

Review

Superhydrophobic Civil Engineering Materials: A Review from Recent Developments

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Abstract: Superhydrophobic surfaces have drawn attention from scientists and engineers because of their extreme water repellency. More interestingly, these surfaces have also demonstrated an infinite influence on civil engineering materials. In this feature article, the history of wettability theory is described firstly. The approaches to construct hierarchical micro/nanostructures such as chemical vapor deposition (CVD), electrochemical, etching, and flame synthesis methods are introduced. Then, the advantages and limitations of each method are discussed. Furthermore, the recent progress of superhydrophobicity applied on civil engineering materials and its applications are summarized. Finally, the obstacles and prospects of superhydrophobic civil engineering materials are stated and expected. This review should be of interest to scientists and civil engineers who are interested in superhydrophobic surfaces and novel civil engineering materials.

Keywords: civil engineering material; superhydrophobicity; anticorrosion; anti-icing

1. Introduction

Civil engineering materials (CEM) are the basis of all construction projects, including dwellings, pavements, viaducts, tunnels, mines, airports, and so on [1–4]. Namely, a construction is an integration of multiple CEMs. However, taking concrete as an example, it has an intrinsic porous structure and is prone to damage by corrosion and freeze–thaw cycles, which will result in the decline of the concrete strength and durability and even bring about safety issues [5–8]. Thus, concrete with better performance such as super durability, high corrosion resistance, and stable freezing resistance is demanded. Moreover, there is a desperate need for some CEMs with excellent properties such as self-cleaning and anti-icing to be developed to satisfy the demand of intelligent buildings.

Fortunately, nature provides solutions. For the past few years, bioinspired superhydrophobic surfaces (water contact angle (WCA) larger than 150° and sliding angle (SA) less than 10°) have attracted attention because of their excellent water-repellent properties [9–15]. In 1997, Barthlott and Neinhuis found that the self-cleaning nature of lotus leaves was related to its microscale structure and epicuticular wax [16]. Subsequently, Jiang et al. disclosed that the main reason of the superhydrophobicity of lotus leaves was due to its hierarchical micro/nanoscale morphology [17,18]. After that, the research on the wettability of solid surface was renewed as a hot topic. In this review, we will summarize the recent progress of bioinspired superhydrophobic CEMs and their

applications. The first section will give the basic theory of wettability. The second section will provide the methods of how to develop the superhydrophobic surface. The third section discusses the recent advances in bioinspired superhydrophobic CEMs and their applications. At last, a personable perspective on the prospects and obstacles of superhydrophobic CEMs is proposed.

2. The Theory of Wettability

The wetting of solid surfaces reveals how the different liquids interact with solid objects based on the balance of cohesion and adhesion forces acting at the three-phase contact line. In the absence of a liquid, the solid surface is characterized with intrinsic surface free energy (the excess energy of the surface of any material compared to its volume), which depends on the surface chemistry of the given material [19]. Once a liquid contacts with the solid, this excess energy drives the liquid molecules to bind to the surface (these are the so-called adhesion forces). However, the intermolecular forces acting within the liquid tend to “compactly hold” all liquid molecules and “coerce” the liquid droplet to minimize its energy and acquire a spherical shape, which is the minimum surface area compared to the volume (these are the so-called cohesion forces, which are also known as surface tension). The competition between adhesion and cohesion forces determines whether the liquid will or will not wet the solid and if wetting it, to what extent (the degree of hydrophilicity, hydrophobicity, etc., if the considered liquid is water).

2.1. Young's Equation

In 1805, Thomas Young gave a classical equation of wettability; it represents a force balance at the contact line between the three interfaces (solid–liquid–vapor) [20]. The balance of interfacial forces at the contact line is γ_{sv} , γ_{sl} , and $\gamma_{lv} \cos \theta$ is shown in Figure 1a: a liquid droplet is spread on a flat surface and forms a CA (Contact angle) with the solid surface. At equilibrium, the contact line is static, and these forces must vanish so that the equation can be written and simplified as follows in Equations (1) and (2):

$$\gamma_{sv} = \gamma_{sl} + \gamma_{lv} \cos \theta \quad (1)$$

$$\cos \theta = \frac{\gamma_{sv} - \gamma_{sl}}{\gamma_{lv}} \quad (2)$$

where γ_{sv} , γ_{sl} , and γ_{lv} are the interfacial energy of solid–vapor, solid–liquid, and liquid–vapor, respectively. These equations elucidate the relationship of the three phases (solid, vapor, and liquid) and the CA was called intrinsic CA.

2.2. Wenzel State

However, a solid surface is often rough and uneven in reality, and the real CA on a rough surface is different from the Young's CA. Thus, Wenzel modified the wettability model of Young's state in 1936 [21]. As can be seen in Figure 1b, the Wenzel model supposed that liquid was completely filled into the rough structure. In this case, the surface areas of the solid–liquid and solid–vapor interfaces associated with the advance of the contact line are increased by a factor of r . As the contact line advances along the surface by a small distance, dx , this leads to the change of the total surface free energy,

$$\Delta E = (\gamma_{sl} - \gamma_{sv}) r dx + \gamma_{lv} \cos \theta dx \quad (3)$$

which for local equilibrium, $\Delta E = 0$. Thus, it can be simplified to:

$$\cos \theta^* = \frac{r(\gamma_{sv} - \gamma_{sl})}{\gamma_{lv}} \quad (4)$$

The above equation can be substituted with Young's equation to obtain the Wenzel equation:

$$\cos \theta^* = r \cos \theta \quad (5)$$

where r is the roughness factor (denoted as the ratio of the actual liquid/solid contact area to the nominal contact area), and θ^* represents the apparent CA. In the Wenzel equation, the roughness factor (r) acts as an amplification of the effect of the surface chemistry determined term, $\cos \theta$. For example, when $\theta < 90^\circ$, increasing the roughness factor r can further reduce the apparent CA (θ^*) toward 0° . However, when $\theta > 90^\circ$, increasing the roughness can further increase the apparent CA (θ^*) toward 180° . Thus, from the Wenzel equation, it can be concluded that the hydrophilicity of the intrinsic hydrophilic surface can be enhanced by increasing the roughness. On the contrary, the hydrophobicity of the intrinsic hydrophobic surface also can be improved by raising the roughness. Hence, the Wenzel equation points out that constructing a rough structure is an effective approach to realize the nonwettability of surface with a high CA even larger than 150° .

2.3. Cassie–Baxter State

The Wenzel equation reveals the relationship between the apparent CA and the intrinsic CA of the homogeneous rough surface. An alternative contact between liquid and solid is that the liquid no longer retains complete contact with the solid at all points below the droplet [19]. In this case, the Wenzel equation is not applicable. In addition, the Wenzel equation cannot explain the reason why the water droplets on lotus leaves show an extreme low SA and low contact angle hysteresis.

Based on the previous studies, Cassie and Baxter proposed the viewpoint that the droplet makes contact with a solid surface by composite liquid/solid and liquid/vapor contacts, as exhibited in Figure 1c [22]. The relationship between the apparent CA and rough structures was analyzed by the thermodynamic equilibrium states of vapor–liquid–solid and displayed as following:

$$\Delta E = (\gamma_{sl} - \gamma_{sv})_1 f_1 dx + (\gamma_{sl} - \gamma_{sv})_2 f_2 dx + \gamma_{lv} \cos \theta dx \quad (6)$$

Combined with Equation (2), at equilibrium, the above equation can be simplified to the following:

$$\cos \theta^* = f_1 \cos \theta_1 + f_2 \cos \theta_2 \quad (7)$$

where θ_1 and θ_2 represent the apparent CA of the liquid on solid and vapor (air), respectively, and f_1 and f_2 are the fraction of the contact area of liquid/solid and liquid/vapor ($f_1 + f_2 = 1$), respectively. Since liquid was totally in contact with vapor, $\theta_2 = 180^\circ$. Thus, the above equation can be rewritten as:

$$\cos \theta^* = f_1 \cos \theta_1 + f_1 - 1 \quad (8)$$

That is the famed Cassie–Baxter equation. It can be applied to commendably explain superhydrophobic phenomenon such as “lotus effect”. Generally, a smaller f_1 among hydrophobic surfaces means a low interfacial fraction at the contact line. In addition, it can be considered as a weighted mean (by interfacial fraction at the contact line) of the Young’s equation contact angle and a contact angle against the vapor. In the Cassie–Baxter state, the liquid bridges between surface features and no longer penetrates between the spaces separating them [19]. It resulted in a high apparent CA and lower SA of liquid on a rough hydrophobic surface. Thus, for the demand of superhydrophobic CEM for super durability, high corrosion resistance, and stable freezing resistance, the microstructure of materials should be matched well with the requirement of the Cassie–Baxter regime. Therefore, the construction methods for superhydrophobic structures are summarized in the next part.

