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Review

Recent advances and emerging trends in multifunctional marine antifouling coatings

Zhen Zhang, Jiaming Wang, Zhanglin Yang, Yuhong Liu, Jianbin Luo*

Key Laboratory of Tribology in Advanced Equipment, Tsinghua University, Beijing 100084, China.

*The correspondence should be addressed to liuyuhong@tsinghua.edu.cn and luojb@tsinghua.edu.cn

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Abstract: Marine biofouling on ships and marine platforms causes economic and environmental problems. Given the ecological toxicity of traditional antifouling systems relying on broad-spectrum biocides such as tributyltin (TBT), coupled with the poor antifouling performance and microplastic emissions of tin-free self-polishing coatings under static conditions, developing green, low-risk, and sustainable antifouling coatings is urgently needed. This review first outlines the mechanisms of biofouling formation, followed by a systematic summary of dynamic (chemical and physical) and static (fouling-release, fouling-resistant, and biomimetic micro-/nanostructured) surface antifouling coatings. Furthermore, it highlights recent advances in emerging fields, including stimuli-responsive coatings, metal-organic framework (MOF)-based coatings, high-entropy alloy (HEA)-based protective surfaces, and triboelectric nanogenerator (TENG)-based self-powered antifouling systems. Finally, future directions are discussed, providing a theoretical framework and forward-looking guidance for

designing next-generation coatings for marine antifouling.

Keywords: marine biofouling; multifunctional antifouling coatings; dynamic surface antifouling; static surface antifouling; emerging antifouling approaches

1 Introduction

Marine biofouling, defined as undesirable colonization and growth of various marine organisms on marine equipment such as vessels and aquaculture cages, has become a widespread issue posing a severe challenge to human ocean activities[1, 2]. The formation and accumulation of biofouling significantly increase surface roughness and disturb near-wall flow, resulting in enhanced hydrodynamic frictional resistance during vessel navigation[3, 4]. Consequently, additional propulsion power is required to maintain a given vessel speed, leading to a substantial rise in fuel consumption and greenhouse gas emissions. It also increases material corrosion, shortens the operational lifespan, as well as enhances the maintenance frequency and cost[5, 6]. Therefore, minimizing adverse effects of biofouling is of critical importance.

Marine biofouling exhibits a dynamic succession process marked by variations in both time and space[7]. This process generally involves four primary stages[8], as illustrated in Fig. 1. In the beginning, organic and inorganic molecules rapidly adsorb onto the submerged surfaces to form a conditioning film. Within several hours, bacteria or other microorganisms subsequently adhere to the conditioning film by releasing extracellular polymeric substances, eventually leading to the irreversible formation of a biofilm[9]. In the third stage, the microorganisms that have attached themselves release a range of substances, such as polysaccharides, lipids and nucleic acids, which trigger the settlement and development of additional small fouling organisms including diatom spores and protozoa on the biofilm surface, thereby forming a highly complex community. Finally, many macrofouling organisms, including barnacles, mussels, macroalgae and tubeworms, gradually attach, grow and cover large areas of the submerged surfaces over extended periods. It is important to note that this process is not strictly linear, and various stages might overlap or happen simultaneously[10].

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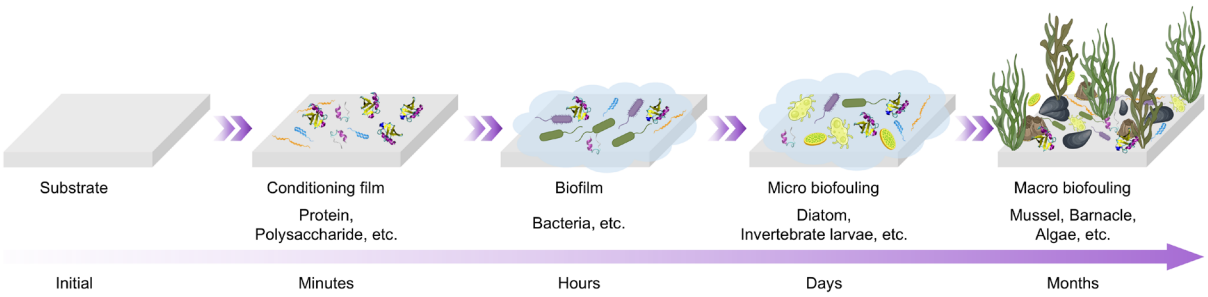

Fig. 1 Schematic diagram of the formation and evolution of marine biofouling.Currently, the primary methods for addressing biofouling include mechanical cleaning[11, 12], ultrasonic treatment[13, 14], ultraviolet (UV) irradiation[15, 16], and electrochemical treatment of seawater[17]. Mechanical cleaning involves the periodic use of high-pressure water jets or underwater robotic scrapers to remove biofouling from submerged surfaces. The force required for effective removal is closely related to the interfacial adhesion strength between fouling organisms and coating surfaces, while excessive force may increase the risk of coating wear and damage during repeated or intensive cleaning[12, 18]. Ultrasonic treatment employs high-frequency sound waves to inactivate or kill attached organisms. UV irradiation eliminates fouling organisms through photochemical damage, and seawater electrolysis generates active chlorine species by electrolytically oxidizing chloride ions in seawater to disrupt cellular structures and inhibit adhesion. However, these approaches still face significant limitations, including high labor and maintenance demands, restricted effective range, and limited applicability in open-water systems due to energy dissipation or the rapid dilution of active agents. In contrast, antifouling coatings[19-21] have become one of the primary strategies for controlling marine biofouling owing to their simplicity, low cost, and high efficiency[22-24].

In the 1950s, tributyltin (TBT) remained dominant due to its highly efficient broad-spectrum biocidal capability and prolonged antifouling duration[25, 26]. However, its lack of specificity resulted in high toxicity to diverse target and non-target marine species at extremely low exposure levels[25]. Besides, the bioaccumulation of TBT compounds raised concerns about possible health risks with the consumption of contaminated seafood. Because of these negative environmental impacts, the International Maritime Organization (IMO) finally implemented a global ban on the application of TBT self-polishing copolymer (SPC) coatings in 2008[27-29]. These regulatory restrictions stimulated the development of tin-free SPC coatings, most of

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which employ functional acrylic copolymers containing silyl, copper, and zinc acrylate units[30-32]. The side groups in these SPCs undergo hydrolytic degradation upon exposure to seawater and are then removed by water flow, demonstrating effective antifouling performance, particularly during high-speed vessel navigation. Nevertheless, they exhibit limited antifouling performance under static conditions. In addition, the main chains of SPCs are non-degradable, causing the generation of marine microplastics upon polymer fragments enter the sea[33]. Annually, an estimated 400 to 1,194 tons of marine coatings are released into European waters, with ~40% utilizing non-degradable polymers as binders. According to the International Union for Conservation of Nature (IUCN), marine coatings contribute 3.7% to the worldwide primary marine microplastic emissions[34, 35]. Therefore, the growing concern about environmental impact and biofouling issues across society has driven the urgent demand for developing eco-friendly and highly effective antifouling coatings[36].

Recent reviews have already covered many marine antifouling materials and strategies[37-42]. However, the broader expansion of marine antifouling from established surface-control strategies toward emerging approaches that introduce responsive regulation, controlled release and catalytic functions, integrated corrosion protection, and self-powered operation has received less focused attention as an organizing perspective. The distinctive contribution of this review is therefore to use this progression as the organizing thread for discussing recent advances in multifunctional marine antifouling. Accordingly, antifouling coatings are first discussed as dynamic and static surface strategies. Dynamic surfaces are discussed in terms of chemical and physical dynamic coatings, while static surfaces are categorized into fouling-release, fouling-resistant, and biomimetic micro-/nanostructured surfaces. The discussion then extends to stimuli-responsive coatings, metal-organic framework (MOF)-based coatings, high-entropy alloy (HEA)-based protective surfaces, and triboelectric nanogenerator (TENG)-assisted self-powered systems, which broaden the design of marine antifouling through triggered interfacial regulation, controlled release and catalytic or ion-mediated defense, corrosion-resistant protection, and energy-assisted operation (Fig. 2). Here, multifunctionality encompasses both the integration of complementary antifouling mechanisms, such as fouling resistance, fouling release, active fouling control, and surface renewal, and the coupling of

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antifouling with mechanical reinforcement, self-healing, corrosion protection, environmental responsiveness, or energy harvesting. Finally, the existing challenges and future prospects are discussed, with emphasis on long-term durability, multi-mechanism synergy, environmental sustainability, field validation, and data-assisted material design and optimization. This review is aimed to offer a thorough perspective for the rational design of multifunctional marine antifouling coatings and related protection systems.

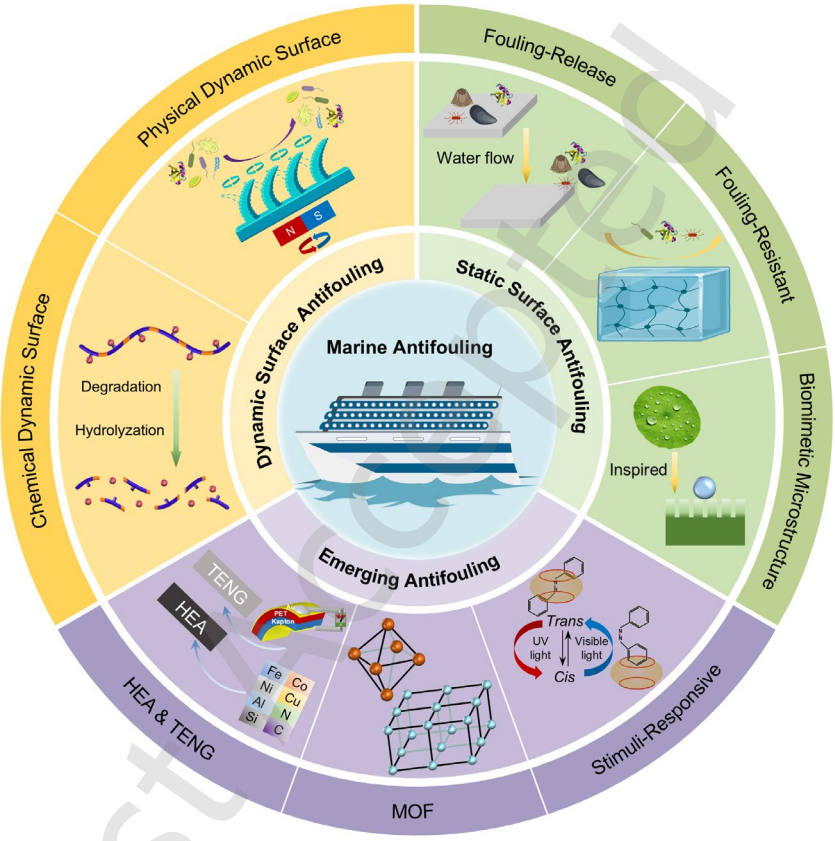

Fig. 2 Overview of marine antifouling approaches.

2 Dynamic Surface Antifouling Coatings

The concept of dynamic surface antifouling (DSA) was proposed by Ma et al.[43] in 2019 with the aim of establishing an interface that can continuously self-renew or undergo dynamic changes in seawater, thereby shortening the effective attachment time of organisms to the surface and weakening stability to inhibit the formation of biofouling. Due to its independence from the release of heavy metals, DSA represents a new paradigm for eco-friendly antifouling with promising potential (Fig. 3a)[44]. This strategy is inspired by nature. In 1984, Masaki et al.[45] explored the antifouling mechanism of the crustose coralline alga, revealing that the

shedding of epithelial cells aids in detaching fouling organisms from its surface. Subsequent efforts have observed similar defensive mechanisms in algae[46, 47], corals[48], and large marine mammals such as dolphins[49] and pilot whales[50]. Accordingly, DSA strategies have developed two technical avenues, namely surface-renewing coatings based on chemical degradation and dynamically unstable coatings driven by physical deformation, which have been confirmed at the microscopic level. Gong et al.[51] employed digital holographic microscopy to investigate the three-dimensional (3D) behavior of bacteria near dynamic surfaces, revealing that bacterial adhesion to the surface significantly decreased as the polymer degradation rate accelerated. Meanwhile, negative chemotaxis induced by degradation products further drove *Escherichia coli* away from the surface, indicating that dynamic surfaces with appropriate rates of variation can effectively inhibit biofouling. The following section offers an overview of recent advancements in dynamic surfaces for marine antifouling.

