

Preparation and Study of Superhydrophobic Materials Based on Natural Waste and Biomimetic Surface Structure

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Abstract

Cellulose fiber derived from agricultural and urban waste are eco-friendly and low-cost sustainable materials. Notably, cellulose fibre powder is hydrophilic originally, while in this document, we discovered a method to convert cellulose fibers into hydrophobic material. Based on the Cassie-Baxter model, we use extracted cellulose fibre powder or other waste material powder that contains cellulose fibre, which features a lightweight, super hydrophobic coating. It's witnessed that the results of extracted cellulose fibre show the best hydrophobic performance. This document will now elaborate on the materials used in the experiments, including their properties, structures, and sources, and then delve into the detailed experimental procedures.

Keywords

Super Hydrophobic, Waste, Hydrothermal Reaction, Sustainable Development, Polymer and Materials Science, Cellulose

1. Introduction

With the growing global emphasis on sustainable development, people start to pay more attention to waste source recycling. The development of green and biodegradable functional materials has become the core research direction in the field of polymer and materials science. As the most abundant natural biopolymers on Earth, cellulose fibers can be extracted from wastes such as coffee grounds and dead leaves [1] [2]. In daily life, people often wear cotton-linen garments (which contain cellulose fiber), they are comfortable to wear, however, they dry more

slowly than other clothes in open air conditions, as cellulose fibres are hydrophilic.

Against this background, we sought to explore strategies to render cotton linen fabrics superhydrophobic.

First of all, we need to understand the basic theory; as a result, we search on the Internet, then we realize the reason that cellulose fiber is hydrophilic is owing to plentiful hydroxyl (OH) groups distributed on the cellulose fibers' molecular chains, which are capable of forming strong hydrogen bonds with water molecule [3]. Therefore, theoretically, the removal of the hydroxyl (OH) groups renders cellulose fiber chemically more inclined towards hydrophobicity.

On the other hand, we found that the lotus-leaf effect can make material more hydrophobic physically (the hydrophobic properties of lotus leaves have been noticed in Chinese literature for centuries. In the poem Zhao Jun Yuan-Yong He Shang Yu, the Chinese poet Yang Wanli described how raindrops behave on a lotus leaf: “却是池荷跳雨，散了真珠还聚。聚作水银窝，泻清波。” The poem describes raindrops bouncing on the lotus leaf, separating and then coming together again like pearls before rolling away. This description reflects the ability of the lotus leaf to keep water droplets from remaining on its surface. The phenomenon has inspired modern research on the lotus effect and development of superhydrophobic surfaces.). The lotus leaf's hydrophobicity can be explained by the Cassie-Baxter model (as **Figure 1** showed); air is locked within the gaps of the rough surface of the lotus surface (while the cellulose fiber's surface is smooth under a microscope) [4].

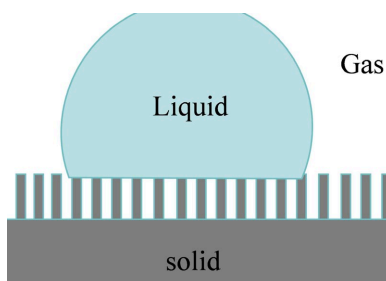


Figure 1. Illustrates that water droplets contact only the elevated solid peaks—a phenomenon significantly influenced by surface tension—rather than penetrating all surface voids.

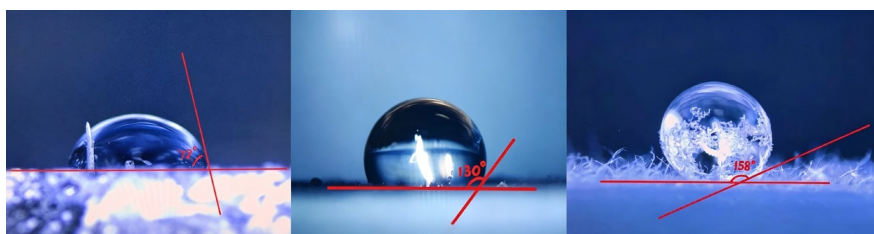


Figure 2. Show the contact angle when it's hydrophilic, hydrophobic and super hydrophobic.