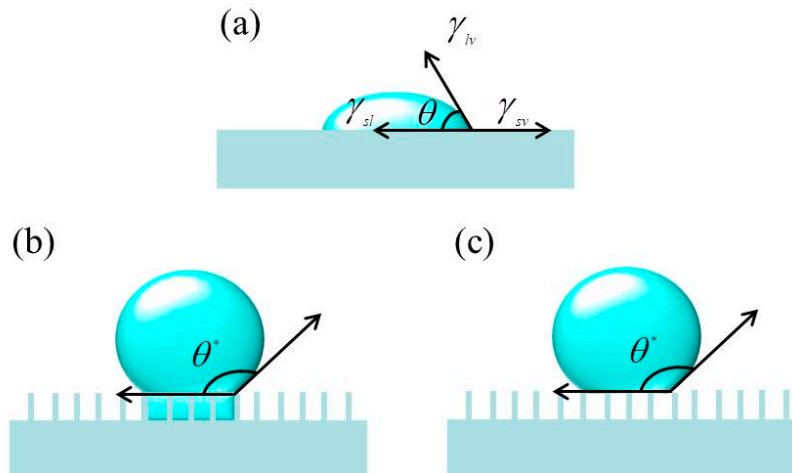


Figure 1. (a) Young's mode on a flat surface; (b) Wenzel mode on a rough surface; (c) Cassie–Baxter mode on a rough surface.

3. Construction of Superhydrophobic Structures

The approaches to fabricate superhydrophobic surfaces can be basically categorized into two directions: construction rough structures on low-surface energy materials and lowering the surface energy of rough structures. Surface energy reduction is relatively easy to achieve [23,24]. Thus, scientists mainly focus on how to construct rough structures, because the SA of superhydrophobic CEMs should be less than 10° for a better performance of anticorrosion, anti-freezing, self-cleaning, and so on. Therefore, the model we refer to is the Cassie–Baxter mode. In this section, the construction methods of Cassie–Baxter structures are addressed in detail.

3.1. Chemical Vapor Deposition (CVD) Method

Chemical vapor deposition (CVD) is a method of producing thin films by chemical reaction on a substrate surface using one or several vapor compounds or elements. Guo et al. fabricated a superhydrophobic surface by aerosol-assisted layer-by-layer chemical vapor deposition (AA-LbL-CVD) of epoxy resins and polydimethylsiloxane (PDMS) polymer films [25]. The obtained surface demonstrated well anti-UV light irradiation and even great mechanical stability. Moreover, due to the shape memory effect of the deposited polymer films, it can be recovered to the previous structure upon heat treatment. Dong et al. synthesized a monolithic three-dimensional (3D) hybrid of graphene and carbon nanotube foam by two-step CVD for the selective removal of oils from water with high absorption capacity and good recyclability [26]. Feng et al. prepared superhydrophobic carbon fibers (SCFs) with a core–shell structure by electrospinning technology combined with a subsequent CVD method; here, the CVD graphitic carbon layer with low surface energy results in a WCA as high as 159.5° [27]. Deng et al. used candle soot as a template which was then shelled with silica by the CVD of tetraethoxysilane (TES) catalyzed by ammonia; the shells were also coated with a semifluorinated silane by CVD to reduce the surface energy [28]. After CVD, the WCA of the droplets on this surface was larger than 165° with a SA less than 1° , demonstrating excellent superhydrophobicity. As shown above, CVD is a powerful technique for constructing superhydrophobic surfaces, regardless of the shape of the substrates. The limitation is that this method needs costly instruments. The deposition rates are also quite slow with the range of tenths of nm/s.

3.2. Etching Method

Etching method is a common method using physical or chemical technology to etch a surface into a rougher structure, which includes chemical etching, laser etching, and plasma etching. Xiu et al. prepared a hydrophobic silicon surface by KOH anisotropic etching with silane treatment [29];

the morphology can be seen in Figure 2a. Afterward, a Cassie state was achieved by further Au-assisted electroless HF/H₂O₂ etching, which is shown in Figure 2b–d with different magnifications; the surface energy was also lowered by silane modification to realize superhydrophobicity. Xue et al. fabricated superhydrophobic poly(ethylene terephthalate) (PET) textile by alkali etching and covered it with polydimethylsiloxane (PDMS) to decrease the surface energy [30]. The water droplet on this treated textile was larger than 150° even after acid/alkaline etching, UV irradiation, and long-time laundering. Jie et al. constructed a micro/nanostructure on a brass substrate by FeCl₃ solution etching combined with a heating treatment approach [31]. The WCA can be as large as 153.6° after modification with stearic acid. Tuo et al. developed an anisotropic superhydrophobic surface on a stainless steel surface by laser etching [32]. They used an ultraviolet laser to etch the steel by rotating the substrate to different angles. The WCA of this surface reached 160° and the SA is 6° after modification by fluoride. Du et al. created a nanotextured surface by oxygen plasma [33]. By modulating the modes of hydrophobic coatings and controlling the hierarchy of nanostructures, droplet mobility can be manipulated easily. As discussed, it can be known that the etching method is efficient for fabricating superhydrophobic surfaces. However, it is not applicable for those substrates that are hard to etch, and it may reduce the strength of substrates.

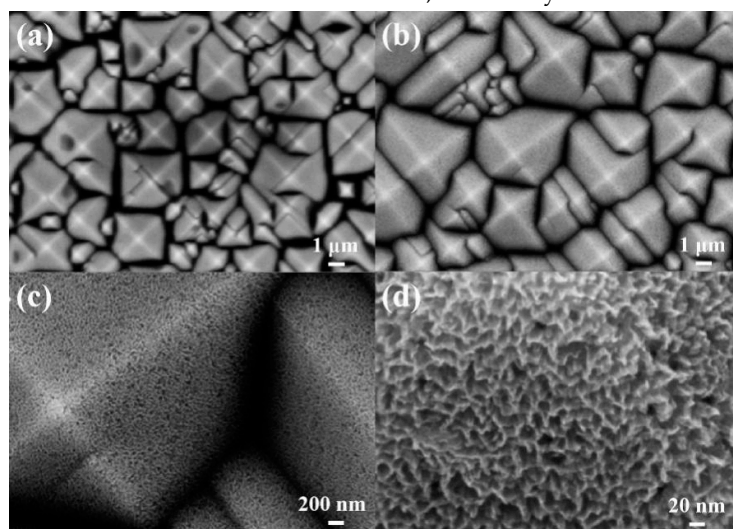


Figure 2. (a) Silicon surface structures after etching in KOH at 80 °C for 25 min; (b), (c), and (d) are the hierarchical structures of a silicon surface from further Au-assisted HF/H₂O₂ etching with different magnifications. Adapted from [29].

3.3. Electrospin Method

Electrospin is a novel and quick fiber manufacturing technology. Polymers can be electrostatically atomized to micropolymer jet flow under a high voltage electric field, and finally solidified to fibers. The morphology of fibers can be regulated by changing the concentration of polymers, the intensity of the electric field, or other parameters. Jiang et al. first utilized this technology to fabricate superhydrophobic polystyrene (PS) fibers [34]; the morphology of PS fibers demonstrated a micro/nanostructure (as shown in Figure 3a,b) while the concentration of PS is 7 wt %. Furthermore, the water droplets on this surface exhibited a spherical shape with a WCA of $160.4 \pm 1.2^\circ$, which can be seen in Figure 3c, and shows a stable superhydrophobicity. Yu et al. also created flexible eco-friendly fibrous membranes with superior waterproofness and extreme breathability via electrospinning, which broadens the application of electrospun fibers [35]. Khayet et al. prepared superhydrophobic nanofibers by in situ surface modification and mechanical strength enhancement via electrospin technology [36]; the morphology is shown in Figure 3d,e with different magnifications. Polysulfone (PSF) was used as a host hydrophilic polymer, and small quantities of a fluorinated polyurethane additive (FPA) were considered for improving the hydrophobicity. It can be seen in Figure 3f that the fibers demonstrated superhydrophobicity, while the concentration of

FPA is higher than 3 wt %. The advantages of the electrospin method for superhydrophobic fields are easy fabricating and handling as well as high efficiency. However, the low strength of the fibers is the fatal defect. The toxic solvent is also a limitation of this method.

