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REVIEW ARTICLE

A systematic review on polymer-based superhydrophobic coating for preventing biofouling menace

Avinash Kumar, Vishal Mishra, Sushant Negi, Simanchal Kar

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Abstract Biofouling is a major issue for many industries including shipping, oil, and gas and can lead to accelerated corrosion, particularly for structures and components in and around salt water. Many efforts are undertaken to lessen its impact and financial losses. One of the promising methodologies is application of antibiofouling coatings to minimize biofouling, and the best results were observed with a superhydrophobic coating. Ample literature reviews on superhydrophobic coating have shown biofouling inhibition on the surface due to high wetting angles similar to the phenomenon on a lotus leaf. The hydrophobic coating can be deposited using multiple techniques such as electroless plating, chemical vapor deposition (CVD), sol–gel, and electrodeposition. In this review, an effort has been made to encompass such experimental work under a single domain and compare the effectiveness of each coating. In addition, mechanical properties and surface characteristics such as wetting angle, surface energy, and morphology were also discussed for various types of polymeric coating. Similarly, the application of nanoparticles such as ZnO, SiO_2, TiO_2, and CeO_2 was found to improve the substrate's mechanical properties, the durability of coatings, improvement in wear properties, adhesion, interlaminar cohesion, and increased wetting angle and above all, improve superhydrophobicity. These improvements are compared for various nanoparticle integrated coatings. In addition, other novel approaches to prevent marine biofouling by various polymer-based coatings such as superhydrophobic, foul-release, and foul-resistant coatings are discussed in this paper.

A. Kumar, V. Mishra, S. Negi, S. Kar (✉)
National Institute of Technology Silchar, Silchar, Assam
788010, India
e-mail: simanchal@mech.nits.ac.in

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Introduction

Biofouling is a natural mechanism of formation of undesirable bacteria, algae, and mussels on any structures and components of machines such as shipyard structures, underwater pipelines, underwater cables, and submarine hulls. Biofouling can accelerate corrosion, degrade structures, and reduce marine system efficiency, which can result in increased fuel consumption and CO_2 emissions. The earliest case of biofouling was reported in papyrus plants in 412 BC, and a mixture of arsenic and sulfur was used to minimize its effect.[1]In the present scenario, biofouling has emerged as a primary issue for structures and equipment used for military or commercial ships, onshore industries, and other marine engineering applications.[2–4] The undesirable adhesion of the marine species (flora and fauna) and its colonization over the substrate leads to the coining of the term called marine fouling. Microorganisms like bacteria, algae, diatoms, sponges, etc., and macro-organisms like mussels, balanus, barnacles, hydroid, etc., are two broad categories of water organisms responsible for marine fouling.[5–11] The shipping industry is considered one of the leading sources of the nation's income, contributing up to 90% in some countries.[12] Biofouling in ships causes tremendous damage to its components and thus promotes losses in the annual economy of the shipping industry. Marine fouling deteriorates the performance of the ship by affecting the ship vessel's surface, increasing the vessel's weight, reducing speed, causing poor ship mileage, and contributing to environmental pollution by emission of SO_2, NO_x, CO_2, and other harmful gases.[13–17] A schematic diagram of marine fouling is shown in Fig. 1. Considering the above parameters, there are two key routes that describe marine biofouling's growth. Initially, when the substrate is immersed in the seawater and gets wet, organic carbon residue (OCR) will immediately adsorb on the substrate's wet surface, generating a conditioned film in a few minutes. The composition of these OCRs solely depends upon the fulvic acids, humic acids, glycoproteins, and ions accessible in the liquid form. After a few hours under electrostatic and Van der Waals force, microbes get adsorbed on the conditioned film to form a biofilm. The adherence of microbes on the conditioned film is promoted by sedimentation, water flow, convective and Brownian motion.[18–21] Finally, after a week of activity, some single-celled algal spores, protists, and marine biological larvae adhere over the biofilm surface to provide nutrition to increase their population to create a vast biological fouling community.[22] The attached organism colonizes onto the ship's hull surface thereafter increasing the ship's drag, reducing the ship's speed, and increasing the hydrodynamic weight of the vessel.[23] For more than 2000 years, humans have been fighting with marine fouling. Early generations used biocidal compounds which are capable of killing fouling organisms to prevent colonization. The developed biocidal compounds vary from simple Cu to Pb sheets on wooden boats for antimicrobial coatings containing arsenic, mercury, and copper on marine hulls. Copper was a popular and effective biocide, but it was only effective for two years. The researchers started to work on antifoul technology to attenuate the harsh effect of biofouling. Spirit-based vanish paint was initially applied to the bottom of the vessels in 1908 to prevent biofouling. Ships were painted with mercuric oxide disseminated in turpentine and alcohol-creating paint, with a nine-month durability rating. TBT (tributyltin)-based polishing and coating technique was developed in the twentieth century, composed of TBT acrylate ester, which acts as an antifouling agent.[24,25] Unfortunately, it was observed that the toxic nature of TBT causes severe damage to aquatic life. After that, many antifouling paints and wax replacing TBT were developed, which inhibit biofouling growth on the substrate.[26,27] The academic and industrial researchers started their research to find the most economical, ecological, and nontoxic coating that can replace the traditional method. The coating techniques were designed based on two prime objectives. First, it is necessary to develop an antifouling strategy to prevent marine organism attacks and secondly, to design a mechanism to create a fouling release strategy to lessen the adhesion force after an attack.[28] Superhydrophobic coating is one of the durable and efficient methodologies recently developed for such purpose.[29] Superhydrophobic surfaces have recently attracted a lot of attention due to their efficacy in the fields of self-cleaning, antifouling, water repellence, and antisticking properties.[29–34] The term superhydrophobicity has been known since the 1940s,[35] but the concept of superhydrophobicity inspired by nature has been utilized in the past decade. This nature-inspired surface consists of a micro–nano-structure and nano-enclosed pores that can repel the adhesion of oil, protein, and bacteria to prevent the surface from biofouling.[36–38] The bio-mimicry of this natural phenomenon helps to suppress biofouling.[39,40] Fouling bacteria or organisms may migrate with ships and could lead to bio-invasion when it reaches different sea water without natural opponents.[41] In the Black Sea, fouling from the jelly comb *Mnemiopsis* arrived in the 1980s that endangered the existence of native anchovies. Hence, the International Organization of Maritime (IOM) circulated the resolution to diminish the impact of biofouling bio-invasion. The corrosion of surfaces can also be exacerbated by biofouling. The accumulation of fouling bacteria on surfaces accelerates regional variations in these types and deliberations of pH values, oxygen levels, and ions which can cause coatings to degrade, increase the conductivity of the liquid, and encourage electrochemical and chemical processes.[42,43] This type of corrosion is known as biocorrosion or microbial-influenced corrosion (MIC).

