

Review

Progress in Marine Antifouling Coatings: Current Status and Prospects

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Abstract: The shipping industry is vital to global trade. Unfortunately, this industry is negatively impacted on a large scale by biofouling, a process whereby unwanted organisms accumulate on submerged surfaces, massively affecting traveling speed and fuel consumption. Fortunately, antifouling coatings have been developed to combat this problem. This review summarizes the process of biofouling and briefly discusses the history of antifouling coating development. Moreover, eight major antifouling coatings are reviewed, including bionic microstructure, self-polishing, fouling and desorption, zwitterionic polymer, self-assembled thin-layer, liquid-smooth surface, conductive, and photocatalytic antifouling coatings. The technical principles, innovation, and advancement of each coating are expounded, and the relevant research progress is discussed. Finally, the remaining issues and challenges in antifouling coatings are discussed, along with their prospects.

Keywords: antifouling coating; self-polishing; progress; polymer

1. Introduction

The shipping industry, accounting for an estimated 90% of global trade, is particularly impacted by biofouling. The agglomeration of marine organisms on ship hulls increases fuel consumption, promotes the emission of environmentally harmful gases (CO₂, SO₂, and NO₂), and diminishes the ship's velocity, engendering adverse economic impacts [1–3]. Biofouling's ecological implications are no less severe, facilitating the invasion of alien species and environmental degradation [3,4], as shown in Figure 1. Indeed, it is reported that the U.S. Navy alone has spent approximately USD 1 billion annually over the past decade to address this problem, while global expenses have exceeded USD 15 billion annually [5,6].

Before discussing the broader implications, it is crucial to delve into the role of antifouling coatings in biofouling mitigation. Antifouling coatings are specialized surface treatments applied to ship hulls, serving as a critical frontline deterrent against biofouling. These antifouling coatings function either by creating a surface that is inhospitable for marine organism adhesion or by releasing biocides that kill or repel these organisms. Given their ability to dramatically reduce the incidence of biofouling, antifouling coatings have gained significant academic and industrial attention. However, the challenge lies in developing coatings that are effective yet environmentally benign, as traditional antifouling coatings have often included harmful toxins that adversely affect non-target marine species [7].



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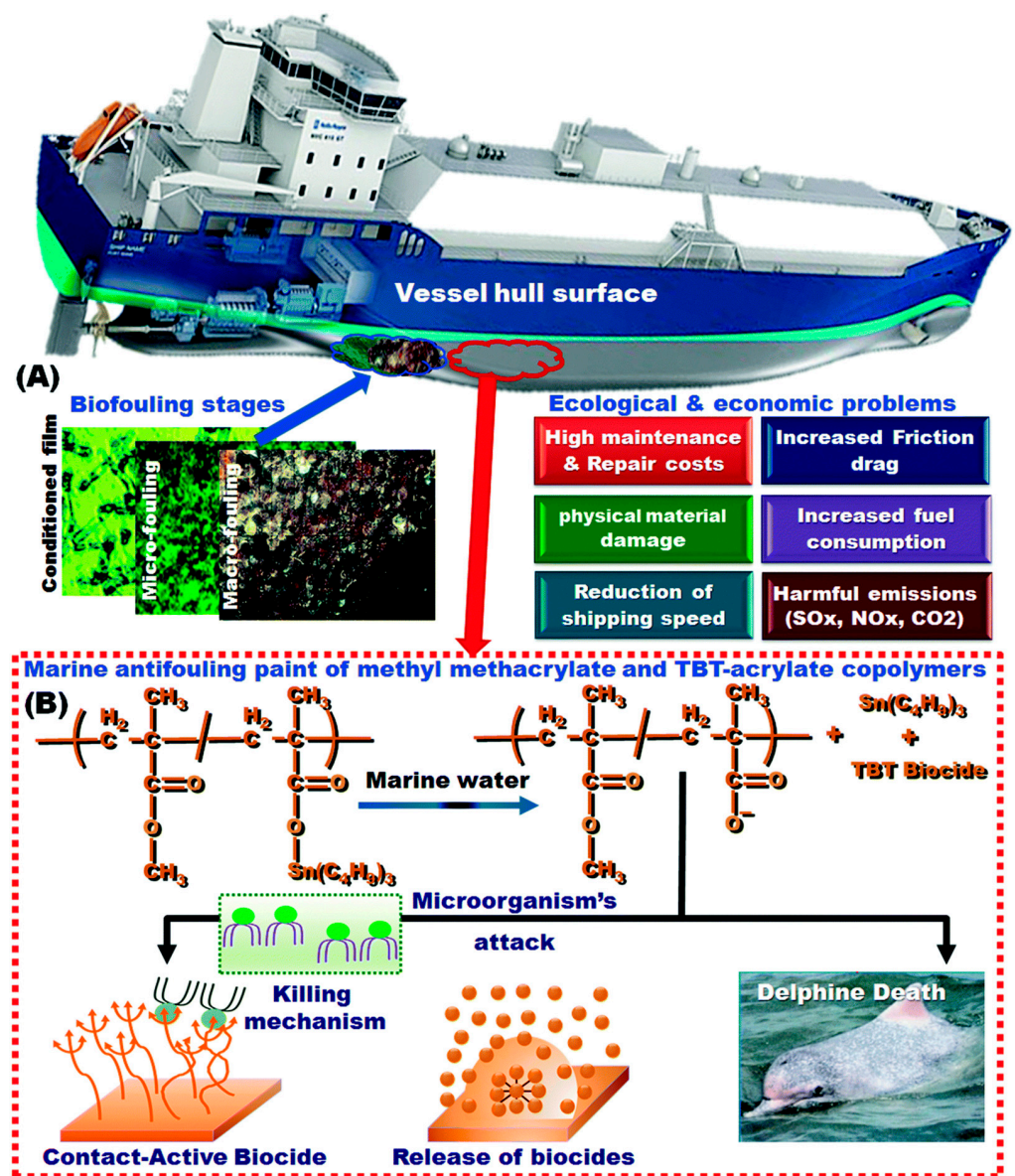


Figure 1. (A) Representative scheme for biofouling stages (Bottom). A series of hazards caused by biological fouling attached to the hull [8–10]; (B) Release mechanism of bactericide in traditional toxic marine antifouling coatings, it can kill not only fouling organisms but also non-target species such as dolphins and fishes (right-side) [11]. Figure reproduced with permission from Progress in Materials Science (Elsevier).

Biofouling mitigation encompasses a range of technologies, including mechanical, ultrasonic, and electrochemical methods, and particularly antifouling coatings. Antifouling coatings have taken precedence in the fight against biofouling, as evidenced by the increasing volume of scholarly publications addressing their development and application [12]. Despite the extensive research focusing on antifouling coatings, including their classification, adhesion mechanisms, and formation processes, there is an apparent dearth of synthesized and up-to-date analyses that amalgamate the existing knowledge while identifying emerging trends, innovations, and challenges [13–16].

This review seeks to address this gap by providing a comprehensive and updated examination of marine antifouling coatings developed over the past two decades. Therefore, this review aims to deliver a comprehensive and updated examination of marine antifouling coatings developed over the past two decades. Building upon the foundations of prior

reviews, this work seeks to highlight recent advancements and delineate the progression of antifouling technologies. This review will also delve into the research progression of eight advanced antifouling coatings, concentrating on their fundamental principles, innovations, and prospective applications. By systematically scrutinizing both established and emergent technologies, this review aspires to provide a comprehensive resource for researchers, industry practitioners, and policymakers engaged in the development of sustainable and effective antifouling solutions. The overarching goal is to contribute to the forward momentum in the field of marine antifouling coatings: their current status and prospects [17–20].

2. Formation of Common Marine Biofouling

Biofouling is a multifaceted process [21–23], typically comprising four distinct stages, as shown in Figure 2: conditioning and biofilm formation (stages I and II), microfouling (stage III), and macrofouling (stage IV). Upon submersion in water, organic and inorganic macromolecules such as polysaccharides and proteins, adsorb to the submerged surface, forming an initial conditioning layer (stage I) [24]. Within hours, bacteria and algae, the primary colonizers, anchor themselves to this layer, giving rise to a mature biofilm (stage II). This biofilm, teeming with extracellular polymers secreted by various organisms, cultivates an environment favorable to the attachment of diverse species. Importantly, the first two stages are generally reversible as bacteria and diatoms can be effectively dislodged [25,26].