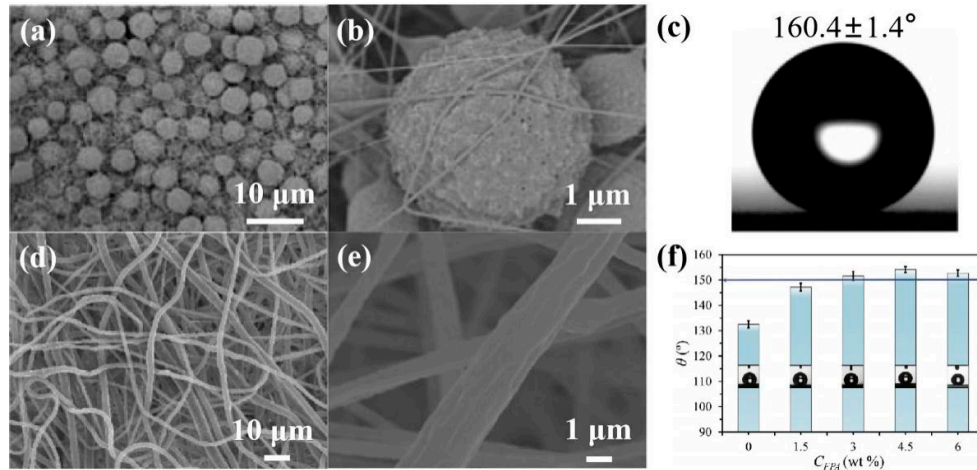


Figure 3. (a) and (b) are the SEM images of polystyrene (PS) fiber prepared from the 7 wt % PS/DMF. Solution with different magnifications, (c) water droplet on this surface; (d) and (e) are the SEM images of PSF-4.5 (weight concentration of fluorinated polyurethane additive (FPA)) and FPA-blended fibers (f) water contact angle (WCA) of droplets on different fibers. Adapted from [34,36].

3.4. Electrodeposition Method

The electrodeposition technique refers to the electrochemical deposition of metals or alloys from aqueous, nonaqueous, or molten salts of their compounds. It is a process of redox reactions. Qing et al. fabricated a rough structure by Zn electrodeposition for superhydrophobic surfaces [37]. Liu et al. fabricated a nickel coating on magnesium alloy taking advantage of the electrodeposition method [38]. They constructed a microstructure by electroless deposition firstly, and subsequently applied a current density of 15 mA/cm² with different time to further create hierarchical micro/nanostructures. After decorating by stearic acid, the WCA was as high as $160.8 \pm 1^{\circ}$ and the SA was lower to $1.8 \pm 1^{\circ}$. Due to the virtues of electrodeposition, including its cost economy, simple process, and easy to control, our team also adopted this method to prepare the superhydrophobic structures. Xiang et al. analyzed the effect of current density on the wettability and corrosion resistance of nickel on low carbon steel and found that the modified nickel coating revealed superhydrophobicity under 6 and 8 A/dm² [39]. In addition, they further deposited a Zn–Ni coating on mild steel as a basic layer and treated with other methods [40], as shown in Figure 4. Three kinds of superhydrophobic coatings such as a porous Zn–Ni–Co coating, a porous ZnO–NiO coating, and ZnO nanorods coating were developed, all of which had WCAs larger than 150° with a low SA. Moreover, a bilayer coating of Ni/Cr was also developed via electrodeposition by our team to enhance the anti-abrasion and microhardness as well as anticorrosion performance [41]. As can be seen above, electrodeposition is an efficient and low-cost method for constructing superhydrophobic structures that are suitable for any metal substrates and can be transferred to industrial production. The disadvantage of this method is that it is not suitable for nonconductive materials.

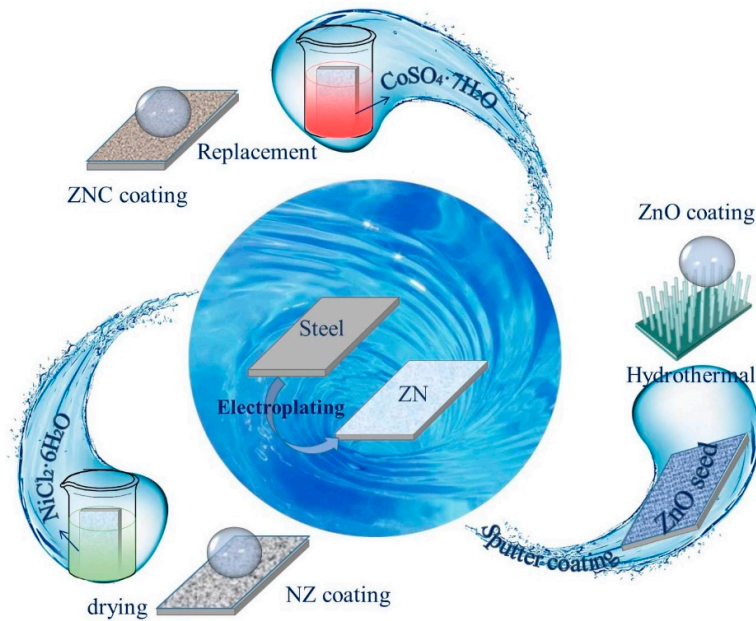


Figure 4. Schematic image of the preparation processes of Zn–Ni–Co coating, NiO–ZnO coating, and ZnO coating, respectively. All of the coatings were based on a Zn–Ni coating. Reproduced from [40].

3.5. Anodic Oxidation Method

Anodic oxidation is the electrochemical oxidation of metals or alloys that are applied as anodes in a cell. Zheng et al. employed this method to develop a stable superhydrophobic coating on an aluminum surface, which was followed by surface modification with inexpensive myristic acid [42]. As shown in Figure 5a, when the voltage is 20 V, the morphology of this coating exhibited a porous structure. Lai et al. reported a robust TiO₂ nanotube array surface composed of vertically aligned side-by-side TiO₂ nanotubes by an anodizing process in fluoride-containing solution [43]. The droplet adhesion can be precisely controlled by regulating the alcohol-based ink removal and patterning as well as UV light. Xiao et al. fabricated a CuO nanoneedle array on a Cu surface by the facile and controllable anodization approach [44]. The Cu(OH)₂ nanoneedles were first synthesized by electrochemical anodization, and could be transform to CuO nanoneedles by a dehydration reaction; the morphology of CuO is shown in Figure 5c,d. This film revealed a very large WCA of 169° and a low SA that was less than 5° after modification. Wang et al. using coaxial anodic oxidation for patterning Ti tubes [45]. An uniform TiO₂ nanotube arrays can be prepared in the inner face of Ti tubes; it demonstrated a WCA of 165° and a SA less than 2° on this surface after lowering the surface energy by trichloro (1H,1H,2H,2H-perfluorooctyl) silane. The disadvantage of this technique is that only can be used for several kinds of metals.

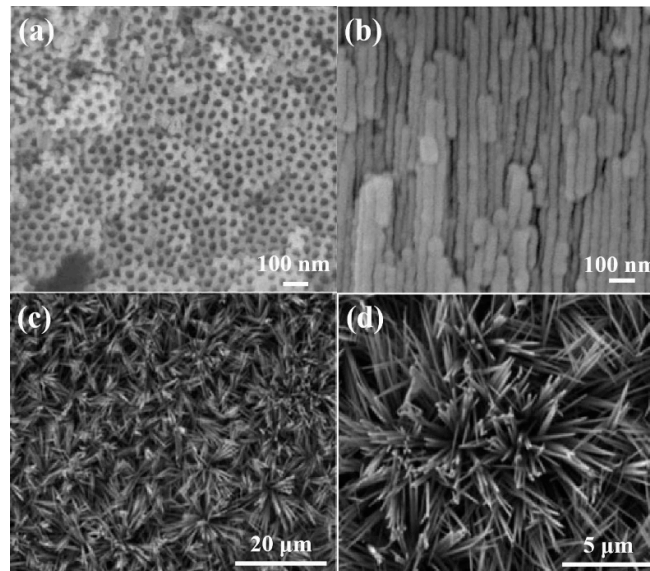


Figure 5. (a,b) The top and cross-sectional view of an anodic oxidation aluminum surface; (c,d) the SEM of a CuO nanoneedle with different magnifications. Adapted from [42,44].

3.6. Sol–Gel Method

The sol–gel method can accurately alter the surface morphology and roughness by adjusting the parameters of the operation process or the raw materials. Latthe et al. synthesized adherent and porous silica film on glass substrates while tuning the ratio of methyltriethoxysilane (MTES) and tetraethoxysilane (TEOS) to 0.43 [46]. This surface displayed a porous structure that can be seen in Figure 6a; the silica film demonstrated a WCA of 160° , as shown in Figure 6b, and a SA less than 3° after modification. Wu et al. combined low and high surface energy SiO_2 nanoparticles to prepare superhydrophobic and superoleophobic coatings via the sol–gel method [47]. The low surface energy SiO_2 nanoparticles were prepared by adding an amount of particles into the hydrolyzed 1H,1H,2H,2H-perfluorooctyltriethoxysilane (PFOTES) solution; the morphology of these particles is shown in Figure 6c,d. They also demonstrated that the hydrophobicity of coatings can be tuned by changing the ratio of low and high surface nanoparticles. Yang et al. fabricated a superhydrophobic cotton textile using fluorinated TiO_2 sols via a sol–gel method [48]. This surface showed a WCA up to 152.5° with good chemical stability. The sol–gel method is one of the most likely methods to be applied in real life because of its high efficiency, low cost, ease of control, and so forth. The disadvantage is the low adhesion between the coating and the substrate. Interestingly, glue might be able to solve this problem [49].