2.1 Chemical Dynamic Coatings

Chemical dynamic coatings are used to prevent biofouling adhesion by maintaining a self-renewing surface in seawater via the degradation of polyester-based polymers. In contrast to conventional SPCs with non-degradable polymer backbones, polyester-based chains are degraded into small molecules by microorganisms or enzymes, which not only helps decrease the accumulation of microplastics but also enables surface self-renewal independent of vessel speed[52, 53], allowing adaptation to both static and dynamic conditions. Previous efforts have demonstrated a positive correlation between polymer degradation rates and antifouling efficacy[51]. On the basis of the principle, various degradable polyester-based polymers has been proposed[33].

The direct application of conventional polyesters, including poly(ε-caprolactone) (PCL), poly(butylene succinate) (PBS) and polylactic acid (PLA), as coatings is limited due to their inadequate film-forming capability, insufficient interfacial adhesion, and high crystallinity that leads to slow degradation[54, 55]. Incorporating polyurethane (PU) segments into degradable polyesters is recognized as an effective approach to enhance film-forming performance and adhesion strength. As an example, Ma et al.[56] developed a PCL-based PU in which PCL served

Just Accepted

as the soft segment, while lysine ethyl ester diisocyanate and 1,4-butanediol served as the hard segment. The coating showed enhanced interfacial adhesion, but the crystallinity remained relatively high. To further reduce crystallinity, ring-opening polymerization was combined with condensation reactions to prepare a range of degradable PUs featuring copolyester oligomers of caprolactone (CL) and glycol acid (GA) as soft segments. Since copolymerization can disrupt the regularity of polymer segments and inhibit soft segment crystallization, the incorporation of GA generally helps reduce crystallinity. Compared with PCL soft segments, these PUs exhibit faster degradation. Experimental results have indicated that the CL/GA10-PU coating without antifoulants exhibits less fouling than the other tested PU coatings after three months of marine field immersion. After the incorporation of the isothiazolinone derivative (DCOIT) as an antifoulant, the duration of antifouling performance was further extended to five months. Similarly, Chiang et al.[57] focused on biodegradable PLA, developing an antifouling coating by incorporating a non-toxic antifouling compound butenolide into a PLA-based PU. Compared with PCL-PU80[58], PLA-PU50 exhibits a higher density of ester bonds, which led to a faster degradation rate and facilitated the effective release of the antifoulant. To address the decline in antifouling performance at later stages caused by the slowing of polymer degradation, water-soluble natural rosin was further integrated into the system, which not only enhanced the self-renewal rate but also significantly improved the sustained release curve of butenolide.

Incorporating hydrophilic groups to promote hydrolysis is another vital approach for enhancing degradation rate. Liu et al.[59] developed a degradable zwitterionic polyurethane coating featuring a PCL backbone and zwitterionic poly(sulfobetaine methacrylate) (PSBMA) side chains within a tannic acid (TA) crosslinked network. This system also incorporated TA/CuO_2 nanoparticles as active antifouling components. The results show that increasing PSBMA concentration can enhance coating hydrophilicity and accelerate hydrolytic degradation. Simultaneously, the hydration layer established by zwitterionic groups suppressed initial adhesion of microorganisms, while the degradation process also drove the sustained release of active components, thereby enabling synergistic antifouling performance (Fig. 3b).

Like PU, polyacrylates are well-suited for applications in adhesives and coatings because of their exceptional film-forming capability[43]. As an example, Xu et al.[60] developed a durable

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and degradable acrylate polymer featuring side chains composed of PLA and natural rosin (Fig. 3c). The coating exhibits robust resistance against protein adsorption, exceptional antibacterial efficacy (>99.9%), and excellent antialgal performance (>87.1%) due to the synergistic effect of the degradable PLA and antibacterial property of natural rosin. Moreover, it provides effective protection after 240 days of exposure in actual marine conditions. Xie et al.[61] synthesized a copolymer composed of 2-methylene-1,3-dioxepane (MDO), 2-(dimethylamino)ethyl methacrylate (DEM) and 3-(methacryloxypropyl)trimethoxysilane (KH570). This polymer exhibits antifouling performance from its degradability and the hydrophilicity of the DEM.

Despite excellent biodegradability of conventional polymers, their synthesis typically relies on ring-opening polymerization of cyclic ester monomers that offer relatively limited sites for functionalized side groups, making further chemical modification or functional design challenging[33]. To overcome these challenges, a hybrid copolymerization approach that integrates functional vinyl units with degradable segments into a single polymer backbone has been proposed. This approach enables the construction of materials featuring both degradable main chains and hydrolyzable side chains, including silyl esters, antifoulants, and hydrolysis-induced zwitterions (HIZs)[62-64]. This coupling effect supports sustained surface renewal of the coating during long-term immersion in seawater. Compared with traditional degradable polymers, these vinyl hybrid systems exhibit more controllable degradation kinetics and more flexible functionalization. For instance, Zhou et al.[64] synthesized polymers featuring both degradable backbones and hydrolyzable side groups through the copolymerization of methyl methacrylate (MMA), tributylsilyl methacrylate (TBSM) and MDO. By modulating the molar ratio between the ester units and the tributylsilyl ester side groups, the synergistic control of degradation and hydrolysis kinetics can be achieved. Besides, incorporating HIZs such as tert-carboxybetaine triisopropylsilyl ester ethyl acrylate (TCBSA)[62] or eco-friendly antifoulants such as N-methacryloyloxymethyl benzisothiazolinone (V-BIT)[63] into the copolymer side chains can maintain dynamic surface self-renewal while further enhancing the antifouling efficiency of coatings (Fig. 3d).

Hyperbranched polymers have been incorporated into degradable coatings to further

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improve the antifouling performance under static conditions. Compared with linear degradable polymers, these can provide a greater number of degradation sites and yield fragments with smaller molecular weights. In addition, hyperbranched polymers promote surface renewal and ensures favorable solubility, enabling the fabrication of coatings containing high solid fraction yet low viscosity as well as exhibiting superior film-forming capability[33, 65, 66]. As an example, Si et al.[67] developed a degradable hyperbranched polymer (h-LLA$_x$) whereas divinyl-functional oligomeric poly(L-lactide) (OLLA) serves as a bridge segment and isobornyl acrylate (IBOA) as a comonomer (Fig. 3e). The microphase separation between IBOA and OLLA within the hyperbranched architecture, combined with abundant degradable ester linkages, enables the coating to exhibit controllable degradation rates and surface fragmentation renewal capability. Furthermore, the synergistic effect between the self-assembled microscale hollow hemispherical micellar structure and the hydrolysis-induced self-renewing surface enables the coating to exhibit stable and excellent antiadhesion performance against *Navicula incerta* (~0 cell mm^{-2}), while also showing effective inhibition against *E. coli* and *Staphylococcus aureus (S. aureus)*. Acute toxicity experiments on zebrafish embryos further confirm the absence of apparent ecotoxicity, supporting its suitability for the development of eco-friendly coatings. In addition, Ai et al.[68] prepared a biodegradable polymer network by integrating hyperbranched poly(*ε*-caprolactone) (h-PCL) with siloxanes. The PCL segments enable biodegradation in the ocean. Meanwhile, the low crystallinity together with the chemically crosslinked structure imparts elastic surface characteristics to the network, whereas the incorporation of siloxane contributes to improved surface smoothness and lower surface energy. This coating that combines a dynamic surface with fouling-release capability is expected to emerge as a highly eco-friendly alternative.

In summary, chemical dynamic coatings demonstrate exceptional antifouling potential by achieving continuous surface self-renewal via main-chain degradation. Integrating main-chain degradation with side-chain hydrolysis, such as antifoulant release and zwitterion generation, or incorporating hyperbranched architectures can further modulate degradation kinetics, enhance mechanical properties and prolong the lifetime of coatings. Moreover, the coatings are primarily composed of degradable polyesters, whose ester bonds easily cleave in the presence

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of water and enzymes to yield products with small molecular weights including acids and alcohols[69]. Upon assimilated by microorganisms, they are further metabolized as sources of carbon and energy and are finally converted into carbon dioxide, water, and biomass without microplastic accumulation, aligning with the development trend of green marine antifouling[69, 70]. However, because surface renewal is achieved through material consumption, the degradation rate simultaneously governs antifouling performance and coating lifetime. Insufficient degradation may fail to renew the interface at a rate sufficient to suppress fouling accumulation, whereas excessively rapid consumption would shorten the effective service period and accelerate the release of incorporated functional components. Therefore, future development should focus on balancing degradation-driven surface renewal with long-term coating durability while maintaining controllable antifoulant release and environmental compatibility.

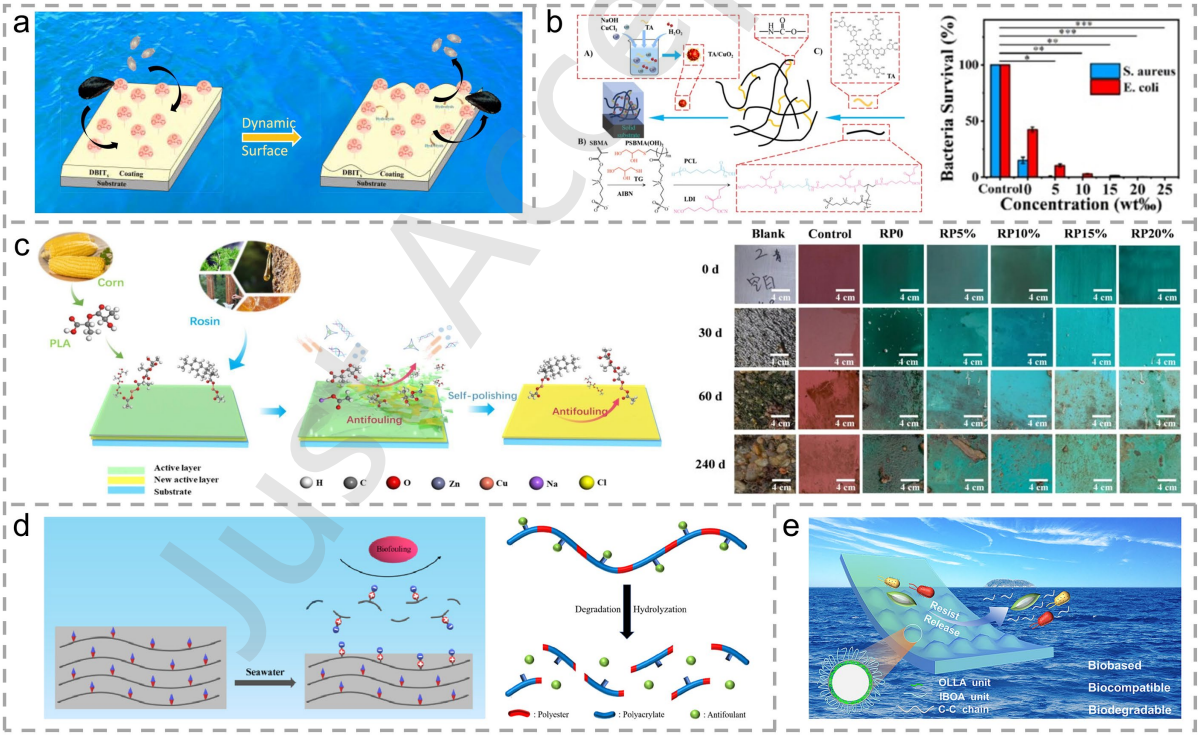

Fig. 3 Strategies of dynamic surface antifouling. a) Schematic of antifouling by a dynamic surface. Reproduced with permission[44]. Copyright 2023, Elsevier. b) Schematic illustration of the fabrication process for antifouling and antibacterial polyurethanes containing nanocomposites and zwitterionic polymers (left) and antibacterial activity of the $(TA/CuO_2\text{-}PU)_n$ coating (right). Reproduced with permission[59]. Copyright 2025, Elsevier. c) Schematic diagram of antifouling mechanism of degradable PLA-reinforced acrylate polymer coating (left) and antifouling effect in actual marine environment (right). Reproduced with permission[60]. Copyright 2024, Elsevier. d) The degradation and hydrolyzation mechanism of coatings containing TCBSA (left) or V-BIT (right). Reproduced with permission[62, 63]. Copyright 2019, American

Chemical Society. e) Schematic diagram of antifouling mechanism of degradable h-LLA_x coating. Reproduced with permission[67]. Copyright 2022, American Chemical Society.

