggregates at low and high magnifications.

3.2. Surface Wettability of the Green Superhydrophobic/Superoleophilic Wood Flour

In order to verify the superhydrophobicity and superoleophilicity of the as-obtained wood flour sample, its surface wettability was investigated by measuring the values of the water/oil contact angles using a contact angle device at room temperature. As shown in Figure 3a, the water contact angle of pristine wood flour was almost 0° , indicating that abundant hydroxyl groups were present on the fiber surfaces. With regards to the sample treated with octadecanoic acid modified-ZnO, the water contact angle reached 120° (Figure 3c) and therefore had a hydrophobic surface. Furthermore, after coating with octadecanoic acid modified-nZnO and polystyrene (Figure 3b), the water droplet on the as-prepared wood flour became spherical, and the water contact angle became as high as 156° . Additionally, the scattering of the contact angle along the sample was 5° , showing that the as-prepared wood flour had excellent superhydrophobic properties. In addition, the oil wettability of the as-prepared wood flour surface was also examined using oil contact angle measurements. When the oil droplets fell onto the surface of the resulting product, all the oil droplets (diesel oil, gasoline and kerosene) were immediately adsorbed onto the treated wood flour at the interface (see Figure 3d), indicating that the oil contact angles of the as-prepared wood flour surface were 0° . A hierarchical structure is presented on the surface of the superhydrophobic/superoleophilic wood flour, including the micron fibers and abundant nanoparticles on each fiber. Due to this structure, the as-prepared wood flour achieved enough roughness to bring about the co-existing features of superoleophilicity. For the same oil or organic solvent, the maximum oil adsorption capacity of the as-prepared wood flour was almost three times that of the pristine wood flour, indicating the enhancement of superoleophilicity. In short, the dual basic properties of the as-prepared wood flour with superhydrophobicity and superoleophilicity were clearly confirmed.

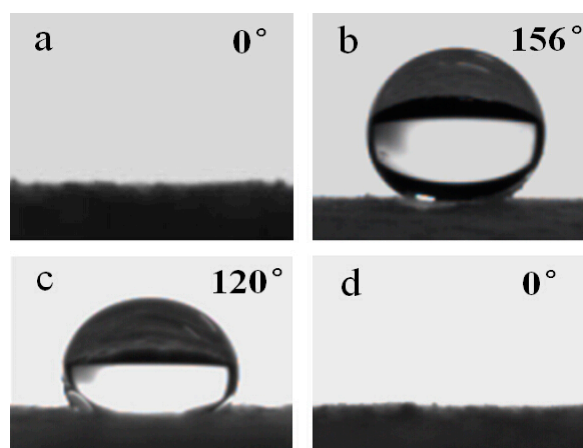


Figure 3. Images of a liquid droplet on different surfaces: typical photograph of a 5 μL water droplet on the surfaces of (a) pristine wood flour and (b) superhydrophobic/superoleophilic wood flour; (c) a water droplet on a wood flour surface treated with octadecanoic acid; (d) an oil droplet on a superhydrophobic/superoleophilic wood flour surface.

On the basis of the theoretical principle of surface wettability, the wettability of a material is a synergistic effect of the chemical composition and surface morphology [37]. The combination of the deposition of numerous nZnO aggregates and the surface decoration through low surface energy octadecanoic acid was required to produce the features of superhydrophobicity and superoleophilicity of wood flour. As a result, when a water droplet was dropped onto the as-prepared wood flour, it bounced off, leaving almost no trace of water. Overall, a novel wood flour with a very good superhydrophobic and superoleophilic performance was successfully obtained in this study.

3.3. Chemical Composition Analysis of the Green Superhydrophobic/Superoleophilic Wood Flour

In order to prove the generation of octadecanoic acid modified-nZnO and polystyrene molecules on the as-prepared wood flour's surface, FT-IR, XPS and EDX were employed to investigate the chemical components of as-prepared wood flour.