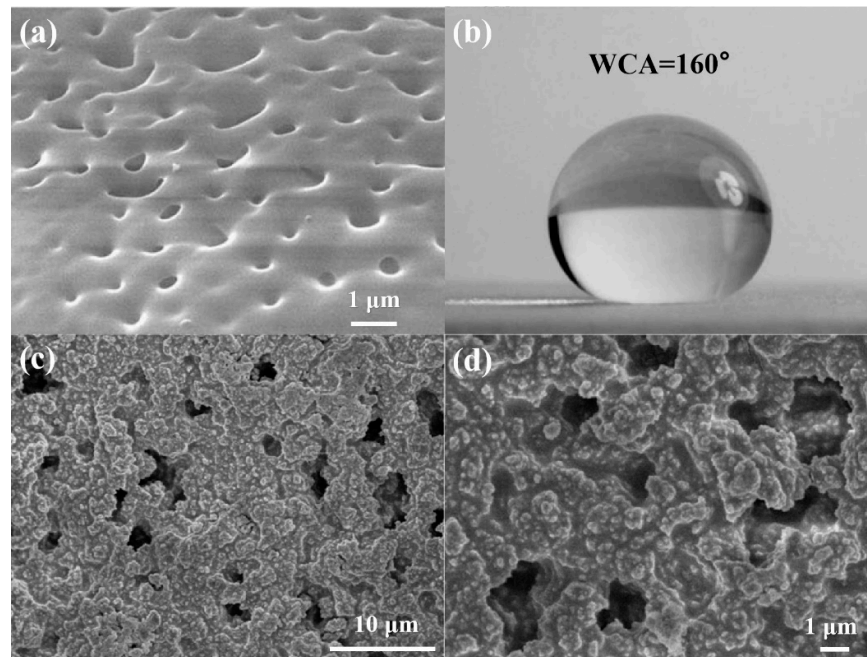


Figure 6. (a) SEM image of porous silica film; (b) water droplet on the as-prepared surface; (c,d) SEM images of superhydrophobic SiO₂ coating with different magnifications. Adapted from [46,47].

3.7. Hydrothermal Method

The hydrothermal method is a method of preparing materials with water as solvent and powders dissolved and recrystallized in sealed pressure vessels such as teflon-lined autoclave. The morphology of products can be precisely controlled by regulating the reaction temperature, reaction time, or the concentration of chemicals. Wan et al. developed a superhydrophobic copper surface for corrosion protection via the hydrothermal method [50]. The copper was etched in ammonia solution for 20 h and then placed in an autoclave containing 60 mL of NaOH solution (0.2 mol/L) to react for 2 h at 200 °C. The surface showed a WCA of $157.7 \pm 1^\circ$ after modification by stearic acid. Jin et al. fabricated a superhydrophobic magnesium hydroxide coating on an AZ31B magnesium alloy [51]. The cleaned magnesium alloy was placed into an autoclave that included 20 mL of deionized water for 2–5 h at 160 °C and formed a porous structure, which is shown in Figure 7a–d. Zhou et al. prepared superhydrophobic TiO₂ films on glass for self-cleaning and anti-icing [52]. The titanium tetraisopropoxide was used as the titanium source and dissolved in the solution of 15 mL of HCl and 15 mL of deionized water; the glass was placed at an angle of 45° on the wall of the autoclave and reacted for a certain time at 150 °C. As a result, TiO₂ nanorods can be generated; the morphologies of different samples can be seen in Figure 7e–g. Chen et al. synthesized Dahlia-like Co₃O₄ nanoparticles on porous nickel foam for water–oil separation via the hydrothermal method [53]. The production procedure can be seen from the reference in detail. As shown in Figure 7h–j, the first image is the morphology of original nickel foam, the second image is the rough foam acidized by hydrochloric acid, and the last one is the superhydrophobic foam with Co₃O₄ nanoparticles (as shown in the insert in Figure 7j). The limitations of this technique include its long reaction time, the few kinds of materials that can be prepared, and mass production, which is difficult to realize.

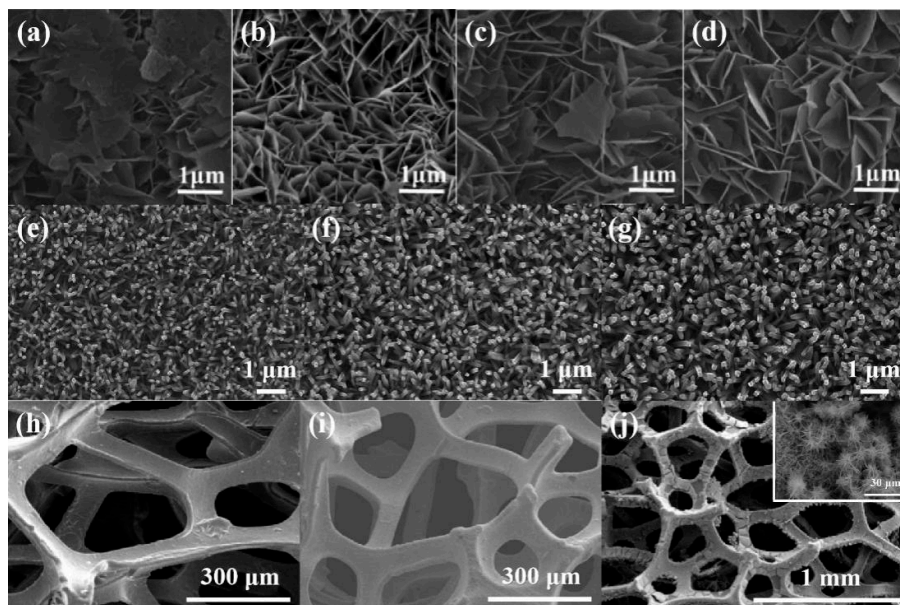


Figure 7. (a–d) SEM images of hydrothermal-treated samples for different times: 2, 3, 4 and 5 h; (e–g) SEM images of TiO₂ nanorods with different reaction times: 6, 8, and 10 h; (h–j) SEM of original, acidized, and superhydrophobic nickel foam; the insert figure is the SEM of Co₃O₄ nanoparticles that were grown on the nickel foam. Adapted from [51–53].

3.8. Flame Synthesis Method

The flame synthesis method is one of the most promising methods for real-world applications. Qu et al. first fabricated a superhydrophobic surface by depositing the soot of burning rapeseed oil [54]; this result opened a new avenue for superhydrophobic research with these easily obtained carbon nanospheres. After that, Yuan et al. developed a self-cleaning flexible infrared nanosensor based on the carbon nanoparticles, which was obtained from alcohol burning [55]. They also used this method to fabricate carbon nanoparticles on a carbon fabric for flexible and high-performance field emitters [56]. As described in Section 3.1, Deng et al. using candle soot as a template for fabricating a superhydrophobic surface [28]. Seo et al. also revealed the possibility of making a superhydrophobic surface via the flame of a candle [57]. As can be seen in Figure 8a,b, they developed a superhydrophobic surface on paraffin wax-coated glass; it can be seen that the candle soot-coated slide demonstrated a large WCA of 162°. Based on the development of the flame synthesis method, large numbers of researchers have tried to employ different approaches for stabilizing the flame-generated coatings, as at that time they possessed low mechanical durability [58–66]. Qahtan et al. fabricated water jet-resistant and thermally stable superhydrophobic surfaces by a spray coating of candle soot dispersion [67]. Gerald et al. sprayed a mixture (containing toluene, polydimethylsiloxane, and inherently hydrophobic rapeseed soot) on mesh for oil/water separation [68]. Esmeryan et al. fabricated inherently robust superhydrophobic carbon soot that does not require any additional stabilizers or chemical treatment; the SEM image of this coating can be seen in Figure 8c [69]. Furthermore, they revealed the possibility of generating carbon materials with a controllable fraction of amorphous graphite-like and diamond-like phases [70]. Moreover, Esmeryan et al. developed an adjustable optical transmittance of superhydrophobic carbon soot coatings by an in situ single-step control of their physicochemical profile [71]. These works reveal the possibility of generating inherently robust superhydrophobic coatings with desirable physicochemical characteristics by controlling in situ the fuel combustion. These findings are of fundamental importance, since they open a new direction in the flame synthesis, namely the opportunity to fabricate and deposit stable and cost-effective superhydrophobic coatings in less than a minute on large-scale objects.

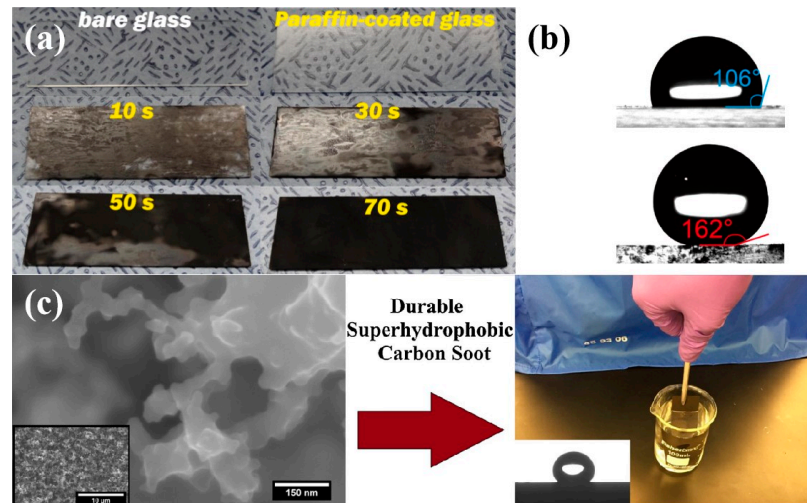


Figure 8 (a) Images of soot on a paraffin wax-coated glass slide with various sooting times; (b) static contact angle on a paraffin wax-coated glass slide (top image) and on a paraffin wax-fixed candle soot-coated slide, respectively; (c) The SEM image of carbon soot and the WCA on the as-prepared surface; the right figure is the optical image of the as-prepared surface immersed in liquid. Adapted from [57,69].

