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A Review of Hydrophobic Materials: From Nature-Inspired Design to Future Technological Applications

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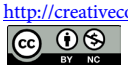

Abstract

Hydrophobic materials have received much interest due to their excellent water-repellent characteristics and various uses such as self-cleaning, anti-corrosion, anti-icing, oil-water separation, biomedical engineering and flexible electronics. This paper gives an overview of the theoretical bases, nature-inspired design rules, artificial materials, fabrication methods and practical applications of hydrophobic materials. In addition to reviewing the theoretical principles, bioinspired designs, material systems and fabrication strategies of hydrophobic materials, this paper includes an illustrative experimental case study on cellulose-based hydrophobic composites prepared through hydrothermal synthesis and biomimetic PDMS replication. The experimental section demonstrates how the reviewed design concepts can be translated into practical material preparation. At last, the present problems and potential developments are also discussed.

Keywords

Hydrophobic Materials, Superhydrophobic Surfaces, Biomimetic Design, Lotus Effect, Wettability

1. Introduction

1.1. Background and Significance

1.1.1. What Are Hydrophobic Materials?

Those items having excellent water repelling characteristics are named hydrophobic. The surface has a low surface energy and a fine/minute roughness, causing

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water drops to be nearly spherical and slide off the surface rather than spreading and wetting the substance. A surface is regarded as hydrophobic when the water contact angle is larger than 90˚; the superhydrophobic materials have a contact angle over 150˚ with very small water sliding angles. Such materials avoid the adhesion of liquids to the surface and separate water from the solid surface effectively [1]-[3].

1.1.2. Why Are Hydrophobic Materials Important?

They provide extensive practical benefits in our daily life, in industries and in environmental protection: [1]-[3].

1) Water droplets which roll off can remove dust, dirt and pollutants, thus decreasing the expense of manual cleaning for buildings, vehicles and optical lenses.

2) Resistance to corrosion and rust: Prevent rainwater, seawater and moisture from contacting metal substrates, reduce oxidation and electrochemical corrosion of steel, ships and pipelines.

3) Anti-icing and anti-fogging: Water does not form a continuous layer on the surface, preventing ice formation on aircraft wings, power transmission lines and fog on glass mirrors.

4) Reduction of fluid drag: Applied to the ship hulls and inner walls of pipelines to decrease the frictional resistance between liquids and solids, thus saving energy in transportation and fluid conveyance.

5) Repel water and absorb oil selectively, which is suitable for quick treatment of marine oil spills and industrial oily wastewater.

6) Reduce water adhesion and protein adsorption on medical catheters and implants to decrease bacterial adhesion and infection risks.

1.1.3. Current Research Hotspots of Hydrophobic Materials

1) Durable superhydrophobic coatings can prevent the loss of hydrophobic layers caused by friction, erosion or UV radiation. People try to bond micro-nano structures firmly to the substrate in order to enhance the mechanical stability, chemical resistance and long service life in outdoor and industrial conditions.

2) Materials which change their wetting properties from hydrophobicity to hydrophilicity according to stimuli such as temperature, pH, light or electric fields are developed. These materials are applied in the control of liquid movement, smart microfluidic chips and adjustable oil-water separating devices.

3) Bio-based and eco-friendly hydrophobic materials substitute expensive and non-biodegradable fluorinated chemicals with renewable resources like plant waxes, cellulose and chitosan. The purpose is to produce low-cost, biodegradable hydrophobic coatings with minimal environmental risks.

4) Mix hydrophobicity with other functions such as flame retardancy, antibacterial property, thermal insulation or electromagnetic shielding. These materials are suitable for use in fire-resistant building coatings, anti-biofouling marine equipment and waterproof electronic devices.

5) Investigate easy and reproducible manufacturing techniques such as spray coating, electrospinning and imprinting, rather than using costly laboratory lithography, to achieve large-scale production of hydrophobic materials for the popularization in civil and industrial fields.

6) Hydrophobic materials with good adaptability to extreme environments have been designed to maintain stability under high temperature, strong acid/alkali, high salt or low-temperature icing conditions for aerospace, deep-sea devices and chemical industrial equipment [1]-[3].

1.2. Scope and Organization of This Paper

This paper combines a comprehensive review of hydrophobic materials with an experimental case study. Chapters 2-5 summarize the theoretical principles of wettability, bioinspired design strategies, representative hydrophobic materials, and common fabrication methods reported in the literature. Chapter 6 presents an illustrative experimental investigation on cellulose/ZnO composites and biomimetic PDMS replicas, demonstrating how hierarchical surface structures and low-surface-energy modification can be applied in practice. Rather than providing a comprehensive mechanistic study, the experimental section is intended to serve as an example that connects the reviewed concepts with practical material preparation. Chapters 7 and 8 further discuss representative applications, current challenges, and future research directions.

2. Fundamentals of Hydrophobicity

2.1. Wettability

Contact Angle

The contact angle is the angle formed at the point where a liquid drop contacts a solid surface, observed from the liquid side. It reflects the wettability of the solid by the liquid. If the contact angle is smaller than 90˚, the solid is hydrophilic; the water spreads and covers the surface. When the contact angle is larger than 90˚, the solid is hydrophobic; the water forms drops and does not spread easily. The surfaces with a contact angle over 150˚ and a small sliding angle are called superhydrophobic. This criterion is usually employed to evaluate the hydrophobicity of materials [1]-[3].

Surface Energy

Surface energy is the additional energy that the molecules on the surface of a material have in comparison with those in the interior of the material. The surface molecules have an imbalance of intermolecular forces, therefore the surface tends to reduce its total energy. Low surface energy substances repel polar liquids such as water, which is the basic chemical basis for hydrophobicity. Examples of low surface energy substances are fluorocarbons and long-chain alkanes. In order to obtain hydrophobic surfaces, researchers often decrease the surface energy by chemical modification, together with micro-nano roughness to enhance the water repellent property [1]-[3].

2.2. Classical Wetting Models

The wetting behavior of liquids on solid surfaces is an important issue in interface science and materials engineering. Many theoretical systems have been established to explain and predict the contact angle and wetting conditions in the last century. The Young model, the Wenzel model and the Cassie-Baxter model are the most famous and commonly used theories. From perfectly smooth surfaces to real rough surfaces, these models take into account the essential effects of surface roughness and multi-phase interfaces gradually. In this section, the basic concepts, assumptions and development of these three fundamental wetting models are described in detail, followed by a summary and comparison of their usages [1]-[3].

2.2.1. Young Model

The Young model describes the equilibrium wetting behavior of a liquid droplet on an ideal smooth, chemically homogeneous and rigid surface. Under these assumptions, the contact angle is determined by the balance of interfacial tensions at the three-phase contact line. The Young equation therefore represents the intrinsic wettability of a material and serves as the theoretical basis for subsequent wetting models. However, because practical engineering surfaces usually possess surface roughness and chemical heterogeneity, the Young model cannot accurately describe the wetting behavior of real superhydrophobic materials [2].

