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A review on bio-inspired antifouling coatings: Applications, challenges and perspectives for the future

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Abstract: *In terms of modulus and scale, there exist various types of fouling, necessitating the development of antifouling coatings to address the risks associated with different fouling phenomena. Nature offers a diverse range of natural anti-pollution strategies, which are elaborated upon in detail for nine distinct pollutants through the synthesis of corresponding anti-pollution materials. Furthermore, apart from considering the antifouling performance of such materials, attention should also be given to their durability, non-toxicity, and economic feasibility.*

Keywords: *fouling; bio-inspired; antifouling mechanism*

1. Introduction

1.1 Definition and effects of fouling, Market size for antifouling coatings

The unwanted adhesion on a surface is known as fouling. This unwanted adhesion can be from a variety of substances, such as ice, natural gas hydrates, waxes and asphaltenes, inorganic scale, bacteria, biofilms, and proteins[1]. Fouling is ubiquitous and causes negative effects. Marine fouling on ships increases the drag force, leads to further fuel consumption, and triggers sonar interference[2]. Based on the estimates of US Nacy Command, marine fouling reduces velocity by around 2% and augments fuel cost from 6% to 45%[2]. In addition, fouling in oil pipelines, including wax, biofouling, and salt, increases fuel waste, reduces heat recovery, and extends the time to shut down manufacturing lines for cleaning, which will lower oil production, and pose safety risks to the workers[3]. Similarly, platelets adhering to implanted devices motivate thrombus formation, causing thrombotic complications and device dysfunction which contribute to the extra death rate for patients[4-7].

Thus, the development of antifouling materials is significant for commercial aspects and human safety. For ice phobic coatings, used in transportation, renewable energy, and communication equipment, it was reported that a Compound Annual Growth Rate (CARG) is 24.5%. If marine fouling was eliminated, approximately \$60 billion of fuel for the shipping industry could be saved annually. This also corresponds to about 384 million tons of CO_2 and 3.6 million tons of SO_2 emissions annually[8]. In the marine industry, according to the data from Allied Market Research in 2021, the self-polishing copolymer coating was the most lucrative one for preventing marine fouling and had the largest global market size, followed by copper-based and hybrid copolymer coatings. Based on revenue, shipping vessels held the largest market share of antifouling coatings around the world[9].

1.2 Main characteristics of fouling

Foulants each have their unique characteristics. Antifouling coatings with different working strategies are used to deal with foulants with different characteristics. Two main characteristics of fouling are as follows.

The modulus of fouling determines its hardness. Typically, fouling with a modulus lower than 10^5 Pa is considered to be soft fouling, whereas fouling with a modulus greater than 10^5 Pa is hard (see Fig. 1a). The length scale of fouling is another physical quantity to consider when choosing antifouling coatings.

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For example, ice and clathrates have a varied length scale from 10^{-10} m to 10^{2} m. The length scale of nine common types of fouling can be seen in Fig. 1b.

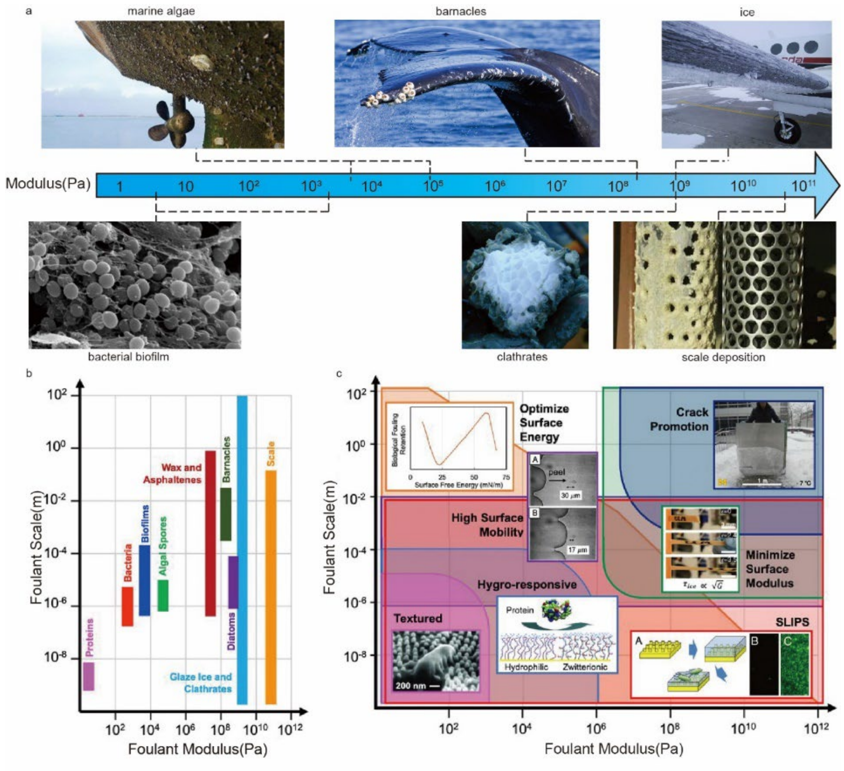

Figure 1: (a) Classification of fouling depending on the modulus (b) Length Scale of Various Foulants. (c) Different antifouling methods for different length scales.[1,10]

Methods used to remove hard fouling and soft fouling are different, because the strain experienced by the foulant under a given applied stress is a direct function of its modulus. The length scale determines the attachment area of the fouling on a surface, and the attachment area will influence the force for fouling removal[11]. Thus, based on the length scale and modulus of foulant, there are multiple different ways to remove fouling, shown in Fig. 1c.