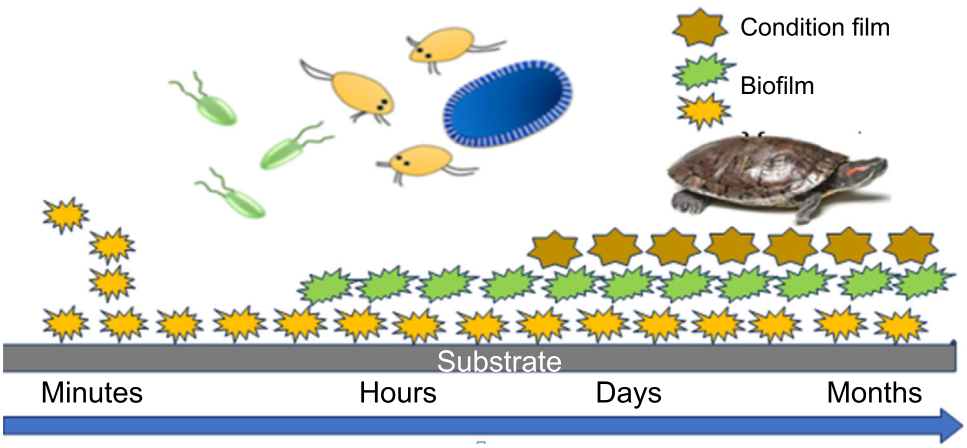

Fig. 1: Graphical representation of the marine fouling method depicting the timeline and causes

MIC accounts for 20% of the corrosion in aqueous systems.[44] Marine corrosion can cause minute surface fissures, much like the proliferation of cancer cells. Over time, this can culminate in large-area corrosion, which weakens the surface and poses major security risks. Every year, the ''cancer of ships'' causes tremendous economic and financial damages that create global problems. According to the National Association of Corrosion Engineers (NACE) survey, the annual cost of corrosion around the world is 3.4% ($2.5 trillion) of global GDP.[45] Thus, reducing the effects of marine biofouling and corrosion is crucial for the growth of the worldwide maritime sector. The previous research articles have suggested that biofouling and corrosive behavior under different field testing like underwater, seawater, river water, and chemical solution have affected the performance of coated materials. Moreover, surface coating methods have recently demonstrated promise as affordable and effective ways to shield subsea surfaces to prevent corrosion and biofouling. Although conventional coatings often serve just one purpose, such as antifouling or anticorrosion. Therefore, merging anticorrosion and antifouling properties into hybridized coating has excellent potential to mitigate fouling.

This review paper compares various environmentally friendly, efficient, and long-lasting antifouling methods for marine applications. Fouling-resistant polymer coatings, superhydrophobic coatings, and fouling-release polymeric coatings are the three main coating methods that have been discussed briefly. Further, the study also envisages the impact of various metal oxide nanoparticles, such as ZnO, SiO_2, TiO_2, nanorod, and others, on the design of superhydrophobic surfaces. Wenzel, Cassie–Baxter, and Young's wetting models are a few analytical models used to determine the appropriate sliding angle and water angle of contact for the superhydrophobic surface. Overall, this review will provide a brief insight to the reader about biofouling's effect and prevention methods.

Methodologies for preventing biofouling

Nature-inspired biomimetic coatings

A methodical replication of biological features on a surface is percieved to give a proficient, efficient, and eco-friendly method to prevent biofouling. Researchers have created and used this nature-inspired concept for engineering applications.[46,47] Biofouling is primarily influenced by surface characteristics such as wettability and microtexture.[48–51] Wettability impacts fouler colonization, ranging from water-loving hydrophilic to water-repelling superhydrophobic. Surface roughness and surface energy are the two characteristics that govern wettability. Siloxanes, stearic acid, and fluorocarbons are the most frequent materials used to reduce surface energy. The concept of superhydrophobicity was inspired by nature, with a WCA of more than 150° and a sliding angle below 10° to reduce the slippery and sticky properties of the surface against water.[52–56] Superhydrophobic surfaces, such as lotus leaves,[57,58] butterfly wings,[59,60] peanut leaf,[61] red rose petal,[62,63] water strider legs,[64] and fish scale,[65,66] have low surface energy to resist wetting. Tables 1 and 2 provide various examples of nature-inspired flora and fauna, as well as their mechanisms which help to understand the superhydrophobic concept from nature and inspire engineers to use concepts of superhydrophobicity to prevent biofouling for engineering applications.