Besides this, there is one more thing that needs to be clear (from **Figure 2**): the criterion for distinguishing hydrophilic, hydrophobic, and super hydrophobic

surfaces lies in the water contact angle. If the water contact angle is less than 90 degrees, it's hydrophilic; if it's more than 90 degrees but less than 150 degrees, it's hydrophobic; if it's larger than 150 degrees, it's superhydrophobic [5].

To sum up, to achieve superhydrophobic cotton-linen fabrics, theoretically, a method is required to disrupt the smooth microstructure of cellulose fibres, generate a rough surface and facilitate the removal of surface hydroxyl groups. In this case, the hydrothermal-reaction system with HMTA and zinc nitrate as reactants is the best solution. High temperature-induced dehydration consumes partial surface hydroxyl groups and lowers the surface energy. Hydrothermal etching disrupts the intrinsic microstructure of cellulose fibres for increased surface roughness [6]. What's more, the reaction produces zinc oxide, which grows in situ to build the micro-nano rough surface on cellulose fibres to trap air pockets for the Cassie-Baxter state and covers exposed hydroxyl groups on the fibre surface and cuts down surface energy, helping the cotton-linen material gain super hydrophobic performance.

2. Experiment Section

2.1. Materials

Microcrystalline cellulose (MCC, 9004-34-6, $C_6H_{10}O_5$)_n powder with a high degree of crystallinity (100% polysaccharide, found by french chemist Anselme Payen in 1838). Hexamethylenetetramine (HMTA, 100-97-0, also known as Methenamine or Urotropin, $C_6H_{12}N_4$, first discovered and synthesized in 1859 by the Russian chemist Aleksandr Butlerov). Zinc nitrate ($Zn(NO_3)_2$, 10196-18-6).

All chemicals were directly used as received without further purification.

2.2. Experiments

2.2.1. Fabrication of Superhydrophobic Coatings from Cellulose and Natural Waste Particles

Cellulose powder (provided by our professor) was used without any further treatment. Several natural waste materials, including coffee grounds, Platanus bark, sunflower stems, white-barked pine needles, ash twigs, and cedar needles, were initially collected and tested. After that, we found that only the coffee grounds and Platanus bark gave us good superhydrophobic results; therefore, we decided to focus on these two for the rest of the study. The coffee grounds were collected from a local coffee shop (Luckin Coffee) and used as received.

To prepare the natural particles, we first dried the coffee grounds and Platanus bark in an electric thermostatic blast drying oven at 80°C for 4 hours to remove moisture, which made the materials brittle and easy to grind. The dried samples were first broken into small pieces manually and then ground in an agate mortar for about 1 hour until fine powders were obtained. The ground powders were used without sieving. Particle-size distribution analysis was also not performed; non-uniform particle size may contribute to experimental variability.

The reaction solution was prepared as follows. First, 100 ml of commercial bot-

tled water was measured with a graduated cylinder and transferred to a beaker. Then, 0.74 g of zinc nitrate hexahydrate $[\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}]$ was weighed on weighing paper using an electronic balance and added to the water, followed by 0.35 g of hexamethylenetetramine (HMTA), which was weighed with a laboratory spoon because of its hygroscopic property. The mixture was stirred with a glass rod until the reagents were completely dissolved.

For the cellulose samples, 3.0 g of cellulose powder was placed in a beaker, mixed with the reaction solution, and stirred with the glass stirrer for 10 min. The suspension was then transferred into a Polytetrafluoroethylene (PTFE) liner. For the coffee grounds and Platanus bark samples, we adopted a slightly different approach to avoid material loss during transfer: 3.0 g of powder was directly placed into the liner, and the reaction solution was poured in afterward. The liner was gently shaken to disperse the powder evenly.

The PTFE liner was then placed inside an aluminum-alloy autoclave body and sealed. The autoclave was put into an electric thermostatic drying oven at 95°C for 12 hours to carry out the hydrothermal reaction.