2.2 Physical Dynamic Coatings

In contrast to chemical degradation or hydrolysis, physical dynamic coatings represent a non-sacrificial approach capable of generating continuously transforming surfaces under physical stimulations such as electric fields, magnetic fields, or oceanic flow[71-74]. These coatings inhibit the settlement of fouling organisms while improving the release of pre-adhered organisms via dynamic deformation[39], thereby exhibiting significant application potential in marine industry. Jin et al. systematically investigated the structure-property relationship between the bulk mechanical properties of the material and dynamic antifouling performance via a graphene-silicone rubber system as a model. By precisely modulating the Young's modulus of the matrix through the control of filler fraction, combined with Kulik equation calculation and hydrodynamic experiments, it has been quantitatively confirmed that a low elastic modulus facilitates higher dynamic deformation under turbulent flow conditions, thereby effectively disrupting the attachment process (Fig. 4a). Additionally, discrete element and computational fluid dynamics simulations further confirmed the influence of elastic modulus on antifouling performance[39, 75].

Building upon this foundation and inspired by the swaying motion of soft tentacles of the coral *Sarcophyton trocheliophorum* to maintain a clean surface in water flow, Tian et al.[76] proposed a graphene-silicone elastomer (TS-GSE) coating with a biomimetic tentacle structure, whose negatively charged surface impacts bacterial adhesion through electrostatic interaction. More importantly, the flexible tentacles undergo harmonic motion at a frequency of ~10 Hz under fluid action, establishing the primary physical barrier and significantly inhibiting bacterial adhesion under dynamic conditions. Liu et al.[77] recently developed a filament-like sweepers array (Fig. 4b). The highlight of this structure lies in the ability to utilize external turbulent energy to penetrate the static viscous sublayer and directly exert shear forces on the attachment interface. Under oscillatory flow, the sweeper array exhibits coordinated sinusoidal motion. A single sweeper enables a contaminant removal rate of ~99.5% via dynamic buckling, while synergistic swaying of the entire array accomplishes complete self-cleaning of the

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covered area within 10 seconds. This dynamic topological surface, based on fluid-structure coupling effects, breaks through the concept of traditional self-cleaning surfaces, providing a novel solution for passive physical antifouling.

The active application of external fields also represents an effective approach for realizing dynamic surfaces to combat biofouling. As an example, Qian et al.[78] developed an approach involving an electric field-induced micromotion nanostructured surface (Fig. 4c). A dielectric polydimethylsiloxane (PDMS) layer incorporating dopamine-modified MXene serves as the actuator, generating an oscillation amplitude of 8 μm and a force of 474 μN under the electric field of 30 V and 1000 Hz. This dynamic surface achieves >90% inhibition of *Pseudomonas aeruginosa* biofilm for 15 days. Meanwhile, the labyrinth effect formed by MXene increases the polarization resistance of Q235 carbon steel by six orders of magnitude by restricting corrosive-medium transport. In another study, Gao et al.[79] fabricated a bioinspired superhydrophobic magnetic-responsive microcolumn array (SMMA) with uniform hierarchical structures via magnetic field-assisted templating and laser etching (Fig. 4d). Magnetic actuation induces continuous microcolumn oscillation, mechanically impeding biofouling attachment and accumulation, while superhydrophobicity minimizes bacterial contact with the surface. Following a 120 h antifouling test, the SMMA inhibits *Paracoccus pantotrophus* and *Bacillus subtilis* by 94.08% and 88.29%, respectively. Its rapid magnetic response and flexible microcolumns further enable directional, damage-free transport of droplets and solid particles under a moving magnetic field, together with high mechanical durability and physicochemical stability.

Moreover, Wang et al.[80] proposed a dynamic interfacial vortex array (DIVA) approach for antifouling, in which a reciprocatively moving magnet array drives micro-dimple arrays to generate enhanced wall shear. This DIVA can maintain surface fouling <2% under high bacterial concentration and achieve >90% bacterium removal within 1 h independent of bacterial species, highlighting potential for maritime applications.

In summary, physical dynamic coatings overcome the limited lifespan of chemical dynamic coatings by establishing unstable interfaces at the macroscopic or microscopic scale. Although actively driven approaches based on electric or magnetic fields offer controllable

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deformation and efficient detachment forces, the dependence on external energy limits large-scale application in marine infrastructure. In contrast, passively driven approaches that employ natural flow shear exhibit broader engineering potential because of their zero-energy consumption and adaptive properties. Future advances in physical dynamic surfaces may focus on integration with other antifouling methods, such as the incorporation of antifouling groups to enhance performance, thereby establishing integrated systems capable of maintaining defensive efficacy even in the absence of external actuation.

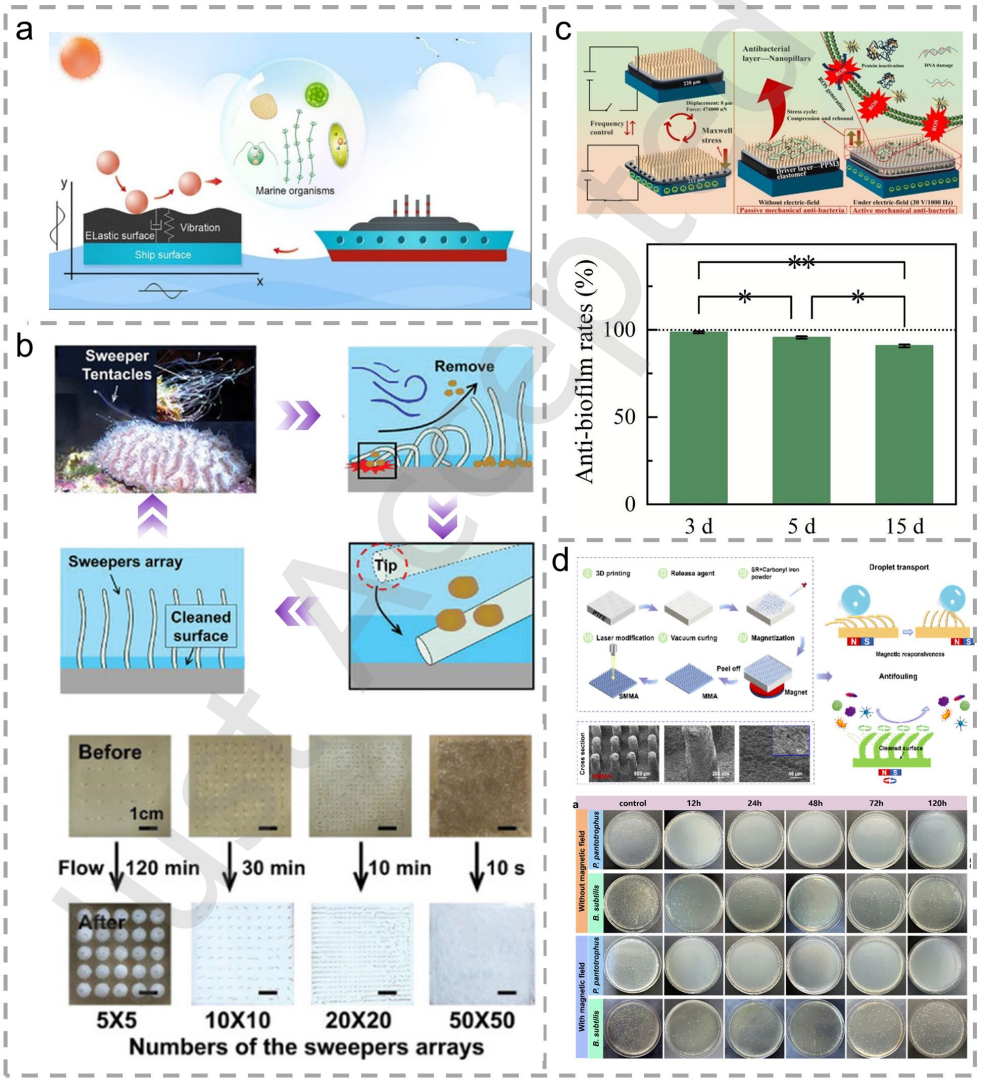

Fig. 4 Strategies of physical surface antifouling. a) Antifouling mechanism of physical dynamic surface. Reproduced with permission[75]. Copyright 2019, Royal Society of Chemistry. b) Active self-cleaning surfaces inspired by sweeper tentacles of coral (top) and the cleaned pattern of sweepers arrays with different numbers (bottom). Reproduced with permission[77]. Copyright 2023, Wiley-VCH. c) Schematic diagram of anti-biofilm mechanism of PPM3/NP surface (top) and the removal capacity of colonizing biofilms (bottom). Reproduced with permission[78]. Copyright 2026, Elsevier. d) Schematic illustration of the fabrication of magnetic-responsive SMMA surface and its dual functionality (top) and bacterial colony counting plate tests of bacterial suspensions with and without magnetic fields (bottom). Reproduced with permission[79].

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3 Static Surface Antifouling Coatings

Distinct from DSA, static surface antifouling aims to establish a chemically stable and structurally durable interface by modulating surface physicochemical properties, such as surface energy and wettability, or engineering specific microstructures, thereby inhibiting biofouling through thermodynamic repulsion or steric hindrance effects. Based on the different mechanisms, static surface antifouling coatings are primarily categorized into fouling-release coatings, fouling-resistant coatings and biomimetic micro-/nanostructured surfaces.

3.1 Fouling-Release Coatings

Fouling-release coatings (FRCs) inhibit marine biofouling by combining low surface energy, low elastic modulus and smooth surfaces to reduce interfacial adhesion of fouling organisms. Therefore, attached organisms can be readily detached by hydrodynamic shear during navigation or simple mechanical cleaning, making FRCs an efficient, economical, and eco-friendly solution[81, 82]. Brady et al.[83] have demonstrated that relative adhesion exhibits a positive linear correlation with the square root of elastic modulus and the critical surface free energy, supporting the widespread use of low-surface-energy silicones and fluoropolymers in FRCs. Among these, silicone rubber elastomers have become the dominant matrix materials owing to their high stability and drag reducing property. However, conventional silicone-based FRCs still remain constrained by weak substrate adhesion, poor mechanical robustness, and insufficient static antifouling performance[81]. Consequently, recent advances in FRCs can be discussed from three main aspects: (1) improving mechanical stability and substrate adhesion, (2) enhancing antifouling performance under static conditions, and (3) extending service life of coatings.