Table 2 presents an example of nature-inspired fauna such as shark fish, dog fish, stone fish, butterflies, blue mussels, dragonflies, pilot whales, and doves. They are mainly found in nature with superhydrophobic characteristics, low surface free energy, and ridge microtexture, which helps them swim on the water's surface. This nature-inspired fauna concept may be helpful for researchers to apply superhydrophobic concepts in engineering materials to prevent biofouling on the ship's hull. The most frequent fouling organisms found at the test sites of the samples are bryozoans, barnacles, calcareous algae, bivalves, tubeworms, and hydroids. The surface can be classified as hydrophilic, hydrophobic, or superhydrophobic based on the constructed contact angle. The varying contact angles of wetting properties are depicted in Fig. 2. Bacterial attachment to ship hulls is reduced due to the low surface energy. It was created to minimize interactions with biomolecules by removing strong polar interactions (like hydrogen or ionic bonding). Due to uneven rough surfaces, the superhydrophobic surface can be beneficial. Air bubbles are trapped on the surface of hills and valleys, consisting of micro–nano-structures and nano-enclosed pores, increasing the water contact angle. Based on the Cassie–Wenzel hypothesis, researchers investigated the chemistry of nonstick and antiadhesion coatings.[83–86] The Wenzel model is applicable for homogenous surfaces having the equation:

$$\cos\theta_w = r\cos\theta \quad (1)$$

where θw = Wenzel angle of contact, θ = Young's angle of contact, r = Surface roughness factor.

For heterogeneous surface Cassie and Baxter's model is applicable by the equation:

$$\cos\theta = f_1\cos\theta_1 + f_2\cos\theta_2 \quad (2)$$

where θ = Baxter and Cassie angle of contact, f_1 = contact area ratio where solid is in contact with liquid, f_2 = contact area ratio with air packets that confine the inner side of surface cavities

Xie et al.[87] fabricated a ZnO/Acrylic superhydrophobic surface on an aluminum substrate via spraying and co-curing to prevent marine biofouling. They obtained a rolling angle of 1.8° and a WCA of

Table 1: Antifouling mechanism inspired by natural flora

S. No	Type	Mechanism	References
1	Lotus	Superhydrophobic self-cleaning surface	57
2	Jewelweed	Hydrophobic surface	68
3	Water fern	Superhydrophobic surface	69
4	Eelgrass (Zosterra marina)	Zosteric acid secretions	70
5	Lady mantle (Alchemilla mollis)	Hydrophobic surface	71
6	Broccoli (Brassica oleracea)	Superhydrophobic surface	71
7	Red Seawood (Delisea pulchra)	Bacteria message manipulation	72
8	Coralline algae (Porhyridium purpureum)	Chemical secretion and shedding	73
9	Seawood (Ulva lactuca)	Hydrophobic surface	74

Table 2: Antifouling mechanism inspired by natural fauna

S. No	Type	Mechanism	References
1	Shark fish (Squalus acanthias)	Low drag, flexion of mucous and scales	75
2	Dog fish (Scyliorhinus canicula)	Ridge microtexture surface	76
3	Stone fish (Synanceia horribilis)	Skin sloughing	77
4	Butterfly (Polyommatus icarus)	Superhydrophobic wing	78
5	Blue mussel (Mytilus edulis)	Microtexture and filter feeding	77
6	Dragon fly (Libellula), Mayfly (Hexagenia)	Hydrophobic wax-covered wings	79
7	Pilot whale (Globicephala melas), Dolphin (Delphinus delphis)	Surface energy and microtexture	80, 81
8	Birds dove (Zenaida), Pigeon (Columba)	Superhydrophobic feathers	82

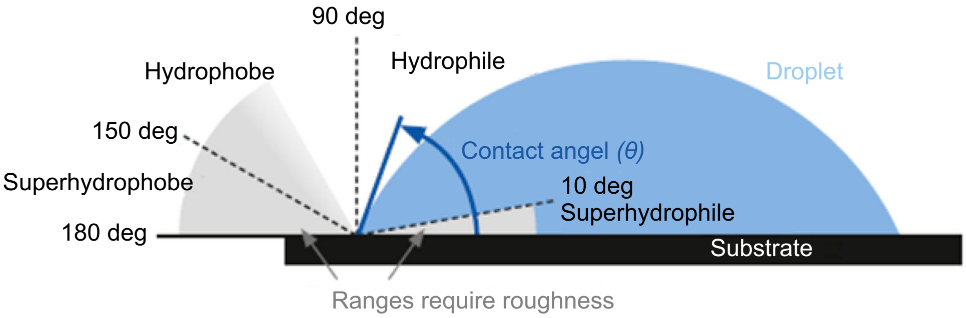

Fig. 2: The contact angle of wetting properties[67]

171°, indicating a superhydrophobic surface after coating. SEM images of the sample shown in Fig. 3 confirmed the results of the antibiofouling test after 150 days in a chlorella-inoculated growth. In Figs. 3a and c, a few chlorellae were adsorbed on zinc oxide pure acrylic polyurethane superhydrophobic coating, indicating it suppresses biofouling. There was a considerable amount of chlorella with a diameter of 5 μm, as indicated by the red circle in Figs. 3b and d, adsorbed on the pure acrylic polyurethane (PUA) coating.

Sun et al.[88] generated an SHS on a stainless steel substrate using a picosecond laser texturing approach. After five weeks of immersion in seawater, a specimen with SHS found considerable antibiofouling impact on a substrate compared to bare stainless steel plates. Figure 4a and b shows a microgroove and micropit array on the surface indicated by the red rectangle that suppresses biofouling.