The typical FT-IR spectra of the wood flour, coated with octadecanoic acid modified-nZnO and the as-prepared wood flour, are presented in Figure 4. In both the spectra, the absorption peaks at 2954 cm^{-1} and 1398 cm^{-1} stemmed from asymmetrical stretching vibrations and symmetrical bending vibrations of $-\text{CH}_3$, respectively. In addition, the peaks at 2916 cm^{-1} and 2846 cm^{-1} were attributed to asymmetrical stretching vibrations and symmetrical stretching vibrations of $-\text{CH}_2$, respectively. All four characteristic peaks confirmed that long alkyl-chains existed on the surface of the wood flour coated with modified-nZnO. The absorption peaks at 1537 cm^{-1} were due to $-\text{COOH}-$ stretching vibrations, whereas those at 1464 cm^{-1} were due to $-\text{COOH}-$ bending vibrations and were induced by the $\text{CH}_3(\text{CH}_2)_{16}\text{COO}-$ groups. The presence of octadecanoic acid on both samples' surfaces was evidenced by the characteristic peaks present in both the spectra. In the high frequency region of Figure 4b, the bands at 3024 cm^{-1} and 3060 cm^{-1} were assigned to the C-H stretching vibrations of benzene ring groups, which were introduced by polystyrene. In the low frequency region of Figure 4b, two typical bands at 696 cm^{-1} and 746 cm^{-1} were due to the C-H bending vibrations of benzene ring groups of polystyrene. Moreover, compared to Figure 4a, in accordance with the characteristic absorption peaks of the benzene rings from polystyrene, the bands at 1599 cm^{-1} and 1452 cm^{-1} were visible in Figure 4b. Based on these observations, it was confirmed that nZnO was successfully functionalized by octadecanoic acid, and that polystyrene was successfully used as an adhesive agent to stick octadecanoic acid-modified nZnO to the surface of the as-prepared wood flour.

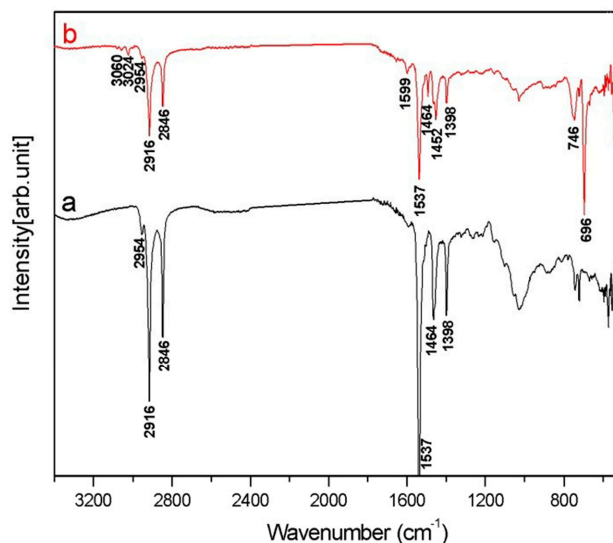


Figure 4. FT-IR spectra of (a) wood flour coated with octadecanoic acid modified-nZnO and (b) as-prepared superhydrophobic/superoleophilic wood flour.

The XPS spectra of the pristine and superhydrophobic/superoleophilic wood flours are shown in Figure 5. For the pristine wood flour (Figure 5a), peaks corresponding to C 1s and O 1s were observed. In comparison, the XPS spectra of the as-prepared wood flour contained a new Zn 2p peak, which accounted for the generation of ZnO. The oxygen content would change when nZnO was introduced into the surface of the wood flour. Based on the XPS spectra, the oxygen intensity in Figure 5a was higher than that in Figure 5b. Therefore, the changes in the C/O proportion were calculated based on the changes in the C and O intensities. In addition, the accurate C/O proportion changed from 65/35% to 83/13%, which resulted in an increase in the C content. In summary, it is inferred that the successful grafting of ZnO with octadecanoic acid was carried out in this study.