2.2.2. Wenzel Model

The Wenzel model extends the Young equation to rough but chemically homogeneous surfaces. It assumes that the liquid completely penetrates the surface asperities so that the entire rough surface is wetted. Under this condition, surface roughness amplifies the intrinsic wettability of the material: hydrophilic surfaces become more hydrophilic, whereas hydrophobic surfaces become more hydrophobic. Nevertheless, the Wenzel model cannot explain situations where air remains trapped beneath the droplet, which commonly occurs on superhydrophobic surfaces [4].

2.2.3. Cassie-Baxter Model

The Cassie-Baxter model describes a composite solid-air interface in which air pockets are trapped beneath the liquid droplet. Because only part of the droplet contacts the solid surface, the apparent contact angle is substantially increased while the sliding angle is reduced. This model is widely used to explain the excellent water repellency observed on lotus leaves and many biomimetic superhydrophobic surfaces. However, external pressure, droplet impact or mechanical damage may destabilize the trapped air layer, resulting in a transition from the Cassie–Baxter state to the Wenzel state [5].

2.3. Factors Affecting Hydrophobicity

The hydrophobicity of a solid surface, particularly superhydrophobicity (which is characterized by a contact angle larger than 150 degree and a sliding angle smaller

than 10 degree), is not attributed to a single factor. It depends on the joint influence of surface chemical composition and the microscale geometric structure. This paragraph explains the four important factors affecting the wetting properties [2] [3].

2.3.1. Surface Chemistry and Surface Energy

Surface chemistry is important for assessing the initial wettability of a material. According to thermodynamics, the surface energy affects the equilibrium state at the solid-liquid-gas interface. Materials with low surface energy (such as fluorocarbons, silanes and some hydrocarbons) have weak interaction with water molecules, leading to a small contact area between the liquid and the formation of droplets. The maximum water contact angle on a perfectly smooth surface is about 120˚ (achieved by coating the surface with -CF_3 groups), which is restricted by the surface chemistry. Therefore, decreasing the surface energy is the key to obtaining hydrophobicity [3].

2.3.2. Surface Roughness

Although low surface energy is essential for hydrophobicity, to obtain superhydrophobicity, surface roughness must be introduced. According to the Wenzel and Cassie-Baxter models, roughness functions as a multiplier: for materials naturally hydrophobic, increasing roughness greatly increases the apparent contact angle. It modifies the energy barriers for droplet spreading by considerably enlarging the actual surface area compared with the projected area [1]-[3].

2.3.3. Microstructure and Morphology

The morphology and distribution of the roughness are determined by the surface microstructure. Single-scale microstructures (either exclusively micro-scale or nano-scale) usually have insufficient air-retention stability or low mechanical durability. Studies show that hierarchical micro/nano-structures, similar to those found on a lotus leaf, are essential for achieving excellent hydrophobicity. In this arrangement, the micro-scale structures act as the main framework to hold the drop, whereas the additional nano-scale structures trap air bubbles to stabilize the Cassie-Baxter state and maintain high mobility of the drop [1] [2].

3. Nature-Inspired Hydrophobic Design

During the long process of evolution, nature has developed various biological surfaces with excellent wettability, serving as useful models for modern materials science to deal with problems such as wetting control, self-cleaning and fluid transportation.

These properties may result from the precise combination of specialized micro/nano-scale structures and surface chemistry. In order to comprehensively survey the recent progress in this biomimetic area, this chapter mainly discusses five typical biological patterns with excellent wetting characteristics: the lotus leaf with superhydrophobic and self-cleaning features, the rose petal showing high-adhesive superhydrophobicity (the “petal effect”), the butterfly wing having anisotropic wetting properties, the water strider able to walk on water with great bearing capacity, and the pitcher plant employing a slippery surface containing lubri-

cant to catch insects. This chapter explains the fundamental correlations between their minute morphologies and biological functions, and emphasizes the advanced techniques for their artificial imitation [2].

3.1. Lotus Leaf

The lotus leaf is generally regarded as an excellent example of superhydrophobicity. The remarkable "self-cleaning" characteristic (lotus effect) is due to its very small contact between droplets. In this part, we mainly study how the cooperative action of microscopic papillae and nanoscopic waxy clusters on the surface of the lotus leaf retains air bubbles, thus forming a typical low-adhesion Cassie-Baxter state [1] [2].

3.2. Rose Petal

Different from the lotus leaf, the rose petal has a special "high-adhesion" superhydrophobic property, called the "petal effect", where water droplets stay fixed even when the petal is turned over. This part studies how the larger microstructures on the rose petal permit some liquid penetration and produces strong adhesive forces while keeping a high apparent contact angle [2] [6].

3.3. Butterfly Wing

In addition to its bright colors, the wings of the butterfly also possess an interesting "anisotropic wettability", which makes water droplets slide off more easily in the outward direction of the wing but remain on the wing in the opposite direction. This part describes the directional scale structure on the butterfly's wings to explain the principles of directional drag reduction and self-cleaning [7] [8].

3.4. Water Strider

Water striders can stay on water because of the tilted microsetae on their legs which have nanoscale grooves. These structures create stable air pockets, enhancing the buoyancy and decreasing the adhesion to the liquid. The arrangement of hairs in various directions can reduce the hydrodynamic resistance and allow them to swim fast on water. The designs based on these structures have resulted in the production of miniature aquatic robots, floating sensors and low-drag interfaces. The strong legs and good flotation of water striders, enabling them to walk easily on water and resist heavy rain, are due to the microsetae and their supercoiled nano-grooves on their legs. This section mainly explains how they take in air to obtain an effective superhydrophobic buoyant force [9] [10].

3.5. Pitcher Plant

Pitcher Plant differs from the air-cushion based superhydrophobic systems; it uses a slippery liquid-infregnated porous surface (SLIPS). The microscale grooves hold a lubricating liquid layer, resulting in an extremely smooth and omniphobic contact with small contact-angle hysteresis. SLIPS have excellent anti-icing, anti-biofouling properties, maintain pressure stability and possess self-repair ability, which

make them suitable for biomedical devices and marine coatings.

The main characteristics are summarized in Table 1.

Different from the organisms which depend on air bubbles, the pitcher plant uses a special method—slippery liquid-infused porous surface (SLIPS)—to catch insects. This part describes how the peristome of the pitcher plant firmly holds a continuous lubricant film by means of its microscopic grooves, forming an omniphobic interface with remarkable droplet mobility especially for low-surface-tension liquids.

Table 1. Summary of common bio-mimetic surfaces: From biological structures to artificial functional patterns [11] [12].

Organism	Microstructure	Function	Artificial Design
Lotus Leaf	Hierarchical micro-pills + wax	Superhydrophobicity, Self-cleaning	Self-cleaning coatings, anti-fouling
Water Strider	Needle-like micro-hairs with nano-grooves	High load-bearing, aquatic mobility	Miniature aquatic robots, floatable devices
Pitcher Plant	Micro-grooves infused with lubricant (SLIPS)	Omni-phobic, anti-icing, pressure-stable	Anti-biofouling coatings, icephobic surfaces

4. Artificial Hydrophobic Materials

4.1. Polymer-Based Materials

4.1.1. PDMS

Polydimethylsiloxane (PDMS) is widely used as a polymer matrix for making artificial hydrophobic surfaces because of its excellent flexibility, optical transparency, chemical stability and low surface energy. The main structure of PDMS includes highly flexible Si-O-Si bonds covered by hydrophobic methyl side chains, which makes the surface of PDMS have an inherent water contact angle about 110˚. In real applications, superhydrophobicity can be easily achieved by forming micro- and nano-scale rough structures through soft lithography, template replication or the combination with nanoparticles. Furthermore, the good biocompatibility of PDMS makes it highly suitable for use in microfluidic chips and biomedical anti-fouling coatings [11] [12].