After the reaction, the autoclave was taken out and cooled under running tap water until it could be safely opened. The PTFE liner was removed, and the product was collected by centrifugation. The solid was washed with tap water three to four times. It was then dried at 73°C overnight.

The dried powders were then subjected to vapor-phase fluorination. The powders were placed on the ceramic plate inside a vacuum desiccator, and a small amount of fluorosilane was dropped into a glass vial at the bottom of the desiccator using a dropper. The desiccator was sealed and evacuated for several minutes to allow the fluorosilane vapor to diffuse. The system was kept at room temperature for 8 hours. Finally, the powders were removed.

2.2.2. Fabrication of Hydrophobic Polydimethylsiloxane (PDMS) Surfaces from Natural Leaf and Artificial Mold Templates

Two types of templates were employed to fabricate micro-structured PDMS surfaces: natural Platanus leaves and a plastic artificial mold featuring surface micro-protrusions.

First, freshly fallen Platanus leaves were collected, rinsed with tap water to remove surface contaminants, wiped dry, and then cut into desired shapes with scissors.

For the leaf-templated samples, PDMS base and curing agent were mixed at a weight ratio of 10:1. A beaker was placed on an electronic balance, and the balance was tared. After that, 54.5 g of PDMS base was weighed into the beaker. The curing agent was then drawn with a syringe, and 5.43 g was added to the same beaker. The mixture was stirred manually with a glass rod for approximately 10 minutes until homogeneous. To eliminate entrapped air bubbles, the beaker was maintained under vacuum for about 40 minutes. Then, the degassed PDMS mixture was poured into a plastic Petri dish close to the bottom, and the pre-cut leaf pieces were gently pressed onto the PDMS surface. The assembly was left at room tem-

perature overnight to allow complete curing. After curing, the leaves were carefully peeled off from the PDMS surface. Among four leaf-templated PDMS preparations, only one yielded an intact surface with minimal bubble defects and was selected for subsequent experiments. Inconsistent sample quality may stem from incomplete degassing during PDMS curing, uneven graphene deposition or template-surface imperfections. No follow-up experiments were carried out to confirm the dominant cause.

For the artificial mold samples, the same PDMS mixture (10:1 weight ratio) was prepared following the identical procedure. First, the plastic white mold with surface microprotrusions was wrapped with transparent adhesive tape around its perimeter for two to three circles to prevent leakage. Then, the freshly mixed PDMS mixture was poured into the mold to a thickness of 3 mm. The mold was placed in the vacuum pump for about 40 minutes to remove bubbles; the sample was then placed in the electric thermostatic drying oven for drying (the drying time and temperature were adjusted based on the experimental conditions). After drying, the PDMS film was carefully peeled off from the mold.

Both the leaf-templated and artificial-mold PDMS samples were then coated with graphene. The graphene powder was applied manually onto the PDMS surfaces before the hydrothermal treatment.

Subsequently, each sample was placed into a PTFE liner, and the reaction solution—prepared by dissolving 0.74 g of zinc nitrate hexahydrate and 0.35 g of hexamethylenetetramine in 100 ml of commercial bottled water—was added. The PTFE liner was sealed inside an aluminum autoclave and placed in an electric thermostatic drying oven at 95°C for 11 hours. After the hydrothermal reaction, the autoclave was cooled under running tap water, and the PTFE liner was removed. The PDMS samples were then taken out, rinsed with tap water four times. Afterwards, the cleaned PDMS samples were subjected to vapour-phase fluorosilane modification following the identical procedure described in Section 2.2.1.

2.3. Characterization

The surface morphology of the 4 samples, including powders and PDMS samples was observed by an optical microscope (China). Digital images were captured by a camera. The water contact angles (WCA) were measured using a protractor at ambient temperature ($22^{\circ}\text{C} \pm 1^{\circ}\text{C}$). A 10 μL water droplet was dropped onto the surfaces. For each sample, we measured water contact angles (WCA) at three positions (once per position), and the average value was reported. To reduce the material waste and water evaporation, the measurement was processed and recorded within 15 seconds after the droplets contacted the surfaces.