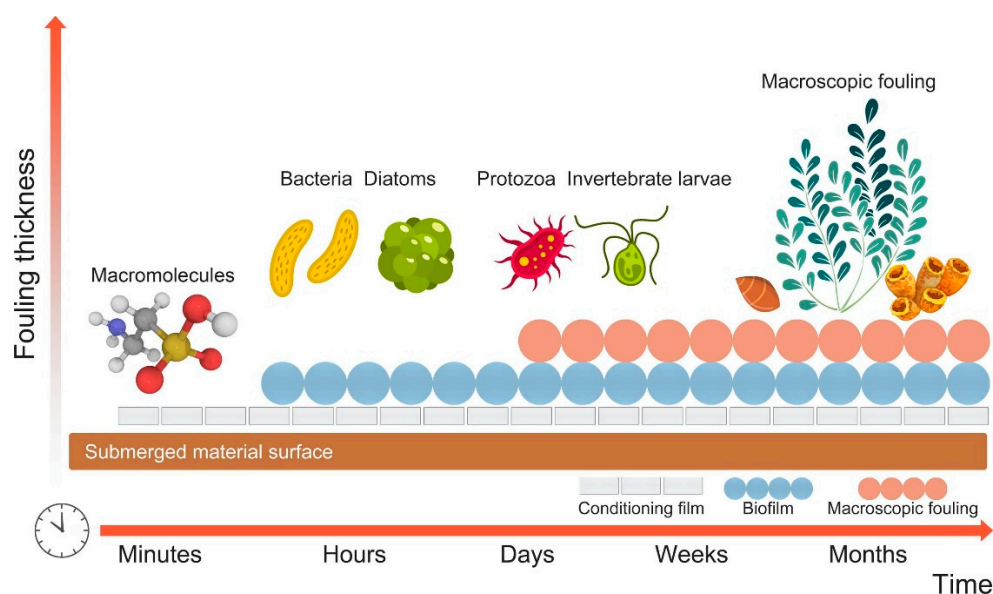


Figure 2. Schematic depicting the four stages of typical marine biofouling [12]. Figure reproduced with permission from Progress in Materials Science (Elsevier).

As the process advances, protozoa, invertebrates, and algal larvae are lured by the substances produced by the bacterial community [27–29], and consequently form distinct microscopic fouling communities (stage III). A few weeks post biofilm formation, larger organisms, such as algae, barnacles, mollusks, and mussels, adhere to the biofilm, culminating in a macrofouling community (stage IV) [30,31]. Despite this linear succession being widely acknowledged, it is critical to note that the formation of biofouling is inherently a dynamic process. It is contingent upon various factors including the type of benthic organisms, as well as the water area’s seasonal and geographical characteristics [32–34].

Macrofouling organisms typically materialize post-biofilm formation, although some organisms, such as bryozoan larvae, may affix to the surface prior to biofilm formation [35]. Barnacles represent a prominent class of fouling organisms. Their sturdy attachments make their removal challenging [36]. When barnacles settle on a surface, they secrete chemical substances, attracting more barnacles for proliferation, leading to increased macrofouling.

Laboratory tests have indicated that biofilms formed during stage II can amplify material friction by up to 70% [37]. Certain macrofouling organisms possess hard, calcareous shells, exacerbating the frictional impact when they cover a surface. In ship resistance tests, severe calcareous biofouling decreased the ship's navigation speed by up to 86% [38].

Biofouling displays spatiotemporal fluctuations, contingent upon factors such as season, temperature, water area, water depth, and light conditions [39,40]. Consequently, it is a complex, dynamic process influenced by a myriad of environmental factors. Researchers have reported on biofouling variations in different seas, including the Red Sea [41], the Persian Gulf [42], the South China Sea [43], Mersin Bay [44], and the Taranto Sea [45]. These spatiotemporal changes present a significant challenge to the development of new antifouling coatings. As a result, there is a pressing need for the development of a broad-spectrum antifouling coating with substantial environmental adaptability.

In order to elucidate the multi-dimensionality of marine biofouling—a phenomenon influenced by a plethora of environmental, biological, and physicochemical factors—we present Table 1 subsequent to this section. This tabular representation synthesizes the most current empirical findings across biofouling stages, influential variables, and their respective impact on material properties. It aims to serve as a consolidated reference that encapsulates the complexities inherent in biofouling processes, thereby providing researchers, industry stakeholders, and policymakers with a structured framework for guiding future innovation in the design and application of antifouling coatings.

Table 1. Summary of Key Aspects and Challenges in Marine Biofouling Across Different Stages.

Biofouling Stage	Main Fouling Organisms	Key Findings	Influencing Factor	Strategies and Challenges
Stage I	Macromolecules, proteins, polysaccharides, etc.	Formation of the initial conditioning layer	Seasons	Development of efficient primary antifouling coatings
Stage II	Bacteria, algae, etc.	Biofilm maturation, extracellular polymer production	Temperature	Regulating coatings to achieve a balance of antimicrobial and anti-algae effects
Stage III	Protozoa, larvae, etc.	Microbial fouling aggregates on biofilm surfaces to form communities	Geographic location	Study of antifouling paint interactions with microorganisms and benthic organisms
Stage IV	Large fouling organisms such as barnacles and clams	Large fouling organisms replace microscopic fouling organisms such as larvae that attach to surfaces and grow continuously	Water depth and light intensity	Development of multifunctional antifouling coatings

3. History of Antifouling Technology Development

The evolution of antifouling coatings, as illustrated in Figure 3, has spanned several centuries. Initially, rudimentary measures were employed to mitigate biofouling in marine vessels. Circa 200 BC, protective layers of hot asphalt, tar, and grease were applied to the wooden hulls of ships. Subsequently, the use of thin lead plates for the same purpose came into practice [46]. With the advent of the Age of Discovery, it was noted that barnacles and seaweeds drastically impeded ship speeds, prompting the use of copper or brass to coat the ship bottoms. A renowned example of such an application is the Cutty Sark, which features a brass coating [47]. Such thin copper layers proved instrumental in combating marine biofouling, inadvertently facilitating Britain's ascendancy as a naval power in the 18th century. By the late 18th century, various toxic compounds such as arsenic, sulfur, and

mercury were used for hull protection [48]. In 1926, the US Navy developed an antifouling coating made from rosin, with copper and mercury oxides as fillers, which could resist biofouling for up to 18 months [49–51].

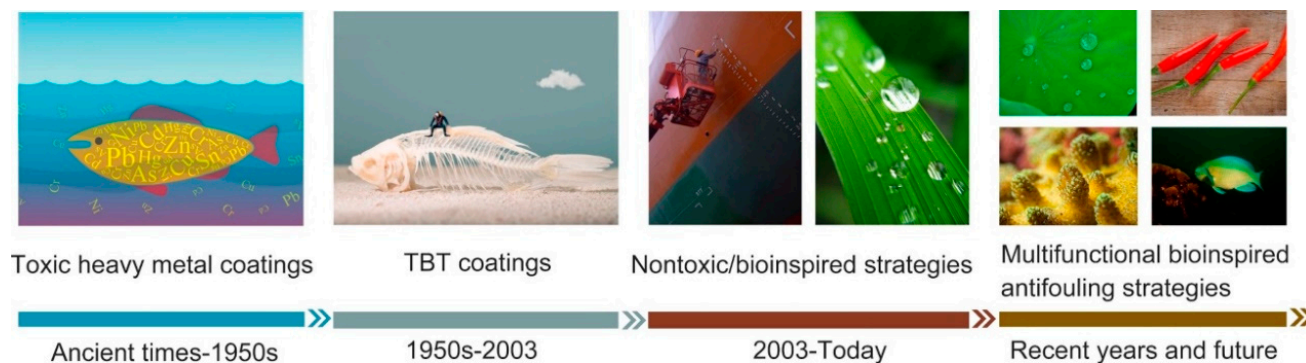


Figure 3. History of antifouling coatings [12]. Figure reproduced with permission from Progress in Materials Science (Elsevier).