For improving the wear resistance of the soot, Zulfiqar et al. stabilized the thin layer of carbon soot by mixing it with sawdust particles [72]. Lin et al. presented mechanically durable superhydrophobic PDMS-candle soot-based composite coatings through a rapid process [73]. Esmerlyan et al. presented novel silver-doped soot coatings with enhanced wear resistance and antimicrobial performance [74]. As summarized above, the flame synthesis method processes large numbers of superiorities for real-world applications. Kelesidis et al. and Mulay et al. reviewed the advantages of flame synthesis and the soot transition from a highly neglected hazardous pollutant to a functional material [75,76]. Compared with other methods, the flame synthesis method exhibits the following advantages. Firstly, the film deposition takes place from a gas phase in the surrounding environment (outside vacuum chambers, CVD, magnetrons, etc.), which allows the superhydrophobization of objects with large dimensions and complex geometry and curvature. Secondly, the flame synthesis is possible using extremely inexpensive precursors such as rapeseed oil, candles, and alcohol. Thirdly, the flame synthesis supports very high deposition rates compared to CVD: around units of $\mu\text{m/s}$. Last but not least, small changes in the fuel's chemistry and/or deposition conditions allow a precise adjustment of the physicochemical properties of the surface (coating) depending on the desired practical application.

3.9. Other Methods

There are a variety of methods to construct hierarchical micro/nanostructures. Qing et al. developed a robust superhydrophobic surface using sandpaper as a template to lock in the fluorinated inorganic/organic film, which demonstrated exceptional mechanical robustness, good pressure stability, and repellency to hot water [77]. Zhang et al. prepared a zeolite antireflective (ZAR) coating via the layer-by-layer (LBL) method [78]; they created micro and macroporous structures to realize antireflective property. Moreover, this coating revealed a transparent superhydrophobicity after being grafted with 1H,1H,2H,2H-perfluorooctyltriethoxysilane (POTS). Xie et al. fabricated superhydrophobic fertilizers via the self-assembly of uniformly dispersed Fe_3O_4 superhydrophobic nanoparticles on the outermost surface of a mixture [79]. Liu et al. synthesized superhydrophobic silica aerogel powders by a phase separation-induced method through the direct hydrophobization of silica alcogels, which was followed by ambient pressure drying [80]. Hensel et

al. fabricated an omniphobic surface by a reverse imprint lithography approach using perfluoropolyether dimethacrylate templates cast from the silicon master [81].

4. Superhydrophobicity on the Application of CEMs

As we described above in the introduction, CEMs with super durability, high corrosion resistance, anti-icing, and other excellent properties are in high demand. Traditional methods such as using seal coating to block the pores of concrete for water repellency would bring about the increase of internal pressure because of the moisture produced within the concrete [82–84]. When the internal pressure goes up to the limit, the seal coating will be destroyed, losing the sealing effect. Hydrophobic treatment is another efficient method to improve the impermeability of CEMs. Based on the rapid development of superhydrophobic materials, here we review and discuss the recent progress of superhydrophobicity on the application of different CEMs in detail. The superhydrophobic CEMs and their applications can be seen in Figure 9.

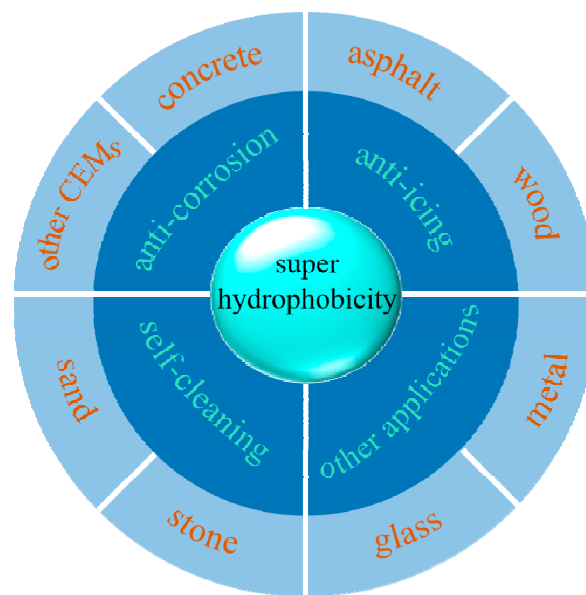


Figure 9. Diagram of the categories of civil engineering materials (CEMs) and their applications.

4.1. Concrete

As it is well known that concrete is a porous hydrophilic material, water and corrosive ions ingress are the two main factors to decrease its durability. In the process of corrosive ions permeation, water acts as the carrier or transport medium. Water itself also can be the reactant in alkali–silica reaction or freeze–thaw deterioration. Thus, the effective approach to prolong the durability of concrete is to prevent water infiltrating into its structure. Flores-Vivian et al. fabricated hydrophobic emulsions (enriched with the polymethyl–hydrogen siloxane oil hydrophobic agent as well as metakaolin or silica fume) and applied them on Portland cement mortar tiles to develop superhydrophobic concrete [85]. Ramachandran et al. controlled entropic molecular interactions to design a novel icephobic concrete. This kind of concrete showed a low ice adhesion strength and could repel incoming water droplets at $-5\text{ }^{\circ}\text{C}$ [86]. Zhao et al. fabricated a superhydrophobic surface on concrete for anti-icing [87]. As depicted in Figure 10a, a fluorine resin solution was first sprayed on the concrete as an adhesive layer and then coated with fluorosilane-functionalized silica to realize the superhydrophobicity. From Figure 10b,c, it can be seen that water droplets spread quickly on the uncoated concrete and formed an ice layer in 5 s at $-20\text{ }^{\circ}\text{C}$, while it showed spherical ice balls on the superhydrophobic concrete (as shown in the insert in Figure 10b); after propagation for a long time, the ice layer covered the superhydrophobic concrete. The as-prepared superhydrophobic concrete surface demonstrated an excellent anti-icing performance with low ice adhesive strength. In

addition, in the light of anti-icing property, Esmerlyan et al. presented a rational strategy for atmospheric icing prevention based on chemically functionalized carbon soot coatings; the strategy is also suitable for anti-icing concrete [88]. Furthermore, they controlled the hydrophilic active sites to delay condensation and frost formation on superhydrophobic carbon soot coatings, which is also a useful perspective for preventing ice condensation on concrete [89]. She et al. developed a superhydrophobic concrete surface by spraying modified silica gel to simultaneously construct a hierarchical micro/nanostructure and lower the surface energy [90]. The water droplets on the surface can be seen in Figure 10d with a WCA of $162 \pm 3^\circ$. Moreover, the water absorption rate of this superhydrophobic concrete was reduced by 95.2% compared with uncoated dry concrete, which is shown in Figure 10e.

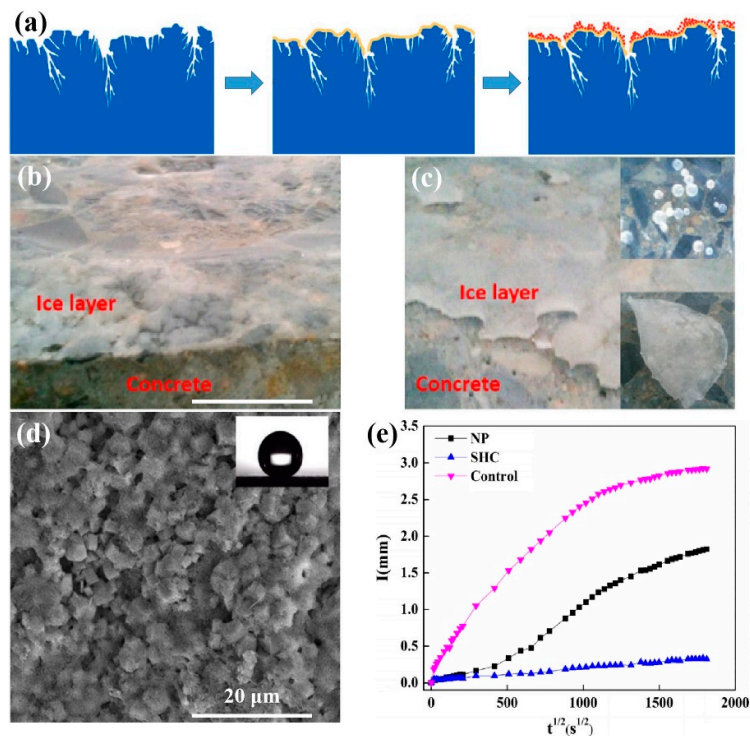


Figure 10. (a) Schematic illustration of the process of fabricating a superhydrophobic concrete surface via the spray method; (b) (c) the ice formation on an uncoated and coated surface at -20°C , the insert figures are the formation of ice balls at the beginning of icing (top image) and the ice piece after removing (bottom image); (d) SEM image of a coated concrete surface, the insert figure is the water droplet on the surface; (e) water absorption results of different concrete surfaces. Adapted from [87,90].