4.1.2. PTFE

Polytetrafluoroethylene (PTFE), often called the "plastic king", has one of the lowest surface energies among the known solid materials. Its molecular chain is fully enclosed by strong carbon-fluorine (C-F)-bonds, which give it excellent chemical inertness, remarkable thermal stability and a natural high hydrophobicity (with a sessile drop contact angle about 114˚). In the fabrication of artificial superhydrophobic materials, PTFE is usually used as powders, aqueous solutions or thin films. By introducing micro/nano-roughness on PTFE through mechanical blasting, plasma etching or electrospinning, the surface can be quickly transformed into a superhydrophobic or even superamphiphobic (repelling both water and oils) state, thus being very useful for durable anti-corrosion and industrial self-cleaning surfaces [11] [12].

4.1.3. PVDF

Polyvinylidene fluoride (PVDF) is a semi-crystalline fluoropolymer which has good mechanical strength, chemical resistance and special piezoelectric/ferroelectric properties. In comparison with PTFE, PVDF has much better solubility in solvents, and can directly make highly porous and interconnected micro/nano-fibrous networks by simple solution casting, phase inversion or electrospinning methods. The natural porosity and rough surface of PVDF, together with its low surface energy, result in excellent hydrophobicity without the necessity of complex post-treatment, which has been studied widely in membrane distillation, oil-water separation and piezoelectric self-cleaning sensors [11] [12].

4.2. Metal-Based Materials

4.2.1. Al (Aluminum)

As the most popular non-ferrous metallic material, aluminum and its alloys possess low density, high thermal conductivity and good machinability; however, they are easily corroded and frozen in moist conditions. Since pure aluminium is a material with high surface energy and is hydrophilic, it is necessary to use a "topography first, chemistry second" approach in order to produce an Al-based superhydrophobic surface. Chemical etching (such as immersion in acids or bases) or anodizing is generally carried out to form micro/nano-hierarchical structures resembling coral or honeycombs on the aluminium base, which is then modified by low-surface-energy silanes or fluorides. This modification can effectively improve the anti-corrosion ability of the aluminium alloys and achieve excellent anti-icing effect [11] [12].

4.2.2. Cu (Copper)

Copper is important in heat exchangers, electronic packaging and condensers because of its excellent electrical and thermal conductivity. However, the condensed water on the rough copper surface usually shows filmwise condensation, which greatly reduces the heat transfer efficiency. By modifying a copper surface into a superhydrophobic one, the condensation process can be changed from the conventional filmwise condensation to the highly efficient dropwise condensation. The common methods for fabrication include chemical oxidation (such as forming dense CuO or Cu_2O nanorod arrays in alkaline persulfate solutions) and electrochemical deposition. After treating with low-surface-energy substances, the superhydrophobic copper surface keeps its excellent thermal conductivity and possesses strong anti-corrosion properties along with automatic droplet jumping behaviour [11] [12].

4.2.3. Steel

Steels like stainless steel and carbon steel are important for modern industrial construction, but they are corroded easily in severe marine or chemical surroundings. Developing superhydrophobic coatings on the steel surface is an important field of advanced physical-barrier anti-corrosion. Usually, robust micro/nano-textures

are produced by laser processing, thermal spraying of hard nanoparticles (for example, SiO_2), or electroplating, and are then combined with fluorosilanes. The stable Cassie-Baxter air cushions contained in these textured parts can effectively isolate the corrosive medium (such as seawater or acid solutions) from the steel base, thus significantly increasing the service life of steel parts in complicated industrial conditions [11] [12].

4.2.4. Ti (Titanium)

Titanium and its alloys are commonly utilized due to their high specific strength and good biocompatibility, resulting in applications in aerospace industry and medical implants. The study on titanium-based superhydrophobic surfaces mainly focuses on two aspects: in aerospace, it exploits the anti-icing and reducing drag effects; in medical implants, the alteration of the surface wettability (such as converting from superhydrophobicity to anisotropic wetting) can effectively control the initial cell adhesion and prevent the colonization and growth of bacteria on the implant surface, thus significantly improving the therapeutic safety and lasting effectiveness of the medical implants [11] [12].

4.3. Ceramic Materials

4.3.1. SiO_2

Silicon dioxide (SiO_2) nanoparticles are mainly used as inorganic ceramic fillers for artificial superhydrophobic coatings in engineering. They can be produced economically and in large quantities by the well-known sol-gel Stöber method, which enables the adjustment of processing conditions to prepare monodisperse SiO_2 spheres with different sizes. These nanoparticles can be conveniently coated on different substrates to form a micro/nano-hierarchical roughness structure. Since the inorganic SiO_2 skeleton has high mechanical hardness, mixing it with flexible polymer matrices significantly improves the mechanical wear resistance and scratch resistance of the resultant superhydrophobic coatings [11] [12].

4.3.2. TiO_2

Moreover, the titanium dioxide (TiO_2) ceramic materials not only possess good mechanical hardness and chemical stability but also have the unique photocatalytic ability. When pure TiO_2 is irradiated by ultraviolet (UV), it becomes superhydrophilic. While the superhydrophobic surfaces based on TiO_2 have the function of being "responsive" or "self-repairing", the hydrophobic TiO_2-based superhydrophobic surfaces can restore their hydrophobicity and cleanability after the organic contamination makes them lose their hydrophobicity. Under the action of UV light, the organic pollutants can be degraded by the photocatalysis of TiO_2, which results in the self-cleaning effect and a smart response. This property is very attractive for intelligent optical window coatings [11] [12].

4.3.3. ZnO

Zinc oxide (ZnO), a common one-dimensional wide-bandgap semiconductor ce-

ramic material, can be easily formed into regular nanorod arrays, nanoneedles or nanoflowers on various complex shapes by the inexpensive hydrothermal method. The orderly and uniform vertical arrangement offers a stable foundation which is suitable for maintaining the Cassie-Baxter wetting state. After efficient low-energy surface treatment, ZnO-based superhydrophobic surfaces show excellent static contact angles and the motion of droplets. Furthermore, the inherent antibacterial and UV-blocking properties of ZnO provide it with a dual function for its use in public health and medical protective articles [11] [12].

4.4. Carbon Nanomaterials

4.4.1. Graphene

Graphene, a single layer of honeycomb lattice composed of sp^2-hybridized carbon atoms, has good mechanical strength, electrical conductivity and thermal conductivity. Although pure graphene has a moderate intrinsic hydrophobicity (the water contact angle is approximately 85˚ - 90˚) theoretically, in order to achieve true superhydrophobicity, macroscopic structure modification is required. By adjusting the reduction of graphene oxide (GO), freezing-drying three-dimensional graphene aerogels or growing wrinkled graphene films by chemical vapor deposition (CVD), the necessary micro/nano-roughness can be introduced successfully. The superhydrophobic graphene membranes not only have great resistance to liquids but also have multi-functional properties such as conductive anti-corrosion, antistatic effect and photothermal deicing [11] [12].