This review classifies fouling with different standards and explains several basic theoretical backgrounds and knowledge in the anti-coating field. The review introduces some antifouling surfaces that emerge in nature and their working strategies. The core of this review is the synthetic antifouling coatings that are currently in use to reduce nine different kinds of fouling. The major concerns; challenges, such as durability, cost, and environmental impacts; and feasibility of new approaches to reduce fouling are also included in this review.

2. Theoretical background

2.1 Contact angle

2.1.1 Static contact angle

Surface wettability is a function of both surface chemistry and texture. This is dependent on the interaction of the fluid with the solid surface [12]. The contact angle is an important quantitative measure of surface wetting. It was originally introduced by Thomas Young in 1805[13]. It describes the relation among three kinds of interface stresses when a drop of liquid rests on a flat, homogeneous surface and creates an angular gradient[14]:

$$cos\theta_0 = \frac{\gamma_{SV}-\gamma_{SL}}{\gamma_{LV}} \quad (1)$$

Here θ_0 is the equilibrium constant angle γ_{SV}, γ_{SL} and γ_{LV} are the interface energies for the solid-gas, solid-liquid and liquid-gas interfaces respectively (see Fig. 2a)[15]. Based on Young's contact angle with water, surfaces can be classified as hydrophobic or hydrophilic. Hydrophilic surfaces are surfaces

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where water has a strong affinity for a substance, while hydrophobic surfaces are surfaces where water repels a substance[16]. As in Fig. 2b, there is also a boundary line and special cases between them.

However, Young's contact angle is the intrinsic contact angle of an ideal solid[18]. Therefore, in 1936, Wenzel[15] worked out a relation between the contact angle on a rough surface (θ^*) and Young's contact angle (θ_0), given as:

$$cos\theta^{*}=r\ cos\theta_0 \tag{2}$$

$$r= S_A\ /S_G \tag{3}$$

Where r is the surface roughness[19], which is a ratio of the real surface area (S_A) to the projection area (S_G)(see Fig. 2c). Fig. 2d shows a strong fluid/solid interaction, and the droplet fills the grooves between the protrusions on the rough surface to the extent of complete wetting. However, the Wenzel condition is not satisfied, and the slot is filled with air in place of the fluid[20]. This state is called the Cassie-Baxter state (see Fig. 2e). It is assumed that the surface is a binary compound, and the contact angle on the rough surface can also be expressed as[21]:

$$cos\ \theta_0 =f_1\ cos\ \theta_1\ +f_2\ cos\ \theta_2 \tag{4}$$

Here f_1 and f_2 are the liquid/solid contact area fractions on the surface, respectively, θ_1 and θ_2 are the contact angles. In special cases, it can also be expressed as:

$$cos\ \theta =f_1(cos\ \theta_1\ +1)\text{-}1 \tag{5}$$

It shows that the cosines of a liquid drop on an inhomogeneous plane are the same as the cosines of their contact angles on both different surfaces, and they are weighed by the liquid/solid contact region fraction (f_1).

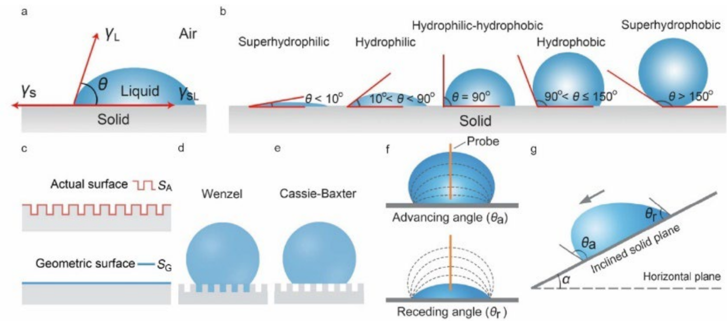

Figure 2: [17] contact angle(a) Diagram illustrating the theory of a stationary contact angle. (b) Various states of a drop in contact with a solid surface. (c) The real surface and geometry of the surface. (d) Wenzel state. (e) Cassie–Baxter state. (f) A schematic representation of an advancement angle (θ_a) and an angular retreat (θ_r). (g) The angular momentum of the contact on the inclined surface.

2.1.2 Dynamic contact angle

On any surface, the maximum possible contact angle measured due to surface texture and chemical heterogeneity is called the advancing contact angle, and the minimum possible contact angle is called the receding contact angle. These angles can be determined by placing a droplet on a horizontal surface and continuing to increase its volume until it reaches a critical point (see Fig. 2f), or by placing a droplet on an inclined surface and changing the angle of inclination (α) to make the droplet start to move see Fig. 2g). In both methods, the contact angle during forward movement is referred to as the advancing angle(θ_a), and the contact angle during backward movement is referred to as the receding angle(θ_r)[22]. The angle of inclination (α) is defined as the sliding angle and the amount of change in contact angle due to roughness, and chemical composition is called(θ_h). The following formula is the relationship between them[23]:

$$\theta_h=\theta_a\text{-}\theta_r \tag{6}$$

$$mg(sin\alpha)/\omega=\gamma_L\ (cos\theta_r\text{-}cos\theta_a) \tag{7}$$

where m and ω are the mass and diameter of the drop, g is the acceleration of gravity, and γ_L is the fluid's surface tension.