The 1950s marked a departure from conventional antifouling coatings, triggering the search for innovative antifouling technology. The Dutch scientist van der Kerk discovered the antifouling properties of tributyltin compounds [52], which were deemed the most effective antifouling technology of that era, leading to widespread global usage. However, while these coatings were highly effective at preventing biofouling, they posed a significant ecological threat [53]. The environmental catastrophe instigated by tributyltin usage included deformities in oyster shells and abnormal larval development, leading to the collapse of the oyster industry in the Bay of Arcachon, France, in the 1980s [54]. Investigations revealed that tributyltin compounds accumulated within various organisms, including crustaceans, fish, birds, mammals, and even humans [55]. These compounds caused hermaphroditism and abnormal growth in many organisms, and due to their slow degradation and significant accumulation in marine environments, their harmful effects persisted for months or even decades [56].

Growing awareness of the harmful effects of tributyltin compounds led to the implementation of legislation in various countries, including the United Kingdom (1987), the United States (1988), Canada (1989), Australia (1989), and the European Union (1989), prohibiting their usage on both small and large vessels [57]. Subsequently, in 2001, the International Maritime Organization (IMO) recommended a global ban on the production and use of tributyltin-containing antifouling coatings, effective from 1 January 2003, and 1 January 2008, respectively. This ban highlighted the pressing need for environmentally friendly alternatives in the field of antifouling coatings [58,59]. In response to this urgent requirement, significant advances have been made in the development of antifouling coatings. Currently, there are eight recognized categories of advanced antifouling coatings, each with unique properties and mechanisms. These categories include biomimetic microstructure antifouling coatings, self-polishing antifouling coatings, fouling-release coatings, zwitterionic polymer antifouling coatings, self-assembled monolayer antifouling coatings, slipper liquid-infused porous surface (SLIPS) antifouling coatings, conductive antifouling coatings, and photocatalytic antifouling coatings, as shown in Figure 4. These advancements in antifouling coating technologies offer diverse approaches to mitigate biofouling problems. Biomimetic microstructure coatings emulate natural surfaces, reducing fouling by incorporating microscale features inspired by marine organisms. Self-polishing coatings release biocides at a controlled rate, preventing excessive buildup of fouling organisms. Fouling-release coatings utilize low-surface-energy materials to minimize adhesion, allowing for easy removal of fouling organisms by hydrodynamic forces. Zwitterionic polymer coatings have hydrophilic and cationic properties, inhibiting the attachment of marine organisms. Self-assembled monolayer coatings form a stable and smooth surface,

detering the attachment of fouling organisms. SLIPS coatings utilize a liquid-infused layer to create a slippery surface, preventing fouling attachment. Conductive coatings employ electrical currents to inhibit biofilm formation and fouling organism settlement. Lastly, photocatalytic coatings utilize light-activated substances to degrade fouling organisms.

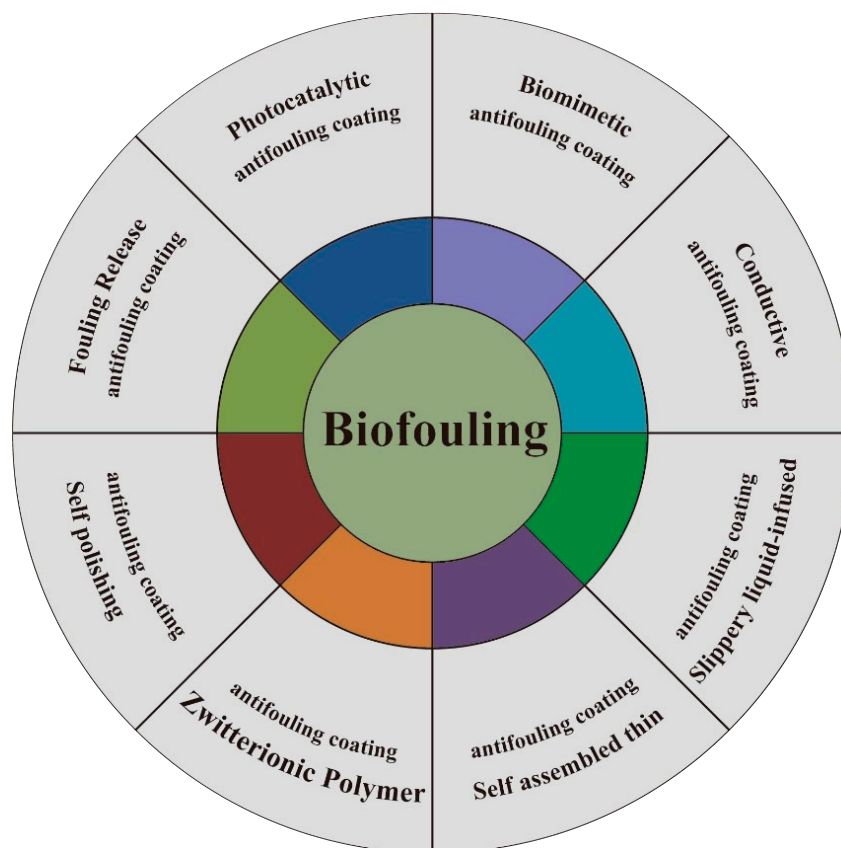


Figure 4. Eight major advanced antifouling coatings. (biomimetic microstructure coatings, self-polishing coatings, fouling-release coatings, zwitterionic polymer coatings, self-assembled monolayer coatings, slipper liquid-infused porous surface coatings, conductive coatings, and photocatalytic coatings).

Following the elucidation of the eight prominent advanced antifouling coatings depicted in Figure 4, it is imperative to delve deeper into the nuances of their evolution over the past decade. To facilitate a comprehensive understanding of their technological trajectory, Table 2 provides an intricate breakdown. This tabulation meticulously delineates the technological innovations, pivotal ingredients, application domains, and environmental and safety implications associated with each coating. By elucidating these aspects, the table aims to underscore the multifaceted advancements these coatings have undergone, highlighting their significance in meeting contemporary challenges and potentially shaping future maritime and industrial applications. Such an examination offers a rich perspective into the sophisticated landscape of antifouling coatings, emphasizing the intricate interplay of science, technology, and environmental stewardship in this domain.

Table 2. Details of the development of advanced antifouling coatings in the last 10 years.

Antifouling Coatings	Technological Innovation	Key Ingredient	Application Area	Environment and Safety
Biomimetic microstructure coatings	Mimics the structure of natural organisms to enhance fouling and dampening effects	Natural materials, surface chemical modification, or surface physical modification	Ship hulls, marine equipment, medical equipment	Friendly to the marine environment and non-polluting
Self-polishing coatings	The coating wears off automatically over time, always maintaining a new surface.	Biodegradable polymers, organometallic compounds	Ship hulls, marine structures	Reduce toxic bioaccumulation, but consider chemical releases
Fouling-release coatings	Reduces surface energy, making it difficult for fouling organisms to adhere	Polysiloxanes, Fluorine compounds	Ship hulls, marine equipment	Non-toxic, reduced impact on marine ecology
Zwitterionic polymer coatings	Using positive and negative charge balance to resist bio-attachment	Phosphate, sulfate polymers	Medical equipment, water treatment equipment	Biocompatible and biologically benign
Self-assembled monolayer coatings	Formation of monomolecular layers on the surface by self-assembly technology	Silane, phosphate	Microelectronics, nanotechnology equipment	Environmentally friendly, but need to control the use of chemicals in the synthesis process
Slipper liquid-infused porous surface coatings	Liquid impregnation of porous surfaces to create an ultra-slip effect	Organic liquids, porous materials	Ship hulls, pipelines	Reduces biological and contaminant attachment and improves cleaning efficiency
Conductive coatings	Provides electrical conductivity for antifouling and electromagnetic shielding	Metal nanoparticles, conductive polymers	Electronic equipment, ship hulls	Ecological impacts of metal particles and currents need to be considered
Photocatalytic coatings	Decomposition of organic biofouling by photocatalytic reaction	Titanium dioxide, zinc oxide	Building surfaces, water treatment equipment	Effective reduction of toxic substances, environmentally friendly

4. Underlying Principles and the Innovation of Advanced Antifouling Coating Technologies

4.1. Biomimetic Microstructure Antifouling Coatings

Biomimetic antifouling coatings have emerged as a promising approach, drawing inspiration from nature's defense mechanisms against biological contaminants. By replicating the micro- and nanostructures found in marine flora and fauna, these coatings aim to imbue various substrates with superhydrophobic and self-cleaning capabilities. The potential for this biomimetic approach was explored by Scardino et al. [60] who investigated a range of natural surfaces, including those of willow coral, marine mammal skin, squash eggshell, echinoderms, algae, shark skin, mollusks, and blue mussels, as shown in Figure 5.