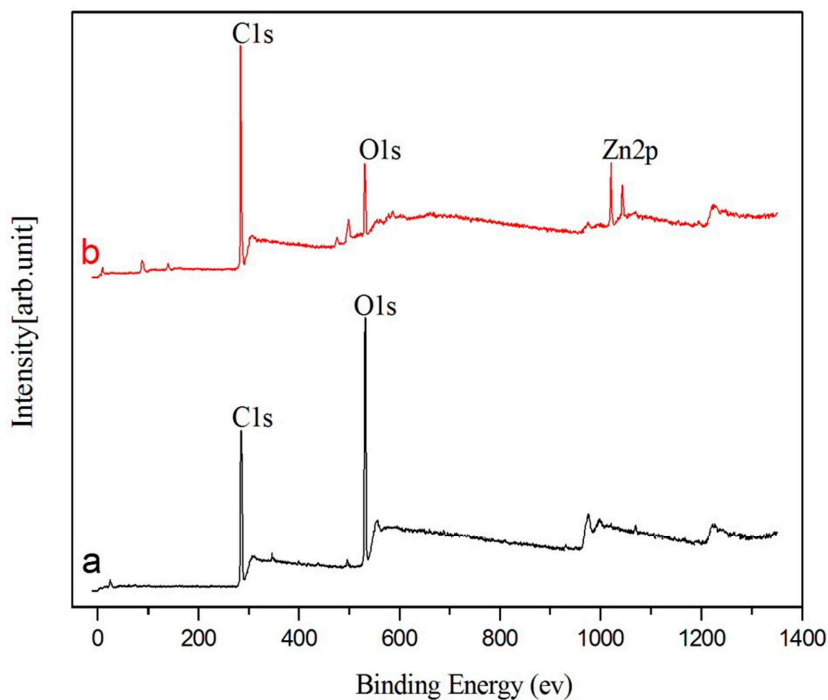


Figure 5. XPS spectra of the (a) pristine wood flour and (b) as-prepared wood flour.

In addition to the characterizations of FT-IR and XPS, the elemental composition of the as-prepared wood flour was analyzed using EDX, and the results are shown in Figure 6. The carbon (C) peak and oxygen (O) peak observed in both the spectra are attributed to the sustainable wooden material. Notably, in comparison with the pristine wood flour, there was a new peak of zinc (Zn) in the as-prepared material, which provided clear evidence of the existence of nZnO on the surface of the superhydrophobic/superoleophilic wood flour.

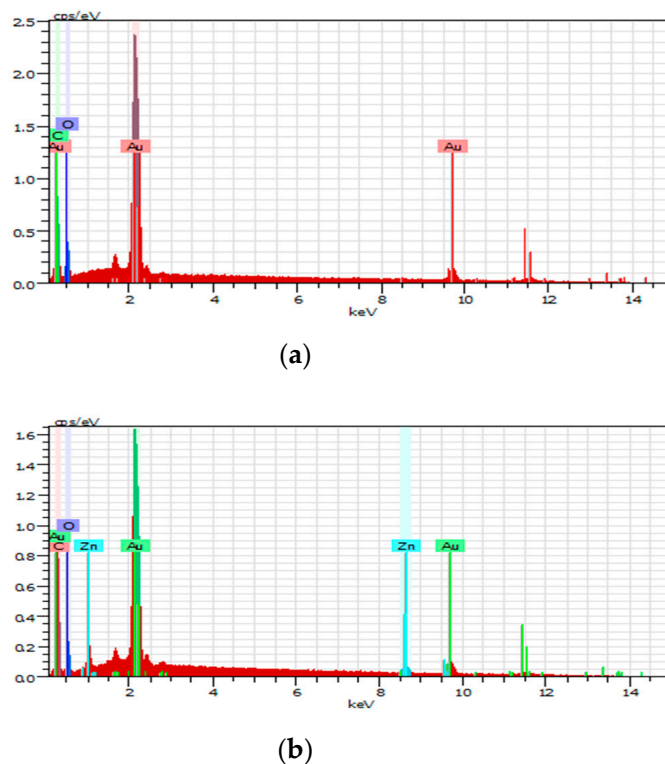


Figure 6. EDX spectra of the (a) pristine wood flour and (b) as-prepared superhydrophobic/superoleophilic wood flour.