Because of the low slip angle, water drops are hard to hold, so the sliding process is easy to remove contaminants.[24], resulting in a self-cleaning surface, often called the lotus effect[25]. Similarly, a rose

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petal displays high contact angles like the lotus leaf, but droplets strongly adhere to it, so they cannot carry away any foulants on the surface. This is often called the "petal effect"[26]. These distinct phenomena are attributed to the different interactions between liquid and solid. The low-contact angle lag is advantageous for the super-hydrophobic surface to remove the fouling. In contrast, the antifouling mechanism of hydrophilic surfaces differs from the lotus effect, as the presence of a hydration layer on a hydrophilic surface acts as a physical barrier to fouling organisms[27]. It increases the energy penalty for the removal of water from a surface[28].

2.2 Energy of surface and modulus of elasticity

The notion of surface energy describes the interference between molecules in the formation of a new surface[29]. In general, drops on high-surface-energy-rich surfaces show excellent wettability, whereas drops on low-surface-energy surfaces show bad wettability (see Fig. 3a). A way to determine the surface energy of a stationary surface may be determined through the Owens–Wendt–Kaelble method[30], the Lifshitz–van der Waals/acid-base method[31], and the Li–Neumann method[32].

Brady et al. have started to apply fracture mechanics to describe the foulant debriding process[33](see Fig. 3b). Fracture mechanics studies the growth of cracks in a material[34]. This field assumes that there are no flaws at all. The presence of microscale cracks creates stress concentration points, which facilitate crack propagation. There are three types of crack opening in the fracture mechanics (see Fig. 3c). In peel, most adhesive joints fail due to requiring less energy for failure compared to shear failure[35]. Fig. 3d depicts a uniform plate with a defect. The fracture will spread when the single axial tension (σ) reaches a certain threshold. In this case, the tensile force P_C (applied to the surface A = πa^2) is computed as[35]:

$$P_C=\sqrt{\frac{\pi E G_C a^3}{1-v^2}} \tag{8}$$

Here, E is an elasticity module, G_C is a critical strain energy release rate (usually referred to as a critical crack energy or a critical surface energy), and in certain situations, it can also be referred to as bonding work W_a, and u is the Poisson's ratio. Likewise, when biological bonding takes place, there is an imperfect relationship between the fouling organism and the surface. These imperfections will expand at the interface under an applied external force. When the defects are large enough, they are removed, which can be interpreted by means of fracture mechanics[36]. Kendall is the first to apply Griffith fracture theory to simulate the adhesive behaviour of elastomers (see Fig. 3e)[37]. In this model, an elastomeric coating was positioned between a rigid substrate and a rigid disk. The critical pull-off force P_C is expressed as [37]:

$$P_C=\pi a^2\sqrt{\frac{2G_C K}{t}} \tag{9}$$

Here K represents the force exerted perpendicular to the surface, while t denotes the thickness of the coating.

By using elastomeric coating ($t \gg a$), Kendall deduce the equation:

$$P_C=\sqrt{\frac{2\pi E G_C a^3}{1-v^2}} \tag{10}$$

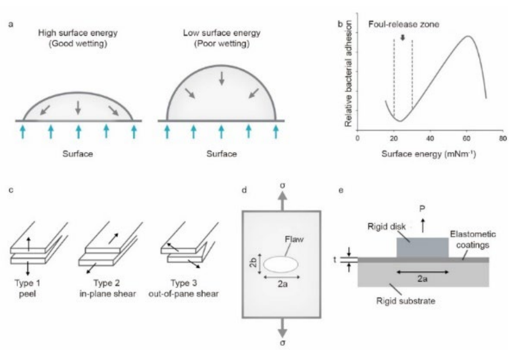

Figure 3:[17] (a) Diagram of high-and low-surface-energy surfaces. (b) A typical "Baier curve"[33] (c) The Crack Initiation Patterns of Fracture Mechanics. (d) The defect occurs in the uniform sheet of 2a wide and 2b thick (e) Elastic cover retained between the solid base plate and the hard disc (P = pull-off force, a = contact radius, t = coating thickness)[35].

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3. Antifouling mechanism of natural organisms

The natural biological system has traditionally served as inspirations to engineer antifouling systems. Due to the intricate nature of fouling, there exist numerous natural instances of antifouling surfaces that apply a unity of physical and chemical control strategies to hinder fouling[38].

3.1 Natural surface coating antifouling mechanism

3.1.1 Chemical secretions as natural antifoulants

Chemical secretions can naturally prevent and remove foulants, serving as an antifouling approach. Reports have documented the presence of inartificial products or extracts with antifouling properties in a diverse array of marine invertebrates, such as sponges, corals, and mussels[39,40]. These natural antifoulants encompass a diverse array of compounds, including alkaloids, diterpenes, indoles, polyketides, sesquiterpenes, steroids, sterols, and terpenoids[41]. Among these invertebrates, the biofouling-combating potential of antifoulants derived from corals has been extensively investigated[42]. Organic extracts from sponges have similarly exhibited potent antifouling capabilities[39].

Meyer and Seegers discovered that the occurrence and localization of peptide group β-defensins, which are produced by the thick integument, can function as a non-specific defensing mechanism against biofouling like algae and bacteria [43]. Additional examples are provided in the table 1.

