

## Review Article

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# Antifouling induced by surface wettability of poly (dimethyl siloxane) and its nanocomposites

<https://doi.org/10.1515/ntrev-2022-0552>

received January 3, 2023; accepted April 19, 2023

**Abstract:** Antifouling technologies have attracted considerable attention in recent years, as numerous fouling phenomena pertaining to inorganic, organic, bio-, and composite foulants substantially affect daily life. Poly (dimethyl siloxane) (PDMS) has several practical applications; however, it possesses limited resistance to inorganic, organic, or biofoulants such as proteins or bacteria. Among the antifouling strategies reported thus far, antifouling induced by surface wettability (AFISW) is an exceptional strategy with considerable potential. It presents numerous advantages such as a physical working

mechanism, eco-friendliness, and facile material fabrication process. To achieve AFISW, PDMS can be modified with several nanomaterials to tune its surface wettability to meet antifouling requirements. This article presents a systematic review of the existing research on AFISW in PDMS to achieve improved antifouling performance. Specifically, we first provide a background on fouling, focusing on the different types of fouling and antifouling mechanisms. Then, we provide a comprehensive review of AFISW based on four types of surface wettability, namely, superhydrophilicity, hydrophilicity, hydrophobicity, and superhydrophobicity. Finally, we discuss suitable AFISW strategies for different types of fouling mechanisms based on PDMS and its nanocomposites. This review will help researchers design and fabricate various polymeric materials and their nanocomposites with tailored surface wettability for AFISW applications.

**Keywords:** antifouling, surface wettability, PDMS

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## 1 Introduction

Antifouling strategies have recently attracted considerable research attention because of the numerous fouling phenomena involving inorganic, organic, bio-, and composite foulants, which substantially affect daily life [1]. The earliest known documentation of fouling is a papyrus dating back to 412 BCE [2]. Foulants such as dust, ice, crude oil, barnacles, bacteria, and blood have tangible impacts, such as the degradation of material surfaces, increased ship drag resistance, and higher probability of infection in hospitals [1–10]. For example, marine fouling, a typical type of biofouling, usually causes severe economic losses of approximately US\$ 150 billion in the transportation industry and necessitates the use of 80,000 tons of antifouling paint per year. In addition, it results in adverse ecological impacts (e.g., the production of harmful compounds due to high fuel consumption and toxic antifouling coatings for marine ecosystems) [11]. Silicone-based polymers such as poly(dimethyl siloxane) (PDMS)

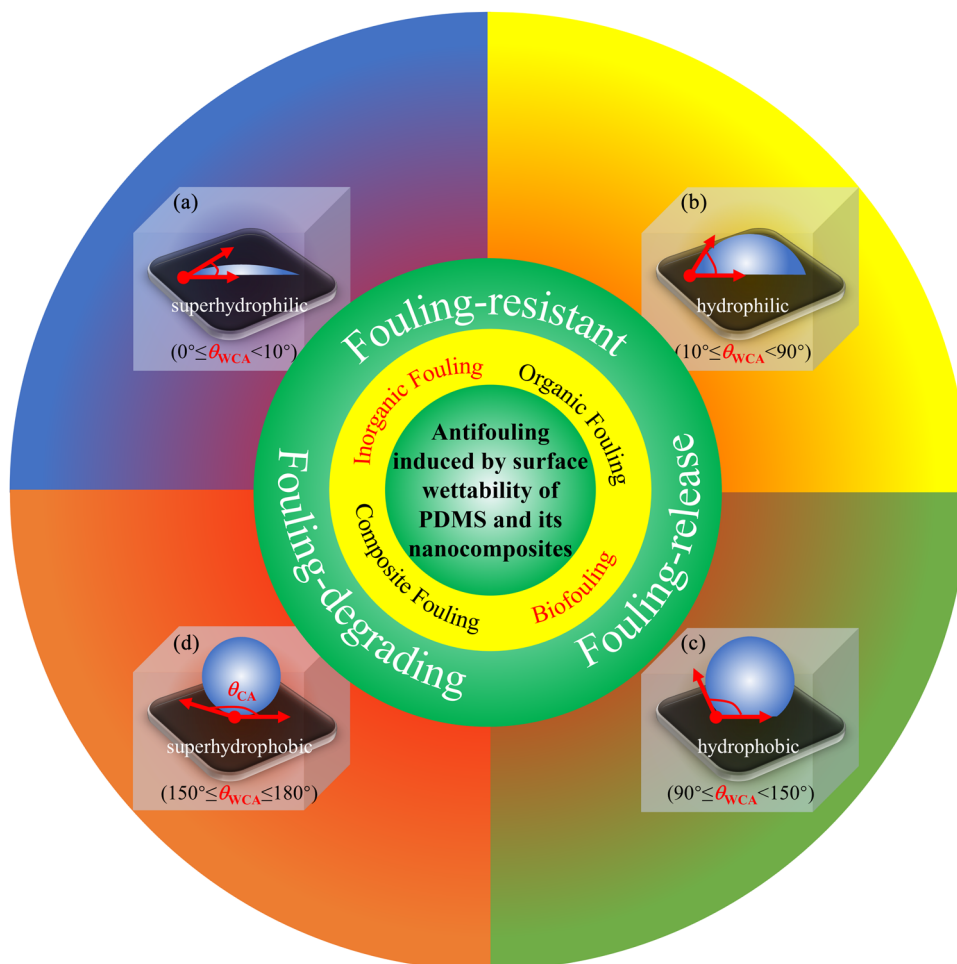
and its nanocomposites are extensively applied in elastomers, stretchable electronics, cosmetics, antifoaming agents, flexible sensors, and biomedical devices [12–16]. PDMS has various advantages owing to its optical transparency, chemical stability, biocompatibility, and acceptable cost [15]. The flat PDMS surface is hydrophobic, with a water contact angle (WCA) of approximately  $100\text{--}110^\circ$  [17–20]. Although it shows some resistance to inorganic foulants, its resistance to organic or biofoulants, such as protein or bacterial attachment, is low [17]. PDMS and its nanocomposites have various applications in everyday life, and their surface wettability can be easily tuned to meet antifouling needs. Therefore, it is necessary to conduct a systematic review on PDMS and its nanocomposites to provide a reference for future improvements in their antifouling performance.

Among the several antifouling strategies reported thus far, antifouling induced by surface wettability (AFISW) has numerous advantages, such as a physical working

mechanism, eco-friendliness, and a facile fabrication process. Figure 1 summarizes the four common types of foulants (the innermost ring, in yellow), three developed antifouling mechanisms (the middle ring, in green), and the corresponding AFISW strategies (the outmost ring) based on various types of surface wettability. As the interactions between the foulant and surface [21] and surface wettability [22] are both determined by the chemical composition and/or physical structures on the surface, it is easy to achieve good antifouling performance by controlling the surface wettability.