Due to equipment limitations, scanning electron microscopy (SEM), energy-dispersive X-ray spectroscopy (EDS), X-ray diffraction (XRD), and Fourier-transform infrared spectroscopy (FTIR) were not performed in the study. Therefore, these mechanisms proposed in this study are based on interpretation of each sample's wettability changes, but they haven't been proved by experiments.

3. Results

3.1. Biomass Route

Superhydrophobic surfaces require both low surface energy materials and micro-nano-particle features [7]-[9]. To find the natural materials that can make superhydrophobic coatings, we evaluated these materials (mentioned in 2.2.1) and selected three (platanus barks, coffee grounds, and cellulose powder) based on literature (Figure 3). Cellulose exhibited the highest WCA (153.4°), which may be attributed to its hierarchical micro-nanostructure that provides sufficient space for ZnO growth and air storage. The coffee grounds showed a lower WCA (130.1°) possibly due to their dense particle structure which contains cellulose/hemicellulose skeleton and a small amount of lignin that limits air storage [10]. The platanus bark powder has moderate hydrophobic performances ($\text{WCA} = 144^\circ$), which may benefit from its porous structure (as observed in similar biomass). All the powders underwent hydrothermal reactions (Figure 4) and fluorosilane modifications (Figure 5).

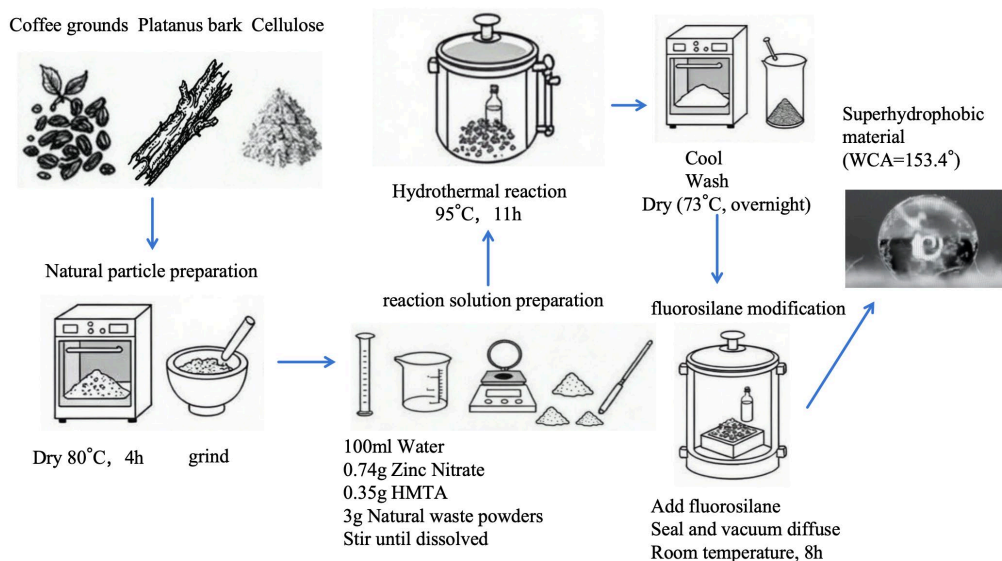


Figure 3. Schematic illustration showing the fabrication process of hydrophobic/superhydrophobic materials using coffee grounds, platanus bark and cellulose as raw materials, including particle preparation, hydrothermal reaction and fluorosilane modification.

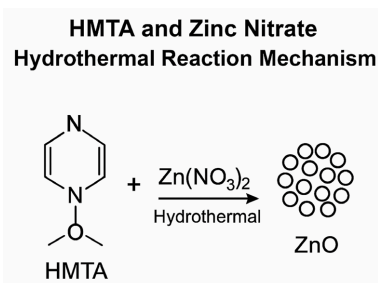


Figure 4. Schematic diagram of hydrothermal reaction mechanism between HMTA and zinc nitrate for the formation of ZnO particles.