Even though superhydrophobic surfaces exhibit plenty of advantages, their weak mechanical robustness severely restricts their application in real-world settings [91]. To this end, Song et al. directly fabricated super robust superhydrophobic concrete without any coatings via adding fluoroalkylsilane (FAS) into the concrete slurry and using Cu mesh to construct a micro/nanostructure [92]. As can be seen in Figure 11a,b, water droplets also showed spherical shapes on the as-prepared concrete and could rebound back after falling down, which was demonstrated by the extreme low adhesion. It can be concluded that the concrete also can protect steel bars for concrete reinforcement; as shown in Figure 11c, the steel bar displayed a bright color in superhydrophobic concrete while exhibiting red rust in the original concrete. In addition, the superhydrophobic concrete also possesses an excellent anti-icing property, which is shown in Figure 11d. Apart from adding FAS, they also developed another superhydrophobic concrete by adding a low-cost commercial water-based stone protector, which also demonstrated outstanding anti-icing

and anticorrosion performance [93]. Although Song et al. and other researchers obtained some excellent results, Kulinich et al. found that surfaces grafted with alkyl groups are prone to the gradual degradation of their hydrophobic and icephobic properties, which is caused by interactions and reactions with both ice and liquid water [94]. As a result, the long-term durability of these materials would be decreased. Thus, in case of civil engineering, we should avoid this phenomenon. Here, some other low-surface-energy materials with unhydrolyzed properties should be developed to address this problem. Grafting with an inactive group may be also useful for increasing the durability.

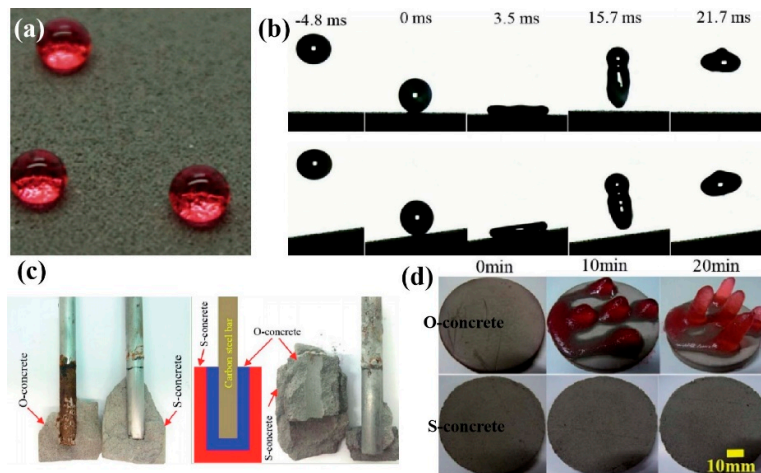


Figure 11. (a) Water droplets on the as-prepared superhydrophobic concrete; (b) dynamic bouncing processes of water droplets on the superhydrophobic concrete flat and tilted surface; (c) corrosion behavior of steel bar in different concrete; (d) the macromorphology of droplets on different concrete at $-5\text{ }^{\circ}\text{C}$ for 20 min. Adapted from [92].

4.2. Asphalt

Asphalt is another widely used CEM for pavement due to its advantages, including low noise and vibration, fine wear resistance, and driving comfort. However, the ice formation on asphalt is a big threat for driving safety. Peng et al. fabricated a superhydrophobic coating on asphalt by spraying modified layered double hydroxides (LDH) and room temperature vulcanized silicone rubber (RTV) with No. 120 solvent naphtha evaporation [95]. This coating demonstrated a long freezing time and low adhesion force between the ice and the surface. Zakerzadeh et al. examined the effect of various superhydrophobic coatings on asphalt pavement for the purpose of reducing the adhesion of the ice and snow to the pavement [96]. They measured the WCAs of different coatings to evaluate the hydrophobicity and found that the as-called W4 exhibited a largest WCA of 156° . Gao et al. studied the effectiveness of the anti-icing and deicing performance of superhydrophobic asphalt concrete and found that the superhydrophobic surface could delay the frost formation and evolution as well as decrease the adhesion force between the ice and surface [97]. Chen et al. reviewed the developments on deicing and verified that the hydrophobicity had high correlation with icephobicity, which indicated that superhydrophobicity could be applied on asphalt for anti-icing [98].

4.3. Wood

Wood is one of the earliest CEMs used by human beings. It was widely employed in structural engineering due to its light weight, high strength, workability, and environmental friendliness. Nevertheless, wood easily absorbs water, which results in the decrease of strength, shortening the service life. Wang et al. fabricated a superhydrophobic coating on poplar lumber substrate [99]. A layer of lamellar ZnO coating was directly grown on the poplar lumber surface by the wet chemical method and followed by stearic acid modification. For improving the mechanical robustness of the

superhydrophobic coating, a polyvinyl alcohol (PVA)/silica (SiO_2) composite polymer coating was also synthesized and coated on the wooden surface [100]. The preparation process is shown in Figure 12a. The water droplets on the as-prepared surface demonstrate superhydrophobicity with a WCA of 159° , which can be seen in Figure 12b. The mechanical property was evaluated via filter paper abrasion; as can be seen in Figure 12c, the surface still exhibited superhydrophobicity even after abrasion for 25 cm under the pressure of 10,000 Pa, indicating an excellent mechanical robustness. Shah et al. prepared a durable superhydrophobic coating by using alumina nanoparticles to create a rough structure and modifying the coating with polydimethylsiloxane (PDMS) to lower the surface energy [101]. As shown in Figure 12d, water droplets displayed spherical shapes on the surfaces even after 1 h of water immersion. Figure 12e,f displayed the changes in the WCAs on two different substrates with a certain holding time and the water uptake ratio of uncoated and coated wood, respectively. The coated woods revealed a very low water uptake ratio, demonstrating efficient water repellency.

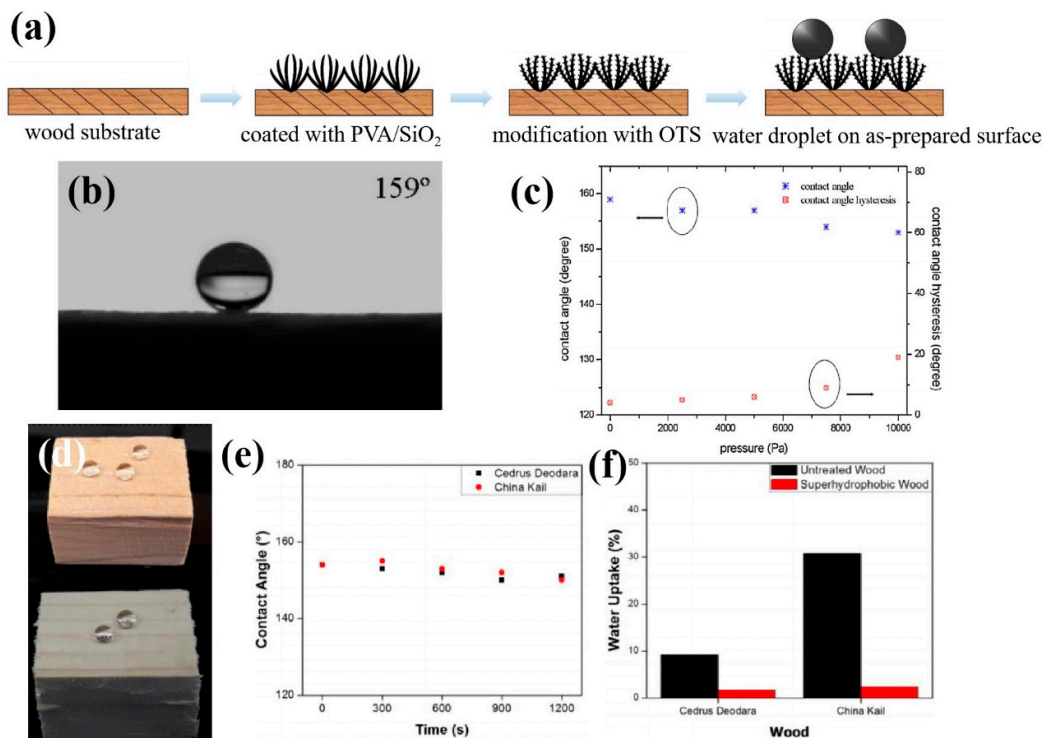


Figure 12. (a) Schematic diagram of the preparation process of superhydrophobic wood; (b) water droplet on the as-prepared surface; (c) the effect of filter paper abrasion on the WCA and contact angle hysteresis under different pressure with a 25-cm abrasion length; (d) water droplets on the as-prepared surfaces after 1 h of water immersion; (e) WCAs of droplets on the surface with different holding times; (f) water uptake ratio of uncoated and coated woods. Adapted from [100,101].