## 1.1 Fouling types

Fouling is commonly classified into four types, namely, inorganic, organic, bio-, and composite fouling, according



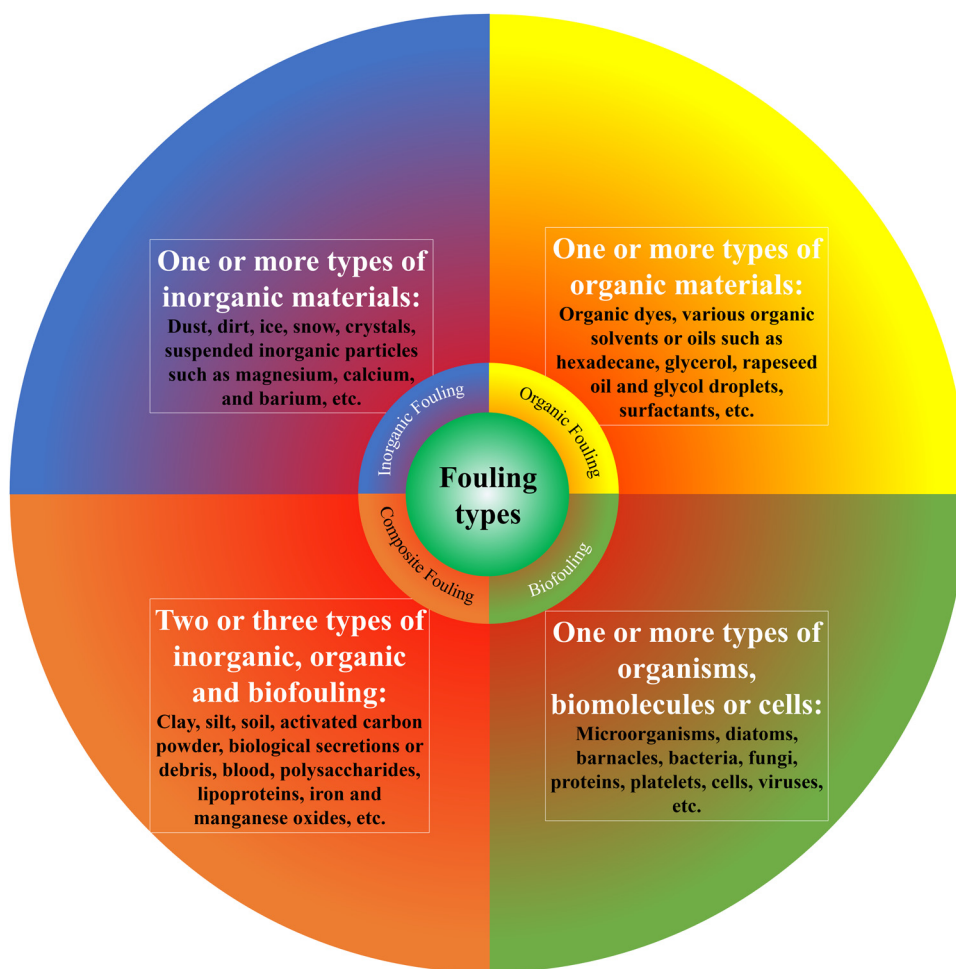
**Figure 1:** AFISW of PDMS and its nanocomposites. Reprinted with permission from He *et al.* [1]. Copyright 2021, Elsevier B.V.

to the foulant [1]. These types and some specific examples of foulants are provided in Figure 2. Inorganic fouling involves one or more types of inorganic materials, and organic fouling refers to the adsorption of one or more types of organic compounds onto a surface. Moreover, biofouling refers to the accumulation of unwanted organisms, biomolecules, or cells on a surface, and composite fouling can be regarded as the combination of two or three of the other previously mentioned types of fouling. In practical applications, fouling rarely involves only one type of foulant; in fact, most of the fouling is caused by different types of inorganic and organic foulants. However, antifouling strategies are easy to design and are usually suitable for any type of inorganic or organic foulant because of their similarities. Conversely, antifouling is more complicated in the cases of bio- and composite fouling because of the diversity of biofoulants and the mixture of two or three types of foulants (inorganic, organic, or bio-), respectively. Therefore, the current research is focused on the

design and fabrication of antifouling materials for bio- and composite fouling [23–25].

## 1.2 Antifouling strategies

Most antifouling strategies can be divided into three types: fouling-resistant, fouling-release, and fouling-degrading strategies (Figure 3) [21,26]. The earliest strategy to combat biofouling was using a coating of biocides or enzymes to degrade the attached foulant [27]. However, chemical coatings containing toxic organotin, copper, *etc.* are currently restricted or prohibited [28]. Next, researchers proposed a fouling-release strategy based on self-polishing coatings. Because these coatings undergo hydrolysis in their side chains or degradation in the main chain, foulants accumulated on them can be easily removed [29,30]. Nevertheless, hydrolysis or degradation reactions still



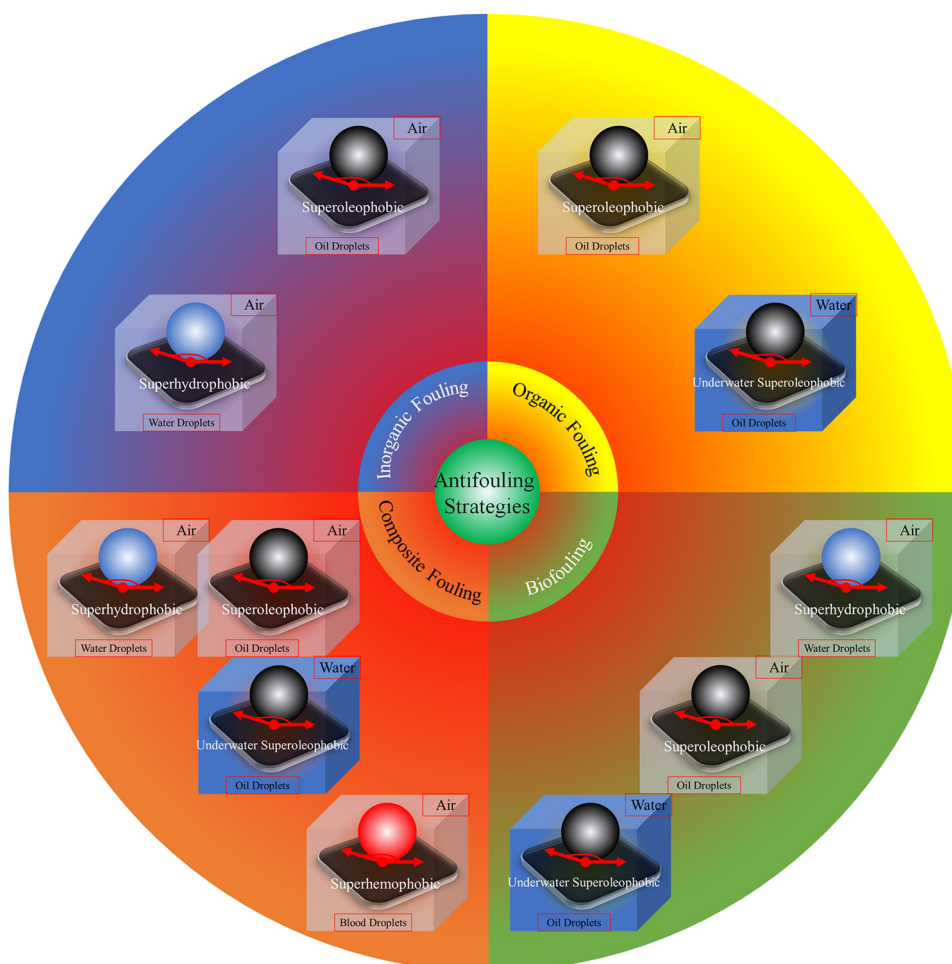
**Figure 2:** Four types of fouling mechanisms based on the nature of foulant. Reprinted with permission from He *et al.* [1]. Copyright 2021, Elsevier B.V.



**Figure 3:** Three common antifouling strategies: fouling-resistant (left), fouling-release (middle), and fouling-degrading (right). Reprinted with permission from Maan *et al.* [21].

have negative environmental effects. Other green fouling-release coatings used low-surface-energy materials, such as silicone- and fluoro-based ones, to reduce the adhesion of foulants [27,31–33], enabling foulant removal by scouring; this is one type of AFISW that usually entails hydrophobicity. Among the fouling-release, fouling-degrading, and fouling-resistant strategies, the fouling-resistant strategy is the optimal one, as it hinders the retention of foulants on

the surface in the first place. Furthermore, the fouling-resistant strategy is another type of AFISW that usually entails superhydrophobicity. Famous natural examples of the fouling-resistant strategy include the lotus leaf, rice leaf, and shark skin effects [34–53]. Inspired by the lotus leaf effect [1], we have comprehensively reviewed the strategies against the four types of fouling in terms of different superphobicities (Figure 4), namely superhydrophobicity



**Figure 4:** Antifouling strategies based on various superphobic surfaces. Reprinted with permission from He *et al.* [1]. Copyright 2021, Elsevier B.V.