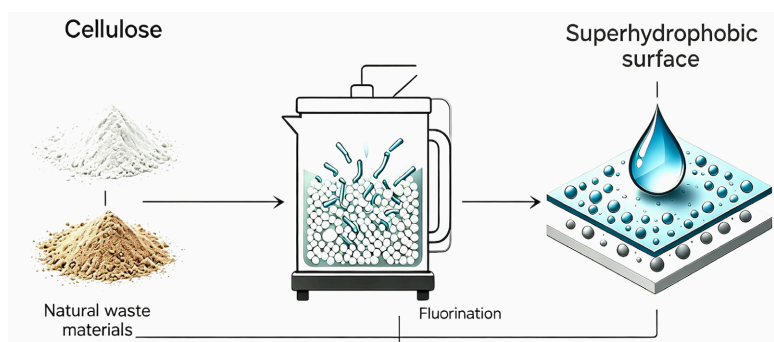


Figure 5. Diagram of fluorosilane modification mechanism aimed at forming a rough, low-surface-energy surface and creating hydrophobic/superhydrophobic surface.

Without direct microscopic characterization (e.g. SEM), we cannot definitively confirm the surface morphology and ZnO growth. The above explanations are based on wettability observations and supported by existing literature.

Figure 6 shows the photos of three materials (cellulose powders, coffee grounds, and platanus bark powders) before and after the hydrothermal and fluorosilane modification. Each material, due to the reaction, changed color slightly. The color of cellulose powders changed from light grey to pure white (**Figure 6(a)**), the colour of coffee grounds changed from dark brown to brown (**Figure 6(b)**), and some white particles appeared on platanus bark powder compared to its original one (**Figure 6(c)**). These color changes may indicate the surface modifications, although we should use further analysis such as EDS or FTIR to prove these observations.

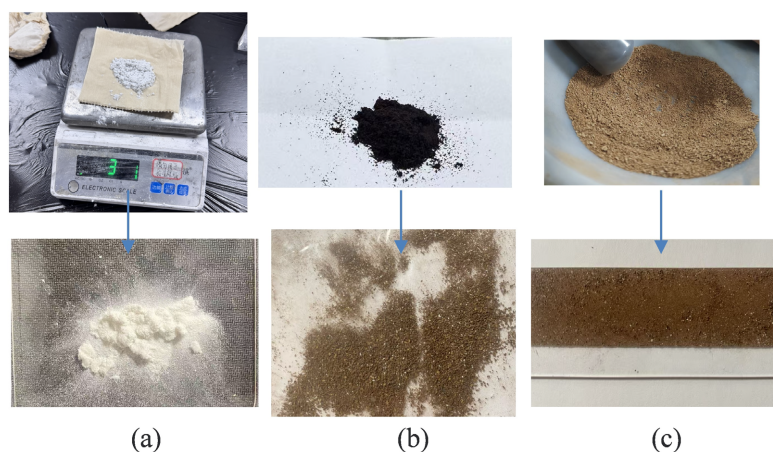


Figure 6. Visual changes of cellulose powders, coffee grounds, and platanus bark powders before and after chemical modifications.

Figure 5 shows the optical images of water droplets that contact three treated materials: cellulose powders, coffee grounds, and platanus bark powders. The water contact angles were measured at three positions (once per position). In **Figure 7(a)**, the water contact angle reached $153.4^\circ (\pm 1.4^\circ)$; In **Figure 7(b)**, the water contact angle reached $130.1^\circ (\pm 2.1^\circ)$; In **Figure 7(c)**, the contact angle reached

144° ($\pm 1.8^\circ$). The droplets on the surface have regular spherical shapes and stand still on the surfaces without spreading, which can indicate that the materials have superhydrophobic/hydrophobic surfaces.