3.4. Steadiness and Durability Analysis of the Proposed Superhydrophobic/Superoleophilic Wood Flour

In order to enhance the economic feasibility of the proposed superhydrophobic/superoleophilic wood flour, its environmental durability and chemical steadiness were analyzed. The chemical steadiness was assessed by recording the changes in the contact angles of corrosion solutions on the as-prepared wood flour (see Figure 7a). The as-prepared wood flour reacted with the aqueous solutions at various pH values ranging from 0 to 14 at room temperature for 24 h, and the changes in its contact angles were detected. From Figure 7a, the water contact angles on the sample surface changed within a very narrow range and remained higher than 150° ($152\text{--}156^\circ$). Meanwhile, the oil contact angle remained at 0° . As such, the great superhydrophobicity and superoleophilicity of the as-prepared wood flour was retained in the corrosive solutions. Moreover, the environmental durability was examined under ambient conditions over 150 days. During the storage period, there were no apparent changes in the values of the contact angles of the as-prepared wood flour, showing that the superhydrophobic/superoleophilic wood flour possessed a remarkable environmental durability (Figure 7b). It is therefore concluded that the superhydrophobic/superoleophilic wood flour prepared in this study has a favorable chemical steadiness and environmental durability, which are due to the viscosity of the polystyrene present on the surface of the as-prepared wood flour fibers.

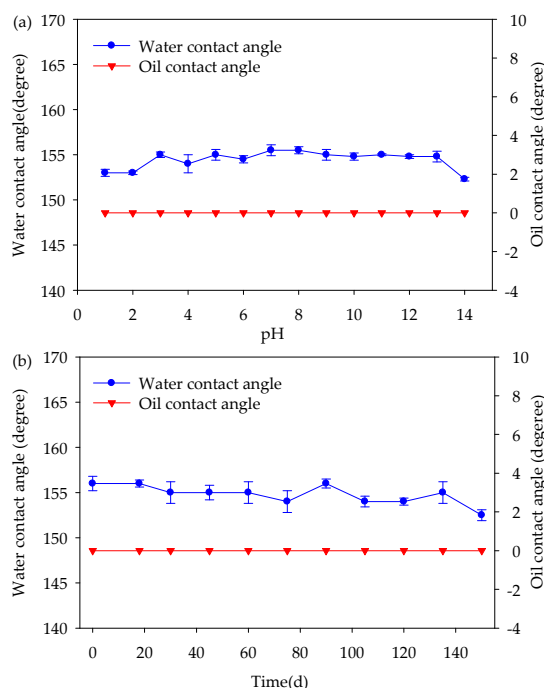


Figure 7. (a) The contact angles of the superhydrophobic/superoleophilic wood flour with different pHs of aqueous solution; (b) the contact angles of the superhydrophobic/superoleophilic wood flour for the storage evaluation in an ambient environment.

3.5. Application of the Green Superhydrophobic/Superoleophilic Wood Flour in Water-Oil Separation

In view of its dramatic chemical steadiness and environmental durability, the as-prepared wood flour has a great potential for application to oil-containing wastewaters. Figure 8 illustrates the procedure for water-oil separation using the superhydrophobic/superoleophilic wood flour as an oil sorbent. A certain amount of as-prepared wood flour was placed in the mixture of water and diesel oil, which was dyed red using Sudan III for ease of observations. As a result, the superhydrophobic/superoleophilic wood flour could effectively adsorb the diesel oil, while completely repelling water. The red diesel oil was fully adsorbed by the as-prepared wood flour within a few seconds. The transparent region on the water surface indicated that the diesel oil was effectively separated from water. After the adsorption, the red wood flour was recovered from the surface of the water.