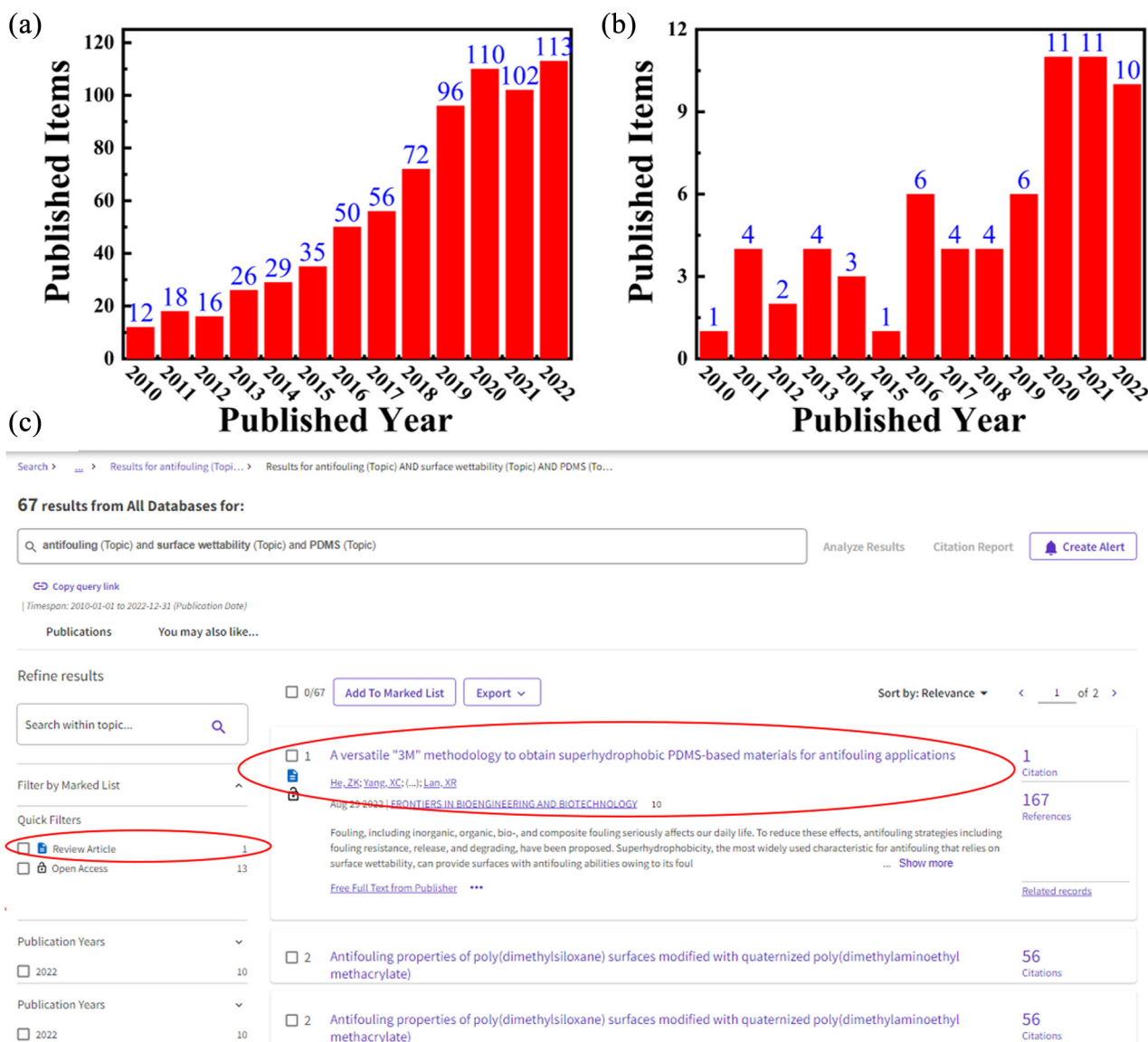


in the air [19,54], superoleophobicity in the air [55,56], superhemophobicity in the air [57,58], and underwater superoleophobicity [59,60]. However, besides superhydrophobicity, it is necessary to further investigate the efficiency of various “surface wettabilities” (i.e., superhydrophilicity, hydrophilicity, and hydrophobicity) on the antifouling ability, referred to here as AFISW.

## 2 Methods

As the interaction between the foulant and surface is determined by the chemical composition and physical structures of the surface [21], careful control of these

two surface characteristics can result in good antifouling performance. Meanwhile, many biological structures such as the lotus leaf, rice leaf, and shark skin have the excellent antifouling ability, as well as a special natural surface wettability that is tunable by controlling the chemical composition and/or physical structures of the surface [22]. Therefore, considerable research has been conducted on AFISW [1,23–25,61,62]. In the last 13 years (Jan 1, 2010–Dec 31, 2022), AFISW has attracted growing attention, as illustrated by the number of publications found in the Web of Science related to “antifouling” and “surface wettability” (Figure 5a). Because the focus of this review is PDMS, a detailed statistical analysis of the publications with the word “PDMS” in the topic was performed. As shown in Figure 5b and c, topics pertaining to “antifouling,” “surface



**Figure 5:** Number of publications in the Web of Science (Jan 1, 2010–Dec 31, 2022) containing the words “antifouling” and “surface wettability” (a), number of publications containing the words “antifouling,” “surface wettability,” and “PDMS” (b and c).

wettability,” and “PDMS” have attracted increasing attention. Most importantly, there is only one review article, published by our group (red circles in Figure 5c), on this topic as of Dec 31, 2022. In the previous review article [61], we focused only on the effect of “superhydrophobic” PDMS-based materials on antifouling applications, whereas in this study, we conducted a comprehensive review of the effect of various surface wettabilities on the antifouling ability.

Surface wettability, usually characterized by a WCA, is affected by the surface free energy of a solid surface, which is based on Young’s equation [63]. Because the WCA ranges between 0 and 180°, the surface free energy of any solid surface is less than 72 mJ/m<sup>2</sup>. In addition to the effect of surface free energy, surface roughness greatly affects surface wettability according to the Wenzel and Cassie–Baxter models [64,65]. Usually, the surface roughness can enhance the surface wettability, that is, the hydrophobicity and hydrophilicity will increase with increasing surface roughness. Therefore, surface wettability is determined by both surface free energy and surface roughness. This review focused on surface wettability based on WCA measurements instead of the surface free energy and/or surface roughness because WCA is visualizable and thus easily measured. The surface wettability can be classified into superhydrophilic ( $\theta_{\text{WCA}} < 10^\circ$ ), hydrophilic ( $10^\circ \leq \theta_{\text{WCA}} < 90^\circ$ ), hydrophobic ( $90^\circ \leq \theta_{\text{WCA}} < 150^\circ$ ), and superhydrophobic ( $150^\circ \leq \theta_{\text{WCA}} \leq 180^\circ$ ) (Figure 6) [1,23]. In the following section, we provide a comprehensive overview of the antifouling of PDMS and its nanocomposites according to the four types of surface wettability.