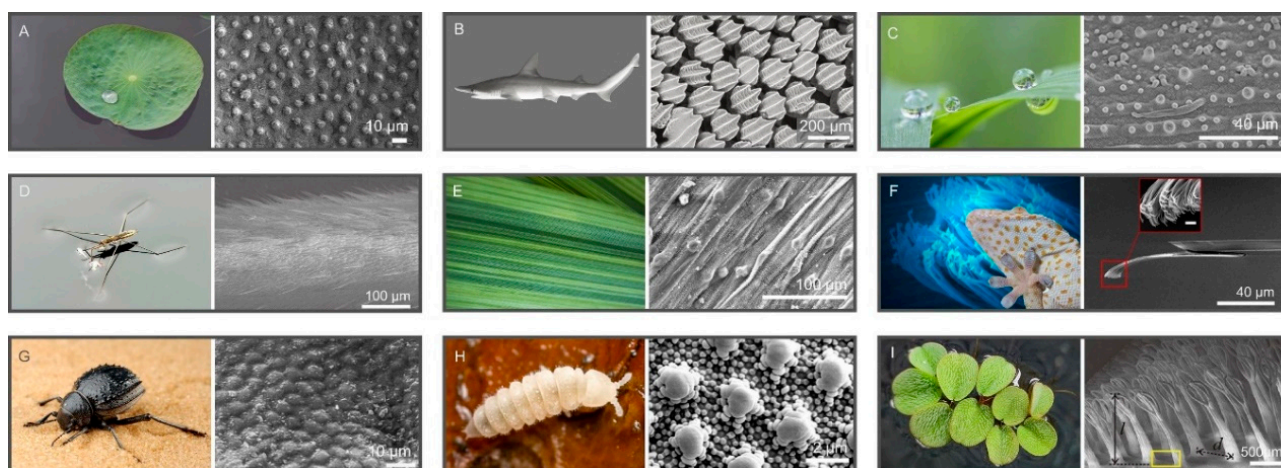


Figure 5. (A) Photograph of a lotus leaf and SEM image of its microstructures [61]; (B) Photograph of a shark and SEM image of shark skin microstructures [62]; (C) Photograph of droplets on rice leaves and SEM image of rice leaf microstructures [63]; (D) A water strider and SEM image of its leg [64]; (E) Photograph of a reed leaf and SEM image of its microstructures [65]; (F) A gecko and SEM image of the setal array of its feet [66]; (G) Photograph of a beetle and SEM image of the textured surface of the elytron regions [67]; (H) Photograph of a springtail and SEM image of the microstructures of its surface [68]; (I) Leaves of *Salvinia molesta* and its egg-beater structures observed by SEM [69]. Figure reproduced with permission from Progress in Materials Science (Elsevier).

A significant milestone in this field was the development of the ‘Sharklet’ surface by Schumacher et al. [70], which replicated the microtexture of shark skin. Remarkably, this surface exhibited antibacterial properties solely through physical alteration of the substrate surface, without the need for chemical additives. This breakthrough paved the way for further innovations in the design of bionic surfaces with enhanced functionalities. Advancing the Sharklet technology, Yin et al. [71] successfully fabricated a superhydrophobic bionic surface on PTFE via femtosecond laser direct writing. The resultant surface exhibited superior self-cleaning capabilities along with substantial mechanical stability. Following this approach, Liu et al. [72,73] engineered intelligent superhydrophobic surfaces that dynamically altered their wetting properties in response to external stimuli such as pressure or light, thereby introducing adaptive antifouling capabilities.

Micro-scale geometries also play a crucial role in antifouling performance. Xu et al. [74] discovered that surfaces with specific micrometric configurations, such as columnar structures measuring 3 μm and grooved surfaces measuring 12 μm , exhibited highly effective resistance against different types of algae adhesion. Furthermore, Schumacher et al. [75] developed a surface with varying nano-force gradients, resulting in reduced algal adhesion by precisely analyzing their adhesion behavior. Notably, a surface with a nano-force gradient of 374 N achieved a 53% reduction in algal adhesion compared to an untreated surface. Additionally, Scott P. Cooper et al. [76] demonstrated that micro-patterned surfaces could reduce the attachment of *Ulva lactuca* zoospores by 70%–80% compared to smooth surfaces, suggesting a synergistic antifouling mechanism involving microtopography.

However, challenges remain in this field of biomimetic antifouling coatings. Scardino et al. [77] developed a coating combining superhydrophobicity and microtopography, significantly reducing diatom adhesion. Nonetheless, this coating suffered from poor mechanical durability and a relatively short lifespan. Therefore, the development of coatings with superior mechanical robustness is essential for their practical application. In summary, the post-Sharklet era saw rapid innovations in bionic surfaces, with features like stimulus responsiveness and tailored micro-geometries. However, mechanical durability remains a challenge that needs addressing for the widespread application of these coatings [78,79].

Recent innovations have made strides in addressing these limitations. Liu et al. [80] introduced a resilient, superhydrophobic composite coating that mimics the structure of a caterpillar's body. This coating exhibited superior mechanical durability, along with self-repairing, self-cleaning, and antifouling characteristics. Similarly, Wang et al. [81] developed a superhydrophobic micro-nano diamond film inspired by the surface properties of plant leaves, showing remarkable mechanical and chemical durability, as well as reducing green algae adhesion by over 95% in marine environments.

These innovative antifouling coatings mimic nature's ability to resist fouling and, in doing so, present environmentally friendly solutions. Moreover, their low surface energy reduces drag for ships, resulting in fuel savings and increased range. Some companies have begun to adopt these bionic superhydrophobic coatings for their vessels. Notably, compared with conventional antifouling coatings, ships utilizing these bionic coatings experienced approximately 6% fuel savings and reduced emissions [82]. However, there are still hurdles to overcome with these coatings. First, they work best when the coated object is in motion. Second, some coatings still suffer from mechanical vulnerabilities. Third, certain coatings contain substances that may leach and cause environmental pollution. Therefore, the effectiveness of bionic superhydrophobic antifouling coatings can be compromised under static conditions and over time due to wear and environmental effects.

To address the issue of static conditions hindering antifouling coating effectiveness, Zhu et al. [83] proposed an innovative solution. They integrated the properties of a stable and non-toxic organic phosphorus scale inhibitor, pentamethylenetriamine methylene phosphonic acid (DTPMPA), into a superhydrophobic coating. This resulted in the development of the DTPMPA Superhydrophobic Anodic Aluminum Oxide (DSAA) coating, which exhibited a novel combination of physical and chemical antifouling mechanisms, promising efficacy in both dynamic and static conditions. The DSAA coating was created through a two-step process. First, an anodic aluminum oxide coating was prepared using a traditional double electrode system, and DTPMPA was then loaded into the nanostructure of this surface. Subsequently, the coating was modified with stearic acid to achieve a superhydrophobic property, resulting in the DTPMPA Superhydrophobic Anodic Aluminum Oxide coating. This coating showcased a synergy of physical and chemical antifouling properties. The nanostructure created an air layer inhibiting the deposition of calcium carbonate, while DTPMPA was continuously released to prevent the bonding of calcium ions with carbonates. This novel approach provides a promising avenue for developing antifouling coatings effective in both static and dynamic conditions [84–86].

In summary, biomimetic antifouling coatings have witnessed rapid innovations in bionic surfaces, incorporating features such as stimulus responsiveness and tailored micro-geometries. However, mechanical durability remains a challenge, and recent research has focused on developing coatings that address this limitation. Efforts towards improved mechanical resilience, such as the caterpillar-inspired composite coating and the diamond film, show promise. Additionally, the integration of stable chemical components, exemplified by the DSAA coating, has shown the potential to enhance antifouling effectiveness in both dynamic and static conditions. Continued research and development in this field are crucial for the widespread application of these coatings and the advancement of environmentally friendly solutions for fouling prevention.

4.2. Self-Polishing Antifouling Coatings

Self-polishing antifouling coatings represent a significant advancement in the field of marine antifouling technology. These coatings are designed to be self-renewing by leveraging the mild alkalinity of seawater to initiate the hydrolysis of their resin component. This process involves a resin and an antifouling agent, where the antifouling agent is gradually released as the resin undergoes hydrolysis and becomes brittle. The shear stress from the flowing seawater then removes the outer layer, exposing a fresh antifouling surface. This sophisticated mechanism ensures the continuous removal of biofouling material while maintaining an effective concentration of the antifouling agent [87–89].