Selim et al.[89] developed a superhydrophobic PDMS/SiC nanowire composite for marine applications. It was observed that there was an excellent antibiofouling property due to micro/nanoscale roughness on their surfaces. The SiC nanowire with (0.5 wt%) showed excellent hydrophobicity with a WCA of 153° and a low surface free energy of 11.25 mN/m after three months in seawater. Selim et al.[90] created superhydrophobic Si/MnO$_2$ nanorod composite coating on

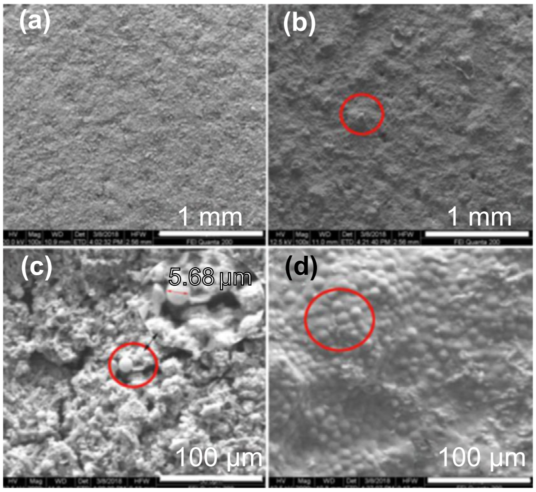

Fig. 3: SEM images of samples after 150 days: (a and c) indicates zinc oxide pure acrylic polyurethane while (b and d) indicates pure acrylic polyurethane coating[87]

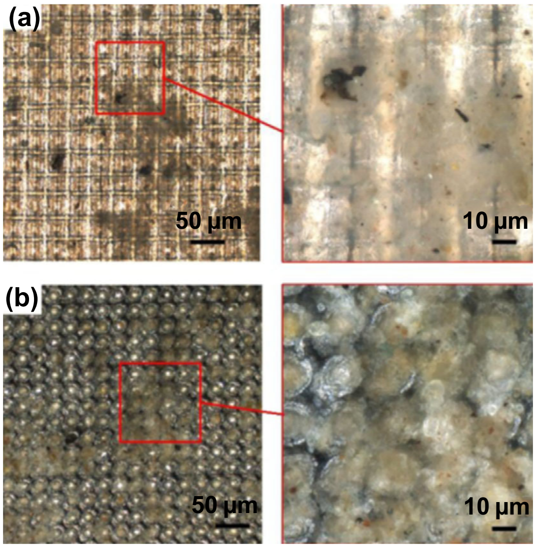

Fig. 4: (a) SEM image of microgroove and (b) Micropit pattern of SHS after 5 weeks[88]

sample and submerged them in seawater for 90 days in the same year to attenuate marine fouling, as illustrated in Fig. 5. It is noticed that uniformly scattered nanorods (0.5 wt%) have a superhydrophobic nature. The nanorod has a WCA of 158° and surface free energy of 12.65 mN/m. This results in an excellent eco-friendly foul-release coating for ship hulls.

Selim et al.[91] considered an in situ approach to produce (PDMS)/Al_2O_3 nanorod composites for marine antibiofouling and found that uniform distribution of Al_2O_3 nanorods inside the PDMS resin gives improved fouling repellence with WCA of 169° and surface free energy of 10.03 mN/m. Similarly, in another study, He et al.[92] used a spin-coating approach to create a superhydrophobic surface using PDMS precursor and silicon dioxide. It is reported that the addition of SiO_2 nanoparticles increased the WCA from hydrophobic (106.8 ± 1.2°) to superhydrophobic (165.2 ± 2.3°) property. This is owing to the presence of micro–nano-scale features of SiO_2 which aids to the increase in superhydrophobicity resulting in slipping of organisms from the substrate surface. An et al.[93] used PDMS coated with fluoroalkyl silane and cerium dioxide (CeO_2) nanoparticles to form a superhydrophobic composite coating with a rough texture. The results showed good superhydrophobicity with a water contact angle of (161 + 2°) and a sliding inclination of (4 + 1°). CeO_2 nanoparticles also provided adequate corrosion protection for metal substrates even in adverse conditions due to their air cushion and corrosion hindrance.

Fouling release coating

Fouling release (FR) coatings are considered environmentally acceptable antifouling coating methods since they have low interface energy and elastic modulus. Low elastic modulus detaches hard foulants like barnacles, and low interface energy reduces relative adhesion.[94–97] The low surface elasticity makes the coated surface flexible or dynamic and creates difficulty for microorganisms to settle on it. It can be described further by the dynamic surface antifouling (DSA) mechanism.[98–102] FR does not get dissolved or decompose in seawater and does not release any hazardous chemicals or gases. They weaken the bond between the substrate and the fouling organism, therefore causing easy removal of the microorganisms with a simple cleaning approach.[103,104] All fouling organisms will fall off automatically when a ship's hull is coated with fouling release coatings, allowing the ship to travel at a speed of 10 to 20 knots in usual conditions.[105] Fluoropolymers and polysiloxanes are now widely utilized fouling-release coating materials to prevent biofouling. Fluoropolymers are nonpolar, with a low surface tension of 10–20 mN m^{-1} to confer the hydrophobic nature of the surface.[106] However, fluoropolymer cannot cover large parts of a ship's hull effectively because it has various defects, including limited structural mobility, which limits fluorine stiffness and rotations in the entire polymer matrix, making fluorine monomers expensive.[107] Polysiloxanes are cheaper than fluoropolymers and have superior physicochemical and mechanical qualities. Polysiloxanes are used in a variety of applications ranging from space to marine, with a market share of 17.2 billion dollars in 2017.[108,109]

Milne was the first to propose using polysiloxanes as antifouling agents.[110] Polysiloxanes have a smooth surface and stable chemical characteristics and are resistant to corrosion. However, siloxane chains have low bonding strength as well as poor mechanical qualities, and also they get easily torn and detached from the substrate when subjected to a dynamic