## 3 Results

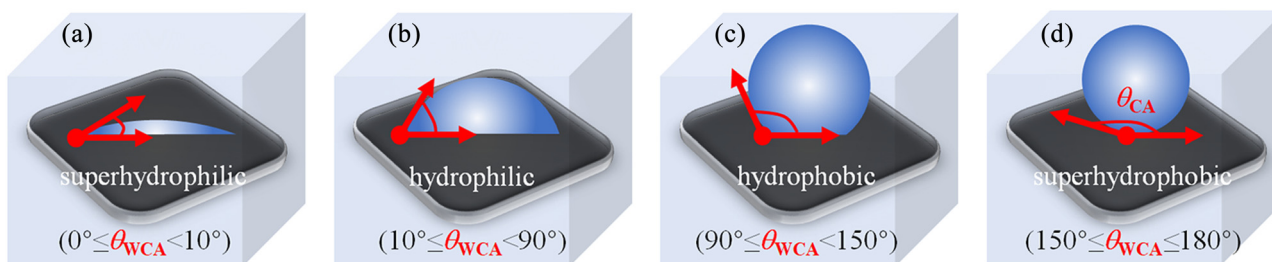
### 3.1 Superhydrophilicity

Because PDMS is naturally hydrophobic, many hydrophilic polymers can be used to modify PDMS and its nanocomposites:

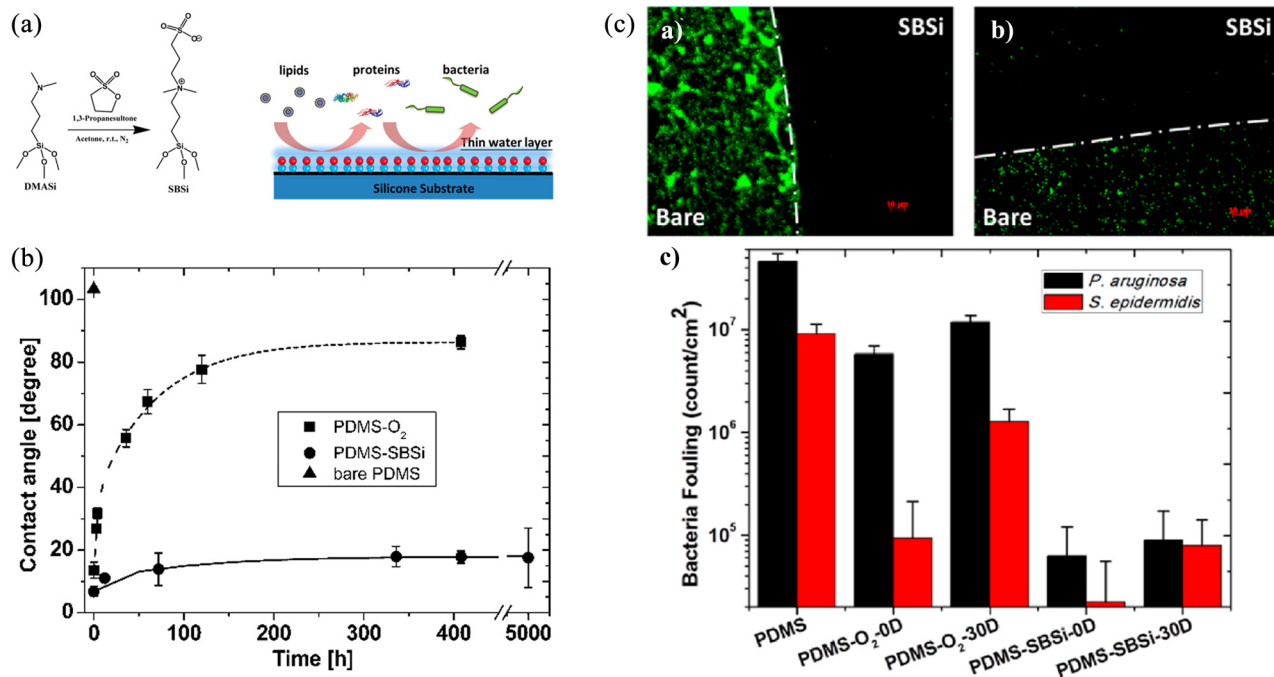
poly(ethylene glycol) (PEG), PEGylated polymers, poly(2-hydroxyethyl methacrylate), polysaccharides, and zwitterionic polymers (e.g., poly(sulfobetaine methacrylate), poly(carboxybetaine methacrylate), and poly(carboxybetaine acrylamide)) [66–85]. Huang and coworkers developed a stable superhydrophilic zwitterionic interface on PDMS by the covalent silanization of sulfobetaine silane (SBSi) [86]. A thin water layer was formed between foulants and this superhydrophilic PDMS because of the hydrophilic zwitterionic polymer, resulting in an excellent antifouling ability (Figure 7a).  $\theta_{\text{WCA}}$  on the superhydrophilic PDMS was approximately 6.8° and remained below 20° after more than 5,000 h of storage (Figure 7b). This surface showed effective resistance to biofouling by both *Pseudomonas aeruginosa* and *Staphylococcus epidermidis* bacteria, even after the surface was stored at room temperature for 30 days (Figure 7c).

### 3.2 Hydrophilicity

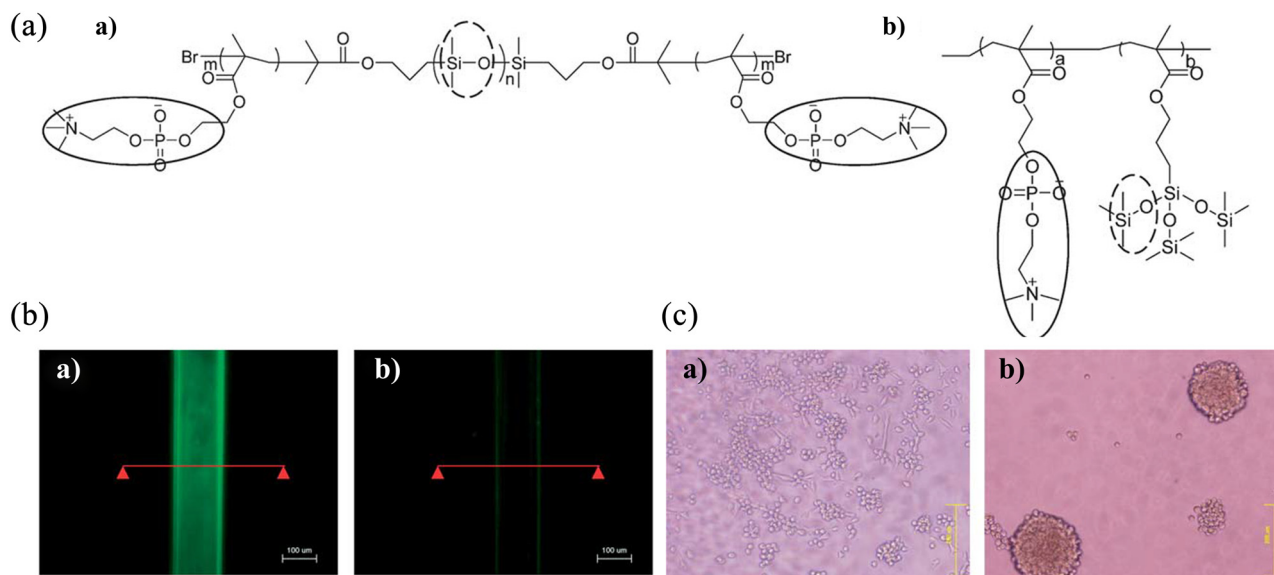
Hydrophilic materials can also be used to modify the surface wettability of PDMS [87–90]. In our previous study, a facile dip-coating strategy was used to fabricate a hydrophilic-coated anti-biofouling bioprosthetic heart valve (BHV) using PDMS and poly(acrylic acid) [25]. Anti-biofouling properties, including anti-coagulation, anti-cell adhesion, anti-calcification, and ability to resist BSA adsorption, were characterized both *in vivo* and *in vitro*. The results showed that BHV with hydrophilic modification had better anti-biofouling abilities than either the control sample or the sample with hydrophobic modification. Ishihara and coworkers modified a PDMS surface with an amphiphilic copolymer composed of 2-methacryloyloxyethyl phosphorylcholine (MPC) and dimethylsiloxane (DMS) units [88]. Block- and random-type copolymers (Figure 8a) with three different compositions were coated on the PDMS surface in a protic solution. The modified