By effectively balancing hydrophobic properties that deter seawater infiltration with a controlled surface erosion rate, self-polishing antifouling coatings achieve durability and consistent antifouling effectiveness [89–91]. A comparison between surfaces with and without the self-polishing coating after a 30-day period in the South China Sea, as illustrated in Figure 6, demonstrates the superior performance of this technology. However, further research is needed to customize and optimize these coatings for specific applications [92,93].



Figure 6. Images of marine antifouling coatings comprising stainless steel plates coating paint after immersion in the South China Sea (N 22°33', E 114°32') for a month. (No. 52) Control plat; (No. 45) Hydrophobic coating; and (No. 48) Hydrophilic coating [93]. Figure reproduced with permission from Progress in Organic Coatings (Elsevier).

Significant advancements in antifouling coatings have emerged from both academic research and industrial applications. For instance, the South China University of Technology has made seminal contributions by devising a copolymer antifouling coating that leverages both side chain hydrolysis and main chain degradation. Their pioneering work, validated in field trials, allows for the tailored control of both the coating's degradation rate and the release rate of the antifouling agent, thereby ensuring long-term effectiveness [94,95]. On the industrial front, Jotun Hull Performance Solutions has set a benchmark with its SeaQuantum brand, particularly with its flagship product SeaQuantum Skate. Having been applied to over 16,000 ships over the past two decades, this product not only withstands rigorous ship maintenance procedures but also contributes to eco-efficiency. It results in up to a 16.2% reduction in fuel consumption and carbon emissions while conforming to international maritime regulations by eschewing the use of harmful substances like tributyltin (TBT) or cyclobutane.

Despite the success of self-polishing antifouling coatings, a notable drawback is their use of ecologically harmful biocides [96,97]. For example, Jalaie et al. [98] evaluated the release rate of biocides from antifouling coatings, the corrosion effects associated with antifouling, and the physical properties of different coating types, and for optimal physical properties and antifouling efficiencies, the total amount of biocides added to the coatings was 30%–50%, which undoubtedly has a bad impact on the water environment. This has prompted the development of alternative, environmentally friendlier antifouling coatings using materials such as copper, silver, metal oxides, and organic fungicides. For instance, Jiang et al. [99] developed an innovative anti-algal nanocomposite hydrogel incorporating silver nanoparticles (AgNPs). The hydrogel combines mechanical resilience with exceptional antifouling performance. The key innovation lies in the use of silver, known for its antibacterial properties, which proved highly effective in preventing the adhesion of marine organisms such as *Phaeodactylum tricornutum* and *Chlorella*. This innovation points to a more sustainable and efficient solution for marine antifouling applications.

In summary, self-polishing antifouling coatings emphasize self-renewal and effective antifouling properties through the controlled release of antifouling agents. While these coatings have shown impressive performance, further research is needed to fine-tune their optimization. Additionally, the industry has witnessed the success of market-leading brands like SeaQuantum, while also exploring environmentally friendly alternatives such as nanocomposite hydrogels incorporating silver nanoparticles. These advancements contribute to the continuous development of antifouling technologies that are ecologically sound and effective for various marine applications.

4.3. Fouling Desorption Antifouling Coating

Commercial antifouling coatings encompass various types, with silicone, organic fluorine, and silicon-fluorine resin coatings being prominent options. These coatings function by reducing adhesion between marine organisms and surfaces, allowing seawater to sweep them away during vessel movement [100,101].

Organic silicone-based coatings are notable for their eco-friendly attributes, as they do not contain biocides, posing no harm to aquatic life. Additionally, they exhibit resistance to weathering and corrosion. However, these coatings have limitations. Their optimal performance is limited to high speeds and hydrodynamic conditions, and they struggle to remove specific types of fouling, such as diatom mud near the waterline [102]. Furthermore, they tend to have poor adhesion and are susceptible to damage. On the other hand, organic fluorine coatings possess a low surface energy and a high water contact angle (reaching 114°), enabling them to resist initial fouling adhesion effectively [103]. However, these coatings require frequent maintenance, exemplified by the USS Parrot, which necessitated hull cleaning every six months due to the porous surface of the coating.

Acknowledging the strengths and weaknesses of individual coatings, researchers have aimed to combine the best features of organic silicone and organic fluorine coatings. By utilizing a siloxane chain as the main chain and incorporating the -CF₃ group into the side chain, a hybrid coating can be engineered to leverage the advantages of both organic silicone and organic fluorine materials. This approach, as studied by Williams et al. [104], shows promise in antifouling applications. Furthermore, Barroso et al. [105] conducted research on coatings made from polysilazane with loaded PTFE particles, thus enriching the field of hybrid systems. These hybrid coatings exhibit lower surface-free energy and improved water contact angles, presenting positive prospects in the realm of antifouling coatings.

However, it is important to note that hybrid coatings are not without their challenges. Combining two different types of materials can sometimes lead to compatibility issues, resulting in reduced long-term stability of the coating. Moreover, the complexity involved in formulating these hybrid systems could potentially make them more costly to produce, posing an economic challenge for widespread adoption.

In summary, commercial antifouling coatings encompass silicone, organic fluorine, and silicon-fluorine resin coatings. While organic silicone coatings are environmentally friendly and resistant to weathering and corrosion, their performance is limited to specific conditions and they suffer from poor adhesion. Organic fluorine coatings possess excellent initial fouling resistance but require frequent maintenance. Researchers have explored hybrid coatings that integrate the advantages of organic silicone and organic fluorine materials to overcome these limitations but face challenges in material compatibility and production cost. This approach shows potential for enhancing antifouling performance and offers opportunities for the development of more effective and durable coatings in the future.

4.4. Zwitterionic Polymer Antifouling Coating

Zwitterionic polymer antifouling coatings, possessing an equal number of anionic and cationic groups, are hydrophilic and effective against fouling. They have shown exceptional performance in deterring biofilm formation, including that of *Staphylococcus aureus* and *Pseudomonas putida* [106–109]. Their potential extends beyond maritime applications to

encompass biomedical implants and other areas. Nevertheless, there are challenges. The performance of these biodegradable coatings in marine environments is still uncertain. One aspect that requires optimization is the balance between the degradable and hydrolyzable chains, which influences water absorption and the coating's mechanical strength [110]. Another concern is the degradation rate; an expedited rate improves anti-adhesion but reduces lifespan [111,112]. Ma et al. [107] devised strategies to regulate the degradation rate, though their practicality in marine environments remains to be seen.

Moreover, the grafting of zwitterionic polymers onto surfaces is cumbersome and constrained by the surface composition [113–115]. Recently, the integration of zwitterionic functionalized nanoparticles has emerged as a solution [116]. These nanoparticles not only bolster mechanical strength but also retain the antifouling properties of zwitterions. One example involves polymethacrylic acid sulfobetaine-functionalized silica nanoparticles, which have demonstrated resilience and effectiveness against fouling [117,118].

Antifouling coatings containing zwitterionic functional nanoparticles can effectively prevent the adhesion of BSA, fungal spores, *Agave cocci*, and *Escherichia coli* [119]. The materials used are cheap and processable, which enables the modification of the coating to achieve more functions. These advantages show that they have potential practical applications. However, the agglomeration of nanoparticles due to high surface energy could compromise the uniformity and effectiveness of these coatings [120]. Hence, refining the proportion and modification of nanoparticles is essential.

Recently, BIMCO and the Safinah group, in collaboration with shipping organizations and hull cleaning companies, have been developing efficient zwitterionic coatings aimed at environmental preservation and emission reduction. Notwithstanding, research on zwitterionic functional nanoparticles is still in its infancy, with only silica-based particles studied thus far. Future advancements necessitate the exploration of a broader spectrum of modified nanoparticles for maritime antifouling applications.

In summary, zwitterionic polymer antifouling coatings exhibit remarkable hydrophilic properties and effectiveness against fouling organisms. However, challenges remain in terms of their performance in marine environments, degradation rate regulation, grafting onto surfaces, and the integration of zwitterionic functional nanoparticles. Ongoing research and development efforts, including collaboration among industry stakeholders, hold promise for the advancement of zwitterionic coatings and the exploration of a wider range of modified nanoparticles for maritime antifouling applications.