4.4.2. CNTs (Carbon Nanotubes)

Carbon nanotubes (CNTs) are the common one-dimensional carbon nano-materials, owing to their large aspect ratio and excellent mechanical flexibility, they are appropriate for constructing "bio-inspired brush" structures. By changing the growing direction, vertical carbon nanotube arrays (VACNTs) can be arranged to mimic the micro-hairs on the legs of water striders. The hollow tube-like structure can store a great deal of air as an insulating layer. The modified surfaces of CNTs show remarkable ultra-high droplet bouncing properties, and because of the good light absorption and electrothermal properties of carbon nanotubes, they can be heated up easily by optical or electrical stimulation, therefore effecting efficient and active de-icing under any weather conditions [11] [12].

4.4.3. Carbon Black

Carbon black is a very economical, easily obtainable and chemically stable zero-dimensional carbon nanomaterial. The nanoparticles of carbon black usually form branched cluster structures with a high degree of aggregation, which develop a multi-scale micro/nano-rough surface during the packing process at the macro level. By adding carbon black as a structural modifier into polymer matrices, superhydrophobic coatings can be prepared over large areas easily through simple spraying methods. Since carbon black has strong absorption for all wavelengths of light, the superhydrophobic coatings based on carbon black show excellent

photothermal conversion efficiency under solar irradiation and therefore can be considered as a promising material for high-valued photothermal de-icing coatings and solar-driven interfacial seawater desalination [11] [12].

4.5. Emerging Materials

4.5.1. MXenes

MXenes form a new type of two-dimensional transition metal carbides, nitrides and carbonitrides, which have become research focuses because of their metallic electrical conductivity, hydrophilic surface functional groups (e.g., -OH and -F), and excellent electromagnetic interference (EMI) shielding ability. Although the pristine MXene sheets show high hydrophilicity due to these numerous polar groups, these active sites can serve as various chemical anchoring places for surface hydrophobic modification. By using surface silanization or combining with low-surface-energy polymers, the MXene sheets have been successfully transformed into superhydrophobic structures. This superhydrophobic treatment not only solves the main drawback of MXene being easily oxidized in humid conditions but also opens up new application possibilities in flexible EMI shielding, wearable electronics and outdoor electronics under all weather conditions [11] [12].

4.5.2. MOFs (Metal-Organic Frameworks)

Metal-Organic Frameworks (MOFs) are three-dimensional porous crystals consisting of inorganic metal atoms and organic ligands connected by coordination bonds. The materials have large specific surface areas, adjustable pore sizes and regular microcrystalline structures. By preparing Fluorinated MOFs (F-MOFs) by using naturally hydrophobic organic ligands (such as fluorinated or long-chain alkyl ligands) directly, or by changing the conventional MOF crystals after the synthesis process, the surface wettability can be precisely controlled at the sub-nanometre scale. These superhydrophobic MOFs can effectively avoid the damage to the crystal structure by water vapor (which is a disadvantage of low water resistance) and exhibit excellent selectivity and capacity in selective fluid adsorption, gas purification and rapid removal of hazardous organic solvents (like oil spill remediation) [11] [12].

4.5.3. Biomaterials

With the worldwide attention on green chemistry and sustainable development, using natural and biodegradable biomaterials, such as cellulose nanocrystals (CNCs), lignin, chitosan and different kinds of plant waxes, to make green superhydrophobic coatings has become an important research direction. These materials can avoid the possible biotoxicity and environmental persistence problems caused by the traditional fluorinated low-surface-energy modifiers (the "forever chemicals" problem). By means of physical emulsification, micro-phase separation or self-assembly processes, hydrophilic cellulose or chitosan can be transformed into rough web-like structural frameworks, which are then coated with a natural plant wax to produce completely environmentally friendly and biode-

gradable superhydrophobic packaging films and bio-based anti-fouling coatings with high practical value in green food packaging and ecological agricultural protection [11] [12].

5. Fabrication Strategies

5.1. Physical Methods

Laser (Laser Texturing)

Laser texturing is a sophisticated energy beam processing technique which employs high-energy density laser beams (like nanosecond, picosecond or femtosecond lasers) to irradiate surface of materials and form micro/nano-rough structures by local instantaneous melting, vaporization and resolidification. The main advantage of this approach is its non-contact processing, high precision and excellent controllability, enabling the computer-aided programming to etch arbitrary micro-arrays (such as micro-grooves, micro-pillar arrays) on metals, ceramics and polymers. Because of their short pulse width and small heat-affected zone (which is called "cold processing"), femtosecond lasers can make fine micro/nano hierarchical textures and serve as an important physical tool for controlling the special wetting property in both research laboratories and high-tech industrial productions.

Lithography

Lithography (including conventional photolithography, electron-beam lithography and nanoimprint lithography (NIL)) plays a crucial role in the microelectronics field and is the most effective method for producing fine, accurate and repeatable geometric structures. By using spin-coating, exposure and development of photoresists on a substrate, together with subsequent etching or deposition processes, scientists can reproduce the geometric features of biological surfaces at the micro- and nano-levels precisely. Lithography is often used in basic theoretical studies (such as quantitatively investigating the critical values of pillar height and spacing during the Wenzel-to-Baxter wetting transition), since it avoids the influence of random roughness and gives an idealized, stable three-dimensional model.

Etching

Etching (which mainly consists of chemical etching, electrochemical etching and reactive ion etching (RIE)) is a common "top-down" physical/chemical subtractive manufacturing method to obtain surface roughness. Chemical and electrochemical etching make use of the selective dissolution of grain boundaries or certain crystal planes of metallic substrates (like aluminum, copper, steel and titanium) to produce naturally rough crystal surfaces. Furthermore, reactive ion etching employs high-energy reactive plasma to bombard solid surfaces for directional anisotropic ablation. Because of its simple processing equipment, fast reaction speed and special ability to form multi-scale roughness directly on large-sized bulk components with complex shapes, etching has been widely applied in industrial metal corrosion prevention and surface treatment.

5.2. Chemical Methods

Sol-gel

The sol-gel process is a well-known "bottom-up" chemical preparation technique. It usually uses metal alkoxides (for example, tetraethoxysilane, TEOS) as raw materials, carries out hydrolysis and polycondensation reactions in the solution to establish a stable sol system, and finally forms a dense three-dimensional inorganic network on the substrate by means of gelation and drying. Its main advantage is the complete homogeneous mixing at the molecular level and high flexibility in processing. By introducing long-chain silane coupling agents (e.g., low surface energy fluorosilanes) directly into the sol system, the structure and chemical modification can be accomplished in one operation. The sol-gel process does not limit the shape of the substrate and can achieve smooth coating over large areas by spraying or dipping.