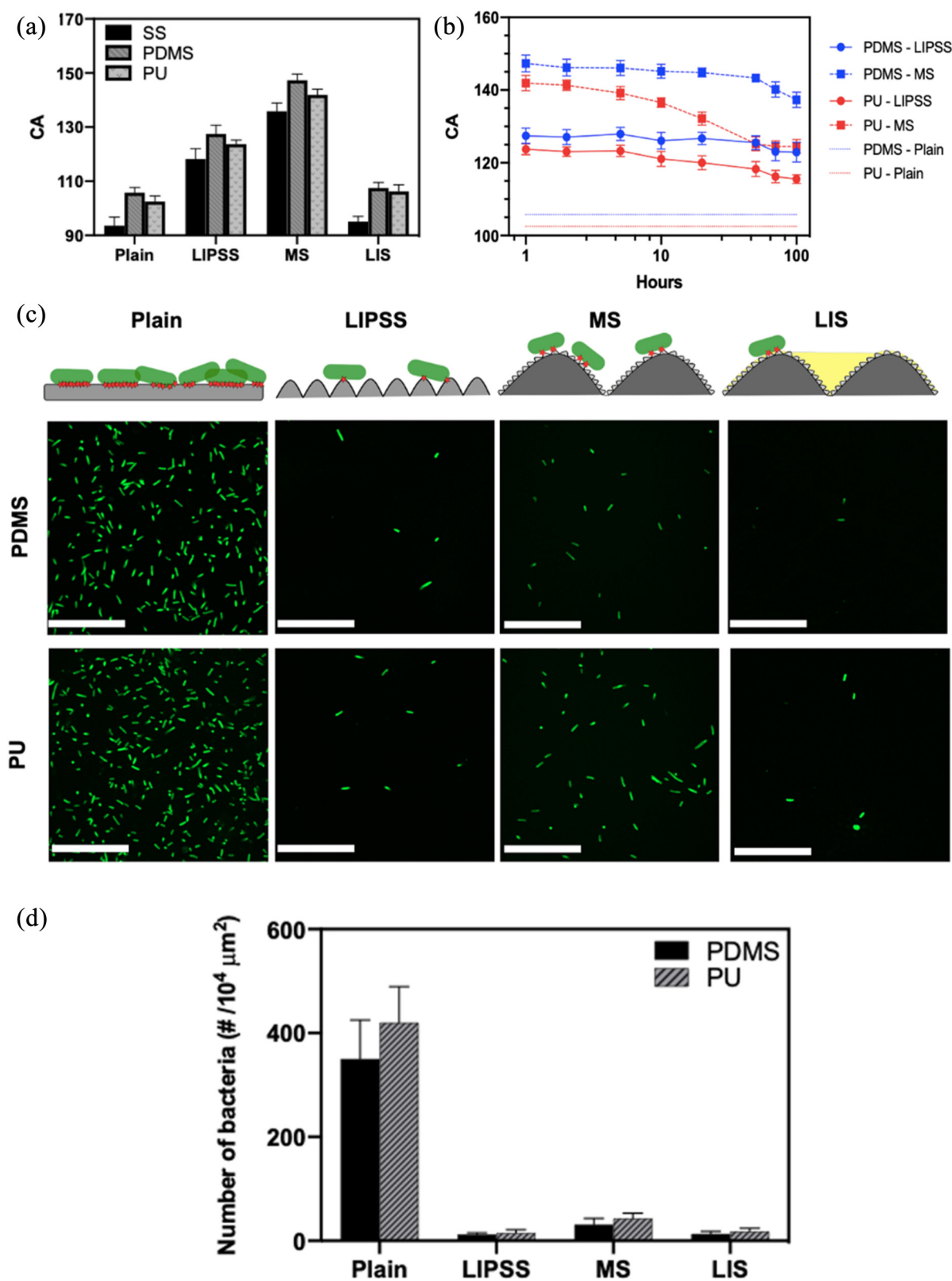
**Figure 6:** Classification of different types of surface wettability, containing superhydrophilic (a), hydrophilic (b), hydrophobic (c), and superhydrophobic (d) based on WCA ( $\theta_{\text{WCA}}$ ) of water droplets in the air. Reprinted with permission from He *et al.* [1]. Copyright 2021, Elsevier B.V.



**Figure 7:** (a) Synthesis of SBSi and antifouling mechanism of the superhydrophilic PDMS surface. (b)  $\theta_{\text{WCA}}$  on different PDMS surfaces over time. (c) Fluorescence micrographs showing the absorption of (a) *P. aeruginosa* and (b) *S. epidermidis* onto partially modified PDMS. (c) Quantification of bacterial adsorption on different PDMS surfaces after 0 or 30 days). Reprinted with permission from Yeh *et al.* [86]. Copyright 2014, American Chemical Society.



**Figure 8:** ((a) (a) Block- and (b) random-type copolymers composed of MPC and DMS units). ((b) Fluorescence microscopy images of FITC-labeled bovine serum albumin adsorption on (a) bare and (b) hydrophilic PDMS microchannels). ((c) Optical microscopy images of adhered cells on (a) bare and (b) hydrophilic PDMS surfaces. Reprinted with permission from Seo *et al.* [88]). Copyright 2011, Royal Society of Chemistry.



**Figure 9:** (a) Static WCA on stainless steel (SS), PDMS, and polyurethane (PU) with various surface structures: plain surface, LIPSS, MS, and lubricant-impregnated surface. (b) Variation of WCA on PDMS and PU with various structures after different immersion times. (c) Schematic representation of bacteria attachment on various surfaces (top) and fluorescence micrographs of bacterial attachment on PDMS and PU surfaces (bottom). (d) Numbers of attached bacteria on PDMS and PU with various structures. Reprinted with permission from Siddique *et al.* [17]. Copyright 2020, American Chemical Society.



surfaces showed obviously reduced protein adsorption (Figure 8b) and cell adhesion (Figure 8c) compared to the unmodified PDMS.