Table 1: Natural antifoulant examples

Natural examples	Anti-fouling mechanism	Reference
Delisea pulchra	Metabolites(halogenated furanones)	Steinberg *et al.*[44]
Red Sea scleractinian coral *Pocillopora damicornis*	allelochemicals	Geffen & Rosenberg[45]
red alga *Delisea pulchra* (Greville) Montagne	secondary metabolites	Nys *et al.*[46]
chili peppers	capsaicin	Xu *et al.*[47]
garlic	allicin	Wu *et al.*[48] Tiwari *et al.*[49]
Ceriops tagal	terpenoids	Chen *et al.*[50]
Guatambú trees	indole	Pérez[51]
black wattle trees	tannin	Peres[52]

3.1.2 Mucus-like hydrogels

Mucus, a biologically dynamic hydrogel consisting of diverse glycoproteins known as mucins, is present at the bio-interfaces that envelop the underlying epithelial cells of miscellaneous organs, including the ocular system, gastrointestinal tract, reproductive system, and respiratory passages and serves as a protective barrier against different fouling[53-56].

The mucous layer of fish and amphibian skins provides a protective barrier[57]. Ritchie's study suggests healthy *Acropora palmata*'s mucus can suppress the proliferation of potentially invasive microbes by up to ten times, indicating the significant role of coral mucus in exhibiting antibiofouling activity[58]. The coral mucus can enhance fouling resistance through a multitude of mechanisms, encompassing the provision of a physical barrier, mucociliary transport for microbial elimination and sloughing to prevent colonization by invasive microbes[59].

Mucus, like synthetic hydrogels, shares hydrophilic and soft attributes, and demonstrates gel-like properties upon hydration [60]. Hydrogels are characterized by crosslinked three-dimensional polymer networks capable of absorbing significant amounts of water[61]. Their matrices exhibit hydrophilic properties. Fig. 4 illustrates the antifouling mechanisms of the hydrogel. Hydrophilic surfaces readily facilitate hydrogen bond formation or electrostatically driven hydration shell formation, creating a physical barrier against fouling organism colonization[27].

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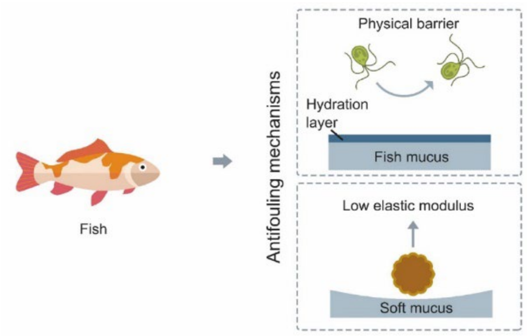

Figure 4: The antifouling mechanism of hydrogel[17]

3.1.3 Zwitterionic coatings

The lipid bilayer forms the structural foundation of biological membranes. Phosphatidylcholine is the predominant lipid component, making up approximately 50% of the entire membrane composition (see Fig. 5a)[62]. The phosphatidylcholine head groups exist as zwitterions, which are composed of an equal number of two species with opposite charges but overall electroneutral and exhibit hydrophilicity due to their polar nature. Consequently, the utility of zwitterionic polymers has been the subject of significant interest in marine antifouling strategies.

The remarkable ability of zwitterionic polymers to resist fouling is widely attributed to their effective hydration and surface charge neutrality[17]. Perspective of Physical/energy barrier is widely employed to elucidate the antifouling mechanism of zwitterionic polymers[63,64]. The extensively hydrated surface of zwitterionic polymers exhibits strong affinity for water molecules that the fouling organisms require significant energy to penetrate the hydration layer. Researchers also attempted to employ ion-coupled adsorption to elucidate the antifouling mechanism exhibited by zwitterionic polymers[65,66]. The adhesion behaviors of fouling organisms, including diatoms and bacteria, are influenced by ion-coupled adsorption due to the production of adhesive proteins by these organisms. However, the zwitterionic surface is not associated with any surface ions, thereby precluding the occurrence of ion-coupled adsorption (see Fig. 5b).

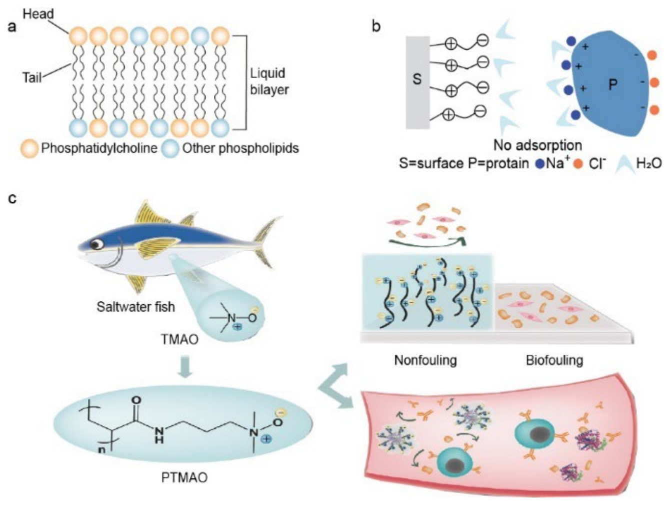

Figure 5: Zwitterionic coatings (a) Arrangements of lipid bilayers in living organisms [17]. Schematic depictions showcasing the ion-coupled adsorption mechanism of a protein interacting with (b) a surface possessing zwitterionic properties[66] (c) TMAO-derived zwitterionic polymers[67]

The natural world harbors numerous other zwitterionic compounds, such as phosphorylcholine within cellular membranes, taurine in animal tissues, and glycine betaine in plants. Their exceptional antifouling performance stems from the electrostatically enhanced hydration that was discussed at the beginning of this section[67,68]. Another category of zwitterionic molecules, known as osmolytes, can be found in marine fish species (see Fig. 5c). Osmolytes are small organic compounds that are soluble and produced by living organisms to regulate cell volume, enabling them to withstand extreme osmotic pressures[69].