CVD (Chemical Vapor Deposition)

Chemical Vapor Deposition (CVD) is a method which uses gaseous starting materials to carry out specific chemical reactions on a heated substrate surface, thus forming high purity solid thin films or nanostructures. In the superhydrophobic field, CVD can grow uniformly nano-scale roughness (like one-dimensional carbon nanotubes or oxide nanowires) or very thin, low energy fluoropolymer layers at the sub-micron level without changing the macroscopic shape of the substrate. The CVD technique has characteristics of being solvent-free, high film purity and excellent adhesion at the interface, which allows it to deal with three-dimensional porous materials with complicated internal pore structures (such as sponges and filters) to accomplish comprehensive omnidirectional hydrophobic treatment of the internal surfaces.

Self-assembly

Self-assembly is a chemical construction method relying on non-covalent intermolecular interactions (such as electrostatic attraction, hydrogen bonding, coordination bonds or Van der Waals forces) to produce spontaneously highly ordered nanostructures. Layer-by-layer (LBL) self-assembly is an example; by repeatedly immersing substrates in solutions containing oppositely charged substances (e.g., positively charged polyelectrolytes and negatively charged SiO_2 nanoparticles), films with accurate thickness can be deposited on any complex surface uniformly. This self-assembly process has very mild operating conditions (usually carried out in aqueous medium at room temperature and pressure), therefore being environment friendly and convenient to operate, which possesses special advantages in the manufacture of precision optical anti-fog coatings and modification of biosensors.

5.3. Hybrid Strategies

Hybrid Approaches and Summary

Since single physical or chemical approaches usually have some disadvantages, like high production expenses (for example, lithography and laser texturing),

lengthy cycles or poor mechanical strength, much research attention has been focused on physical/chemical combined techniques. The main idea of hybrid techniques is to combine the high efficiency of physical structure roughening with the long-lasting stability of chemical low-surface-energy treatment or (self-sustained) nanostructure growth.

For example, mechanical blasting or chemical etching are used initially to quickly form large micro-skeletons on a metal surface (physical/subtractive), then the hydrothermal or sol-gel process grows dense secondary nano-structures (chemical/additive), and finally CVD or coupling agents are applied to achieve the low-surface-energy modification. This combined engineering method not only considerably reduces the manufacturing complexity but also significantly improves the mechanical wear resistance, anti-peeling ability and thermal stability of the superhydrophobic micro/nano-structures when exposed to actual industrial conditions, which can be considered the basic principle for promoting superhydrophobic surfaces to practical large-scale commercial use.

The fabrication methods are compared in (Table 2).

Table 2. Integrated comparison of fabrication methods.

Method	Cost	Complexity	Scalability	Typical Materials	Key Advantages/Limitations
Laser	High	Medium	Medium-Low	Metals, Polymers, Ceramics	Highly precise and environmentally friendly; however, expensive equipment has difficulty dealing with intricate inner structures.
Lithography	Very High	High	Low	Silicon, Polymers	Perfectly regular and controllable structures; however, they are too costly, time-consuming and not feasible for large-scale industrial use.
Etching	Low	Low	High	Metals (Al, Cu, Steel), Ti	Simple process, highly efficient, low cost; but involves acid/alkali wastewater, random morphology.
Sol-gel	Low	Medium-Low	High	SiO_2, TiO_2, Polymers	Supports one-step processing with little equipment requirement; however, it has a long curing time and is likely to crack over extensive areas.
CVD	Medium-High	High	Medium	Carbon (CNTs, Graphene)	Extremely high coating purity and strong adhesion; however, it needs vacuum/high-temperature chambers and the substrate is limited.
Self-assembly	Low	Medium	High	Biomaterials, Multi-polymers	Mild and eco-friendly conditions, appropriate thickness regulation; however, low stacking efficiency and poor wear resistance.

6. Experimental Investigation

6.1. Experimental Materials

To investigate the preparation of hydrophobic materials, two experimental methods were used in this research. One method consisted of synthesizing a cellulose-based hydrophobic composite by means of hydrothermal processing and fluorination treatment, while the other aimed at forming a biomimetic hydrophobic

surface by replicating a natural hierarchical structure with polydimethylsiloxane (PDMS). Both methods were intended to examine the influences of hierarchical surface structures and low-surface-energy modification on the wetting behavior of solid surfaces.

Cellulose was chosen as the main substrate since it is a renewable, biodegradable and environmentally friendly substance with a large number of hydroxyl groups which are suitable for chemical modification and the formation of inorganic nanosystems [13]. Graphite powder was added to the composite to enhance the structural stability and to offer more nucleation points during the hydrothermal synthesis, and the cellulose-to-graphite mass ratio was maintained at 20:1. Zinc nitrate hexahydrate ($Zn(NO_3)_2{\cdot}6H_2O$) and hexamethylenetetramine (HMTA) were used as the precursors for the hydrothermal formation of ZnO nanostructures. The hydrothermal precursor solution was prepared by dissolving 0.35 g of HMTA in 100 mL of deionized water, followed by the addition of 0.74 g of $Zn(NO_3)_2{\cdot}6H_2O$ under continuous magnetic stirring until complete dissolution. ZnO nanostructures were selected due to their controllable shape and chemical stability, which make them applicable in hydrophobic surface engineering.

For the biomimetic surface, PDMS prepolymer and curing agent were employed to reproduce the hierarchical microstructures of a fresh lotus leaf, which was used as the natural template. PDMS is frequently used in soft lithography due to its flexibility, good replication capacity and low surface energy. After hydrothermal processing, the PDMS samples were dried in a forced-air drying oven at 75˚C for 3 h, and 1H,1H,2H,2H-perfluorodecyltriethoxysilane (PFDTES) was used as the fluorinating agent to decrease the surface free energy of the prepared materials. For the fluorination treatment, 2 mL of PFDTES was placed at the bottom of a vacuum desiccator, and the sealed desiccator containing the PDMS samples was transferred to a vacuum drying oven and maintained at 65˚C for 6 h.

The essential experimental instruments consisted of an analytical balance, a magnetic stirrer, a 100 mL Teflon-lined hydrothermal reactor, a vacuum desiccator, a drying oven, a vacuum drying oven, and a contact angle goniometer.

6.2. Experimental Procedures

6.2.1. Preparation of Cellulose-Based Hydrophobic Composite

The procedure for preparing the cellulose-based hydrophobic composite is shown in **Figure 1**. First, 0.35 g of hexamethylenetetramine (HMTA) was dissolved in 100 mL of deionized water by continuous magnetic stirring to obtain a homogeneous precursor solution. After complete dissolution, 0.74 g of zinc nitrate hexahydrate ($Zn(NO_3)_2{\cdot}6H_2O$) was added and stirred continuously until a homogeneous solution was obtained. Then, cellulose fibres and graphite powder were added gradually into the solution at a cellulose-to-graphite mass ratio of 20:1 and stirred thoroughly to achieve uniform dispersion.

The suspension was put into a 100 mL Teflon-lined hydrothermal reactor and was heated at 95˚C for 10 h. The hydrothermal treatment was intended to

promote the formation of ZnO nanostructures on the surfaces of cellulose fibres and graphite particles. Cellulose served as a supporting framework, whereas graphite facilitated heterogeneous nucleation, which was expected to contribute to the formation of a hierarchical micro/nanostructure beneficial for hydrophobicity.