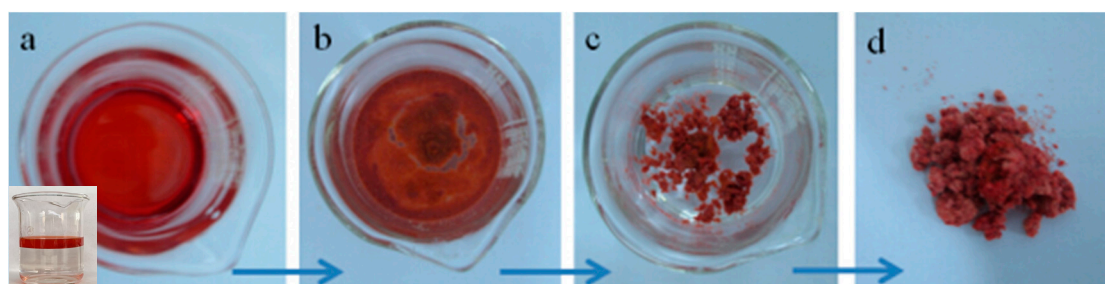


Figure 8. Photographs of the procedure for water-oil separation using the superhydrophobic/superoleophilic wood flour as an oil sorbent: (a) water and diesel oil mixture (diesel oil was dyed with Sudan III); (b) right after the addition of the as-prepared wood flour; (c) after a few seconds, the wood flour filled with red diesel oil floated on the surface of the water; and (d) the red oil-adsorbing wood flour recycled from the surface of the water.

The adsorption capacities of the pristine and as-prepared wood flours for oils and organic solvents are shown in Figure 9a.

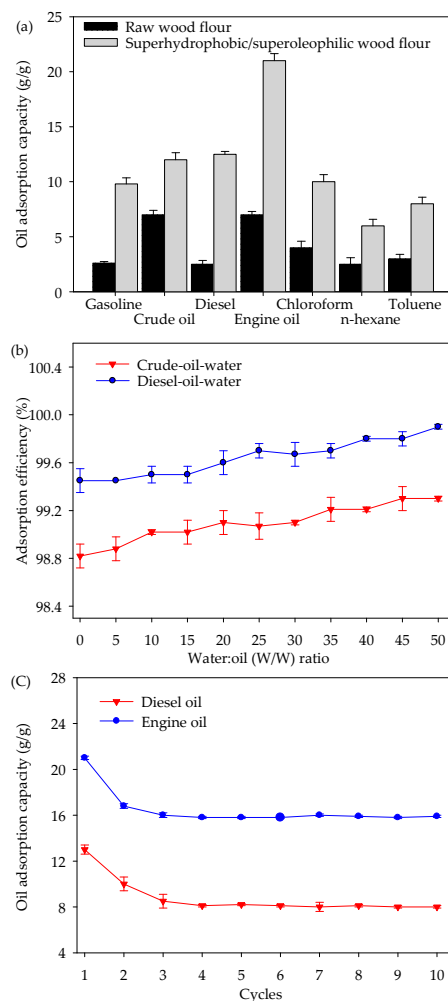


Figure 9. (a) The maximum adsorption capacities of the pristine and superhydrophobic/superoleophilic wood flours for various oils and organic solvents at room temperature. (b) The variation in adsorption efficiency (%) for diesel oil and crude oil with various mass ratios of water-to-oil. (c) The reusability of the superhydrophobic/superoleophilic wood flour for diesel oil and engine oil applications.