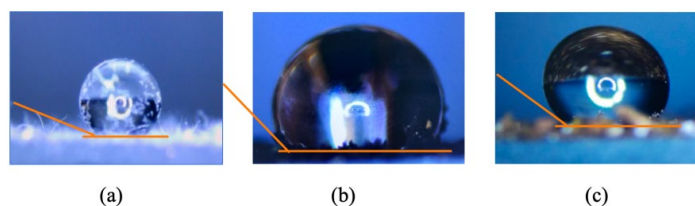


Figure 7. Photographs of water droplets on each material (cellulose powders, coffee grounds, and platanus bark powders), showing hydrophobic/superhydrophobic performances.

Among these materials, cellulose powder showed the highest water contact angles $\text{WCA} = 153.4^\circ (\pm 1.4^\circ)$, which exhibited a superhydrophobic effect ($\text{WCA} > 150^\circ$).

The coffee grounds and platanus bark powders exhibited hydrophobic but not superhydrophobic effects ($\text{WCA} < 150^\circ$). The cellulose powder's excellent performance may be attributed to its hierarchical micro-nanostructure which may provide sufficient space for storing air.

3.2. Polydimethylsiloxane (PDMS) Surfaces from Natural Leaf and Artificial Mold Templates

Figure 8 and **Figure 9** illustrate the soft-lithography method and the replicated structures. This method is divided into two separate routes: (i) natural leaves template and (ii) artificial molds with micro-protrusions [11] [12]. We made four leaf-templated PDMS preparations, but only one yielded an intact surface. Other leaf samples contain numerous air bubbles which may be due to incomplete degassing (**Figure 9(a)**). We made two PDMS samples from artificial molds (**Figure 9(b)**). All the samples were successfully replicated without bubbles. Samples from both routes underwent graphene coating (**Figure 10**), hydrothermal reactions and fluorosilane modifications.

We tested one of the PDMS samples which were replicated from artificial molds. **Figure 11(a)** shows the morphology of a PDMS sample which replicated from templates. It exhibited micro-protrusions on the PDMS surface. The droplet on the surface has a regular spherical shape and stands still on the surface without spreading. The water contact angle reached approximately $130^\circ (\pm 2.2^\circ)$. In **Figure 11(b)**, when we slightly tilted the materials, the water droplets rolled down quickly without obvious deformation. This characteristic suggests the successful micro-structure replication that potentially aids the hydrophobic performance.

Based on the definition stated in the introduction, ($90^\circ - 150^\circ$: hydrophobic; $>150^\circ$: superhydrophobic), the PDMS is classified as hydrophobic rather than superhydrophobic. Although it underwent fluorosilane treatment, it did not reach superhydrophobic threshold likely due to uneven coverage of graphene reducing the exposure area for fluorosilane.