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3.2 Natural surface structural antifouling mechanism

3.2.1 Slippery liquid-infused porous surfaces (SLIPS)

The pitcher plant, an example of a carnivorous flora, possesses the unique ability to entrap its prey by utilizing a slick, fluid film encircling its peristome (see Fig. 6a)[70]. A "slippery surface" is physically and conceptually distinct from the lotus effect, as it is characterized by a surface infused with a lubricating fluid. This attribute is observed in the micro-structured gastrointestinal tract of humans[71]. The mucus layer secreted by the earthworm across the textured skin generates a lubricious surface, which confers superior antifouling properties[72].

Typically, a slippery surface is composed of a porous or textured substrate immersed in a lubricating fluid. Figure 6b illustrates the fabrication process of SLIPS. It demonstrates the entrapment of a lubricating fluid within a micro-/nano-porous matrix, resulting in the formation of a continuous liquid layer[73]. SLIPS possess the capability to deter a multitude of liquids, to promptly re-establish their hydrophobic properties following mechanical damage, to resist the adherence of ice and contaminants, and to withstand elevated pressures[73].

Over the past decade, the academic community has advanced a multitude of antifouling strategies for SLIPS like physical barrier, decreasing attachment strength, blocking signals, self-cleaning and forming molecularly smooth surface[17].

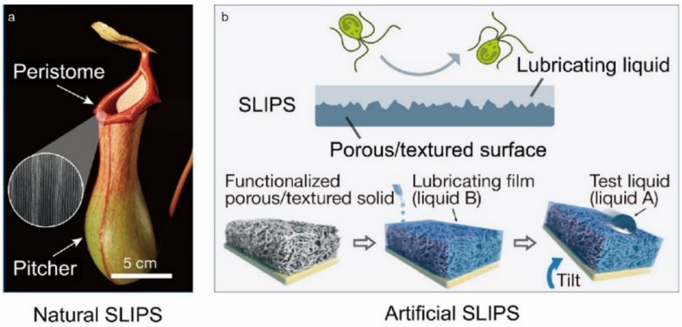

Figure 6: Natural(a) and artificial(b) SLIPS[17]

3.2.2 Dynamic surface antifouling (DSA)

A dynamic surface is described as an ongoing renewal process occurring in seawater, which effectively eliminates fouling organisms, according to Ma et al. [27]. Some marine creature like crustose coralline algae renew their surface by epithelium shedding[74]. Additionally, taking into account the fact that certain aquatic creatures like dolphins and soft corals possess delicate skin surfaces that can create unstable conditions in the presence of fluid flow, this dynamic mechanism serves as a preventive measure against biofouling.

The antifouling mechanism of dynamic surface can be categorized into grooming and sloughing. The process of grooming involves physically eliminating the presence biofouling on the host, thereby effectively managing both slow- and fast-growing biofouling. The crustaceans and decapods utilize specialized brush structures to groom other organisms, effectively removing foulers from their gills and appendages. Chinoderms and bryozoans utilize specialized structures known as pedicellariae to engage in the grooming of macroepibionts, while crayfish rely on *Branchiobdellid annelids* to facilitate the removal of foulers from their gills. The process of sloughing refers to the gradual shedding of an organism's outermost layer, thereby effectively controlling the proliferation of slow-growing biofouling. This mechanism is utilized by various organisms including crustaceans, seaweed, and stonefish *S. horribilis*[38].

The colonization of fouling organisms necessitates the identification of a suitable substrate[75]. The presence of a continuously changing dynamic surface poses challenges to the identification process. Additionally, the surface can effectively remove attached fouling organisms by undergoing dynamic deformation. Consequently, a range of dynamic surfaces have been developed following this approach. The instability of such surfaces makes it challenging for fouling organisms to adhere to them[17].

3.2.3 Natural antifouling effect of micro/nano-structured surfaces

To withstand natural stress, numerous organisms have developed diverse micro/nanotextures that possess desirable wettability properties, such as superoleophobic self-cleaning and low-adhesive superhydrophobic surfaces. These surfaces effectively resist biofouling by facilitating the removal of contaminating particles through water washing[76]. Natural creatures like lotus leaves, sharks, water striders, rice leaves, reed leave, springtails, geckos, beetles, and peanut leaves all have remarkable self-cleaning and antifouling properties on each surface, enabling organisms to seamlessly adapt to their surroundings[17].

The dermal denticles, which are the minute scales resembling teeth on shark skin, possess ribbed longitudinal grooves. The design of shark skin surface effectively prevents marine organisms from fouling it, as the rough nanotexture of the shark skin minimizes available surface area for organism adhesion[77]. It has been reported that both genetic and morphological evidence have been presented to support the presence of periostracal adventitious hairs on spat of the mussel *Mytilus edulis*, which may serve as an inhibitory mechanism against fouling, during the primary adhesion phase, particularly by conspecifics. The microtextured skin of pilot whales (*Globicephala melas*) and common dolphins (*Delphinus delphis*) serves as a deterrent to biofouling[43,78].

The mystery of the lotus effect has recently been unraveled, with the discovery of microstructures and waxes on the surface of lotus leaves, which have now become a prominent example of hydrophobic materials[79]. The microstructures possess the ability to create an air layer between a liquid droplet and the leaf surface, while the epicuticular waxes demonstrate hydrophobic properties. Both characteristics effectively prevent liquid infiltration into the surface[25]. Later, the presence of nanostructures was observed on each micropapilla covering the surface of the lotus leaf[80]. The presence of micro/nano hierarchical composite structures guarantees the manifestation of minimal adhesive characteristics on lotus leaves, while simultaneously establishing a physical obstruction on the surface substrate to combat biofouling organisms[81].