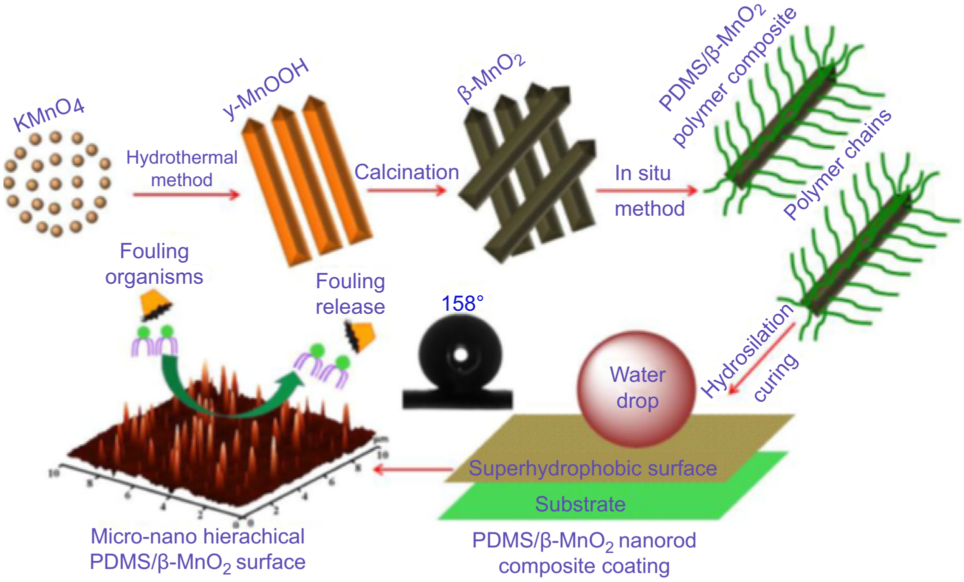

Fig. 5: In situ synthesis of PDMS/β-MnO$_2$ nanorod composite coating[90]

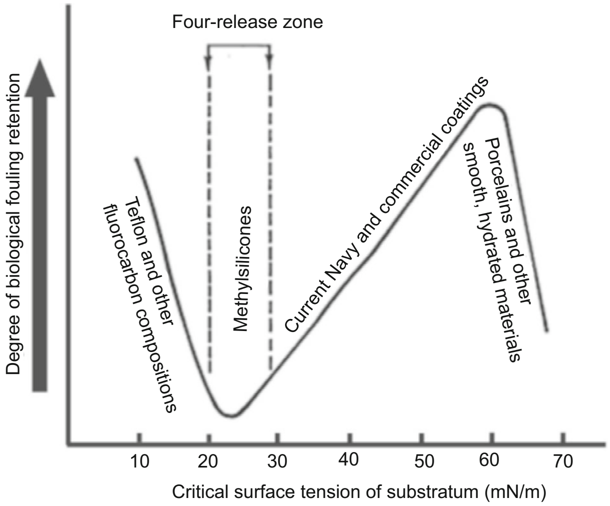

Fig. 6: Baier Curve[112]

environment.[111] Polysiloxanes exhibit poor antifouling performance in static conditions. PDMS (polydimethylsiloxane) with low surface energy and small Young's modulus is more efficient than polysiloxanes. PDMS is employed as a marine fouling release because it fits in the fouling release zone criteria of the Baier curve. In the late 1960s, Baier gave the correlation between polymer surface properties and relative adhesion of fouling organisms, as shown in Fig. 5. According to the curve, relative adhesion is lowest when surface energy is 20–30 mN m^{-1}, often known as the fouling release zone. Moreover, PDMS meets these conditions as it reduces microfouling settlements on the coated surface.[112–114] Nontoxicity, cost-effectiveness, good heat resistance, nonleaching property, and long-term durability against ultraviolet irradiation are all advantages of PDMS coatings.[115] However, its main drawback is poor mechanical potential, which can be enhanced by adding nanofillers (Fig. 6).

Cavas et al.[116] designed a PDMS-based FR coating with MWCTs and graphene oxide (GO) reinforcement. The antibiofouling performance and mechanical properties of such nanocomposite coating were improved with 0.5 wt% reinforcing elements. The characteristics of PDMS with varying wt% of multiwall carbon nanotubes (MWCNT) were investigated by Koumoulos et al.[117] They discovered a steady diffusion of carbon nanotubes in the PDMS matrix which improved the composite's surface and mechanical properties. As the wt% of (CNT) gets increased, the surface's mechanical capabilities get reduced due to agglomeration of CNT in the polymer matrix, whereas the hydrophobicity of the material gets enhanced. Carl et al.[118] investigated the incorporation of TiO$_2$ and carbon nanotubes (CNTs) in PDMS to ameliorate (FR) characteristics against the mussel. Nanofillers make larvae difficult to adhere onto the surface and reduce adhesive bonding. As per the investigation, the dispersion of TiO$_2$ and CNTs also improved the mechanical characteristics of the coatings. Roy et al.[119] used octa-methyl-cyclo-tetra-siloxane to make a PDMS-sepiolite nanocomposite. The findings revealed that the consistent interaction of PDMS-sepiolite with the polymer matrix improves the polymer's thermal, dynamic, and mechanical properties while reduces the relative adhesion of fouling communities.

In another study, Irani et al.[120] produced a PDMS coating with MWCNTs varying wt% of 0.05, 0.1, and 0.2. As nanotubes were applied in small amounts, the surface chemistry of the coating gets changed, therefore significantly improving fouling release properties

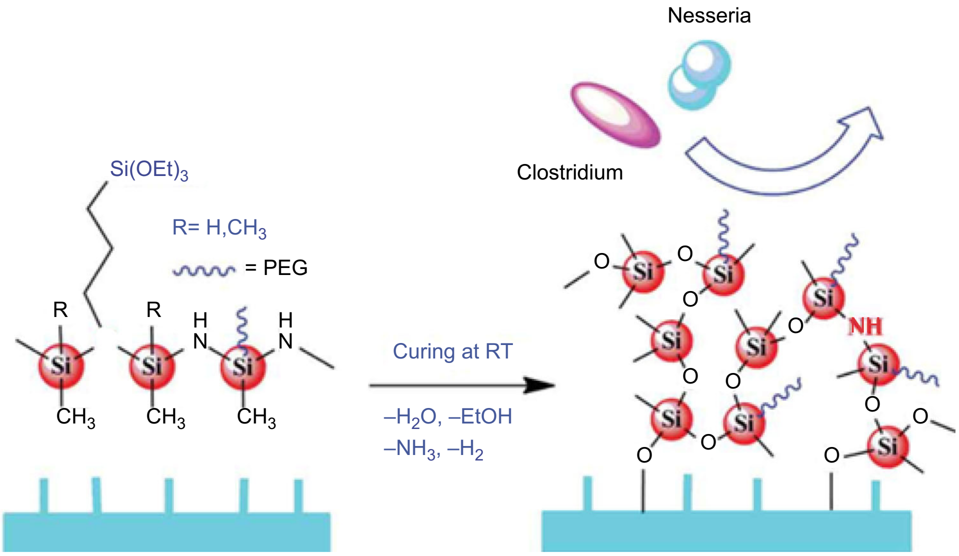

Fig. 7: PEG grafted network for marine biofouling[133]