After the reaction, the samples were cooled naturally to room temperature, washed three times with deionized water to eliminate the remaining reactants and then dried in an oven. Lastly, the dried samples were treated with 1H,1H,2H,2H-perfluorodecyltriethoxysilane (PFDTES) to decrease the surface free energy. The combined effect of hydrothermal treatment and fluorination was expected to enhance the hydrophobicity of the cellulose composite. The observed wettability is consistent with the Cassie-Baxter wetting model [3] [5].

6.2.2. Fabrication of Biomimetic PDMS Replica

A hydrophobic surface was produced by means of a PDMS replica moulding method. The PDMS prepolymer and curing agent were mixed in a ratio of 10:1 and stirred evenly. Subsequently, the mixture was put into a vacuum chamber to expel the air bubbles before pouring.

The degassed PDMS was then coated on a fresh lotus leaf and was dried by means of heating. After drying, the PDMS replica could be detached from the model without altering its hierarchical surface. A thin layer of graphite was next laid on the reproduced surface as the nucleation sites for the hydrothermal growth of ZnO.

The coated samples were also subjected to the same hydrothermal treatment as described in Section 6.2.1. After the hydrothermal treatment, the PDMS samples were dried in a forced-air drying oven at 75 ˚C for 3 h. Subsequently, 2 mL of 1H,1H,2H,2H-perfluorodecyltriethoxysilane (PFDTES) was placed at the bottom of a vacuum desiccator, and the desiccator containing the PDMS samples was sealed and transferred into a vacuum drying oven at 65 ˚C for 6 h to complete the fluorination process. This preparation technique combines biomimetic surface replication with chemical surface modification and provides a feasible approach for fabricating biomimetic surfaces with hierarchical structures [2] (**Figure 1**).

6.3. Characterization Methods

The wettability of the prepared cellulose-based composite was assessed by determining the static water contact angle with a contact angle goniometer. A water droplet of a constant volume was gently placed on the sample surface, and the contact angle was calculated by means of image analysis software. To increase the accuracy of the measurements, each sample was examined at three different sites, and the average value was reported. Furthermore, the sliding angle was determined by slowly tilting the sample until the water droplet began to move, providing an indication of the adhesion between the water droplet and the sample surface (**Figure 2** & **Figure 3**).

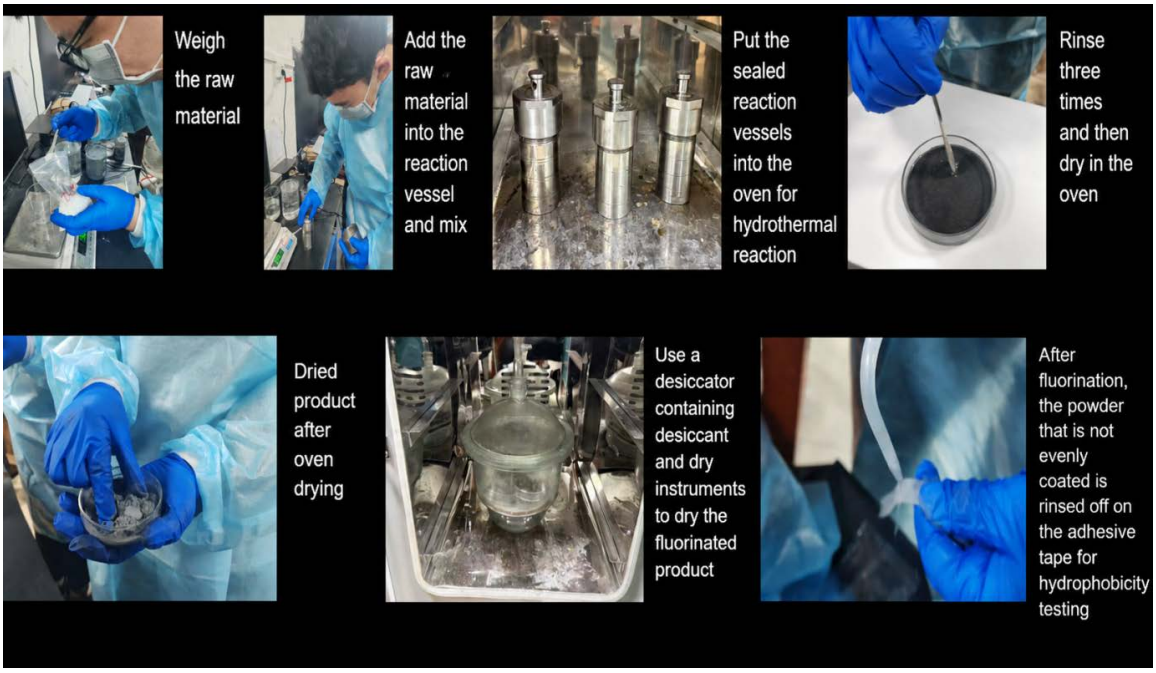

Figure 1. Flow chart of the experiment.

Figure 2. Measuring the water contact angle of the PDMS materials.

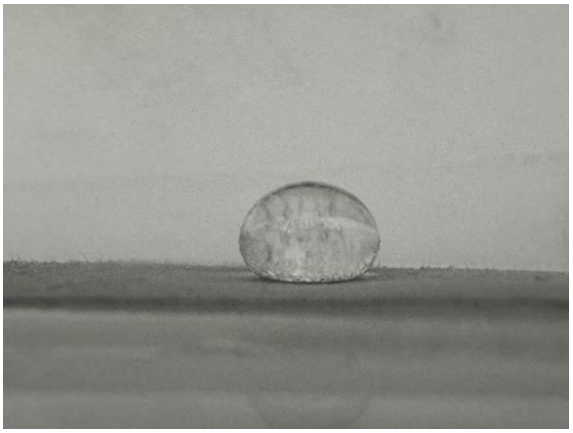

Figure 3. Measuring the water contact angle of the cellulose-based materials.

6.4. Experimental Results and Discussion

The experimental results for the cellulose-based composite show that it exhibited enhanced hydrophobicity after hydrothermal treatment and fluorination. The prepared sample exhibited an average water contact angle of 165˚ and a sliding angle of 5.4˚. These measured values indicate that the modified cellulose-based composite exhibited excellent water repellency and satisfied the criteria for a superhydrophobic surface.

The improved hydrophobicity may be attributed to the combined effects of the surface roughness generated during hydrothermal treatment and the reduction in surface free energy introduced by fluorination. According to the Cassie-Baxter wetting model, the measured wettability is consistent with a surface state in which trapped air reduces the effective solid-liquid contact area, resulting in a high water contact angle and low droplet adhesion [3] [5]. The measured water contact angle of 165˚ and sliding angle of 5.4˚ indicate that the prepared cellulose-based composite exhibited superhydrophobic behavior. Moreover, the biomimetic PDMS replica successfully retained the structural features of the fresh lotus leaf template, demonstrating the feasibility of the biomimetic surface replication process.

According to the experimental results, the combination of hydrothermal synthesis, biomimetic surface replication, and fluorination provides a feasible approach for producing hydrophobic materials. The cellulose-based composite shows good application prospects in self-cleaning coatings, anti-fouling surfaces, moisture-resistant materials, and functional protective coatings. Although the biomimetic PDMS replica was successfully fabricated, its water contact angle and sliding angle were not quantitatively measured in the present study. Therefore, its hydrophobic performance cannot be quantitatively discussed. In the future, efforts may be directed towards enhancing the mechanical strength of the coatings and examining environmentally friendly substitutes for fluorine-containing substances to improve the long-lasting properties and environmental friendliness of the prepared materials.