Some other organisms also possess micro/nano-structured surfaces, enabling them to effectively prevent fouling. Bixler and Bhushan[82] observed that various natural structures, such as rice leaves and butterfly wings, exhibit a unity of properties that prevent fouling. The flow characteristics of droplets on rice leaves and butterfly wings exhibit a unique anisotropy, with droplets flowing along the surface of the rice leaf blade and moving away from the butterfly's body in an axial direction.[82]. The hierarchical structures of rice leaves, confer anisotropic flow, superhydrophobicity, and low adhesion properties. The amalgamation of anisotropic flow, superhydrophobicity, and minimal adhesion has been observed to augment the self-cleaning capabilities[82,83]. The micro/nano hierarchical surface structures facilitate the retention of water droplets, which readily migrate and encapsulate contaminants, thereby enhancing the antifouling performance significantly.

4. The technology and synthetic surfaces used to reduce different types of fouling

The phenomenon of fouling is prevalent in nature and gives rise to numerous crucial issues across various industries as forementioned. Nine different types of fouling classified based on the modulus and length scale (Fig. 1c) and several methods have been developed and applied to synthetic coatings that aim to mimic the physical and/or chemical effects displayed by living organisms are discussed in this section.

4.1 Soft and low length scale

4.1.1 Non-structural proteins

Protein fouling impacts many fields, especially the medical field, where it commonly occurs on implanted medical devices. It shortens the service life of devices, contaminates the body's immune system, and may lead to thrombosis[84-86]. Also, it brings danger and risks to food storage and adds up the cost in the marine industry[87,88]. Due to the outcomes of protein fouling, it is important to develop antifouling surfaces that restrict the formation of protein fouling.

Three kinds of antifouling surfaces can be effective against protein fouling[89]. The first type is non-amphoteric hydrophilic materials, which include Poly(ethylene glycol) (PEG)-based grafted polymers. These surfaces are hydrophilic and electrically neutral. A hydroxyl group that can form a hydration layer with water molecules is at the end of the polymer chain [90,91]. The long molecular chain and side-linked branches of PEG can increase the chain mobility and steric hindrance, and thereby have better resistance [92,93]. When proteins adsorb onto PEG, the hydration layer between the polymer and the protein must be removed. This process is energetically unfavorable, and thus, proteins are typically unable to adsorb on PEG surfaces[94]. However, PEG surfaces can lead to the denaturation of proteins[95], and PEG hydration layer is vulnerable to the external condition such as salinity and acidity.

Another category of anti-protein fouling surfaces is zwitterionic materials mentioned inb 3.1.3 Zwitterionic materials like phosphorylcholine, sulfobetaine, and carboxy betaine have excellent biocompatibility[96,97]. Among them, carboxy betaine is the most valuable one because it has a low synthesis cost, biocompatible, and a very strong anti-protein adsorption ability[98,99].

The last type of surface uses mixed materials[89]. The mixed materials combine a group which can form a hydrogen bond and a zwitterionic group[99]. Therefore, it can prevent the formation of foulants of protein by a synergistic action of hydrogen bonding and ion solvation[89].

4.1.2 Bacterial fouling

Bacteria influence many aspects of daily life and hurt the health of humans. Superhydrophobic antifouling surfaces are used for bacterial fouling. This kind of surface can reduce the effective contacting area with water. Since most bacterial contamination scenarios involve planktonic bacteria dispersed in aqueous media, using superhydrophobic surfaces can helpfully lower bacterial adhesion on a surface. Two textures are used for superhydrophobic antifouling surfaces: patterned and random. However, producing this kind of surface with tiny textures at large scales is of great challenge in commercial aspects[100]. Thus, other methods for producing hydrophobic surfaces are used. For example, the deposition of NPs, and the nucleation and growth of nano/microcrystals on a surface via a deposition process are established[101-103]. The superhydrophobic surfaces formed in this way are extremely resistant to bacterial attachment and fouling. However, textures are often destroyed by external mechanical forces, which shortens the durability of surfaces.

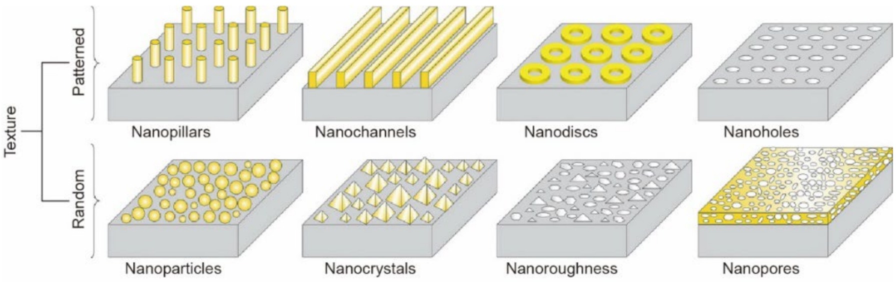

Figure 7: Two types of textures for superhydrophobic surfaces[100].

Another antifouling surface is the repulsion-based surface. This kind of surface uses the repulsive force between bacterial cells and the surface to prevent attachment. Repulsion-based surfaces tend to be very hydrophilic[100]. Some other surfaces are also used to prevent bacterial attachments. It has been proved that SLIPS can greatly reduce the adhesion of *E. coli*, *S. aureus, L. monoHcytogenes*, and S. *enterica* bacteria as well as other biofilm forming organisms[100]. Another type of antibacterial surface uses sharp, needle-like nanoscale structures to break down cell walls[104-107]. By destroying bacterial cell walls, the inactivation of bacteria quickly occurs. Therefore, bacteria are unable to accumulate on the surface. However, this surface is only useful for certain types of bacteria, since its efficacy depends on the weakness of bacterial cell walls.