4.5. Self-Assembled Thin-Layer Antifouling Coatings

Self-assembled thin-layer antifouling coatings are engineered through the layer-by-layer assembly of multiple substances. For instance, Ren et al. [121] developed a self-healing coating composed of chitosan and dialdehyde starch, demonstrating excellent adhesion to diverse materials such as plastics, metals, and glass (as shown in Figure 7). Another innovative example is the nanocomposite coating crafted by Fu et al. [122], which boasts a double-layer structure consisting of a quaternary ammonium salt functionalized fluorinated copolymer and polyurea formaldehyde nanoparticle functionalized fluorinated copolymer. The amalgamation of fluorinated segments and quaternary ammonium ions lends the coating superhydrophobic and robust antibacterial properties.

One of the standout attributes of self-assembled thin-layer coatings is their eco-friendliness compared with that of traditional biocide-based coatings. They possess the ability to intervene early in the biofouling process, obstructing bacterial attachment and hindering the development of biofilms. With advancements in stability and performance across temperature ranges, these coatings have the potential to emerge as versatile antifouling solutions [123,124]. A noteworthy development in this domain is by Lipocoat, which has devised a repertoire of coatings exploiting these properties. Lipocoat's coatings are distinct in that they simulate biofilms with regenerative, antifouling, wetting, and lubrication properties through the chemical control of interface components. These coatings are non-covalently bonded to substrates, requiring no curing but possibly necessitating

surface pre-treatment. Their application is simplified through a dip-coating process, and their ultra-thin profile (approximately 5 nm) ensures negligible weight addition while preserving corrosion resistance.

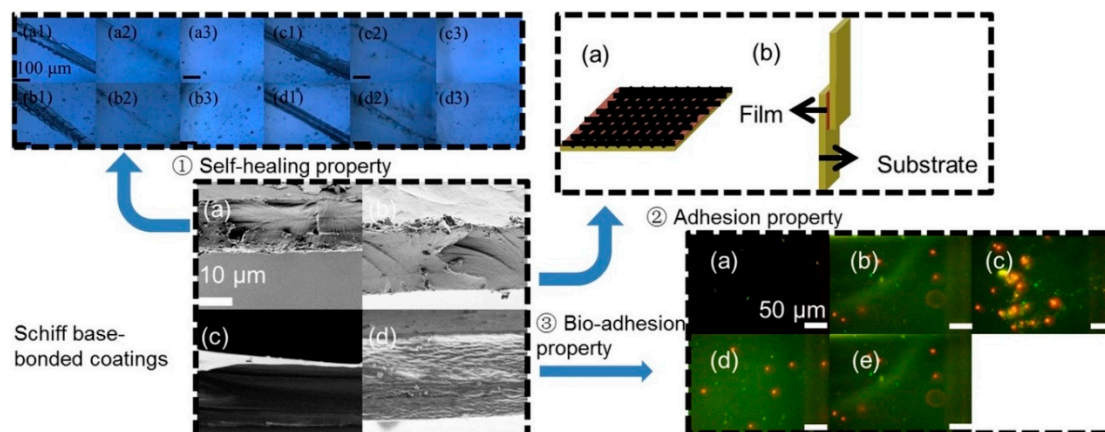


Figure 7. The principle of adherent self-healing chitosan/dialdehyde starch coating [121]. Figure reproduced with permission from *Colloids and Surfaces A: Physicochemical and Engineering Aspects* (Elsevier).

However, challenges persist, chiefly the long-term stability of these coatings. Those modified with polyethylene glycol, in particular, suffer from thermal instability, oxidation, and functionalization difficulties [125]. The grafted polyethylene glycol will lose its ability to repel proteins at temperatures >35 °C [126] or lose its resistance to proteins after a certain period, thereby losing its antifouling effect. These limitations have propelled research into alternative materials [127,128]. Strategies to enhance stability encompass the selection of more stable substances or polymer brushes and the utilization of polymers with inherently stable chemical structures and networks [129,130].

In summary, self-assembled thin-layer antifouling coatings offer unique advantages in terms of eco-friendliness and early intervention against biofouling. Lipocoat's innovative coatings exemplify these properties with their biofilm-mimicking characteristics. However, challenges persist, necessitating research and development efforts to improve the long-term stability of these coatings. Strategies include exploring alternative materials and enhancing stability through the selection of stable substances and polymer structures.

4.6. Slippery Liquid-Infused Porous Surface (SLIPS) Antifouling Coatings

Since 2011, when the concept of SLIPS was introduced [131], its potential for drag reduction, antifouling, and self-cleaning has garnered significant interest. SLIPS technology involves using a nano/microstructured substrate to hold a lubricating liquid, creating a stable and inert "smooth liquid film" on the material's surface. One approach involves injecting eco-friendly lubricants into a porous surface [132–134]. Yuan et al. [135] demonstrated this by injecting environmentally friendly lubricants into porous surfaces, as shown in Figure 8. They evaluated the coating's stability through contact angle and sliding angle tests and its antifouling performance by measuring the adhesion of different seaweeds. While silicone oil injection showed promising antifouling effects, algae coverage occurred over prolonged immersion. Notably, surfaces injected with grease evolved to a state where biofilms could easily be removed, revealing a fresh antifouling surface.

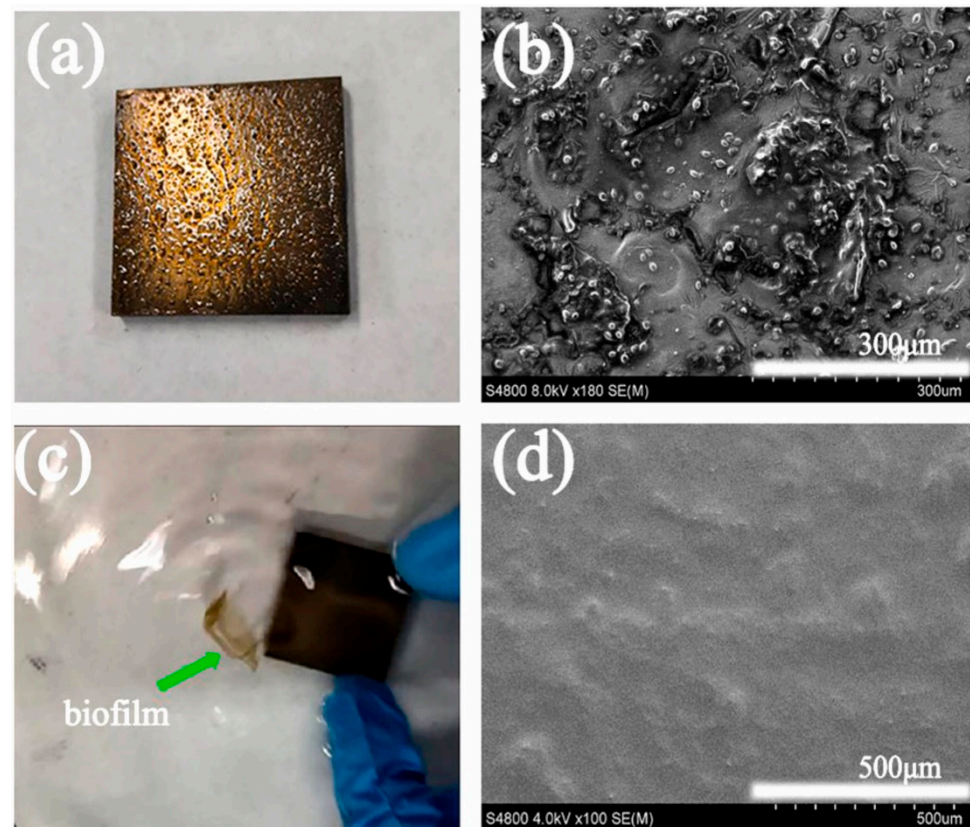


Figure 8. (a) Optical picture of the GIPS-Ca surface after being immersed in *N. exigua* solution for 24 days; (b) SEM image of the *N. exigua* biofilm on the GIPS-Ca surface after being immersed for 24 days; (c) The biofilm was separated from the surface after gently rinsing in water; (d) Re-exposed grease surface after the biofilm fell off [135]. Figure reproduced with permission from Colloids and Surfaces A: Physicochemical and Engineering Aspects (Elsevier).