(1) Improving mechanical stability and substrate adhesion. High mechanical strength is crucial for preventing damage and extending the service life of flexible silicone-based FRCs. Currently, reinforcement approaches mainly include physical blending and chemical structural modification. In terms of physical blending, the incorporation of nanomaterials including carbon nanotubes (CNTs), graphene oxide (GO), as well as metal nanoparticles effectively

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enhances the mechanical performance of the matrix[84-89]. For instance, Qiu et al.[90] proposed a PDMS/polythiourethane (PTU) composite coating modified with tetrapodal-shaped zinc oxide particles (t-ZnO). The results demonstrate that the incorporation of t-ZnO (1 wt%) particles substantially improves the mechanical performance of the coating while simultaneously endowing it with excellent fouling-release capability. Selim et al.[91] synthesized β-MnO_2 nanorods via a hydrothermal method and incorporated them into a silicone matrix as a physical reinforcing phase (Fig. 5a). It is found that a nanofiller loading of 0.5 wt% effectively improves the mechanical stability of the coating, allowing it to withstand impact at 100 cm/s without obvious cracking while maintaining excellent adhesion to the substrate at a 5B rating. In addition, the composite coating achieves superhydrophobicity with a water contact angle (WCA) of 158° through micro and nanoscale hierarchical roughness induced by the nanorods, demonstrating outstanding self-cleaning and fouling-release capabilities. They further investigated the impact of reduced graphene oxide (RGO) and graphene oxide/boehmite nanorods (GO-γ-AlOOH) nanofillers on the performance of PDMS coatings. At 3 wt%, GO-γ-AlOOH hybrid nanofiller exhibits superior dispersibility than pristine RGO, markedly enhancing coating durability and preventing fracture under an impact energy of 15 J[86]. In terms of chemical structural modification, the introduction of epoxy, PU, or urea groups represents an effective method for enhancing coating adhesion[92-94]. For instance, Liu et al.[92] prepared a polydimethylsiloxane-based polyurea (PDMS-PUa-x) through polycondensation, as shown in Fig. 5b.This coating maintains low surface energy and hydrophobicity of PDMS for robust fouling-release, while dense hydrogen bonding between hard-segment urea groups and substrate functionalities enhances substrate adhesion (>1.0 MPa), superior to conventional PDMS coatings (~0.4 MPa). Building on this work, Xie et al.[95] further combined physical and chemical approaches by incorporating fluorinated nanodiamonds into the PDMS-PUa matrix (Fig. 5c). The results demonstrate that the tensile strength of the PDMS-PUa is enhanced from 1.2 MPa to 1.6 MPa, and its adhesion from 1.5 MPa to 2.1 MPa. Furthermore, facilitated by the dynamic noncovalent network formed by the polyurea hard segments, the composite coating exhibits self-healing capability at room temperature and maintains excellent fouling-release performance as well as integrity during a real-sea immersion for 4 months. These results

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indicate that substrate adhesion in silicone-based FRCs has been progressively improved from approximately 0.4 MPa for conventional PDMS to over 1.0 MPa by introducing PUa segments and further to 2.1 MPa through nanofiller reinforcement.

(2) Enhancing antifouling performance under static conditions. Despite improved mechanical robustness, the antifouling performance of silicone-based FRCs remains limited under static conditions. Accordingly, antifoulants or hydrophilic groups have been introduced to enhance antifouling performance. Martinelli et al.[96] evaluated the impact of various surface-active side chains on the antifouling performance by incorporating copolymers composed of PDMS and polymethacrylate blocks containing either hydrophilic oxyethylene (EG) or hydrophobic fluoroalkyl (AF) side chains into a PDMS matrix through physical blending. The findings reveal that the composite coating containing Si-EG primarily enhances fouling-release capacity with superior removal rates for *Ulva linza* and *Balanus amphitrite* larvae compared with pure PDMS, whereas the coating containing Si-AF exhibits exceptional antiadhesion performance by effectively preventing the settlement of *B. amphitrite* larvae.

Zhao et al.[97] developed an environmentally sustainable silicone coating bearing natural borneol fluorinated side chains (PDMS-F-PIBO) to improve static antifouling capability (Fig. 5d). By leveraging the low surface energy of the fluorocarbon segments, the coating drives migration and enrichment of antibacterial borneol groups toward the surface during film formation. Experimental results have shown that this coating achieves an antibacterial efficiency of 95.6% against *Pseudomonas* sp. and reduces the biofilm coverage by 86.9% while efficiently inhibiting diatom adhesion. Field tests in the Yellow Sea have further confirmed sustained antifouling efficacy for >6 months. Qin et al.[98] recently fabricated an epoxy-poly(dimethylsiloxane)/capsaicin (EP-PDMS/CAP) coating featuring a semi-interpenetrating polymer network (SIPN) structure between the epoxy and PDMS layers (Fig. 5e), which increases interfacial adhesion from 0.4 MPa to 2.04 MPa. Additionally, coupling PDMS-mediated fouling release with capsaicin-induced biorepellence enables synergistic antifouling performance under both static and dynamic conditions, yielding an antibacterial efficacy of 96.8% and anti-algal adhesion rate of 95.7% against *Chlorella*. To address antifouling failure caused by the rapid release of antifoulants, Lu et al.[99] fabricated a composite coating by chemically

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immobilizing capsaicin onto the surface of $CoFe_2O_4$/gelatin magnetic nanoparticles and embedding them into a PDMS-based block copolymer. The findings indicate that this method effectively prevents the excessive release of capsaicin, thereby extending the lifespan of the coating.

Furthermore, amphiphilic surfaces with nanoscale hydrophilic-hydrophobic mosaic structures can be developed by incorporating hydrophilic groups into silicone coatings including polyethylene glycol (PEG), zwitterions, and poly(N-vinylpyrrolidone) (PVP)[100-102], effectively inhibiting the biofouling adhesion[103]. As an example, Guo et al.[104] synthesized a highly stable PDMS-PVP amphiphilic network coating through the chemical crosslinking of triethoxysilane-functionalized PVP with PDMS. The coating can effectively reduce the adhesion of bacteria and *Navicula* diatoms by 99% and inhibit the attachment of mimetic barnacle by 82%. On this basis, Sun et al.[105] fabricated a transparent silicone hybrid coating via simple sol-gel method, integrating amphiphilicity, nanocellulose-mediated lubrication, and electrostatic repulsion. The microscale hydrophobic/hydrophilic heterogeneities from amphiphilic telomers disrupted microbial orientation, while castor oil-modified cellulose nanocrystals along with cholic acid are introduced to provide lubrication sites on the surface and negatively charged electrostatic repulsion, respectively. This synergistic strategy obviously reduces both the friction coefficient (~0.08) and bacterial adhesion force (~0.29 nN), sustaining >90% macrofouling resistance during 180 days of marine exposure.

zwitterions have gained widespread attention owing to their capacity to form a more stable hydration layer compared to conventional hydrophilic polymers. Shu et al.[101] developed a fluorine-free amphiphilic copolymer by initiated Chemical Vapor Deposition, combining hydrophobic 1,3,5,7-tetramethyl-1,3,5,7-tetravinyl cyclotetrasiloxane with hydrophilic pyridinium-based zwitterions (Fig. 5f). The coating decreases *P. aeruginosa* adhesion by >90% compared to aluminum surfaces and achieves a removal rate of 99.7% for *N. incerta* after water jetting, outperforming a commercial paint 1100SR. However, high surface energy of zwitterions often causes them to become entrapped within the bulk matrix, thereby restricting their antifouling efficacy. Besides, their tendency to swell upon water absorption frequently damages the mechanical properties[106]. Consequently, Tan et al.[107] developed a composite

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coating by simply blending hydroxyl-terminated PDMS with a zwitterionic silica sol (h-MAPS) synthesized via the co-hydrolysis of tetraethyl orthosilicate (TEOS) and a zwitterionic precursor (MAPS). The results show that this design substantially enhances the mechanical properties of the matrix through a dense silica sol network (tensile strength from 3.8 MPa to 15.6 MPa), and induces *in situ* formation of a zwitterionic hydration layer upon seawater immersion, thereby enabling mechanical stability with static resistance to bacteria, proteins, as well as marine slime. Moreover, the barnacle removal strength remains within the typical range for commercial FRCs (<0.4 MPa), indicating the excellent fouling-release characteristic.

(3) Extending service life of coatings. To further extend the lifespan of FRCs and decrease maintenance costs, self-healing techniques based on dynamic bonds have gained significant research attention[108, 109]. For instance, Cui et al.[108] constructed a self-healing PU composite coating with a multiple hydrogen-bonding network formed by abundant carbonyl sites. The coating recovers 85% of its tensile strength after 48 h at room temperature with faster self-healing process at elevated temperature. Additionally, incorporation of lubricant PDMS and antifoulant medetomidine further yields antibacterial and algal-adhesion rates of 91.7% and 90.9%, respectively. Hu et al.[110] developed a new silicone-based coating by exploiting the coordination interaction between zinc ions and 2-(2-benzimidazolyl)ethanethiol (BET). The coating achieves self-healing at room temperature through reversible zinc-imidazole coordination bonds, with the resulting metal complex as a non-leaching antifoulant to efficiently prevent the adhesion of *Pseudomonas* sp. and *N. incerta*.

However, single dynamic bonds typically fail to balance mechanical strength with self-healing efficiency. Sun et al.[109] constructed a multidynamic-network silicon coating via disulfide exchange between the functionalized monomer lipoic acid-benzothiazole (LA-BTZ) and PDMS-based polyurea-urethane with LA modification (Fig. 5g). Synergistic disulfide and hydrogen bonding enable excellent self-healing efficiency (98.5%), as well as enhanced mechanical strength (2.58 MPa) and substrate adhesion (3.33 MPa). Moreover, non-leaching benzothiazole group provides exceptional antibacterial, antifouling, and anticorrosive properties to the coating, thereby achieving the multifunctional integration of self-healing, antifouling, and anticorrosion capabilities. Building on this foundation, Liu et al.[111]

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constructed a Cu^{2+}-coordinated polymer network utilizing hydroxypropyl-terminated PDMS and polytetrahydrofuran (PTMG) as the main chain, 2-hydroxyethyl disulfide (HEDS) and dimethylglyoxime (DMG) as chain extenders and silver nanoparticles (AgNPs) as functional nanofillers. The coating achieves a self-healing efficiency of 91.7% at room temperature via the interaction of disulfide bonds, oxime urethane bonds, metal coordination bonds, and hydrogen bonds while maintaining robust substrate adhesion (>1.3 MPa). More importantly, the incorporation of AgNPs enhances the tensile strength to 2.75 MPa while endowing it with excellent antibacterial efficacy (97.8% inhibition rate against *P. aeruginosa*), thereby reconciling mechanical properties with healing efficiency and extending the lifespan in complex marine environments.

Although the physical reinforcement and self-healing approaches described above have substantially improved the durability of conventional flexible FRCs, they still remain vulnerable to sharp object scratches and high-intensity wear. To overcome this limitation, recent efforts have gradually focused on polymer-ceramic hybrid coatings, which reconcile high hardness of inorganic ceramics with the flexibility of organic polymers through molecular-level structural design and the modular assembly of multiscale building blocks, thereby enabling the precise tailoring of mechanical and surface properties[112-115] and significantly broadening their applicability in complex marine engineering environments[116, 117]. Sun et al.[118] developed a silicone/ZrO_2 sol hybrid antifouling coating through a sol-gel strategy by covalently integrating rigid zirconia-derived structures with a flexible antibacterial amphiphilic telomer (Fig. 5h). The resulting organic-inorganic network exhibits high hardness (4-6H), good flexibility (~5 mm bending diameter), strong substrate adhesion (~2.89 MPa), and high optical transparency (>98%). Meanwhile, surface enrichment of the non-leaching amphiphilic telomer endowed the coating with remarkable self-cleaning, and fouling release ability against proteins, bacteria, diatoms, and biofilms. To impart anticorrosion capability while enhancing hardness, Cheng et al.[119] developed a fluorinated silicone-based polymeric film (FSPF) by blending liquid polysilazane preceramic precursor (PSZ) with fluorinated polysilazane resin at 200-500 °C. It is found that the coating exhibits high corrosion resistance (0.0899 V) and possesses excellent antifouling performance along with a high elastic recovery rate (77.3%), which is primarily

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attributed to the fluorine-based modification significantly reducing surface energy (28.43 mJ/m^2) and enhancing surface toughness.

However, the static antifouling performance of hard coatings remains limited when it relies solely on silicone segments of low surface energy. Therefore, introducing hydrophilic groups including PEG or zwitterions into the hybrid network to establish a hydration layer has emerged as an effective method[120, 121]. As an example, Huang et al.[121] constructed a zwitterionic coating by integrating polysilazane with a FT telomer bearing low-surface-energy fluorine groups and hydrolysis-activated zwitterions (Fig. 5i). The coating undergoes self-adaptive interfacial rearrangement, exposing fluorinated groups in air to minimize liquid adhesion, while zwitterionic groups form a stable hydration layer underwater that suppresses swelling and confers antibiofouling, low liquid adhesion, and antiscaling properties. Furthermore, FT toughens the highly crosslinked network, endowing the coating with a hardness of up to 7H, superior flexibility, and robust interfacial adhesion of 2.06-7.67 MPa. In another work, Zhang et al.[117] constructed a self-reinforcing gradient antifouling coating through a modular design concept. Specifically, covalent coupling between thiol-terminated polysilsesquioxane (POSS-SH) and isocyanate-terminated polyurethane (NCO-PU) forms a cohesive crosslinked network that improves the mechanical stability of the coating, while reactive amphiphilic polymer self-enriches on the surface under thermodynamic and kinetic driving forces to form a gradient antifouling interface, effectively exerting resistance against biofouling.