4.1.3 Algal spores

Algae spores attach to surfaces and grow into mature algae, forming biofilms and fouling. The production of adhesive polymers[108,109] facilitates the settlement and attachment of algal spores. And algae spores form, release, survive, disperse and settle all contribute significantly to the fouling process [110]. Studies have shown that the number of spores attached to biofilms increases with biofilm age[111]. Natural bacterial biofilms were observed to both reduce and increase the adhesion strength of settled spores and other organisms. Algal fouling can be concerning in marine environments due to its negative impacts on infrastructure, such as reduced ship performance and increased maintenance costs[112].

The solution to Algal spore fouling involves developing effective methods to prevent the settlement and growth of algal spores on surfaces. A key challenge is the transition of motile spores to sessile-adhered spores, which triggers the formation of biofilms and subsequent fouling[113]. Biofouling is a process where macromolecular films are colonized by biofilm bacteria, followed by the settlement of motile spores of fouling marine algae[111]. Additionally, it has been observed that different algal species exhibit preferences for specific surface characteristics, such as channel dimensions[114]. Therefore, designing surfaces with unfavorable characteristics for algal settlement can be a potential solution. Testing solutions against settled spores is also important, and techniques like transferring spores to test

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solutions can be employed[115].

4.2 Soft and high length scale--Biofilms

Biofilm fouling is a significant problem and over a billion-dollar / year industry of biocides, cleaners, and antifouling materials worldwide. The biofilm fouling accumulation occurs in multiple steps, and factors can be considered as the transport of suspended material from the fluid to the surface and its attachment to the surface, microbial metabolism within the film, fluid shear stress at the surface of the film, and the roughness and material composition of the surface[116], which gives some perspective to solve fouling.

After a certain amount of research, it has been found that fluoride can be used as a main material to control biofouling. Periodic or continuous chlorination is effective in controlling biofilm fouling. It works by oxidizing biofilm polymers, causing disruption and partial removal, and inactivating parts of the microbial population. Mechanical cleaning can also physically remove parts of the attached film.

4.3 Hard and low length scale

4.3.1 Barnacles

Barnacle fouling refers to the process of barnacle organisms attaching themselves to surfaces, such as ship hulls or underwater structures, and forming a layer of biofouling. Barnacles are known for their strong adhesion abilities, and their fouling can cause various issues. It can increase drag on marine vessels, leading to decreased fuel efficiency and higher operating costs[117]. Barnacle fouling also contributes to the accumulation of other types of fouling organisms, such as algae[118], and can negatively impact the performance and lifespan of submerged equipment[119].

To address barnacle fouling, scientists have been studying the adhesion mechanisms of barnacles and their larvae, exploring the role of the larval nervous system in controlling barnacle settlement and metamorphosis, which can provide insights into strategies to disrupt fouling processes[120].The solution to barnacle fouling is to use of antifouling coatings or paints contain biocidal compounds. These coatings release chemicals that deter barnacles from settling on the surface[121]. Another approach is the development of non-toxic or environmentally friendly coatings, such as polymer films, that prevent barnacles from attaching[122,123]. Physical methods like regular cleaning or scraping can also be effective in removing barnacles from surfaces[116]. Additionally, understanding the biology and behavior of barnacles can help in designing strategies to reduce fouling, such as manipulating surface roughness or using materials that discourage barnacle settlement[119,120].

4.3.2 Diatoms

Algal spores are reproductive cells produced by various algae, while diatoms are a specific type of microscopic algae. Algal spores are mobile and can choose where to settle[124]. They can sense chemical cues in the environment to identify suitable surfaces for attachment[113]. Once a spore finds a suitable surface, it settles and causes fouling[109]. Diatoms are known for their strong adhesion to surfaces and their ability to colonize resistant surfaces. They play an important role in fouling and have unique properties, such as silica or calcium components, that contribute to their fouling behavior[113].

Studies have demonstrated that diatoms possess strong adhesion capabilities to surfaces[125]. The attachment strength of diatoms varies among different species, with some showing tolerance to antifouling paints and the ability to colonize them[126,127]. Diatom fouling tends to occur during microalgal blooms in the summer[128]. There is limited information on the contribution of diatoms to the biofilm ecology formed on fouled surfaces[129]. It is known that antifouling coatings are somewhat effective in limiting the growth of fouling diatoms[127]. Further research is needed to address the role of fouling in the biofilm ecology.

4.4 Hard and high length scale

4.4.1 Waxes

Wax fouling not only decreases the oil transportation rate and reduces productivity, but also cause huge extra costs and brings significant risks. The cost for cleaning 400m-underwater-pipelines can reach $1 million/mile [130]. Most methods used to remove wax fouling focus on deposit-inhibiting additives. However, this action leads to more cost as many of these additives need to be removed before the oil can

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be sold for commercial use.

According to earlier studies, the modulus of wax varies between 61 and 250 MPa[131,132]. Based on the classification of fouling in this review, wax fouling can be removed by minimizing surface modulus. (Fig. 2c) A group tested the degree of wax fouling on low-energy surfaces, including polyvinylidene fluoride, poly (vinylidene fluoride)-chlorotrifluoroethylene, and a methyl acrylate-styrene. They compared the results with fouling on high-energy surfaces, polyurethane surfaces and epoxy surfaces, and proved that wax fouling will decrease as the surface energy decreases[133].