Adaptive Surface Technologies estimates that SLIPS technology can reduce global energy consumption by approximately 27 million tons of oil and cut CO₂ emissions by approximately 107 tons annually. Following 2 years of collaboration with the US Naval Research Office, the team is now testing SLIPS technology at five biofouling-prone locations. The effectiveness of SLIPS has spurred increased investment and product development in this area. For example, Adaptive Surface Technologies has developed SLIPS Foul Protect, SLIPS SeaClear, and SLIPS Dolphin coatings for ships, which have exhibited remarkable antifouling abilities, as shown in Figure 9.

However, SLIPS antifouling coatings share a significant drawback with bionic antifouling coatings—they require additional energy to remove biofilms when heavily fouled, making them unsuitable for ships with extended docking periods. Future developments should focus on integrating the strengths of SLIPS with other dynamic and static antifouling coatings to overcome these challenges.

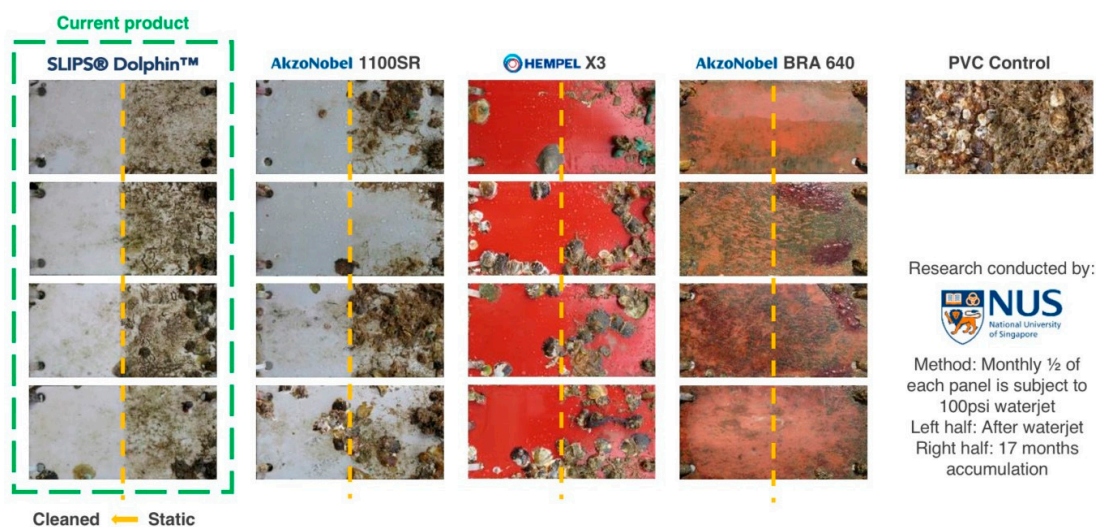


Figure 9. The comparison of SLIPS Dolphin coating. Image courtesy of Adaptive Surface Technologies Ltd. <https://www.adaptivesurface.tech/>.

4.7. Conductive Antifouling Coatings

In the 1990s, Mitsubishi Heavy Industries in Japan pioneered conductive antifouling coatings by integrating conductive agents into the coatings [136]. This innovation involved the generation of hypochlorite ions via seawater electrolysis through a mild electric current on the coating surface, thereby deterring fouling. Gelest, a subsidiary of Mitsubishi, advanced this technology by designing materials with varying conductivities, such as copper, aluminum, and molybdenum. These materials found applications in high-temperature scenarios.

Recently, the inclusion of carbon nanotubes and graphene as fillers has imparted conductivity to the coatings. Moreover, Gaw et al. [137] demonstrated that applying a low-voltage pulse to a conductive surface could create a hydrogen bubble layer that acts as an isolation barrier, preventing bacterial adhesion. This method proved highly effective, reducing 99.5% of surface bacteria.

Further, Zhang et al. [56] developed a structure using carbon nanotube polyvinylidene fluoride (CNT-PVDF) as a porous non-latching cathode, generating negative charges on the surface through capacitive charging. This structure hindered the attachment of certain organic substances. The key innovation of conductive antifouling coatings is their environmentally friendly nature compared with that of traditional copper-containing coatings, along with their prolonged antifouling effects. However, the complexity of the technology and the requirement of additional currents limit its wide-scale application [138].

Nowadays, Mostafaei A et al. [139] explore a new epoxy-based paint made from a blend of epoxy and conductive polymer polyaniline (PANI), along with an additive consisting of a PANI nanocomposite with ZnO nanorods. This coating demonstrates significant marine antifouling and antibacterial properties due to the presence of the emeraldine salt structure in PANI, which maintains surface pH at 4–5, and the generation of hydrogen peroxide by ZnO nanorods. Moreover, Huang et al. [140] present a biomimetic antifouling interface coating that mimics the antifouling properties of biological films and overcomes the low conductivity of traditional coatings. This coating uses a polyethylene glycol–Au gel as a support structure and electron transfer layer, covered by a hydration layer made from phospholipids and ampholytes. This innovative coating has low absorption in biological matrices and has been successfully used in multimodal clinical testing systems. Both studies represent significant breakthroughs in the development of antifouling coatings, offering new possibilities for practical applications.

In summary, conductive antifouling coatings, pioneered by Mitsubishi Heavy Industries, leverage conductive agents and electric currents to deter fouling. Recent advance-

ments have incorporated carbon nanotubes, graphene, and innovative structures to enhance conductivity and antifouling properties. Further research has explored new approaches, such as epoxy-based coatings with PANI and PANI-ZnO nanocomposites and biomimetic interface coatings with unique support and hydration layers. These developments contribute to the advancement of antifouling technology and open new avenues for practical applications in various fields.

4.8. Photocatalytic Antifouling Coatings

Photocatalytic antifouling coatings are based on titanium dioxide particles, which are semiconductors. When exposed to ultraviolet (UV) light, electrons move to higher energy levels in titanium dioxide particles (photocatalytic reaction), generating negatively charged electrons and positively charged holes [141,142]. They can react and form a strong oxidant on the surface to decompose any attached organic matter.

Titanium dioxide is a naturally occurring mineral and is considered harmless. The oxidant formed on the surface of titanium dioxide in the photocatalytic process has a short life span and will not pose a further threat to the environment [143]. This is because it uses the photocatalysis technology of sewage treatment as a basis. Researchers found that ship surfaces treated with titanium dioxide can inhibit the attachment of diatoms and bryozoan larvae. After 24 h cycle (14 h of light and 10 h of dark environment), more than 80% of larvae died before attachment. However, more importantly, after 48 h, the remaining larvae still attached to the material did not develop into viable larvae, proving that this type of coating had a strong inhibitory effect at the early stage of biofouling formation [144,145].

An important issue for this coating is whether the underwater UV intensity is enough to activate the photocatalytic titanium dioxide surface, especially in the shaded part of the hull. Therefore, scientists have chemically modified titanium dioxide so that it can be activated by visible light (in the blue region of the spectrum), which will improve its performance under poor light conditions. In addition to laboratory tests, researchers are conducting long-term field experiments on a large number of commercial coating formulations that have adopted photocatalytic technology [146,147].

Photocatalyst Coatings launched ecotio₂, a commercial marine coating based on the principle of photocatalysis. The coating has the following characteristics: (1) it converts light energy (from the sun or an electric light source) into chemical energy, which is transferred to water vapor to produce reactive oxygen species on the surface; (2) after the coating is excited by light, it will cause several reactions on the surface of the coating; (3) this light stimulus will change the surface of the coating, produce a purifying effect, and bring about self-cleaning; and (4) the coating is transparent and designed for various surfaces. The active ingredient, titanium dioxide, is insoluble in water. It is a safe substance that has been applied to various cosmetics and food additives. In brief, when the nano titanium dioxide coating (ecotio₂[®]) is exposed to light, it will decompose pollutants into harmless by-products, as shown in Figure 10.