7. Applications

7.1. Self-Cleaning

Self-cleaning is the most distinctive and profitable use of artificial superhydrophobic surfaces, similar to the “lotus effect”. On such a surface, where the adhesion is very low, water drops do not flatten; they maintain nearly a spherical shape. If the surface is slightly inclined, the gravity can cause the drops to roll off quickly. During this rolling motion, some loose dust, dirt and solid impurities adhere to the surface of the moving water drop and are transported along with it to the solid base. This ability to clean the surface naturally by means of rainwater or simple washing is beneficial for the self-cleaning maintenance of high-rise building windows, large-scale solar photovoltaic panels (to avoid dust accumulation which might decrease the power efficiency) and outdoor high voltage power line insulators requiring long-term no-maintenance operation.

7.2. Anti-Corrosion

Metallic corrosion causes serious and lasting economic and structural losses worldwide, and superhydrophobic surfaces provide a new approach different from the conventional physical barrier coatings. When metallic parts (such as ship hulls of sea-going vessels, the foundations of offshore wind turbines and oil transmission pipelines) are in contact with severe environments which contain large amounts of acidic corrosive substances, the special micro/nano structure of the superhydrophobic surface can trap and keep a thick, macroscopically continuous layer of air. According to the Cassie-Baxter model, this air layer functions as a natural "physical barrier" which can prevent the high-corrosion liquid phases from contacting directly with the metal base, thus reducing the actual solid-liquid electrochemical contact area by more than 90%. This separation not only breaks the electrochemical circuit completely but also effectively inhibits the diffusion and penetration of harmful ions, giving strong and long-lasting anti-pitting and anti-electrochemical corrosion properties to the metallic materials.

7.3. Anti-Icing

The heavy accumulation of ice and snow on the wings of high altitude airplanes, the blades of wind turbines and the high voltage power lines usually causes serious mechanical and electrical failures. These problems can be effectively solved by using superhydrophobic surfaces in two aspects: "active ice preventing" and "easy deicing". On one hand, because of the low adhesion to water molecules and the high interfacial thermal resistance caused by the micro/nano structures, when supercooled water droplets strike the surface at high altitudes, the heterogeneous ice nucleation delay time of these droplets is greatly increased, which enables most of them to slide off or bounce away under the influence of ambient wind shear before freezing. Thus, active ice preventing is achieved. On the other hand, the presence of many air cavities between the solid and ice interfaces guarantees that the ice-adhering strength of the superhydrophobic surface is less than 20 kPa (much lower than the several hundred kPa of ordinary materials) under severe icing conditions. Therefore, the ice deposits can be easily removed and shed under small wind forces or slight mechanical vibrations, leading to efficient de-icing.

7.4. Oil-Water Separation

Frequent severe marine crude oil leakage and large amount of industrial oily wastewater discharge cause serious harm to the global aquatic ecosystem. It is of great importance to develop high-efficiency and large-flow oil-water separating materials. By carrying out superhydrophobic and superoleophilic surface treatments on porous substrate materials (for example, stainless steel mesh, polymer sponge and cotton fabric), a highly efficient "intelligent selective fluidic switch" can be successfully designed. When an oil-water mixture is poured onto the sur-

face of these porous materials, the extremely large water contact angle together with the Cassie wetting state and strong capillary pressure firmly prevents water from infiltrating, causing it to remain above the mesh. At the same time, the low-surface energy property ensures the zero contact angle superoleophilic wetting characteristics towards oils, which makes organic solvents and oils pass through the porous network quickly by capillary action. This efficient "water repelling and oil permeable" mechanism achieves a separation purity over 99% while maintaining continuous and high flow rate operation.

7.5. Biomedical Engineering

In the field of modern biomedical engineering, controlling the wettability of the material surface is an effective method to regulate the interfacial biological interactions. The superhydrophobic surfaces have a significant protective function in both clinical and experimental work due to their excellent property of repelling liquids. They can prevent the attachment and residue of medical devices such as surgical knives, medical catheters and blood storage containers by means of the air cushion on the superhydrophobic surface, thus decreasing the possibility of thrombosis (good biocompatibility). At the same time, this surface can also hinder the initial adhesion of bacteria and inhibit the development of harmful strains and the formation of biofilms, which has promising applications in antibacterial implants. Furthermore, in microfluidics, alternating superhydrophobic and superhydrophilic "micro-patterned arrays" are commonly used for the accurate direction control of droplets, the capture of single-cell microarrays and the execution of sensitive *in-vitro* biochemical diagnosis.

7.6. Flexible Electronics and Energy

Due to the increasing use of wearable devices, flexible electronic skins and new types of green energies, the outdoor installation of electronics in various weather conditions should possess good stability against sweat corrosion, short circuits from rain and moisture failures. At present, embedding superhydrophobic properties into flexible electronic and energy devices is a developing research field. In flexible sensors, by uniformly combining conductive carbon nanotubes or graphene with flexible superhydrophobic polymers (like PDMS or PVDF) in depth, the resulting stretchable sensors can precisely detect human joint movements with excellent resistance to sweat, durability and self-cleaning functions. In the energy field, superhydrophobic coatings play crucial roles as charge-enhancing and protective layers for triboelectric nanogenerators (TENGs), which can effectively prevent the loss of surface charges caused by atmospheric humidity, thus greatly enhancing the power generation in high temperature and humid regions; they can also be applied directly as efficient "raindrop energy-collecting devices". In fuel cells, superhydrophobic microporous layers function as necessary gas diffusion and water management channels, solving the current problem of system failure due to internal water flooding.

8. Challenges and Future Perspectives

Although significant theoretical progress has been made and various types of artificial hydrophobic materials have been developed in the last few decades, it is still an ongoing task to apply superhydrophobic surfaces in practical industrial conditions. In order to provide a detailed plan for the future wetting-control materials, this chapter thoroughly describes the five main obstacles and development directions which characterize this field [11] [12].

8.1. Mechanical Durability

The main difficulty in the actual use of superhydrophobic surfaces is their susceptibility to mechanical destruction. According to the definition, the extreme water repulsion mainly depends on the high aspect ratio and delicate micro/nano-scale hierarchical roughness at a very small scale. In actual operating conditions, these fragile structures are frequently exposed to severe physical stresses such as scratching by solid particles, linear wear, impact of high-speed liquid droplets and adhesive force from tape-peeling. When the microscopic pillars or nanostructures are broken or flattened by external forces, the enclosed Cassie-Baxter air film will collapse irreversibly, resulting in a transition to the highly pinned Wenzel state or complete wetting failure [11] [12].

To solve this problem, recent studies have changed from weak and short-lasting coatings to the construction of "bulk" superhydrophobic materials and strong architectural structures. Possible methods are mentioned in [11] [12].

Using hard inorganic ceramic matrices uniformly dispersed in a highly elastic polymer matrix to absorb impact energy.