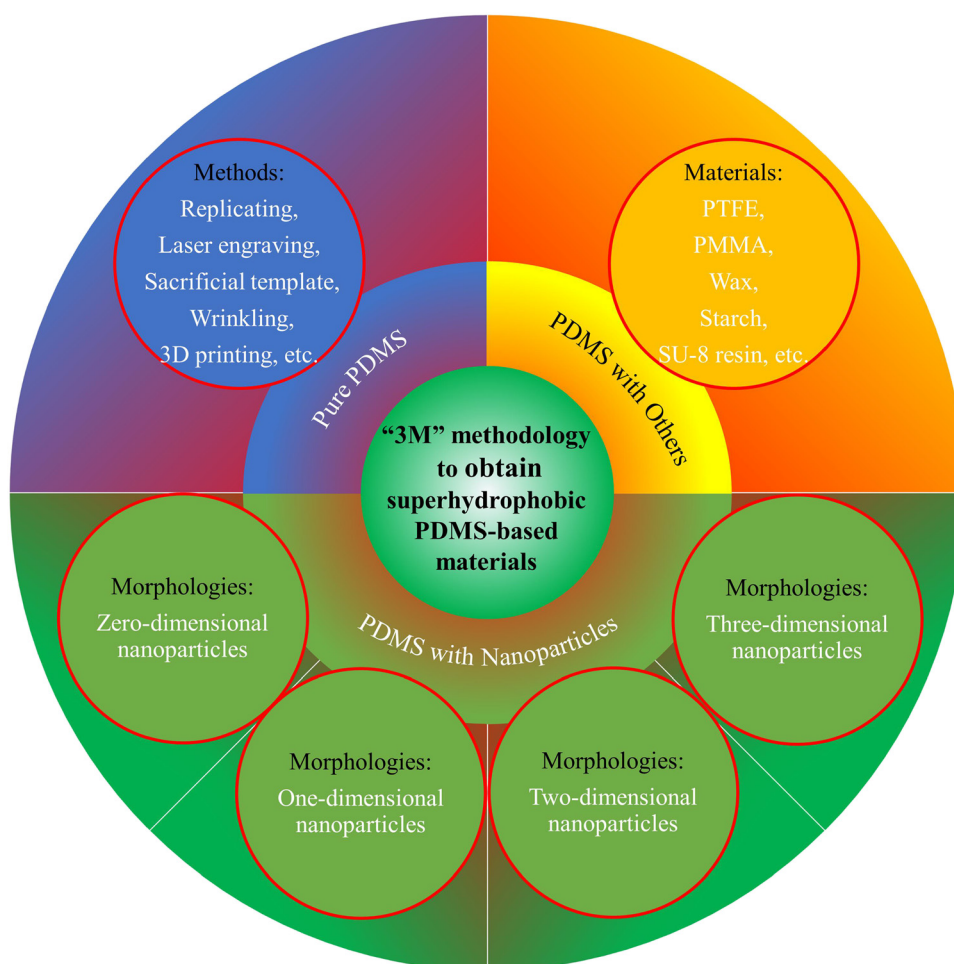
### 3.3 Hydrophobicity

Hydrophilic antifouling materials easily swell in water [91,92], while the hydrophobic ones can avoid this issue. Although silicone materials are hydrophobic in most cases, they do not display satisfactory antifouling ability. Joshi and coworkers investigated the effects of surface wettability on the antifouling ability of PDMS and other substrates [17]. Plain PDMS is hydrophobic with  $\theta_{WCA} \approx 106^\circ$  (Figure 9a and b) but it entails the substantial attachment of *Escherichia coli* (Figure 9c and d). Roughing the surface using a laser-induced periodic surface structure (LIPSS) or multiscale structure (MS) can improve the hydrophobicity and decrease bacterial attachment.

### 3.4 Superhydrophobicity

Section 3.3 demonstrated that improving surface hydrophobicity is an efficient strategy to enhance antifouling performance. Then, one would expect a superhydrophobic surface to have similar antifouling effects [93–95]. PDMS-based superhydrophobic materials have been extensively studied in this regard. Our previous review proposed a versatile “3M” (i.e., materials, methods, and morphologies) methodology to design superhydrophobic materials containing pure PDMS, PDMS with nanoparticles, and PDMS with other substances (Figure 10) [61].

Among various superhydrophobic antifouling materials based on PDMS, PDMS combined with nanoparticles has attracted considerable attention. Nanoparticles can be classified into the following four types: zero-dimensional nanoparticles such as spherical  $\text{SiO}_2$ ,  $\text{TiO}_2$ , and  $\text{Ag@SiO}_2$  core-shell nanocomposites [18,19,96–98]; one-dimensional nanoparticles such as linear ZnO nanorods and CNTs [99–101]; two-dimensional nanoparticles such as laminar



**Figure 10:** A versatile “3M” methodology (materials, methods, and morphologies) to obtain PDMS-based superhydrophobic materials. Reprinted with permission from He *et al.* [61].

graphene and montmorillonite [102–104]; and three-dimensional nanoparticles such as single tetrapod-shaped ZnO or composite nanoparticles comprising one or more types of nanoparticles of other dimensions [105–107].

The superhydrophobicity of PDMS nanocomposites can be explained according to the Wenzel model (equation (1)) and/or the Cassie–Baxter model (equation (2)) [64,65]:

$$\cos \theta = r \cos \theta_e, \quad (1)$$

$$\cos \theta = f_s \cos \theta_e + f_s - 1, \quad (2)$$

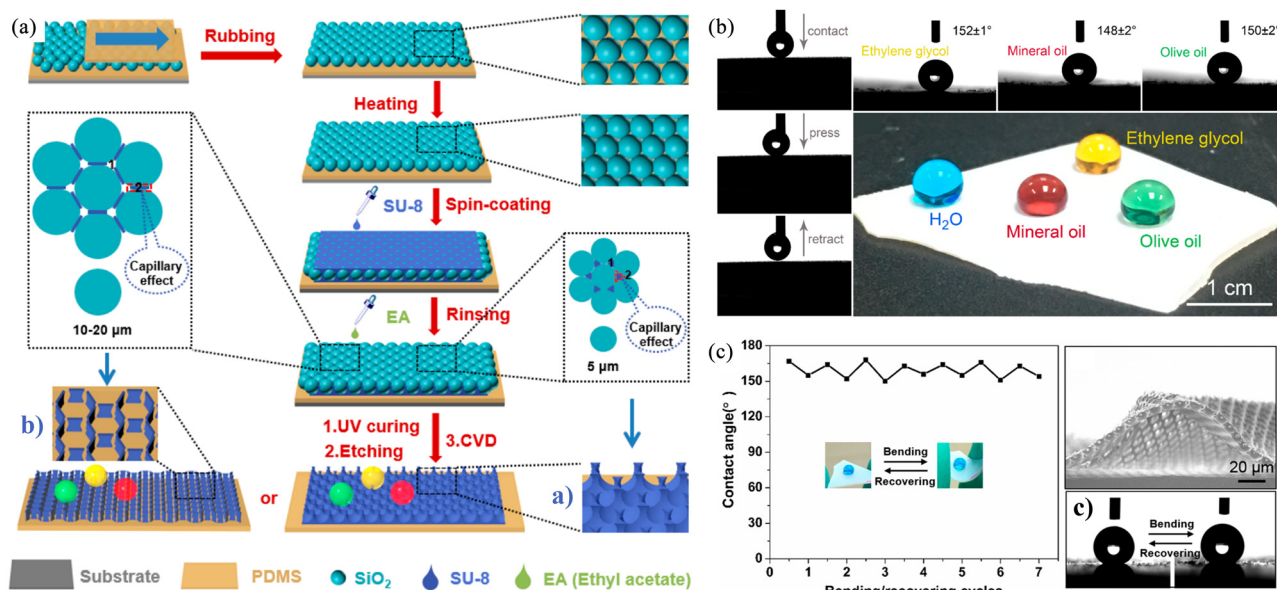
where  $\theta$  is the measured WCA for PDMS nanocomposites;  $r$  is the surface roughness of PDMS nanocomposites;  $\theta_e$  is the equilibrium WCA on the smooth PDMS surface; and  $f_s$  is the solid fraction of the solid–air compound surface [18]. Due to the inherent hydrophobicity of PDMS, superhydrophobicity can be achieved simply by roughening the surface of pure PDMS or its nanocomposites to increase the  $r$  value and/or decrease the  $f_s$  value, using various techniques: spin coating [108], electrospinning [109], drop-casting or spray-coating [110–113], replication [17,114–119], laser engraving [120–125], introducing a sacrificial template [126], wrinkling [127], 3D printing [20,128], and other methods [129–147]. Although the exact method or resultant morphology may differ, they have the same goal of introducing micro- and nanoscale or hierarchical roughness (higher  $r$  and/or lower  $f_s$  values) into the hydrophobic PDMS-based material to achieve superhydrophobicity.

In our previous article, a facile and universal strategy was proposed to fabricate superhydrophobic PDMS and SiO<sub>2</sub> nanoparticle surfaces *via* spin coating [18,19]. Multi-scale physical structures with microscale nanoparticle aggregates and nanoscale single nanoparticles were obtained through the spontaneous aggregation of nanoparticles [148–150]. Owing to the low surface energy of PDMS and hydrophobic SiO<sub>2</sub> nanoparticles, the final coating exhibited superhydrophobicity (WCA higher than 150°) and good antifouling ability against inorganic and organic powder foulants because of its self-cleaning ability [151].