However, in 2022, another group investigated the developable to use tethered liquid-like layers as the surface for removing wax fouling [134]. They studied surfaces of silicon and silicon grafted with different polymers. According to their measurement, a decrease in surface energy actually cannot lead to significance decreased fouling. C-12 (23.74 mN/m), *t*-BCSE (26.82 mN/m), and *t*-PDMS (23.31 mN/m) have similar surface energies, but they have quite different fouling behavior. Compared to F-17 (14.47 mN/m) and C-12, the higher grafting density, high stability, and high conformational entropy led to a greater degree of polymer antifouling for *t*-PDMS and *t*-BCSE. Between two better antifouling surfaces, *t*-PDMS has the best ability of fouling resistance. Their discoveries show that highly disordered or flexible grafted chains might be immensely effective to eliminate polymer adsorption, including wax[134].

4.4.2 Ice

Ice fouling has negative effects in many fields, especially when combined with algae fouling in the marine industry. Ice accumulation on the vessels and the freezing water droplets on exposed surfaces affect the ship's center of mass and weaken its stability [135,136].

The most investigated surfaces for anti-ice fouling are the superhydrophobic surfaces[137,138]. However, the surface properties and micro/nanostructures degrade as they are kept in use. This makes superhydrophobic surfaces exhibit poor anti-icing stability[139,140]. Recently, hydrophilic and amphiphilic anti-icing surfaces have also been developed to prevent ice fouling[141]. The presence of hydrophilic groups can lower the freezing point[142], while hydrophobic groups can repel incoming water droplets. These two properties work together to promote the anti-icing abilities of amphiphilic surfaces[143,144]. However, these two surfaces also have drawbacks when considering the commercial problems during the production process.

In 2023, *Z. Zhang et al.* studied underwater self-layering interpenetrating polymer-network (IPN) coatings comprising fluorocarbon resin/polyacrylate (F/PAA-X, F/PBA-X) via *in situ* polymerised acrylate monomers in a fluorocarbon resin system[145]. The structure extends the nucleation time of ice crystals and makes it easy to remove the ice adhesion[145,146]. The study introduced different types of acrylic monomers into the fluorocarbon system via free-radical polymerization to cross-link the polyacrylate with the fluorocarbon resin, and a series of IPNs were prepared. The IPNs were named F/PAA-X and F/PBA-X. Among all these surfaces, F/PAA-4 shows the lowest freezing point for water. *Z. Zhang et al.* also learnt the stability of wetting of coating and found that F/PBA-3 has the smallest change in surface energy (32.01 J/m^2). This indicates that F/PBA-3 is the most stable surface. Moreover, by carrying out 20 icing/de-icing cycle tests in 20 days, the group found that F/PBA-3 has a more stable de-icing capacity than other surfaces. Therefore, considering the manufacturing cost, F/PBA-3 is more valuable to develop further.

4.4.3 Inorganic scale

Inorganic scale fouling is a serious problem in many fields, such as oil, water desalination, and power generation[147]. It causes an increased cost of production, a loss of productivity, and health problems to humans. In pipeline applications of the oil industry, examples of inorganic scale fouling include $CaCO_3$, $BaSO_4$, and $SrSO_4$. There are typically two types of inorganic-scale-fouling formation. The first one is when temperature and pressure change, carbonate scales form. The second one is that sulfate scales take place where two incompatible brines mix[148-150].

In 2016, a group studied how different properties of surfaces influence the fouling of inorganic scale under different conditions. They found that in the deposition process, no direct relationship is between the surface roughness and the scale size[151]. Also, due to daily applications, the surface modulus cannot determine which kind of surface is useful in preventing the formation of inorganic scale fouling. Another group in 2018 studied the lotus-like superhydrophobic surfaces to prevent the formation of inorganic scale fouling. They found that this kind of surface is not as effective as the lubricated slippery surfaces, which have more development potential for restricting inorganic scale fouling[152].

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5. Conclusion

The fouling phenomenon is a dynamic and intricate process which encompasses multiple length scales and contains a diverse array of molecules and organisms. This review classifies fouling according to length scale and elasticity modulus. Nature offers valuable insights into the significance of these factors through examples of antifouling and fouling-release surfaces and their working strategies. The core of this review is the synthetic antifouling coatings that are currently in use to reduce nine different kinds of fouling.

The development of effective antifouling strategies necessitates the integration of both chemical and physical principles, no single chemical or physical technique has been universally proven to be effective in preventing fouling or facilitating the release of fouling. In recent decades, there has been a global gradual prohibition of conventional toxic antifouling coatings. In real-world scenarios, these materials may encounter a range of constraints such as poor mechanical strength, antifouling durability, and weak bonding strength. Furthermore, the practical implementation of these surfaces often requires precise material control at multiple stages or aspects, highlighting the limitations of traditional surface design methodologies. Given the collective desirable criteria, it remains highly difficult to achieve a fabrication method for antifouling coatings that is both non-toxic, cost-effective, easy-applicable, and adaptable while maintaining efficiency and durability. The utilization of bioinspired coatings offers a multitude of advantages, including heightened efficiency, environmental compatibility, and cost-effectiveness. It might remain essential to enhance the antifouling effectiveness by exploring novel polymers and/or alternative materials, with a particular emphasis on their enduring performance in both stationary and moving settings. The multi-functional anti-fouling coating can significantly improve the anti-fouling effect and extend the service life, and is non-toxic to the environment. It can also be further improved to achieve more functions, such as reducing resistance, reducing noise, anti-corrosion and so on.

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