4.4. Structural Steels

Structure steel, whether used for producing the various sections or as a reinforcement material in Portland cement concrete structure material members, is another commonly used CEM because of its outstanding features such as its excellent physical, mechanical, and process performance. However, metals are susceptible to corrosion in humid environments, and may bring about huge safety hazards. Constructing a bioinspired superhydrophobic surface is one of the most promising techniques to solve this issue. Latthe et al. fabricated a superhydrophobic surface on steel by sulfuric acid etching with silane modification [102]. As shown in Figure 13a, the treated steel displayed superhydrophobicity even after bending for 90° . Moreover, this superhydrophobic steel exhibited a low corrosion current density, which indicated its corrosion protection. Zhang et al. developed a

superhydrophobic coating on steel by two steps [103]. Firstly, a porous and hierarchical silica film was directly fabricated on steel by electrochemical deposition. Afterwards, silane was used to modify the surface to endow its superhydrophobicity. This coating exhibited excellent corrosion resistance and good mechanical abrasion resistance as well as thermal stability. Shen et al. prepared a superhydrophobic coating on Al via the hydrothermal method for enhancing the self-propelling dynamics of condensed droplets; this coating displayed a huge potential for inhibiting the attachment of condensed droplets [104]. Zheng et al. discussed the mechanism and impact factors of an anti-icing/icephobic surface and presented a new performance evaluation method for anti-icing/icephobic, which can be used for metal surfaces. They suggested that the anti-icing surfaces can not only delay the freezing time, but also can reduce the freezing point [105]. Huang et al. also summarized the anti-icing coating for aerospace application, which demonstrated the ability to repel water droplets, delay ice nucleation, and significantly reduce ice adhesion [106]. Ruan et al. prepared rough alumina surfaces with a large WCA of 157.6° ; this surface can delay the icing time and reduce the freezing temperature to 65 min and -8.3°C , respectively [107]. Kulinich and Farzaneh studied the effect of wetting hysteresis on ice adhesion strength, and they found that the ice adhesion on rough hydrophobic surfaces has no correlation with the values of WCA but is related to wetting hysteresis—that is, the ice–solid contact area [108]. Kulinich et al. also discussed whether the superhydrophobic surface can really be used for ice repellent [109], and the results showed that the ice-repellent properties of the materials deteriorate during icing/deicing cycles, as surface asperities appear to be gradually damaged, and water condensation both on top of and between the surface asperities takes place, leading to significantly larger values of ice adhesion strength. Thus, this work demonstrated that superhydrophobic surfaces cannot always be used for anti-icing. Briefly, considering the different test methods of ice adhesion, Ronneberg et al. compared the ice adhesion using different methods at two different laboratories and presented that the repeatability should be considered a key parameter in determining the ideal ice adhesion test method [110]. Zang et al. created a superhydrophobic coating on Mg alloy that was inspired by the lotus seedpod [111]. As can be seen in Figure 13c, the TiO_2 film was grown via an in situ hydrothermal method; the electroless plating combined with a self-assembling technique was subsequently applied to generate the superhydrophobicity. The fabricated coating revealed an extremely low SA and adhesion, which is shown in Figure 13d. This coating also demonstrated superior corrosion resistance. As shown in Figure 13e, the superhydrophobic coating exhibited the lowest corrosion current density and largest corrosion potential, revealing the greatest corrosion protection performance.

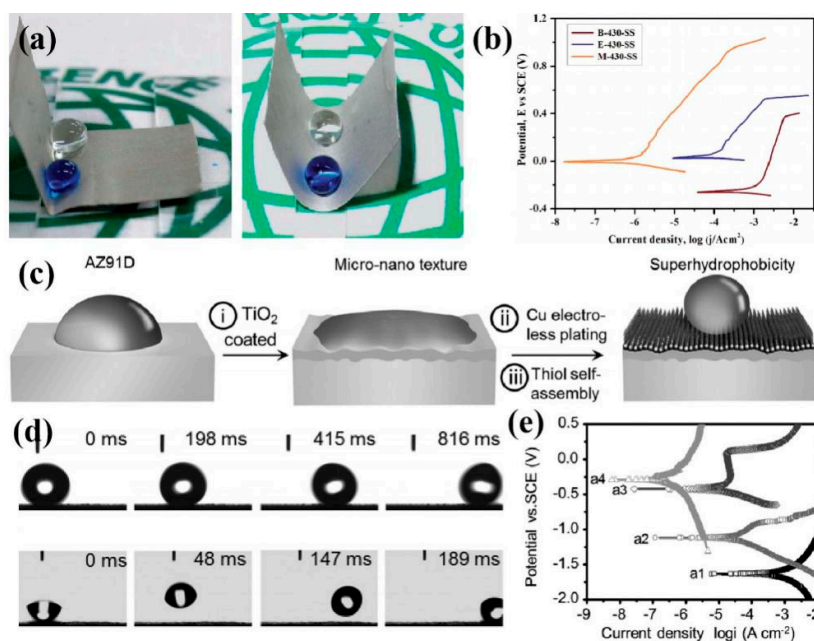


Figure 13. (a) Water droplets on a bent superhydrophobic steel; (b) Tafel plots of different surfaces after immersion in 5 wt % NaCl aqueous solution; (c) schematic illustration of the fabrication process of superhydrophobic Mg alloy; (d) the sliding and bouncing behavior of a water droplet on a superhydrophobic surface; (e) Tafel plots of different surfaces. Adapted from [102,111].

4.5. Glass

Glass has been widely applied in building windows for its good light and broad vision. Thus, it is significant to maintain its transparency. Generally, the way to keep glass clean and transparent is to wash by the “spider man” technique, which is time-consuming and fraught with danger. A superhydrophobic coating can help glass maintain its transparency based on its self-cleaning property. Shang et al. fabricated a transparent porous silica coating on a glass substrate by the layer-by-layer method [112]. The image of Figure 14a displays water droplets on the superhydrophobic glass, which indicates its transparency. Zuo developed a superhydrophobic coating on glass via the magnetron sputtering of nano ZnO particles [113]. As shown in Figure 14b, this coating exhibited preminent thermal stability even after being treated under 100 °C. Furthermore, it revealed its great self-cleaning property, which can be seen in Figure 14c. Saxena and Paria first created fractal patterns on glass and then immersed it into the PTFE nanoparticles solution to form a superhydrophobic coating [114]. As depicted in Figure 14d, a water droplet on the PTFE-coated glass showed a spherical shape while exhibiting wetting behavior on plain glass. Moreover, the coated glass revealed a low adhesion, which can be seen in Figure 14e. Lin et al. created transparent glass by an ultrafast laser technique [115]. As shown in Figure 14f,g, the as-prepared glass displayed super transparency as well as hydrophobicity; the transparency is over 92% in the visible light. With the exposure time increasing, the glass still exhibited a large WCA and low SA, which is shown in Figure 14h, indicating a sensational stability.

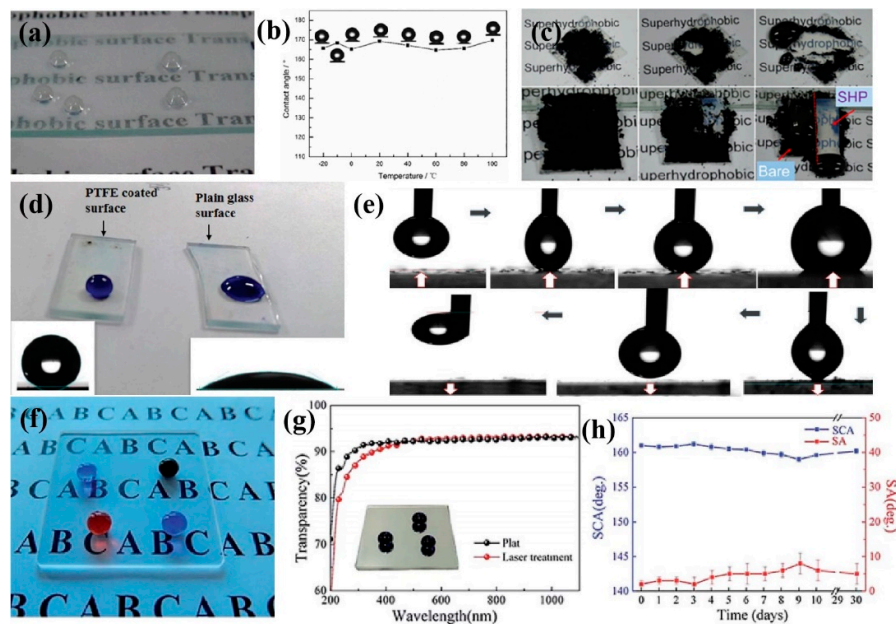


Figure 14. (a) Water droplets on superhydrophobic glass; (b) WCA of glass after being treated at different temperatures for 24 h; (c) self-cleaning property of superhydrophobic glass; (d) water droplets on PTFE-coated and uncoated glasses; (e) the adhesion testing process of the prepared surface; (f) water droplets on a designed superhydrophobic glass lying on paper; (g) transparency of the optimal designed surface in visible light; (h) WCAs and sliding angles (SAs) of the superhydrophobic glass surface with different exposure times. Adapted from [112–115].