and lowering adhesion strength by 67% compared to the uncoated sample. Yang et al.[121] developed FR coating by reinforcing alumina in PDMS containing phenyl methyl silicone oil. Upon testing it was found that inclusion of alumina significantly improved the coating elastic modulus and shore hardness but sacrificed the antifouling performance. Zhao et al.[122] used an epoxy silicone prepolymer combined with PDMS to make epoxy-modified silicone. The coating's adherence and mechanical properties are improved by changing epoxy to silicone. Ba et al.[123] used phenylmethyl silicone oil to create an FR coating on PDMS (PSO). The addition of PSO improved the coating's antifouling capabilities by increasing its hydrophobicity and reducing its elastic modulus. Padmavathi et al.[124] used a wet chemical precipitation process to make CuO nanoparticles, which were placed into a PDMS FR surface to improve micro–macro-fouling. The findings suggested that incorporating nanofillers into a PDMS matrix could be a feasible antifouling method to prevent algal fouling and larval colonization by imparting toxicity at the surface through metal ion release. Seo et al.[125] proposed an environmentally friendly oleamide-PDMS copolymer (OPC)-based coating for antifouling and drag reduction. It is reported that algae spores and mussels could not adhere to the developed OPC surface due to its slippery characteristics. The suggested OPC surface coating can be used in various commercial products, such as biomedical equipment, marine vehicles, and water management.

Fouling-resistant coatings

Algae, bacteria, and protein discharge from marine species can be prevented or inhibited by fouling-resistant coatings.[126–128] It is used mainly to modify the surface of marine components to prevent unwanted adherence. It involves high interfacial hydrated surfaces with a strong water network that inhibits foulants from adhering and is usually formed by hydrophilic polymers like polyethylene glycol (PEG) and zwitterions.[129] However, they swell in maritime environments, resulting in poor mechanical qualities that limit their usage.

Polyethylene glycol (PEG)-based polymers

PEG is a nontoxic, extremely hydrophilic, and neutrally charged compound. It is used for coating because of its antifouling solid potential to minimize unwanted protein adsorption and cell adhesion. By forming hydrogen bonds with water, PEG lowered interfacial energy and inhibits the adhesion of larvae and spores.[130,131] PEG decomposes on the coated surface due to autooxidation in the presence of oxygen and transition metal ions.[132]

Perrin et al.[133] produced an antifouling coating by using a hydrosilylation procedure to covalently graft PEG chains onto silazane polymers, as illustrated in Fig. 7. The PEG-grafted coatings are very resistant to Gram-negative *Neisseria* sp. and Gram-positive *Clostridium* sp. bacteria compared to the pristine polysiloxane surface. Similarly, Leckband et al.[134] described two methods for creating PEG layers on silicone surfaces, focusing on protein adsorption with the macroscopic surface and adjusting the molecular interaction between adsorbing proteins and the surface. The modified surface shows antifouling properties, which can resist massive protein molecules. Kingshott et al.[135] found that PEG layer grafting reduces Gram-negative bacteria adhesion and prevents protein adsorption and consequent biofouling. Yang et al.[136] used the Michael addition reaction to make

PEG-based hydrogels by incorporating polycarbonate. These hydrogels were grafted on silicon rubber to evaluate *S. aureus* antibacterial activity for a day. The results demonstrated the presence of different *S. aureus* on the rubber surface without coating. After coating, nothing was found on the surface, indicating that coating prevents surface fouling. Zhou et al.[137] combined PEG of various molecular weights with alkyl alcohol to create amphiphilic surfactants. The results obtained show an amphiphilic-coated surface can be employed for antifouling and foul-release coating, potentially paving the way for a fluorine-free, ecologically acceptable foul-release polymeric coating. Due to ether linkages, PEG is chemically unstable and prone to degradation. In contrast, zwitterionic polymers are chemically stable and most effective for fouling applications to attach water molecules through ionic interactions.

Zwitterionic polymers

PEG can be replaced with zwitterionic polymers. Zwitterionic, unlike amphiphilic PEG, is a neutral molecule with both positive and negative charges in its structure, making it very hydrophilic.[138,139] It has been identified as a promising antifouling material due to its ability to build a hydration shell via electrostatic interactions, stronger than hydrogen bonds.[140,141] Nonspecific adsorption at the solid/liquid interface can be reduced thin or thick coating of zwitterionic compound films.[139] These materials are chemically stable and inexpensive. Due to its remarkable antifouling properties, it has been used to develop antifouling surfaces for biosensors, medical devices, and marine coatings applications.[142,143]

Dai et al.[101] created antifouling copolymers incorporating hydrolysis-induced zwitterionic monomer tertiary carboxybetaine triisopropylsilyl ester acrylate (TCBSA) copolymerized with methyl methacrylate (MMA). It was noticed that the surface is resistant to marine bacteria, algae, and diatoms in terms of antifouling and protein resistance. Zhang et al.[144] employed atom transfer radical polymerization (ATRP) to graft zwitterionic sulfobetaine methacrylate (SBMA) on glass surfaces to create a polymer layer. Zwitterionic polymers completely prevent spores from settling and reduce adhesion as well as form biofilm in bacteria. Zwitterionic polymers have weak mechanical properties and high water absorption and are not directly employed as homopolymers. Wu et al.[145] developed copolymers with zwitterionic side chains attached to the glass substrate via a reverse addition-fragmentation chain transfer (RAFT) polymerization technique to solve this problem. The study has outlined a simple but effective process to produce nonfouling antifouling surfaces. Niu et al.[146] designed copolymers with adhesion and antifouling properties for surface modification by combining the dopamine methacrylamide (DMA) and the zwitterionic monomer 2- methacryloyloxyethylphosphocholine (MPC). It was found that copolymer-modified substrate has outstanding antifouling performance in resisting protein adsorption and repelling oil attachments.