Due to the environmental protection and high efficiency of the photocatalytic antifouling coating, it has now become a heavily researched topic in the antifouling field. Selim et al. [148] prepared an environmentally friendly, UV-visible, intelligent, silicon-rich, spherical TiO₂ nanocomposite suitable for ships. Through polymer solution pouring, nanofillers of different concentrations were blended into silicone nanocomposites for comparative research. It was found that the vinyl polydimethylsiloxane matrix at the enrichment end of the single crystal TiO₂ optical film enhanced photocatalytic activity. After UV-Vis radiation, the nanocomposites showed the best self-cleaning performance with 0.5% nanofiller. Scandura et al. [149] made a functionalized methyl-modified silica dry gel encapsulated by Bi₂WO₆ into an antifouling coating, which can effectively prevent biomass accumulation. The principle here is that when the coating receives light, it will react (Formulas (1)–(5)) to produce H₂O₂, thereby inhibiting the attachment and deposition of microorganisms. Kim et al. [150] synthesized carbonized fluorescent particles by acidic dehydration of catechol-q-poly dimethylamino ethyl methacrylate and made a coating with

TiO₂ as the photocatalytic active agent. The coating enhanced the absorption of photons and promoted the decomposition of organic pollutants on various substrates.

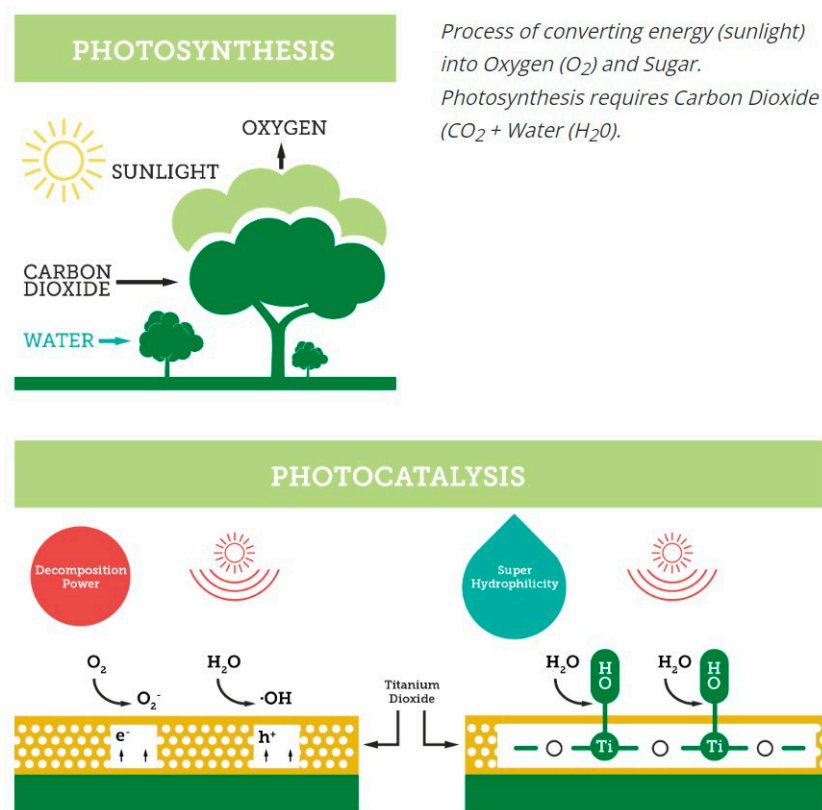
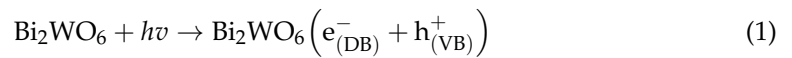


Figure 10. The principal of ecotio₂ coating. Image courtesy of Photocatalyst Coatings NZ Ltd. <https://photocatalyst.co.nz/>.

To further strengthen the advantages of photocatalytic coatings, Trávníčková et al. [146] investigated eight complex surfaces with different contents of TiO₂, alkoxysiloxane, and a hydrophobic agent and compared them to select the surface components that can inhibit the growth of photooxy filamentous Kerry algae. The results showed that photocatalysis combined with superhydrophobic surfaces during the early attachment of biological stains could effectively prevent a large number of fouling spores from gathering and forming a biofilm.

Although photocatalytic antifouling coating has many advantages, it needs to be exposed to strong UV light to have a good antifouling effect. While the ship is sailing, the UV light intensity received at the bottom is poor, which limits its large-scale commercial use.

5. Obstacles and Future Perspectives

5.1. Obstacles in Antifouling Coating Development

The journey towards the development of effective antifouling coatings is replete with challenges that span material science, chemistry, and environmental engineering. One of the foremost hurdles is the complexity involved in the production of micro/nanostructure coatings, which are often complicated, inefficient, and costly to manufacture. These limitations may be alleviated by leaning on advancements in polymer and laser processing, along with the potential of 3D printing technologies. Coupled with this are the concerns surrounding quasi-static water environments where existing coatings show suboptimal performance. A promising avenue to navigate this issue may involve the use of hybrid coatings, which amalgamate different technologies to enhance efficacy. Moreover, the current antifouling agents often lack broad-spectrum effectiveness, necessitating research focused on identifying more versatile, natural antifouling agents.

In terms of the long-term functionality of these coatings, the controlled release of antifouling agents is a pivotal factor that relies on a nuanced understanding of the underlying molecular mechanisms. Dynamic surface coatings, another subset, demand further refinement, especially in terms of degradation rate control and improving the substrate-coating binding forces. Adding another layer of complexity are transparent coatings, which are indispensable in specialized applications like sensors or optical equipment. Achieving high transmittance in these coatings without sacrificing their antifouling properties calls for extensive research and innovation.

5.2. Future Perspectives in Antifouling Coatings

Given the increasing challenges and considerations around marine environmental protection, the development of new antifouling coatings is inevitably moving toward eco-friendly solutions. Sustainable approaches are becoming central to the innovation process, as they not only meet antifouling requirements but also aim to mitigate long-term environmental impacts. This imperative for sustainability has catalyzed the development of multifunctional coatings, marking a new frontier in antifouling technology.

Some pioneering institutions are leading the charge, innovating coatings that meet not only superior antifouling requirements but also stringent environmental safety standards. These coatings can be engineered for additional functionalities like drag reduction. To enhance their utility further, there is an emerging trend of incorporating smart materials and sensors, providing real-time adaptability and extending both the lifespan and effectiveness of these coatings.

However, the actual applicability of these advancements hinges on extensive field testing. Laboratory conditions often fail to capture the spectrum of real-world challenges, including varying water conditions and material degradation over time. Looking ahead, the zenith of antifouling technology appears to be an integrated approach. The future likely holds a blend of various biological or non-biological coatings, smart materials, and adaptive technologies that will work in synergy to achieve breakthrough performance across a multitude of applications, all while maintaining an unwavering focus on environmental sustainability.

6. Conclusions

(1) Transition to eco-friendly solutions: With the refinement of various maritime regulations and the increasing emphasis on marine environmental protection, the development of antifouling coatings is increasingly leaning towards environmentally friendly formulations. Most of these coatings incorporate DCOIT (4,5-Dichloro-2-n-octyl-4-isothiazolin-3-one) to ensure long-lasting antifouling properties. However, DCOIT poses irreversible threats to marine ecosystems and aquatic life. Consequently, future work should focus on optimizing the biocide dosage in antifouling coatings and, more ambitiously, on the development of commercially viable antifouling coatings that are entirely free of biocides, to mitigate their environmental impact.

(2) Collaboration between academia and industry: As the global demand for sustainable, environmentally friendly, and efficient antifouling coatings increases, it is particularly important that academia and industry work together. Academic research provides industry with theoretical support and new coatings, while industry provides academia with field data and feedback through applied research and large-scale production. However, the development of new antifouling coatings in the laboratory can often overlook the cost of mass production, such as bionic microstructure antifouling coatings and amphoteric coatings. Cost is often a priority for industry. In general, future research and development of new coatings in the laboratory will need to focus on antifouling effectiveness, durability, ecological impact, ease of application and cost. In this way, the results of laboratory research can be brought to industry more quickly.

(3) Persistent challenges and future considerations: Currently, the development of novel antifouling coatings continues to face persistent challenges. For example, single antifouling agents lack potential for marine applications, biomimetic microstructure coatings are easily damaged in harsh marine environments, and there is a loss of the lubricating liquid in SLIPS surfaces, among others. A promising future direction would be the integration of advantages from multiple antifouling strategies, both biomimetic and non-biomimetic. For instance, combining the static antifouling advantages of self-polishing coatings with the dynamic benefits of biomimetic microstructure coatings could achieve a synergistic antifouling effect, extending the lifespan and reducing toxicity to the environment. Moreover, additional functionalities can be incorporated into antifouling coatings, such as corrosion resistance, anti-icing properties, and drag reduction, paving the way for multi-functional antifouling solutions.

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