Building inter-connected, small scale porous sacrificial frameworks (similar to large scale underground matrices) which protect delicate nano-scale secondary structures in their internal hollows.

Developing "self-healing" or autonomous structural-reconstruction surfaces which can move low-surface-energy chains or restore hierarchical roughness in response to external thermal, chemical or optical stimuli [11] [12].

8.2. Environmentally Friendly Materials

In the past, the preparation of high-efficiency hydrophobic materials has mainly relied on fluorinated substances like per- and polyfluoroalkyl compounds (PFAS) and long-chain fluorosilanes to reduce surface energy. However, because of the great chemical stability of these substances, they show serious bioaccumulation and environmental persistence and are therefore called "forever chemicals". The strictification of global environmental laws has resulted in an immediate requirement to remove toxic fluorinated compounds from functional coatings.

Therefore, the approach has changed mainly to green, sustainable and fluorine-free materials. Scientists are now using natural biomaterials, such as cellulose nanocrystals (CNCs), lignin, chitosan and plant derived biodegradable waxes

(e.g., carnauba wax, beeswax) as structural supports and hydrophobic agents. The main scientific problem is to obtain similar water repellence, especially for complicated or low surface tension liquids, by employing only hydrocarbon based polymers or eco-friendly inorganic frameworks without affecting the durability of the material or increasing the high manufacturing costs.

8.3. Large-Scale Manufacturing

Although sophisticated techniques like photolithography, electron-beam processing and femtosecond laser texturing produce fine and even micro/nano-structures in the laboratory, they are greatly limited by large initial costs, complicated operating procedures, low production speed and restrictions on the size of the substrates. These obstacles make them impractical for large-scale, economical industrial mass production, such as covering extensive areas of solar panels, huge ship hulls or long high-voltage power transmission lines [11] [12].

Bearing in mind the difference between precise laboratory scale and industrial large scale, it is necessary to improve the high throughput "substrate-independent" processing techniques. Mainly, scalable methods include scalable roll-to-roll (R2R) nanoimprint, automatic industrial spraying, dip coating and co-assembling, and fast ambient chemical etching. The main objective of the engineering is to control the spatial distribution and statistical uniformity of multi-scale roughness on extensive, non-flat surfaces accurately, and to guarantee high reproducibility and economic efficiency among different batches.

8.4. Multifunctional Surfaces

Real-world operating environments are usually complex and unfavorable, so single-purpose liquid repellency is not enough to meet the sophisticated engineering demands. Presently, it is required to combine superhydrophobicity with other supplementary functions in a single unified material system [11] [12].

For example: In aerospace and high-altitude energy structures, a material should integrate excellent mechanical strength with the ability to provide active photothermal/electrothermal heating to cope with anti-icing in various weather conditions [11] [12].

In the fields of flexible electronics and smart wearable skins, the superhydrophobic protective matrix should keep a high level of electrical conductivity, great mechanical flexibility and quick signal transmission without being affected by the erosion of sweat or physical changes [11] [12].

In the fields of marine and chemical engineering, the advanced surfaces must have excellent optical transparency or anti-reflection properties together with their strong anti-fouling and long-lasting anti-corrosion characteristics [11] [12].

Achieving this combined function needs a careful adjustment in the material composition to prevent any trade-offs, wherein improving one functional characteristic may lead to a decrease in wetting performance [11] [12].

8.5. AI-Assisted Material Design

The conventional method for identifying and improving superhydrophobic materials has mainly depended on Edisonian "trial-and-error" experiments, where chemical compositions, mixing ratios and microstructural shapes are adjusted manually. This empirical approach is very time-consuming, consumes much resources and is restricted by human cognitive biases, thus hindering the rapid development of new materials [11] [12].

To promote progress, Artificial Intelligence (AI) and Machine Learning (ML) are being used in advanced wetting-control engineering. By training deep neural networks on large data sets which include chemical compositions, surface energies, geometric roughness and the corresponding contact/sliding angles, AI models can quickly forecast the wetting characteristics of potential materials. In addition, generative algorithms can perform high-speed virtual screening and reverse design of optimal topographic structures or polymer structures suitable for certain targeted environments. This computational revolution greatly shortens the R&D period, changing the field from empirical investigation to data-based, predictive engineering.

9. Conclusions

This detailed study examines the advancement of artificial hydrophobic materials, relating basic thermodynamic laws, natural structures, advanced production techniques and possible industrial usages. In short, the main subject of this field can be indicated by referring to three important aspects: [11] [12].

9.1. What Has Been Achieved?

Recently, the research on surface wetting has developed from qualitative experimental results to accurate theoretical studies. Based on the classical wetting theories like Young's, Wenzel's and Cassie-Baxter's, the researchers have elucidated the complex thermodynamic connection between the surface energy and the micro/nano roughness. By making a comparison with natural phenomena such as the self-cleaning lotus leaf, the sticky rose petal, the anisotropic butterfly wing, the floating water strider and the slippery liquid-infused pitcher plant, the material scientists have successfully manufactured different kinds of artificial superhydrophobic surfaces. Various top-down physical techniques and bottom-up chemical procedures have been set up, which enable these surfaces to be used in important areas such as self-cleaning buildings, anti-corrosion in seawater, anti-icing devices, high-purity oil-water separation and biocompatible medical instruments [11] [12].

9.2. What Are the Remaining Challenges?

Although some important results have been obtained, several important obstacles still prevent the commercial use of these technologies. Physically, the delicate structure at the micro/nano level is easily damaged by friction and wear, resulting

in an early wetting change and failure. From the chemical point of view, the dependence on long-lasting fluorinated substances causes serious environmental and legal problems, thus requiring immediate development of fluorine-free substitutes. Additionally, the high expense, low production rate and size limitations of precise manufacturing techniques limit the large scale application. Moreover, it is difficult to develop multifunctional surfaces which can maintain their good performance under various severe and changing conditions is also a complicated problem [11] [12].

9.3. Where Will Hydrophobic Materials Go?

For the future, the development of hydrophobic materials will mainly concentrate on the aspects of sustainability, intelligence and data-driven design. The present trend is to change from the rigid and single-functional fluorinated coatings to the eco-friendly and bio-based bulk materials which have self-healing ability. In the structure of surfaces, there will be more and more intelligent and responsive architectures which can adjust their wettability according to the external stimuli. What is important is that the application of artificial intelligence in material informatics and high-speed machine learning algorithms will take the place of the conventional empirical method of trial and error. This will enable the prediction and automatic reverse-design of the next generation interfaces which are suitable for solving various practical problems.

Author Contributions

Conceptualization, Hanyu Ren and Tianxi Li; methodology, Hanyu Ren and Tianxi Li; validation, Hanyu Ren and Tianxi Li; formal analysis, Hanyu Ren and Tianxi Li; investigation, Hanyu Ren and Tianxi Li; data curation, Hanyu Ren and Tianxi Li; writing—original draft preparation, Hanyu Ren (Abstract), Tianxi Li (Keywords and manuscript framework), and Hanyu Ren and Tianxi Li (main text); writing—review and editing, Hanyu Ren and Tianxi Li; visualization, Hanyu Ren and Tianxi Li. All authors have read and agreed to the published version of the manuscript.

Conflicts of Interest

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

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