Role of inorganic nano-fillers in preventing biofouling

Incorporating inorganic nano-fillers is often advantageous to the coating mechanical properties such as wear and scratch resistance and has more potential to prevent biofouling. It increases cost savings while improving mechanical, self-cleaning, and lotus effect properties. [147,148] Self-cleaning nano-surfaces could achieve fouling prevention on ship hulls with the lotus effect, superhydrophobic surfaces with greater WCA, and low surface-free energy parameters.[149] Selim et al.[150] used an in situ approach to create a polymethyl/ZnO nanorod composite for a superhydrophobic surface and foul release (FR) antifouling coating surface. The rough topography inhibits water from pervading the surface. Including 0.5 wt% ZnO nanofillers in the silicone matrix promotes nonwettability and surface roughness and reduces surface-free energy. More significantly, nano-filler concentrations (up to 5 wt%) result in low water and fouling repellence due to the clustering of nano-fillers, which generates gaps, pinholes, and crevices on the matrix surface. It affects coating adherence and performance against marine organisms. The study developed a cost-effective and environmentally friendly alternative for marine antifouling coatings using nanofillers at a concentration of 0.5 wt%. They offer favorable FR characteristics with a maximum WCA of 158° and minimum free energy of 11.25 mN/m. The bare silicone and dispersed ZnO NR composites (0.5–5 wt%) were tested in sea water for six months. The results are presented in Fig. 8 which shows high fouling prevention at 0.5 wt% due to low free surface energy and homogenous filler dispersion.

Arukalam et al.[151] coated Q235 steel with a perfluoro-decyl trichlorosilane (FDTS)-based hydrophobic polydimethylsiloxane (PDMS)-ZnO nanocomposite coating to achieve antifouling and anticorrosive properties. Due to their antibacterial characteristics and hydrophobic nature, ZnO nanoparticles provide antifouling and minimize hydrophobicity. The surface energy of the coating was modified via FDTS, which reduced the contact angle while enhancing antibiofouling and anticorrosive efficacy. Because the trichloro group of FDTS grows and forms a hydroxyl bond on the steel surface, the adhesion strength of Q235 steel improves. Zhang et al.[152] successfully used a simple dipping approach to fabricate superhydrophobic TiO_2 nanowire coating. The coated surface has antifouling properties for organic solvents with a low boiling point. The TiO_2 nanowire allows damaged coating surfaces to be easily regenerated and repaired.

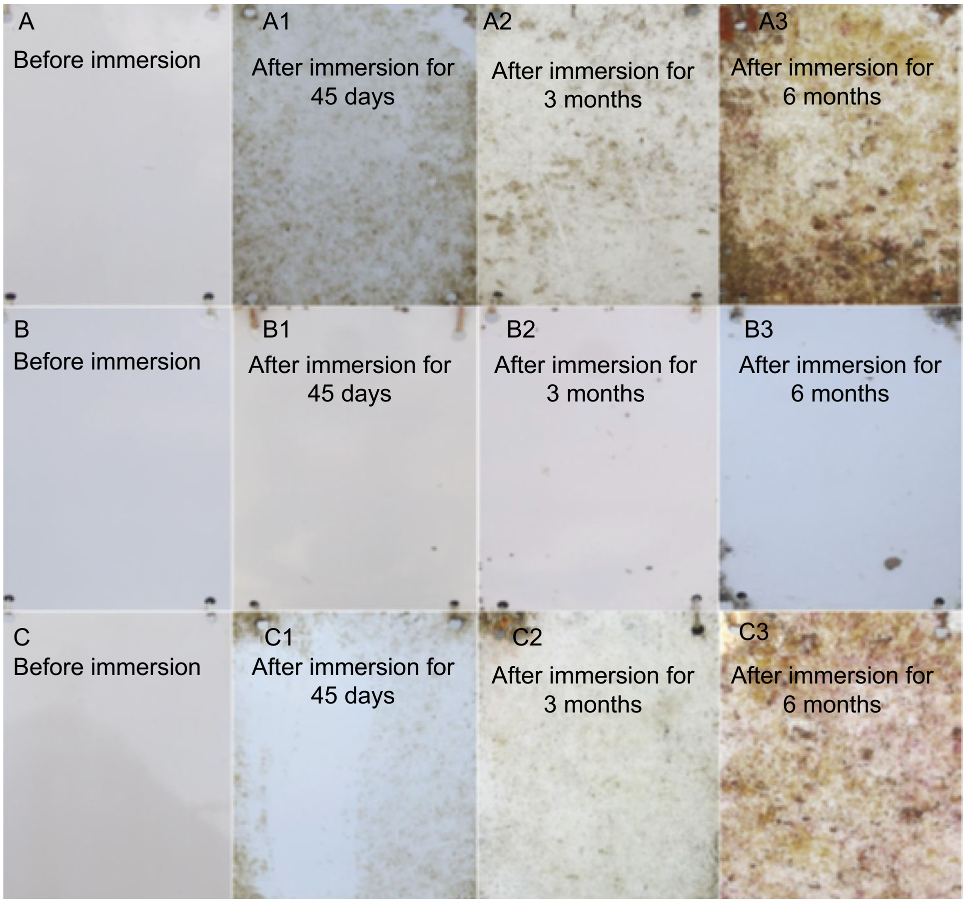

Fig. 8: (A, A1, A2, and A3) show foul release coating of bare PDMS; while (B, B1, B2, and B3) show (0.5 wt% NRs) for silicone/ZnO nanocomposites and (C, C1, C2, and C3) show (5 wt% NRs) silicone/ZnO composite coating immersion in seawater for six months[150]