In summary, recent FRC research has moved beyond simply lowering surface energy toward integrated coating architectures that simultaneously reconcile fouling release with mechanical durability, static antifouling performance, and service life. In particular, the low elastic modulus favored for efficient fouling release introduces an inherent mechanical trade-off. A compliant interface facilitates the detachment of adhered foulants under hydrodynamic shear, whereas insufficient bulk strength and substrate adhesion increase susceptibility to abrasion and mechanical damage during long-term service. Reinforcement through nanofillers, hard polymer segments, or organic-inorganic networks can improve coating strength and adhesion. However, excessive stiffening, particularly when transferred to the outer interface, may weaken the compliant, low-adhesion characteristics required for fouling release. Therefore,

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next-generation FRCs should decouple bulk mechanical support from interfacial antifouling functions, allowing a mechanically robust coating body to provide substrate adhesion and damage resistance while preserving a compliant, low-surface-energy outer interface for fouling release. This functional separation also allows amphiphilic or zwitterionic surface functionalities to improve static fouling resistance and dynamic-bond-mediated networks to restore mechanical damage and extend service life, while minimizing interference with the fouling-release interface. Degradability or recyclability should also be considered during material design to improve long-term sustainability[122].

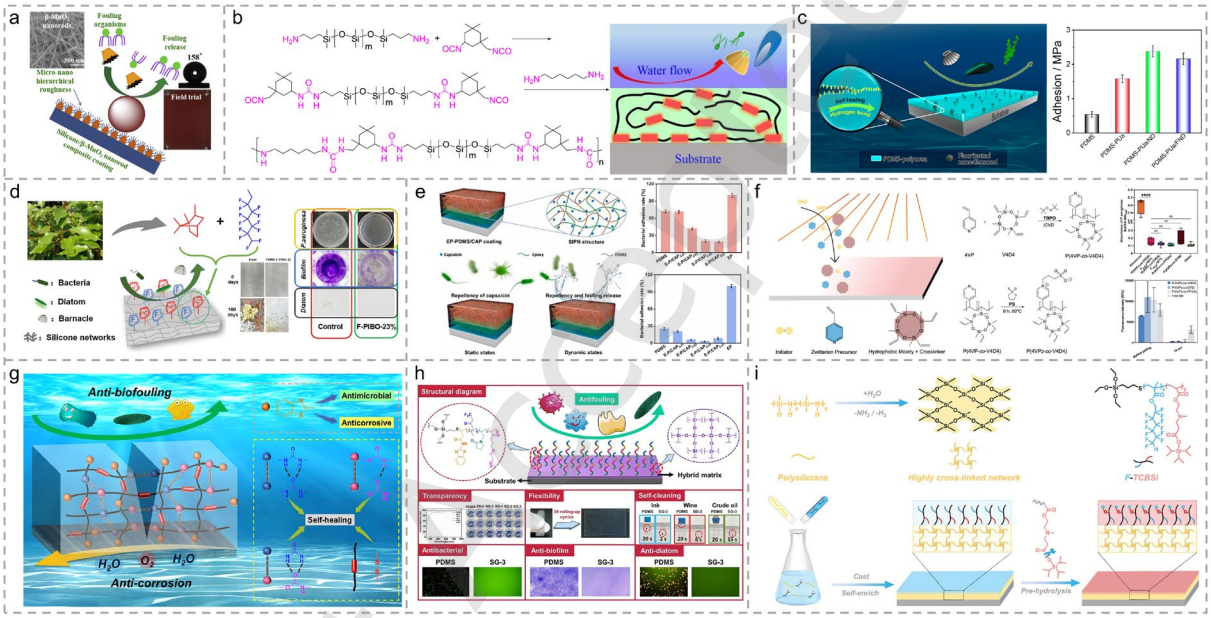

Fig. 5 Strategies of fouling-release coatings. a) Antifouling mechanism of organosilicon/β-MnO_2 nanorod composite coatings. Reproduced with permission[91]. Copyright 2019, Elsevier. b) Synthesis of PDMS-PUa-x (left) coating and corresponding antifouling mechanism (right). Reproduced with permission[92]. Copyright 2016, American Chemical Society. c) Schematic diagram of antifouling mechanism of PDMS coatings modified with fluorinated nanodiamonds (left) and adhesion strength of PDMS-based coatings (right). Reproduced with permission[95]. Copyright 2020, American Chemical Society. d) Antifouling mechanism of PDMS-F-PIBO coatings via surface self-enrichment of natural borneol. Reproduced with permission[97]. Copyright 2023, Elsevier. e) Synergistic antifouling mechanism of EP-PDMS/CAP coatings via SIPN-enhanced interfacial adhesion (left) and the bacterial adhesion rates of coatings under static or dynamic state (right). Reproduced with permission[98]. Copyright 2023, Elsevier. f) Synthesis of an amphiphilic surface (left) and relative antibacterial and anti-algal performance (right). Reproduced with permission[101]. Copyright 2025, Wiley-VCH. g) Schematic illustration of a self-healing PDMS-PUU-LA polymer coating via disulfide bonds and hydrogen bonds. Reproduced with permission[109]. Copyright 2022, Elsevier. h) Antifouling mechanism and excellent properties of silicone/ZrO_2 sol hybrid coating. Reproduced with permission[118]. Copyright 2024, Elsevier. i) Schematic of the preparation of self-adaptive zwitterionic polysilazane coatings. Reproduced with permission[121]. Copyright 2025, Wiley-VCH.

3.2 Fouling-Resistant Coatings

Unlike fouling-release coatings, fouling-resistant coatings aim to prevent initial attachment of biofouling by constructing highly hydrated interfaces. Since marine biofouling is typically initiated by nonspecific adsorption of proteins and organic molecules, inhibiting this process is critical for preventing subsequent microbial settlement and biofilm formation[123]. Currently, PEG, zwitterionic polymers and hydrogels coatings are representative material systems for constructing fouling-resistant interfaces[124-126].

Linear PEG, a classic antifouling material, is widely grafted onto surfaces to prevent protein adsorption because of its high hydrophilicity, biocompatibility, and non-toxicity. However, weak interfacial binding between PEG and the substrate, coupled with the complexity of grafting processes involving surface chemical reactions, has constrained its large-scale application in marine antifouling coatings[41, 127]. To address this challenge, Zhang et al.[128] prepared an antifouling coating through physically blending polystyrene microspheres compounded with PEG-based hydrogel into a PDMS matrix. PEG-based hydrogel with hydration layer and PDMS with low surface energy and elasticity synergistically inhibit the adhesion of proteins, bacteria and benthic diatoms. Although this hybrid design offers an effective approach, the long-term stability of PEG-based coatings remains a critical challenge[129]. Yang et al.[130] fabricated a hydrophilic surface with excellent antifouling properties by three-dimensional (3D) grafting of poly(ethylene glycol) diacrylate (PEGDA) on a bisphenol A epoxy acrylate (BEA) sheet via UV curing technique (Fig. 6a). Despite this structural design, the antifouling performance was lost after exposure to the ocean for one month. In contrast, Esteves et al.[131] introduced PEG segments as a reservoir of hydrophilic dangling chains into a crosslinked PU network composed of polycarbonate, methoxy-PEG, and a triisocyanate crosslinker. The as-prepared PC-mPEG PU coatings exhibit low levels of protein adsorption and a self-replenishing surface hydrophilicity, which remain stable after one year of water immersion, highlighting their potential for long-term marine antifouling.

Zwitterionic polymers have emerged as promising next-generation marine antifouling materials owing to their strong hydration via ionic solvation[132, 133]. These polymers carry equal numbers of anionic and cationic groups along their molecular chains, thereby maintaining

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overall electrical neutrality. Typical structures include phosphocholine (PC), sulfobetaine (SB), and carboxybetaine (CB). Unlike PEG, whose hydration is mediated by hydrogen bonding, zwitterionic polymers form a more stable hydration layer through electrostatic interactions[134]. Wu et al.[135] further demonstrated that this enhanced hydration affords greater hydrophilicity and resistance to protein adsorption than PEG. However, the high hydrophilicity often causes insufficient mechanical strength, restricting application for marine antifouling[136]. To solve this problem, integrating zwitterions with PU, a material known for its excellent adhesion and biocompatibility, has emerged as an effective approach. Nevertheless, the pronounced polarity mismatch between zwitterions and hydrophobic PU chains hinders their effective integration. Researchers have employed methods such as surface grafting, polymer blending, and interpenetrating polymer networks (IPN) to improve the compatibility between PU and zwitterionic polymers[137-139]. For example, Liu et al.[140] mitigated the compatibility conflict between zwitterions and PU by introducing zwitterionic precursors containing PDMS segments into a PDMS-PU and forming a semi-interpenetrating network through the chain entanglement of shared PDMS segments (Fig. 6b).

Hydrogels are 3D crosslinked networks composed of hydrophilic polymer chains with abundant hydrophilic groups and low elasticity, exhibiting excellent antifouling performance in water[126, 141, 142]. However, conventional hydrogels typically possess insufficient mechanical strength, making them highly susceptible to damage, swelling or detachment under prolonged seawater immersion and shear stress. Furthermore, a single physical barrier is inadequate to inhibit explosive colonization of biofouling[143]. Consequently, dual-network architectures or nanomaterials have been introduced to develop mechanically robust hydrogels with multifunctional antifouling properties, enhancing their applicability under harsh marine conditions[126, 144].

Gong et al.[145] first proposed the concept of dual-network (DN) hydrogels in 2003, which are interpenetrating network structures cotaining two fully or partially IPNs[146]. Li et al.[147] subsequently combined a chemically crosslinked polyacrylamide network with a Cu^{2+}-crosslinked alginate network, where Cu^{2+} coordination simultaneously enhanced mechanical strength and antibacterial activity. Moreover, Zhang et al.[148] prepared a new hybrid ionic-

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covalent crosslinked sodium alginate/carboxybetaine acrylamide (SA/CBAA) DN hydrogel by a simple one-pot process (Fig. 6c), achieving an elastic modulus of 0.28 MPa, a tensile strength of 0.69 MPa, and effective self-healing. The team further constructed a similar DN hydrogel by replacing CBAA with SBMA[149]. The results show that the introduction of SBMA effectively prevents the adhesion of proteins, cells, bacteria, and algae while maintaining excellent mechanical properties (elastic modulus of 0.19 MPa and tensile strength of 0.73 MPa), owing to the superior hydration capability of the zwitterionic groups.

In addition to DN, the incorporation of nanomaterials is also an effective approach. In nanocomposite (NC) hydrogels, carbon-based nanomaterials including CNTs, graphene, and GO, two-dimensional materials including MoS_2 and MXene, and metal oxides or metallic nanoparticles including Fe_2O_3 and Ag can function as crosslinking nodes that adsorb multiple polymer chains, optimizing stress distribution within the network and suppressing the propagation of macroscopic cracks[126, 150-152]. For example, Xiong et al.[126] proposed a poly(vinyl alcohol) (PVA) DN hydrogel (DN-M-P) reinforced with MXene and carboxylated cellulose nanofibers (CNF), as depicted in Fig. 6d. The synergistic reinforcement of CNF and MXene affords a toughness of 16,050 $kJ \cdot m^{-3}$ and an elongation at break of 4554%. More importantly, the hydrogel exhibits 100% inhibition of protein and algal adhesion and 99.97% antibacterial efficacy by integrating the bactericidal effect of MXene and PHMB along with the antiadhesion ability brought by hydrophilicity. This synergistic design successfully addresses the fundamental limitations of traditional hydrogels, which suffer from poor mechanical properties and single defense mechanism.