4.6. Stone and Sand

Facio and Mosquera applied a superhydrophobic coating on building sandstone. The as-prepared coating produced a rough structure in which the air is trapped so that water droplets cannot penetrate into the coating [116]. As shown in Figure 15a, water jet flow wetted the untreated stone while bouncing back from the coated stone. Besides, the product provided a promising adhesion with the substrate and preserved the aesthetic qualities of the substrate. Karapanagiotis et al. prepared organically modified silica gels and used them to coat several stones even on monuments [117]. The WCA and SA can reach 165° and 4° , respectively. This is the first report of a superhydrophobic organically modified silica gel film that does not contain nanoparticles. Aslanidou et al. sprayed siloxane emulsion, which included silica nanoparticles on white marble and sandstone to protect and conserve buildings and monuments of cultural heritage [118]. In Figure 15b, it can be seen that water can be repelled after applying the film coating, which exhibited an excellent self-cleaning property. The preparation process of this coating provides a significant improvement without the use of any organic solvent, revealing exceptional environmental friendliness.

Furthermore, scientists also developed superhydrophobic sand for civil engineering and other applications. Men et al. fabricated superhydrophobic sand by adding octadecyltrichlorosilane into the preparation solution [119]. These sands demonstrated a WCA of $154.8 \pm 1^\circ$ and an SA of $4.5 \pm 0.9^\circ$, which can be used for oil/water separation. Chen et al. fabricated superhydrophobic sands by cladding SiO_2 and metal (Ag and Cu) on sand surfaces and then modified them to lower the surface energy [120]. As shown in Figure 15c, water droplets could wet the raw sand while displaying spherical shapes on the other three kinds of sands. Based on the authors' viewpoints, these sands can be used to address the issues of desert water storage and transportation.

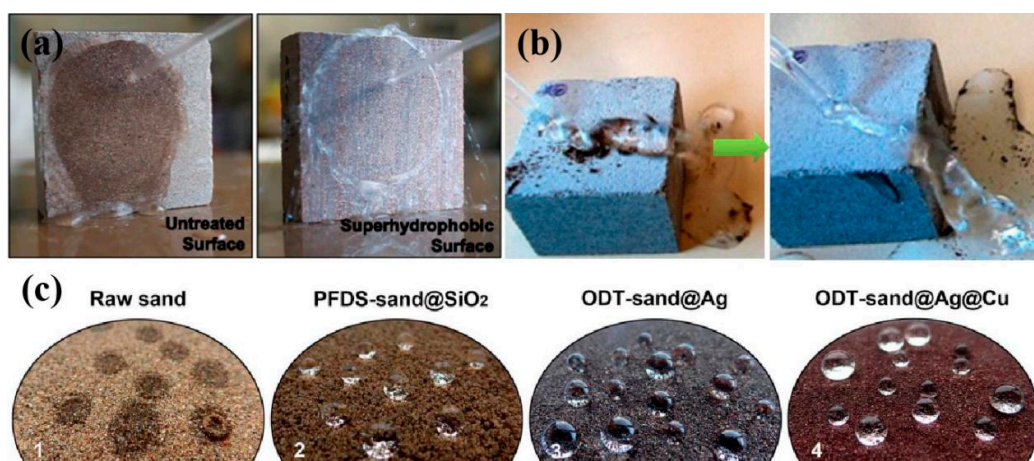


Figure 15. (a) Optical images of water jet flow on untreated and treated stone surfaces; (b) the self-cleaning property of the treated sandstone surface; (c) the wetting states of water droplets on different sand surfaces. Adapted from [116,118,120].

4.7. Other CEMs

Superhydrophobicity also has been applied on other CEMs such as aerogels for building thermal and acoustic insulation, glass fibers for enhancing the strength of building components, coatings for building fire protection, etc. Malakooti et al. prepared polybenzoxazine aerogels via a facile method that is significantly low cost [121]. This aerogel demonstrated a good mechanical strength with superhydrophobicity. Moreover, it still kept the stability and robustness even after heat treatment at 250°C , so it must be a good candidate material for building thermal insulation. Thai et al. developed a novel rubber aerogel from car tire waste for the first time via the freeze-drying method [122]. This aerogel also showed an excellent heat insulation property and good thermal stability even up to 500°C . It had a better sound insulation performance than a commercial sound foam absorber at 2000–3000 Hz and exhibited a noise reduction coefficient of 0.41.

It can be used for building thermal and sound insulation. Lv et al. grafted polysiloxane onto the surface of glass fibers to improve their hydrophobicity [123]. Their study revealed that the interfacial shear strength increased by 36.52% compared with that of the original glass fiber composites. Zhao et al. fabricated a superoleophobic dry chemical agent for the re-ignition of pool fires [124]. The dry chemical agent can be splashed onto oils to prevent oil evaporation, which can quickly quench pool fires. Lin et al. constructed superhydrophobic and flame-retardant coatings on cotton fibers by a one-pot treatment which included tetraethoxysilane (TEOS), hydroxyl-terminated polydimethylsiloxane (PDMS), and ammonium polyphosphate (APP) [125]. When this cotton fiber was exposed to fire, it quickly charred to extinguish the fire because of the physical barrier effect of the PDMS–silica and the flame-retardant effect of APP. This coating can be an excellent substitute for building waterproofing and antifiaming. Arianpour et al. studied the hydrophobic and ice-retarding properties of doped silicone rubber coatings; they also obtained the result that the delayed ice formation and lower adhesion strength on superhydrophobic samples were related to small wetting hysteresis, which is similar to the above results in Section 4.4 [126]. Lazauskas et al. studied the water droplet behavior on superhydrophobic SiO₂ nanocomposite films, in which the substrate was lithium aluminosilicate glass ceramic [127]. Some other superhydrophobic materials with special applications may be used on CEMs in the future. For example, Lv et al. proposed a strategy combining the material structure design with a voltage control for a better selectively to receive the specific frequency of electromagnetic wave with less loss [128]. They also conducted other works for excellent electromagnetic absorption, which can be used on outer walls [129–132]. Esmeryan et al. developed superhydrophobic soot-coated quartz crystal microbalances for human spermatozoa quality assessment [133,134]. They also found that the superhydrophobicity is beneficial for the cryopreservation of human spermatozoa [135]. These exciting findings may be useful for the assessment and storage of CEMs. In addition, Cao et al. designed superhydrophobic and superoleophilic copper mesh, which was assisted by candle soot for oil–water separation [136]. Nazhipkyzy et al. impregnated magnetic soot particles with cobalt, nickel, and iron salts for removing oil contamination from water [137].

5. Conclusion and Perspective

In this review, we have described the development history of the wettability theory, explained the application scopes of these theories, summarized the newest methods for constructing hierarchical micro/nanostructures, and concluded the recent progress of superhydrophobicity on CEMs and its applications. It is self-evident that superhydrophobic surfaces have attracted a significant amount of attention, promoting the development of surface and interface science. However, surface wettability is a complex phenomenon involving many disciplines, including material science, chemistry, physics, mechanical engineering, and so forth. The relationships between contact angle hysteresis, surface morphology, and surface energy are still not clear. Most fabrication methods are complicated and hard to apply in industry settings. For the CEM field, the low-cost and fast methods are still in demand. Moreover, the mechanical stability is an important issue that needs to be addressed for CEMs. Recently, a slippery omniphobic surface has been created, which was inspired by *Nepenthes* pitcher plants, and gave us a new insight for solving the stability of water-repellent surfaces for self-cleaning, anticorrosion, anti-icing etc. From our perspective, the stimuli-responsive superhydrophobic surfaces on CEMs such as smart windows that can adjust the brightness or color, smart concrete, and asphalt that can self-heal should be brought into more research fields since they may provide some interesting results. Last but not least, broadening the applications of superhydrophobicity to other CEMs is also a promising direction.

In summary, benefitting from the advancing micro and nanofabrication techniques, appropriate methods should be developed for real life. Furthermore, numerous magical wetting properties in nature such as mussels with high adhesion will give us endless inspiration to develop novel CEMs. We must note that superhydrophobic surfaces can highly impact other fields that are related to building, such as air conditioner external units and refrigerator for better heat exchange, outdoor

antennas for anti-icing, interior conduits for drag reduction, and so on. We believe that superhydrophobic materials will dramatically advance CEMs and our lives.

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