For the same oil or organic solvent, the maximum oil adsorption capacity of the as-prepared wood flour was almost three times that of the pristine wood flour, which may be due to the pretreatment of the wood flour and the composites' coating on the surface of the as-prepared wood flour. As such, the oil sorption capacity of the superhydrophobic/superoleophilic wood flour was greatly enhanced. In addition, the oil adsorption efficiency of the as-prepared superhydrophobic/superoleophilic wood flour was examined. The variation in the adsorption efficiency (%) for diesel oil and crude oil for various mass ratios of water-to-oil is shown in Figure 9b. The adsorption efficiency ranged from 98% to 100%, which was mainly potentially due to the wastage of oil with a high viscosity decreasing with the increase in the proportion of water.

The reusability of the proposed material is an important issue with regards to its practical applications. After the adsorbed oils were removed using acetone, the superhydrophobic/superoleophilic wood flour could be reused for many cycles. As seen in Figure 9c, the sorption capacity of the reused wood flour for both diesel and crude oil decreased to 83% and 77% for the second and third cycles, respectively, which was probably due to the trace residual oils left in the fibers of the as-prepared wood flour. The oil adsorption capacity of the superhydrophobic/superoleophilic wood flour remained almost unchanged after three cycles, exhibiting a good reusability. Moreover, after several cycles, the as-prepared wood flour can still be used as an environment-friendly and low-cost material with satisfactory removal efficiencies for diesel and crude oils. Finally, a comparison

of the oil adsorption capacities between the proposed superhydrophobic/superoleophilic wood flour and the bio-materials reported in the literature is presented in Table 1. It can be seen that the green superhydrophobic/superoleophilic wood flour had a relatively high oil adsorption capacity, demonstrating that the as-prepared wood flour has a great potential for being used as an oil adsorbent due to its excellent superhydrophobic/superoleophilic characteristics.

Table 1. A comparison of the oil adsorption capacities of various adsorbents.

Sorbent	Adsorption Capacity (g Crude Oil/g Fiber)	Reference
As-prepared wood flour	16.89	This study
Acetylated peat moss	8.00	[2]
Sphagnum Dill	5.80	[38]
Black rice husk ash	6.22	[39]
Chitosan based polyacrylamide	2.30	[40]
Carbonized rice husks	6.00	[41]
Recycled wool-based nonwoven material (RNWM)	11.50	[42]
Superhydrophobic/superoleophilic polyurethane sponge	44.00	[43]

4. Conclusions

In this work, green superhydrophobic/superoleophilic wood flour with a WCA of 156° and OCA of 0° was prepared for an efficient oil removal from contaminated wastewater. The water-resistance and oil-adsorption properties of the as-prepared superhydrophobic/superoleophilic wood flour were comprehensively ascribed to the synergistic effect of octadecanoic acid modified-nZnO on the micron-fiber surface and glutinous polystyrene employed to attach the modified-nZnO aggregates to the surface of fibers. In addition, the as-prepared wood flour could effectively adsorb oil, while completely repelling water, demonstrating that it has a great potential for use as an effective oil adsorbent from wastewater. For the same oil or organic solvent, the maximum oil adsorption efficiency of the proposed wood flour was almost three times that of the pristine wood flour. The adsorption efficiency ranged from 98% to 100% for the proposed wood flour. After the adsorbed oils were removed using acetone, the green superhydrophobic/superoleophilic wood flour could be reused many times. Notably, the superior chemical steadiness and environmental durability of the proposed wood flour with environment-friendly characteristics add to its commercial feasibility.

Author Contributions: S.-H.H. conceived designed the experiments. D.Z. performed the experiments. X.T. performed the experiments and wrote the paper. H.Q. and G.C. analyzed the data. F.L. contributed in materials preparation and their surface analysis. All the experiments were performed under the supervision of S.-H.H. All authors read and approved the final manuscript.

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Conflicts of Interest: Declare conflicts of interest or state “The authors declare no conflict of interest.”

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