Insufficient substrate adhesion of hydrogels represents another major bottleneck that limits the application in marine antifouling. Upon hydration, the resulting interfacial water layer weakens physical and chemical interactions between the hydrogel polymer and the substrate, leading to coating delamination under harsh marine conditions[153, 154]. To tackle this challenge, Shen et al.[155] successfully anchored a PSBMA and polyacrylic acid (PAA) bicomponent hydrogel coating to the substrate by using a silane coupling agent (Fig. 6e). Despite strong interfacial adhesion, hydrolytic cleavage of silane bonds in aqueous solution compromises long-term durability. In addition, its reliance on organic solvents, stringent processing

Just Accepted

conditions, and expensive silane reagents constrain scalable fabrication and marine deployment, highlighting an urgent need to explore alternative approaches for durable hydrogel adhesion to metals and other substrates.

Jiang et al.[156] proposed a surface-treatment-free strategy combining segment orientation with covalent anchoring to construct durable underwater superoleophobic hydrogel coatings on diverse substrates. Hydrophilic PEG segments form an antifouling hydration layer at the hydrogel/water interface, whereas hydrophobic segments and epoxy groups enriched at the hydrogel/substrate interface enable covalent anchoring and hydrophobic association. This Janus interfacial structure endows the coating with strong underwater adhesion, with a shear strength of up to 5 MPa, as well as stable antiswelling, crude-oil self-cleaning, and resistance to protein and diatom attachment. Similarly, Xu et al.[127] proposed a hydrogel coating (HSOI) featuring spontaneous segment-oriented interfaces, where catechol groups enhance substrate bonding and PEG segments form a water-facing antiadhesive hydration layer (Fig. 6f). The coating is fabricated by brushing, rolling, or spraying without chemical pretreatment, maintaining a shear strength exceeding 100 kPa within 30 days of water immersion and effectively repelling crude oil, proteins, polysaccharides, bacteria, and diatoms.

As one of the most abundant biomolecules in nature, proteins have been explored as an emerging platform for hydrophilic coatings due to their structural tunability and abundant functional groups[157, 158]. However, maintaining stable protein-based interfaces remains challenging as environmental variations such as pH and salinity can affect protein conformation and surface properties, thereby influencing antifouling performance[159]. Zhao et al.[160] designed an amphiphilic coupling protein by grafting SBMA fragments onto native bovine serum albumin (BSA) through thiol-ene click chemistry. The SBMA segments maintain strong hydration capability under varying salinity and pH conditions, thereby improving the environmental adaptability and fouling resistance of the protein-based coating.

Another strategy exploits protein conformational transition and self-assembly. Protein phase transition induced by disruption of intramolecular disulfide bonds can promote β-sheet-rich amyloid-like assembly, enabling robust coating formation and adhesion on diverse substrates[161, 162]. This phase-transition strategy has also been combined with zwitterionic

Just Accepted

components to introduce hydrated interfaces and enhance resistance to nonspecific adhesion[163]. In addition, polyphenol-mediated protein assembly can strengthen coating cohesion through multivalent hydrogen-bonding, hydrophobic, and ionic interactions[164]. Yin et al.[165] constructed hydrophilic coatings through ovalbumin-tannic acid co-assembly, in which protein–polyphenol interactions reinforced interfacial assembly and improved coating stability. However, the marine applicability of these protein-derived interfaces remains to be established through long-term seawater exposure and field validation against marine foulants.

In summary, fouling-resistant coatings have developed from classical PEG-based hydration layers to a broader range of hydrophilic interfaces involving zwitterionic polymers and tough hydrogels, while protein-derived interfaces provide an emerging source of transferable anchoring and hydration concepts. Among these systems, antifouling performance is governed by the balance between interfacial hydration and structural stability rather than by hydrophilicity alone. Highly hydrophilic components can effectively strengthen the hydrated interface that suppresses nonspecific adsorption, whereas excessive water absorption can induce swelling and progressively weaken mechanical properties and compatibility with the relatively hydrophobic coating substrate. Meanwhile, complex grafting, crosslinking, or self-assembly strategies can improve laboratory performance, yet their scalability and long-term durability in complex and variable marine conditions remain insufficiently validated. Therefore, the next generation of antifouling coatings should form a stable hydrated interface while controlling its swelling and maintaining mechanical properties and underwater adhesion.

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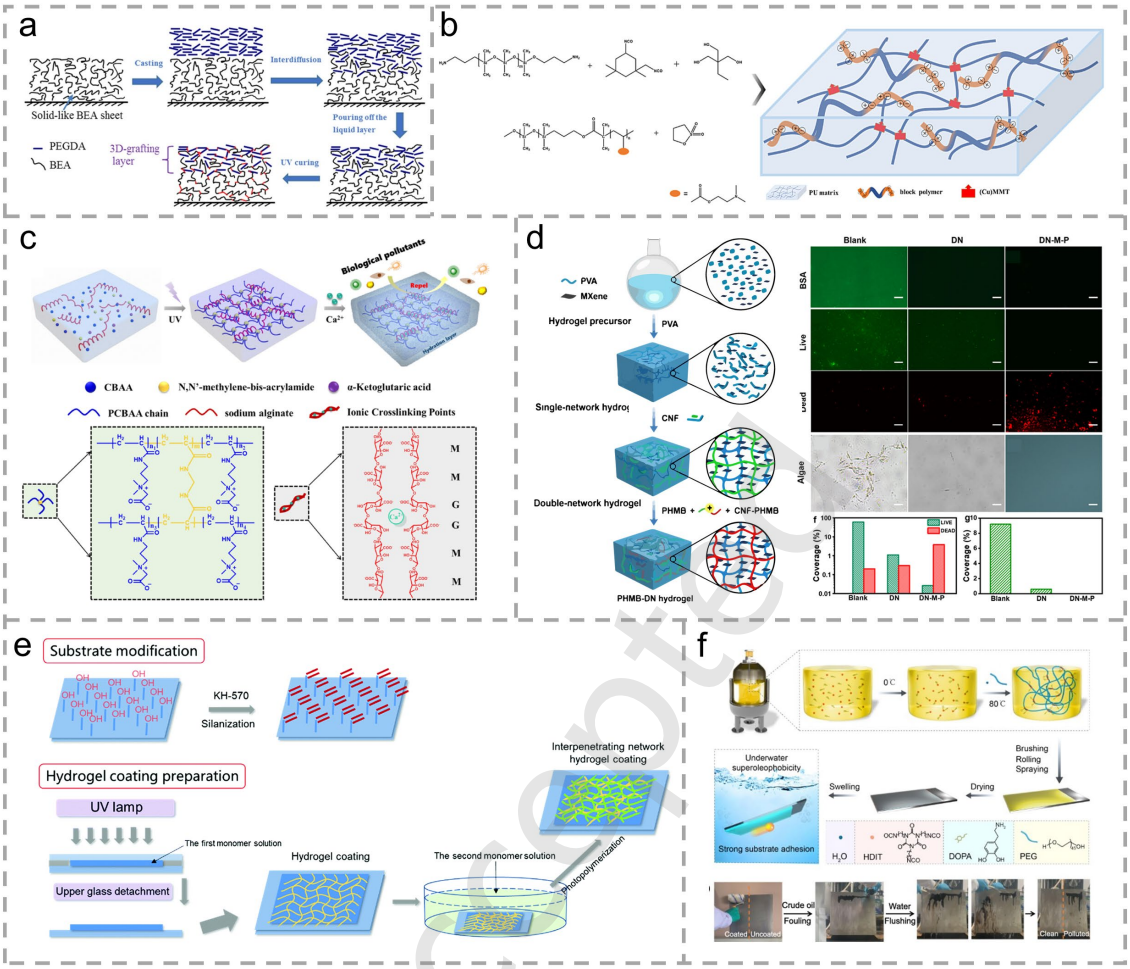

Fig. 6 Strategies of fouling-resistant coatings. a) Schematic diagram of preparation of PEGDA-grafted BEA sheets. Reproduced with permission[130]. Copyright 2016, Wiley-VCH. b) Synthesis of PDMS-PU coatings. Reproduced with permission[140]. Copyright 2023, Elsevier. c) Preparation of the hybrid ionic-covalent PCBAA/SA-Ca^{2+} DN hydrogel using a "one-pot" method (left) and corresponding performance of the DN hydrogel (right). Reproduced with permission[148]. Copyright 2021, Elsevier. d) Fabrication of DN-M-P hydrogel (left) and corresponding antifouling performance (right). Reproduced with permission[126]. Copyright 2025, Elsevier. e) Preparation of PSBMA/PAA hydrogel coatings via surface silanization treatment. Reproduced with permission[155]. Copyright 2019, Royal Society of Chemistry. f) Fabrication of the HSOI coating and its antifouling performance. Reproduced with permission[127]. Copyright 2025, Elsevier.

3.3 Biomimetic Micro-/Nanostructured Surfaces

Beyond FRCs and fouling-resistant coatings, biomimetic micro-/nanostructured surfaces have emerged as an effective method for marine antifouling by imitating the topographies of natural organisms to inhibit the attachment of fouling organisms[166-168]. Representative natural prototypes include lotus leaves with remarkable self-cleaning properties[169] and shark skin with drag-reducing and antifouling structures[170-172].

It should be highlighted that the antifouling mechanisms of micro-/nanostructures remain

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unclear. Current studies primarily attribute them to wetting state regulation[173], alterations in near-wall fluid dynamics[174], and reduction of effective attachment sites[175]. For instance, inspired by lotus leaves and coral mucus, Liu et al.[176] proposed an eco-friendly and durable superhydrophobic antifouling coating (SHAC) via one-step spray deposition (Fig. 7a). The biomimetic design couples physical antiadhesion of superhydrophobicity with chemical repulsion of antifouling agents, retaining a WCA of > 150° after 100 abrasion cycles and achieving 92% bacterial and 96% algal inhibition. A 30-day marine field exposure further confirms markedly reduction in bifouling attachment, demonstrating an effective and sustainable approach for marine antifouling. Zhao et al.[177] replicated the surface microtopography of *Laminaria japonica* as a template (Fig. 7b). Experiments have shown that the microstructure itself can reduce diatom adhesion compared with smooth PDMS, indicating its ability to decrease available attachment sites to a certain extent. Following the sequential introduction of hydrophilic components via layer-by-layer self-assembly, the synergistic effect between the surface-formed hydration layer and micro-morphology results in enhanced inhibition efficiency against diatoms (9±5 diatoms·mm^{-2} adhered) and *E. coli* (96.2±1.3% bactericidal rate).