As another typical example, Wu and coworkers reported a robust, transparent, and superhydrophobic PDMS film with SU-8 resin (Figure 11a) [138]. After chemical vapor deposition of 1*H*,1*H*,2*H*,2*H*-perfluorooctyl-trichlorosilane, the film showed excellent repellency to water droplets and other types of organic fouling droplets (Figure 11b). Moreover, the superhydrophobicity was mechanically stable, as demonstrated by surface wettability measurement and SEM imaging after several bending/recovery cycles (Figure 11c).

## 4 Discussion

As previously discussed, although there are four different types of surface wettability, the antifouling efficiency of AFISW strategies changes according to different types of fouling.



**Figure 11:** (a) Fabrication of superhydrophobic PDMS. (b) Behavior of water and other types of organic foulant droplets on superhydrophobic PDMS. (c) Mechanical stability of surface wettability during several bending/recovery cycles. Reprinted with permission from Wu *et al.* [138]. Copyright 2018, American Chemical Society.

**Table 1:** Comparison of AFISW strategies, materials, methods, morphologies, antifouling efficiency (excellent: 75–100%; good: 50–75%; medium: 25–50%; poor: 0–25%), and antifouling applications for inorganic, organic, and biofouling

| Categories                              | AFISW strategies    | Materials                                                                                         | Methods                                                                                           | Morphologies                        | Efficiency                                 | Applications                                                             | Ref.                     |
|-----------------------------------------|---------------------|---------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------|-------------------------------------|--------------------------------------------|--------------------------------------------------------------------------|--------------------------|
| Inorganic fouling                       | Superhydrophobicity | PDMS and PTFE powder                                                                              | Spin coating                                                                                      | Honeycomb-like structures           | Excellent (almost 100% of reduction)       | Anti-icing                                                               | [108]                    |
|                                         |                     | PDMS                                                                                              | Laser engraving technique                                                                         | Various columns, holes, grooves     | Excellent (almost 100% of reduction)       | Anti-icing                                                               | [120]                    |
|                                         | Organic fouling     | Superhydrophobicity                                                                               | PDMS and 1,1,2,2-tetrahydroperfluorodecyltrimethoxysilane-modified TiO <sub>2</sub> nanoparticles | Spraying on sandpaper               | Spontaneous nanoparticle aggregates        | Excellent (almost 100% of reduction)                                     | Anti-snow and anti-icing |
| PDMS and Ag nanowires                   |                     |                                                                                                   | Spray coating                                                                                     | Nanowire                            | Excellent (almost 100% of reduction)       | Olive oil, <i>n</i> -propanol, <i>n</i> -hexadecane, dimethylsulfoxide   | [55]                     |
| PDMS and SiO <sub>2</sub> nanoparticles |                     |                                                                                                   | Spin coating                                                                                      | Spontaneous nanoparticle aggregates | Excellent (almost 100% of reduction)       | Carbon powder                                                            | [151]                    |
| Biofouling                              | Superhydrophobicity | PDMS and 1,1,2,2-tetrahydroperfluorodecyltrimethoxysilane-modified TiO <sub>2</sub> nanoparticles | Spray coating on sandpaper                                                                        | Spontaneous nanoparticle aggregates | Excellent (almost 100% of reduction)       | Oil                                                                      | [152]                    |
|                                         |                     | PDMS and SBSi                                                                                     | Covalent silanization                                                                             | Thin film                           | Excellent (>99% of reduction)              | <i>P. aeruginosa</i> and <i>S. epidermidis</i>                           | [86]                     |
|                                         |                     | PDMS and poly(carboxybetaine methacrylate)                                                        | Surface-initiated atom transfer radical polymerization                                            | Thin film                           | Excellent (>98% of reduction)              | Nonspecific protein                                                      | [156]                    |
|                                         | Hydrophilicity      | PDMS and poly((2-dimethylamino) ethyl methacrylate)                                               | Surface-initiated atom transfer radical polymerization                                            | Thin film                           | Excellent (>97% of reduction)              | <i>E. coli</i>                                                           | [157]                    |
|                                         |                     | PDMS and PAA                                                                                      | Dip coating                                                                                       | Thin film                           | Excellent (>83% of reduction)              | Anti-coagulation, cell adhesion, calcification, and bovine serum albumin | [25]                     |
|                                         |                     | PDMS and poly(2-methacryloyloxyethyl phosphorylcholine)                                           | Copolymerization and coating                                                                      | Thin film                           | Good (approximately 70% of reduction)      | Protein and cell adhesion                                                | [88]                     |
|                                         |                     | PDMS and poly(2-methacryloyloxyethyl phosphorylcholine)                                           | Copolymerization and coating                                                                      | Thin film                           | Excellent (approximately 90% of reduction) | <i>Staphylococcus aureus</i>                                             | [89]                     |
| Hydrophobicity                          |                     | PDMS                                                                                              | Femtosecond laser-induced submicron topographies                                                  | LIPSS                               | Excellent (>89% of reduction)              | <i>E. coli</i>                                                           | [17]                     |
|                                         |                     | PDMS                                                                                              |                                                                                                   |                                     |                                            |                                                                          | [42]                     |

(Continued)

Table 1: Continued

| Categories          | AFISW strategies | Materials                                                                             | Methods                                          | Morphologies                        | Efficiency                                      | Applications                                                                                                                                 | Ref.  |
|---------------------|------------------|---------------------------------------------------------------------------------------|--------------------------------------------------|-------------------------------------|-------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------|-------|
| Superhydrophobicity |                  | Hydroxy-terminated PDMS, isophorone diisocyanate trimers, and polycaprolactone triols | Elastomeric stamping imprinting                  | Biomimetic shark skin surface       | Excellent (approximately 90% of reduction)      | Bovine serum albumin, ovalbumin, and algae                                                                                                   | [158] |
|                     |                  |                                                                                       | Copolymerization and coating                     | Thin film                           | Medium (50% of reduction in protein adsorption) | Bovine serum albumin                                                                                                                         |       |
|                     |                  |                                                                                       | Femtosecond-laser-induced submicron topographies | MSs                                 | Excellent (>89% of reduction)                   | <i>E. coli</i>                                                                                                                               | [17]  |
|                     |                  |                                                                                       | Casting                                          | Spontaneous nanoparticle aggregates | Excellent (>75% of reduction)                   | <i>Bacillus subtilis</i> , <i>S. aureus</i> , <i>P. aeruginosa</i> , <i>E. coli</i> , <i>Candida albicans</i> , and <i>Aspergillus niger</i> | [98]  |
|                     |                  | PDMS and Ag@SiO <sub>2</sub>                                                          | Casting                                          | Hybrid nanofillers                  | Excellent (>75% of reduction)                   | Gram-positive, Gram-negative, and fungi                                                                                                      | [107] |

As shown in Table 1, in the case of inorganic fouling, the AFISW strategy mainly involves improving the surface hydrophobicity to impart superhydrophobicity to PDMS and its nanocomposites, because inorganic foulants are usually hydrophilic and can be easily cleaned owing to the well-known self-cleaning effect of a superhydrophobic surface. Thus, the superhydrophilic and hydrophilic AFISW strategies are excluded [108,120,152].