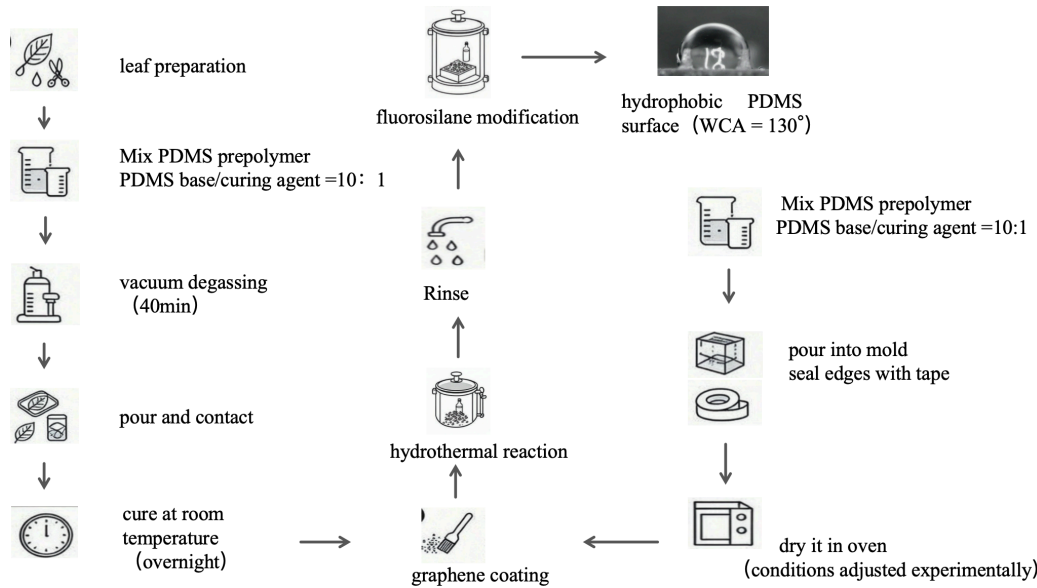


Figure 8. Fabrication process of hydrophobic/superhydrophobic PDMS surfaces by using soft-lithographic method (replicating microstructures from tree leaves and artificial molds).

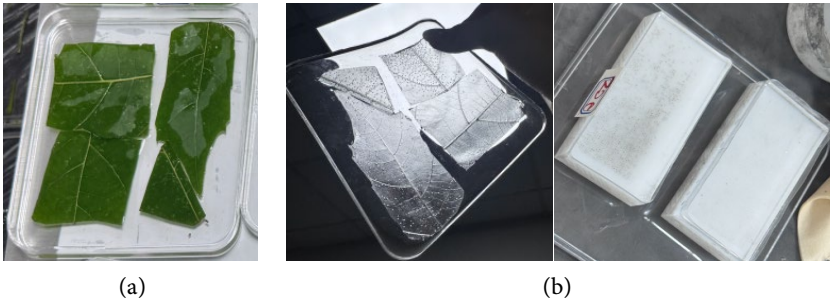


Figure 9. Replicated structures from tree leaves and artificial molds.

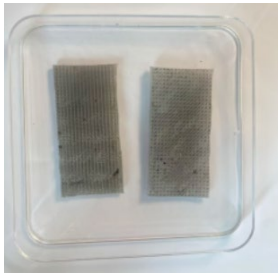


Figure 10. Graphene-coated replicated structures from artificial molds.



Figure 11. Photographs of water droplets on treated PDMS showing contact angles and hydrophobic/superhydrophobic performance testing.

4. Conclusions

In this study, superhydrophobic particles were prepared from cellulose powder and several natural waste materials through a hydrothermal process followed by fluorination. The samples were observed using an optical microscope.

Among the tested materials, cellulose powder and Platanus bark showed the best hydrophobic performance. The measured water contact angles were 153.4° for cellulose powder and 144° for Platanus bark, while coffee grounds showed a lower contact angle of 130.1°. Hydrophobic PDMS surfaces were fabricated by replicating textures from natural Platanus leaves. After graphene-coating, hydrothermal treatment and fluorination, the PDMS surfaces became more hydrophobic. However, surface coverage was not uniform, leading to variable hydrophobic performance across different regions. Only one out of four leaf-templated PDMS samples yielded good-quality surfaces, whereas the other three contained substantial bubble defects.

Based on measured wettability and published literature, it is hypothesised that hydrothermal treatment and fluorination may introduce favourable surface roughness and low-surface-energy species to enhance water repellency. Direct structural and surface-chemical characterization (e.g. SEM, XPS or FTIR) was not performed in this work, so these proposed mechanisms cannot be confirmed by our experimental data. This study demonstrates that cellulose-containing waste materials are promising low-cost, environmentally friendly candidates for fabricating superhydrophobic surfaces.

Several limitations should be acknowledged. Although contact-angle values were averaged from multiple measurement positions, control experiments were not conducted to separate the contribution of each processing step. The observation that coatings are mechanically fragile comes only from qualitative manual handling, without quantitative durability tests. In future work, further experiments are required to investigate coating stability and real-world practical performance.

Author Contributions

Lei Wang supervised this project. Ziyang Guo wrote the experimental section and the conclusion of the manuscript, and completed the collection and sorting the references. Haonan Wang was responsible for writing the abstract and introduction of the manuscript. Shuning Zhang performed material characterization and drafted the results section. All three authors jointly contributed to figure preparation.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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