For shark-skin-inspired surfaces, riblet-like denticles can modulate near-wall flow and reduce biofouling attachment. Mawignon et al.[178] fabricated a sharkskin-inspired ZnO/PDMS surface combining microscale patterns with ZnO nanostructures, which exhibits a WCA of 151°, a drag reduction of up to 12.2%, and a 96.5% reduction in bacterial adhesion compared with smooth PDMS. Pan et al.[171] further developed stretchable shark-scale-patterned surfaces that reduced the number of adherent bacteria from approximately 6×10^5 to 1×10^5 CFU mL^{-1}. At an elongation ratio of 0.5, bacterial contamination was further reduced by ~40%. Recently, Hsueh et al.[179] developed hierarchical wrinkle arrays inspired by the oriented microwrinkles of caudal shark scales. The optimized surface-functionalized wavy taco-shell architecture reached a WCA of 145.1° and exhibits a 15.1° difference in sliding angle between directions parallel and perpendicular to the wrinkles, facilitating directional liquid transport and achieving >96% bacterial removal relative to the unpatterned substrate when rinsed along the wrinkle direction. These studies highlight multiscale topology, structural deformability, and

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orientation as crucial factors influencing biofouling attachment and release on shark-skin-inspired surfaces.Additionally, some micro-/nanostructured surfaces exhibit mechano-bactericidal capacity by causing morphological damage on bacteria. Ivanova et al. first found that nanopillars on cicada and dragonfly wings can exert lethal effects on attached Gram-negative bacteria, Gram-positive bacteria and spores by physically stretching bacterial cell membranes[180, 181]. Following this mechanism, Qi et al.[182] rationally designed TiO_2 nanowire (NW) arrays with mechano-photocatalytic antimicrobial activity (Fig. 7c). Closely packed TiO_2 NWs with diameters below 100 nm can inactivate >90% of bacteria and prevent >70% of algal attachment via mechanical disruption alone. Benefiting from the inherent photocatalytic activity of TiO_2, the nanowire arrays further generate reactive oxygen species (ROS) under UV irradiation. This synergistic mechanical-photocatalytic approach can eliminate >99% of bacteria within 30 min and inhibit >94% of marine algal fouling. However, purely mechanical bactericidal surfaces encounter the challenge of dead bacteria and cellular debris accumulation during long-term use which masks the nanostructures and then leads to functional failure. Therefore, integrating nanostructures with low-adhesion hydration interfaces has offered an effective route to achieve both fouling reduction and fouling release. Wang et al.[183] developed a bioinspired zwitterionic nanowire coating (ZNW) by grafting PSBMA onto $ZnO@Al_2O_3$ nanowire arrays (Fig. 7d). The high-aspect-ratio nanowires provide a mechanical biocidal effect, while the zwitterionic PSBMA shell forms a nanostructured hydration layer with low interfacial adhesion. In marine algae fouling tests, the ZNW coating reduced algal coverage to less than 30% after 14 days and enabled over 60% fouling release under a mild wall shear stress of 0.4 Pa for 30 min that is four orders of magnitude lower than regular water jet cleaning. Together, these effects reduce the total adhesion energy toward algal biofilms by approximately 60-fold compared with uncoated planar surfaces. This work demonstrates that coupling biomimetic mechanical biocidal nanostructures with zwitterionic hydration layers can simultaneously suppress marine fouling accumulation and promote foulant release under hydrodynamic shear, providing an important design framework for non-toxic and renewable marine antifouling coatings.

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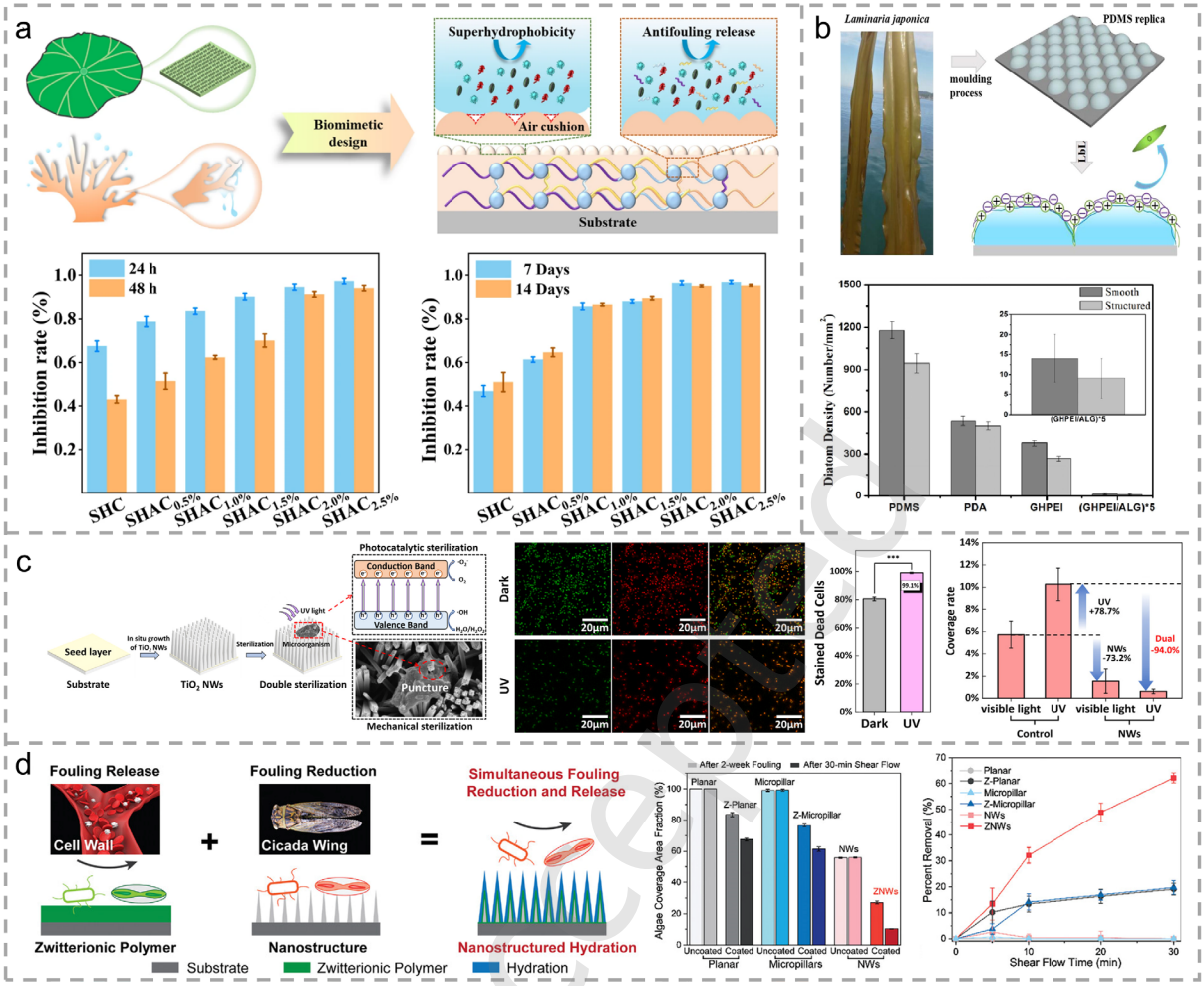

Fig. 7 Biomimetic micro-/nanostructured surfaces for antifouling. a) Schematic diagram of synergistic physicochemical antifouling mechanism (top) and antifouling performance of SHAC (bottom). Reproduced with permission[176]. Copyright 2025, Springer Nature. b) Schematic illustration of *Laminaria japonica*-mimetic surface combining micro-topography and hydrophilic hydration layer to synergistically inhibit diatom settlement (top) and statistical chart of *N. closterium* adhesion (bottom). Reproduced with permission[177]. Copyright 2020, Elsevier. c) Rationally designed TiO_2 nanowire (NW) arrays with mechano-photocatalytic antimicrobial activity (left) and synergistic bactericidal capability (middle) as well as comparison of algae coverage (right) of TiO_2 NWs. Reproduced with permission[182]. Copyright 2025, Wiley-VCH. d) Bioinspired ZNW coating with simultaneous marine fouling reduction and fouling release (left) and algae coverage area fraction on the tested surfaces (middle) as well as percent removal on the tested surfaces as a function of shear flow time (right). Reproduced with permission[183]. Copyright 2024, Wiley-VCH.

Notably, despite the considerable application potential demonstrated by biomimetic micro-/nanostructured coatings, significant challenges remain in transferring them from the laboratory to industrial implementation. Current manufacturing methods for such high-precision microstructures primarily rely on techniques such as photolithography, laser processing, or 3D printing, which are typically expensive, time-consuming, and difficult to implement on large-scale and complex ship surfaces. Future efforts should focus on developing low-cost, scalable

microstructure fabrication techniques with high mechanical durability while integrating stable chemical modifications or multi-mechanism synergistic designs to advance their practical application in marine antifouling.

4 Emerging Antifouling Coatings

Beyond the dynamic and static antifouling coatings discussed above, several emerging antifouling strategies and coating-related platforms have been actively explored. Stimuli-responsive coatings represent an emerging mechanism-level approach that enables adaptive regulation of surface properties or antifouling activity in response to external or local environmental stimuli. MOF-based coatings are then summarized as representative multifunctional material platforms capable of controlled release, catalytic activity, and coating reinforcement. In addition, HEA-based protective surfaces and TENG-related self-powered systems are briefly discussed because of their potential relevance to next-generation multifunctional marine antifouling design, although they are not conventional antifouling coatings in a strict sense.

4.1 Stimuli-Responsive Coatings

Stimuli-responsive coatings exploit specific external or local environmental signals, such as temperature, pH, light, or ionic strength, to regulate interfacial physicochemical properties or antifouling functions, thereby enabling adaptive or on-demand fouling control[184, 185]. This mode of regulation differs from the dynamic surface strategies discussed in section 2, in which chemical surface self-renewal or physical interfacial motion or deformation itself directly contributes to antifouling. In stimuli-responsive coatings, these signals instead alter material or interfacial states and functional outputs that regulate antifouling performance. Examples include changes in polymer conformation, wettability, or surface charge, as well as activation of photothermal or photocatalytic processes and controlled release of antifoulants.

Temperature is one of the most common stimuli that can trigger reversible transitions in polymer chain conformation, interfacial wettability, and surface topography, making temperature-responsive polymers widely recognized in the field of marine antifouling[186, 187]. Poly(N-isopropylacrylamide) (PNIPAAm) is a classic thermoresponsive polymer whose

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hydrogen-bonding interactions vary across its lower critical solution temperature (LCST) of 32 °C. Crossing this threshold induces a reversible transition between swollen and collapsed states, enabling temperature-triggered removal of surface foulants[184, 188]. Nevertheless, a single mechanism is insufficient to sustain antifouling efficacy under complex marine conditions[189]. Furthermore, natural seawater temperature fluctuations cannot provide controllable, on-demand thermal activation, necessitating external sources. Liu et al.[186] successfully developed a smart antibiofouling composite coating with the combination of photothermally-active polyaniline (PANI) and thermoresponsive PNIPAAm (Fig. 8a). It is found that the PNIPAAm/PANI coating can reach 89.3±1.6°C under light irradiation, achieving an antibacterial rate of 99.99%. Subsequently, the WCA of the PNIPAAm/PANI coating reduces from 81° to 35°, whereas the roughness of coating surface enhances upon lowering the temperature to below the LCST of PNIPAAm. These changes in surface wettability and morphology weaken adhesion of dead bacteria to the coating surface, leading to a 99.99% release rate for both *E. coli* and *S. aureus* and achieving a dynamic self-cleaning effect. Moreover, the as-prepared PNIPAAm/PANI coating inhibits protein and algal adhesion by >99%.

pH is another important stimulus for on-demand antifouling because localized acidification can be generated by the metabolic activity of colonizing microorganisms, particularly bacteria, during the early stage of biofilm formation. Therefore, pH-responsive coatings can use these biologically generated microenvironmental changes to trigger antifoulant release or interfacial property switching. Inspired by the smooth-skin and toxin-mediated defenses of the blue-ringed octopus, Zhang et al.[190] developed a pH-responsive antifouling coating (OC-MCSX), as shown in Fig. 8b. The coating constructs a low-surface-energy interface by grafting PDMS brushes onto an epoxy matrix through thermal crosslinking and introducing DCOIT-loaded double-shell microcapsules, which can sensitively detect localized acidification signals generated during the initial stages of microbial colonization, triggering an on-demand release mechanism to form a smart closed-loop system of trigger-eliminate-feedback. The optimized OC-MCS3 coating achieves antibacterial efficiencies of 98.25% and 99.56% against *E. coli* and *S. aureus*, respectively, and inhibits *Chlorella* and diatom adhesion by 88.22% and 93.13%. Its pseudo-barnacle removal strength is only 0.053 MPa. In addition,

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field photographs obtained after 60 days of marine test show lower biofouling coverage on OC-MCS3 than on the control samples. Tan et al.[191] developed a pH-responsive hydrogel (PANA-Cu@His) by coupling a PAA network with histidine-coordinated cupric nanoclusters (Cu@His). Density functional theory simulations reveal a specific coordination configuration between histidine and Cu^{2+} characterized by imidazole coordination and assisted by carboxyl groups. This pH-responsive coordination enabled precise on-demand Cu^{2+} release, with an approximately 127-fold increase in release rate compared with that under neutral conditions.