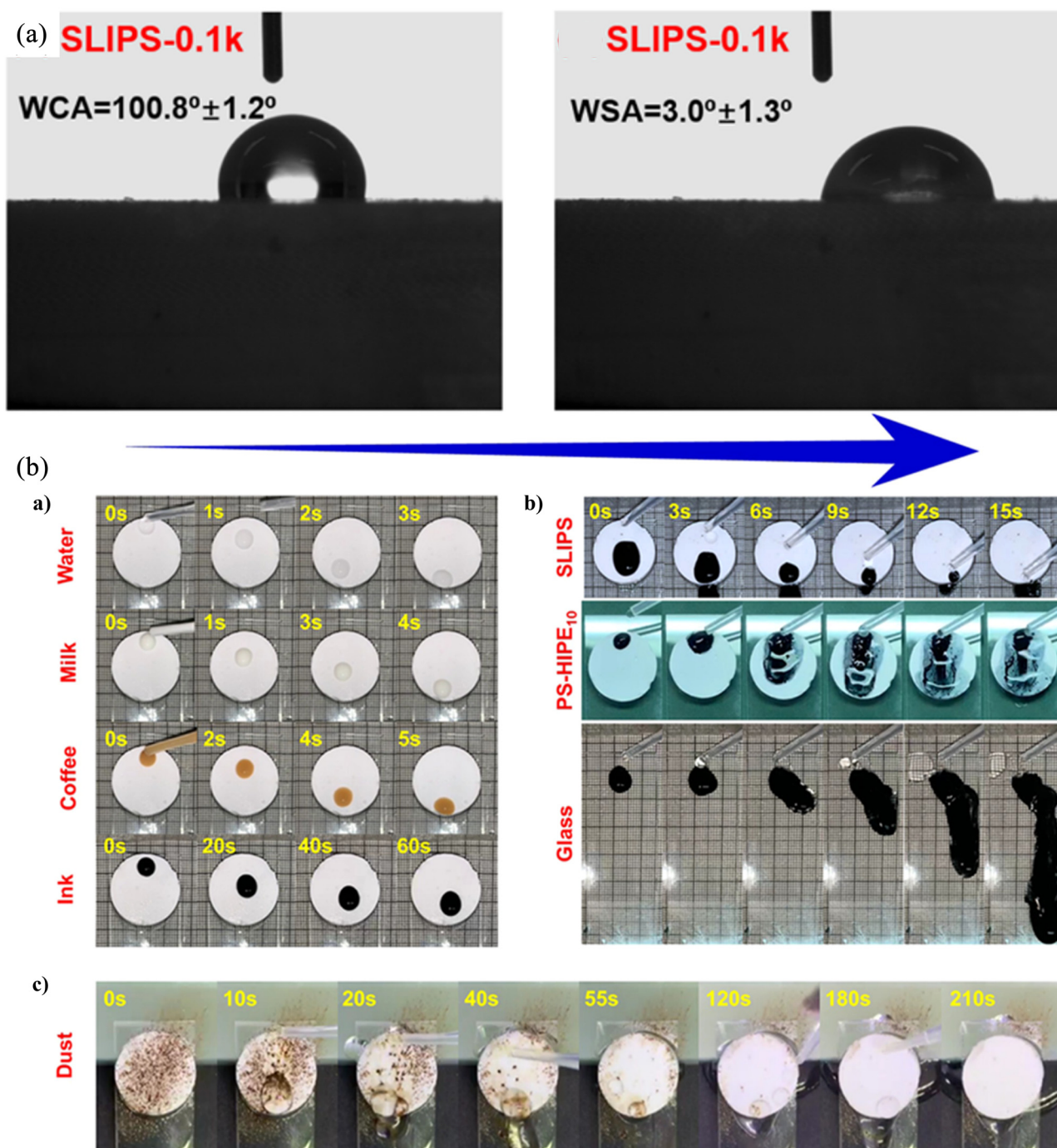
Similarly, the superhydrophobic AFISW strategy can be used for organic fouling, especially for organic foulants with a powder morphology [151]. If the organic foulants are oils or solvents, it is highly challenging to achieve good antifouling ability because of the organic characteristics of PDMS. However, in this situation, the superhydrophobic AFISW strategy can be made more effective by introducing fluoro-based materials [55,152]. It is worth mentioning that the superhydrophilic surface usually has underwater superoleophobic characteristics and can be used for organic fouling [1]. However, using PDMS-based materials is not the optimal superhydrophilic AFISW strategy for organic fouling due to the organic characteristics of PDMS; therefore, hydrophilic materials are usually adopted [153–155]. Moreover, the hydrophilic and hydrophobic AFISW strategies are rarely used in the case of organic fouling because of their low antifouling efficiency.

Superhydrophilic [86,156,157] and superhydrophobic [17,98,107] AFISW strategies are extensively used for biofouling because of the good chemical stability and biocompatibility of PDMS. Hydrophilic [25,88,89] and hydrophobic [17,42,158] AFISW strategies are also usually reported to achieve anti-biofouling. Overall, superhydrophilic and superhydrophobic AFISW strategies are more efficient than hydrophilic and hydrophobic AFISW strategies. Moreover, superhydrophilic and hydrophilic AFISW strategies may be more suitable than superhydrophobic and hydrophobic AFISW strategies, especially for potential biomedical applications.

Although composite fouling is more complex than inorganic, organic, and biofouling, it can be treated as the combination of any of these two/three types of fouling. Consequently, AFISW strategies for composite fouling can be based on the individual strategies corresponding to the specific combination that constitutes composite fouling.

Moreover, it is worth mentioning that there is another special hydrophobic AFISW strategy to improve the anti-fouling ability, *i.e.*, to make the surface slippery [159]. For example, Lei *et al.* constructed a slippery surface by infusing liquid hydrophobic PDMS into a porous poly (high internal phase emulsion) substrate [160]. The porous substrate infused with PDMS lubricant became slightly





**Figure 12:** (a) WCA and WSA values on the slippery surface. (b) Antifouling behavior against different foulants. Reprinted with permission from Zhang *et al.* [160]. Copyright 2019, American Chemical Society.

hydrophobic with  $\theta_{\text{WCA}} = 100.8^\circ$  and displayed an extremely low water sliding angle (WSA) of  $3.0^\circ$  (Figure 12a). This slippery surface exhibited excellent antifouling properties against various liquid foulants such as water, milk, coffee, ink, and dust (Figure 12b), revealing its excellent potential in various antifouling applications.

## 5 Conclusions

This study represents the first systematic review of AFISW strategies (superhydrophilicity, hydrophilicity, hydrophobicity, and superhydrophobicity) based on PDMS and its nanocomposites. The surface wettability of PDMS and its

nanocomposites can be easily tuned to satisfy antifouling needs, especially for organic and biofouling, which is difficult to achieve with pure PDMS. Antifouling strategies for inorganic and organic foulants are easy to design and usually mutually compatible because of their similarities. Conversely, it is difficult to formulate antifouling strategies for bio- and composite fouling. Therefore, more attention should be paid to the design and fabrication through superhydrophilic and superhydrophobic AFISW strategies, of antifouling materials for these two types of fouling. Moreover, the slippery AFISW strategy, which showed extensive potential in various antifouling applications, should be further investigated. Finally, we believe that this review may help researchers to design and fabricate various polymeric materials and their nanocomposites with tuned surface wettability for AFISW applications.

**Funding information:** The authors would like to acknowledge the financial support from the Key Research and Development Programs of Luzhou (No: 2022-GYF-12), the Sichuan Science and Technology Program (2022YFS0634), the National Natural Science Foundation of China (No: 51873240), the Talent Introduction Program of The Affiliated Stomatological Hospital of Southwest Medical University (No: 2022BS02), the Talent Introduction Program of Chengdu University (No: 2081920001), the High-level Talent and Cultivation Program of Chengdu University for the National Natural Science Foundation of China (No: Z1332), and Innovative leading talents program of The Affiliated Stomatological Hospital of Southwest Medical University (No: 2022LJ02).

**Author contributions:** Zhoukun He, Junlong Li, Fei Deng, and Xiaorong Lan conceived and designed this review paper; Zhoukun He, Na Wang, and Xiaochen Yang wrote and revised this review paper; Linpeng Mu, Zhuo Wang, Jie Su, Mingdong Luo, Junlong Li, Fei Deng, and Xiaorong Lan revised this review paper. All authors have accepted responsibility for the entire content of this manuscript and approved its submission.

**Conflict of interest:** The authors state no conflict of interest.

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