Using fluorinated silica (F-SiO_2) particles as raw material, Liu et al.[153] designed an inorganic/organic composite coating with good mechanical features. The addition of F-SiO_2 to composites results in a 27.7% drag reduction. The application of graphene for preventing marine biofouling is reviewed by Jin et al.,[154] who observed that graphene incorporation on coated surfaces improved mechanical strength and antifouling characteristics, and created superhydrophobic surfaces that are environmentally friendly.

Anticorrosive paints to prevent biofouling and corrosion

The depletion and failure of structures and components are caused by physical, chemical, and certain biological phenomena known as wear, erosion, and corrosion. Several intricate elements frequently contribute to marine corrosion. For instance, rapid liquid flow and sand impact on surfaces cause erosion and corrosion, and high-speed propeller rotation causes rusting due to cavitation. In sea water, different salts, such as magnesium chloride, potassium sulfate, magnesium sulfate, and sodium chloride, are present that significantly increase corrosive behavior due to the increment of the electrical conductivity of seawater. This phenomenon has been observed because the dissolved salt conducts more ions in seawater, releasing positively charged cations and negatively charged anions. Furthermore, under the influence of waves and seawater scouring, the surface seawater is sufficiently rich in CO_2 and O_2 to touch the metal surface thoroughly, enhancing the chemical and electrochemical processes between the metal and seawater.[155]

Anticorrosive paint on superhydrophobic surfaces is another superficial method to prevent corrosive behavior and avoid fouling. Anticorrosive paint is a system developed in which different layers are coated using various coatings processes to influence the corrosive properties.[156] These individual coatings can be metallic, inorganic, or organic, consisting of primer, topcoat, and one or several intermediate coats, preventing high material corrosion in marine environments. Nurani et al.[157] coated ($20 \times 25 \times 0.3$ cm^3) mild steel specimen with anticorrosion and antifouling paint. Before coating, the mild steel specimen was sandblasted. The test specimens were immersed in splash and tidal zone (0, 1, and 3 m from sea level) for a month, considering various seawater parameters. It was found that after antifouling coating, no fouling and corrosion took place, whereas the bare mild steel sample got corroded by 17 MPY. Similarly, in other studies, proactive water grooming was used to improve the performance of ship hull antifouling coating. Before coating, the surface was groomed (cleaned) to free it from fouling. Furthermore, an ablative copper antifouling coating, silicone fouling release coating, epoxy coating, and coating with PTFE solid sheer upon sample underwent exposure to static seawater immersion situated at East West of Florida for 120 days and grooming intervals of 3, 6, 12, and 24 days. It was found that grooming reduces the biofilm development on ablative copper-coated samples.[158] Various laboratory tests can be performed on the samples to analyze the corrosion behavior on coated samples, such as the physical, chemical, and acid–alkali tests. The physical examinations determine the degree of adhesion, hardness, and flexibility. The chemical tests are based on the presumption that the pace at which paint pigments dissolve in artificial aging solutions in the laboratory accelerates at a rate similar to that of seawater. The acid–alkali test depends on hydrochloric acid's leaching rate and pH value**.**

Conclusions

Marine biofouling is an issue that has received special attention from scientists, engineers, biologists, and other experts in diverse sectors. Collaborative work with numerous advancements has led to the development of antifouling technology based on various surface treatments. Inspired by nature (like flora and fauna), a researcher discovered antifouling technology and applied it to remove biofouling. In this review, various novel methods of advanced coating technology to prevent biofouling are discussed, and the following conclusions were drawn:

1. Biologically inspired superhydrophobic surface can curtail biofouling by forming nonadhesive micro–nano-grooves and nano-enclosed pores on the surface.
2. Several foul-release coating materials such as fluoropolymers, polysiloxanes, and PDMS can prevent biofouling, but PDMS is found to be efficient with low surface energy and small Young's modulus, and hence, it is highly effective in preventing biofouling.
3. The addition of nanofiller to PDMS surfaces resulted in a broad-spectrum antibacterial and antifouling property, which improved biofouling prevention in the marine environment.
4. PEG has been found to be effective as a surface coating to minimize unwanted protein adsorption and cell adhesion. In contrast, zwitterionic polymer is slowly replacing PEG polymers because of its better antifouling properties**.**
5. Inorganic nanoparticles are effectively used in coatings which act as different foul release agents and increase the coating mechanical properties, wetting angle, and surface energy reduction.
6. Finally, widespread application on the basis of the coating's nature and level of toxicity is still a major setback since the residues of these coatings are entering the marine environment and directly/indirectly may harm the ecosystems. Hence, additional research is recommended to determine the best eco-friendly strategy that will benefit humans and ecosystems.
7. It is also observed that corrosion in a laboratory environment is limited to a small number of tests, such as electrochemical, algae related and salt bath tests. However, field test has a wide variety of corrosion, such as sea water temperature, dissolved oxygen, chloride concentration, and water flow velocity.

Nevertheless, the present review has shown that coatings can be tailored to improve the biofouling ability and other mechanical properties. Further, a wide range of nanoparticles are available that have shown specific improvement in mechanical properties, and structural integrity of composite nanocoatings. Further, the composite coatings have shown remarkable improvement in preventing biofouling, increase in adhesion, superhydrophobicity, and adhesion property. This review will also help the present researchers working in this field and coating industries to explore the advantages of composite layers and consider the critical results mentioned in this paper.

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