This mechanism effectively overcomes the issue of excessively rapid initial release in conventional coatings while substantially enhancing the utilization efficiency of the antifoulant. The experimental results have demonstrated that the PANA-Cu@His hydrogel achieves strong bactericidal effect and anti-algal adhesion performance. In addition to controlled release, pH response can also be utilized to drive adaptive reconfiguration of interfacial physicochemical properties. Wei et al.[192] fabricated a pH-responsive amphiphilic smart coating (PDHM) by coupling protonated zwitterionic-tertiary amine polymers with PDMS through condensation (Fig. 8c). By leveraging the differences in pKa values of various functional groups, the surface charge of this coating undergoes pH-responsive transformation. Under seawater conditions (pH ~8.3), negatively charged groups cooperates with the hydration layer to suppress biofouling adhesion. Colonization-induced acidification subsequently protonates the tertiary amines, rendering the surface positively charged, impairing foulant viability, and facilitating detachment under hydrodynamic shear forces. Compared with PDMS, the PDHM coating achieves adhesion inhibition rates of 99.54% 99.59% and 98.23% for *B. subtilis*, *Pseudoalteromonas ruthenica* and *Phaeodactylum tricornutum*, respectively. 16S ribosomal deoxyribonucleic acid (16S rDNA) sequencing results further confirm its outstanding antifouling properties.

Although temperature- and pH-responsive coatings can regulate antifouling performance in response to thermal or biologically generated chemical signals, their activation is often limited by trigger controllability. In contrast, light-responsive antifouling coatings provide a non-contact and remotely controllable stimulation strategy, enabling on-demand activation with high spatiotemporal resolution and pollution-free operation[193, 194]. Over the past decades, titanium dioxide (TiO_2) has attracted widespread interest due to its exceptional photoactivity,

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high physicochemical stability, low cost, ease of availability and low toxicity[195-197]. Its antifouling activity is primarily attributed to light-induced ROS[198]. Vijayan et al.[199] prepared molybdenum disulfide (MoS_2)/Ag-TiO_2 heterostructures (MAT) via a liquid-phase exfoliation method and incorporated them into PU coatings (Fig. 8d). Facilitated by the electron-mediating role of Ag and the interfacial charge separation at the MoS_2/TiO_2 heterojunction, this system effectively suppresses electron-hole pair recombination to enhance photocatalytic activity, achieving an 81% degradation efficiency for crystal violet dye. Under light irradiation, the MAT-based coating demonstrates exceptional antifouling properties, reducing bacterial adhesion by ~95%. Furthermore, to address the functional failure of antifouling coatings caused by the accumulation of bacterial debris and extracellular polymeric substances, Zhou et al.[200] incorporated aminopropyltriethoxysilane-modified Ag-TiO_2 (APTES-Ag@TiO_2) into silicone coatings. It is found that this design leverages photocatalytic activity to degrade the accumulated organic foulants, facilitating surface self-renewal and substantially extending the lifespan of the coating.

Beyond single-stimulus regulation, multi-stimuli-responsive coatings have also been developed to integrate antifouling, anticorrosion, and self-healing functions. Lin et al.[201] recently proposed an awakenable smart-responsive hydrogel armor coating (HZC) that integrates a polyamide-phenol (PAE) self-healing network with a PVA/borax dynamic network to form a DN hydrogel matrix (reinforced by CNF for mechanical properties) and incorporates a pH- and photothermal-responsive ZIF-8@BTA@MPN smart release unit modified with a metal-phenol network (MPN), as shown in Fig. 8e. Localized acidification induced by microorganisms can trigger the on-demand release of the corrosion inhibitor BTA from ZIF-8 nanocontainers. Under near-infrared irradiation, the MPN shell undergoes photothermal conversion, accelerating BTA release. Simultaneously, the temperature increase activates the shape-memory effect of the epoxy matrix to promote crack closure, synergizing with the reversible restructuring of dynamic boronate bonds to achieve self-healing.

Notably, the stimuli-responsive approaches discussed above mainly operate through solid-phase coating surfaces, where fouling organisms, regarded as soft or hard solid foulants, adhere via solid-solid contact. Another method is to replace this solid adhesion substrate with a mobile

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liquid layer, thereby converting the dominant fouling-contact pathway from solid–solid to solid–liquid contact. This interfacial conversion is achieved by slippery liquid-infused porous surfaces (SLIPS). Inspired by Nepenthes[202, 203], SLIPS forms a continuous and smooth liquid barrier layer by trapping lubricants within its porous structure. Owing to this lubricant-mediated low-adhesion interface, SLIPS exhibits outstanding fouling-release properties against foulants and can rapidly regain hydrophobicity after physical damage, resist ice adhesion, contaminant attachment and maintain pressure stability[204, 205]. Similar evidence has also been provided by liquid-entrenched smooth surfaces (LESS), in which a molecularly grafted PDMS layer stabilizes a thin lubricant film on smooth substrates and repels liquids, bacteria, and viscoelastic solids with dynamic viscosities spanning over nine orders of magnitude[206], further confirming the importance of stable liquid interfaces in reducing foulant adhesion. However, conventional SLIPS still suffers from durability issues as gradual the lubricant depletion can weaken the low-adhesion of liquid interface, thereby reducing long-term antifouling performance. To address this challenge, stimuli-responsive mechanisms have been incorporated into SLIPS to regulate lubricant retention, release and interfacial mobility, thereby improving the durability and adaptability of slippery antifouling interfaces. Inspired by hagfish mucus defense, Tong et al.[207] introduced a host-guest supramolecular interaction between Azo and α-cyclodextrin (α-CD) to modulate the lubricity of SLIPS. Under visible light or heating, Azo undergoes isomerization and combines with α-CD, driving shrinkage of polymer chains to squeeze stored lubricant to the surface and then enabling the antifouling mode to intelligently switch between enhanced and conventional modes to cater to different conditions. Additionally, disulfide and hydrogen bonds are incorporated to improve self-healing efficiency (91.73%). Drawing on the toxic-mucus defense of *P. pavoninus*, the team subsequently developed a coumarin-functionalized SLIPS (CmSLIPS)[20]. Reversible coumarin photodimerization controlls locking and unlocking mode between lubricant and matrix, while its intrinsic antimicrobial activity strengthens biocidal antifouling effect and prolongs antifouling durability.

Despite their lubricant self-replenishment capability, SLIPS undergo a transient lubricity loss before surface recovery, increasing their susceptibility to biofouling and compromising long-term protection. Inspired by the skin of a poison dart frog which contains venom glands

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and mucus, Tong et al.[208] infused a ZIF-8-based porous liquid lubricant into a silicone polyurethane (SPU) matrix to construct slippery porous-liquid-infused porous surfaces (SPIPS) for marine antifouling (Fig. 8f). SPIPS employs an intelligent switching mechanism between defensive and offensive modes. Under normal conditions, PL, as a stable lubricating layer, provides physical defense (defensive mode). When exposed to specific light stimuli, the photosensitizer (NBA)-induced microenvironmental acidification triggers the disassembly of the ZIF-8 framework, releasing the encapsulated eco-friendly antifoulant (Tralopyril) to actively kill fouling organisms (offensive mode). This on-demand response approach significantly reduces antifoulant waste while effectively extending the lifespan of the coating in complex marine environments. Moreover, it maintains effective antifouling performance after 360 days in actual marine fields during boom seasons.

Beyond the regulation of lubricant behavior, responsive SLIPS can be further integrated with mechanically dynamic surface structures to achieve multimodal antifouling. Inspired by the wielding poisonous tentacles of sea anemones, Tong et al.[209] proposed a 4D SLIPS coating (FSLIPS) with biomimetic cilia for marine antifouling. The bionic cilia can sway in 3D space driven by a magnetic field, creating physical disruption against the attachment of microorganisms (physical defense). Simultaneously, the light-sensitive nitric oxide (NO) donor (BNN6) integrated within the coating responds to light in one-dimensional (1D) time to release NO, thus realizing a controllable biocide impact on microorganisms (chemical defense). This 4D dynamic defense system, which combines field-driven interfacial motion with light-triggered chemical release, demonstrates tremendous potential for marine industrial applications.

In summary, early studies primarily focused on stimulus-induced regulation of individual interfacial properties or antifouling functions, whereas recent systems, such as 4D SLIPS and awakenable hydrogel armor, increasingly integrate responsive regulation with complementary mechanisms, including field-driven dynamic interfacial motion and self-healing, to achieve multimodal protection under complex marine conditions. However, increasing functional integration places greater demands on the long-term stability and reliability of responsive functions. Future efforts should concentrate on improving the robustness of multi-responsive

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systems and validating their reliability under long-term marine immersion, rather than simply increasing the number of stimuli or functional components.

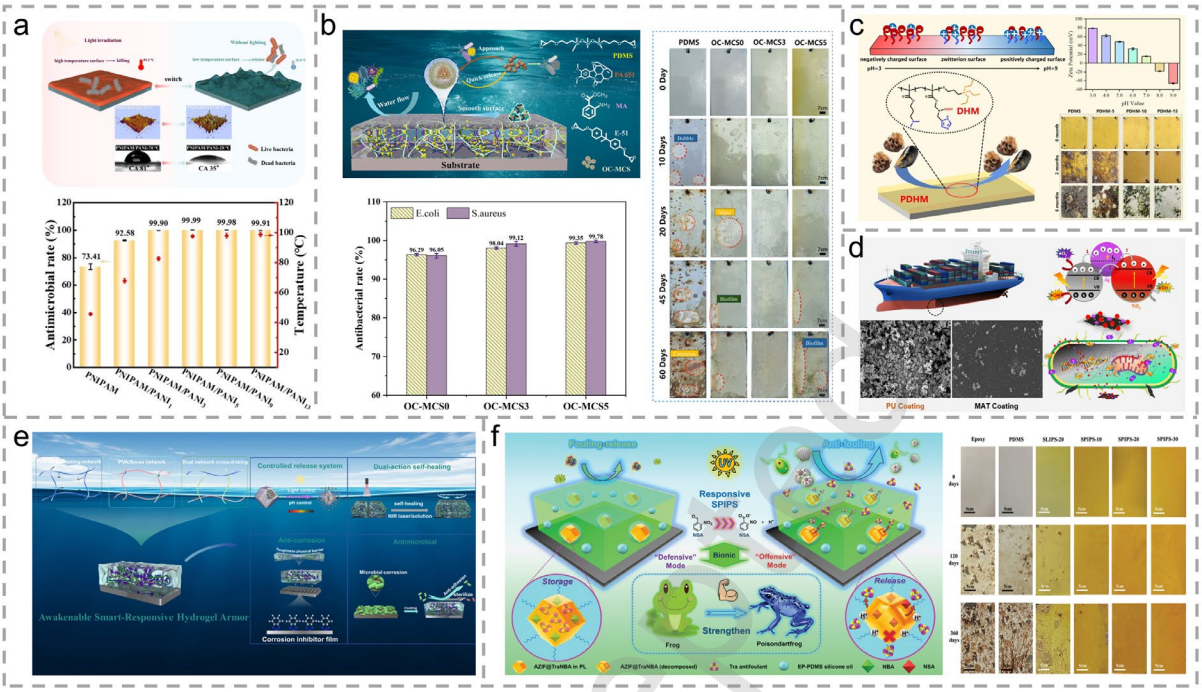

Fig. 8 Stimuli-responsive coatings for antifouling. a) Schematic diagram of the antifouling mechanism of the PNIPAAm/PANI coating (left) as well as photothermal antibacterial rates and the corresponding photothermal temperature of the PNIPAAm/PANI coating (right). Reproduced with permission[186]. Copyright 2024, Elsevier. b) Schematic diagram of the antifouling mechanism of OC-MCSX composite coating (left top) and corresponding antibacterial rate (left bottom) as well as antifouling effect of seawater immersion (right). Reproduced with permission[190]. Copyright 2026, Elsevier. c) Schematic of the antifouling mechanism and pH response of the PDHM coating. Reproduced with permission[192]. Copyright 2026, Elsevier. d) A scheme showing the plausible antibiofouling mechanism of the PU coating with the MAT nanocomposites. Reproduced with permission[199]. Copyright 2024, American Chemical Society. e) Schematic illustration of the antifouling mechanism of the awakenable HZC coating. Reproduced with permission[201]. Copyright 2025, American Chemical Society. f) Design and response mechanism of SPIPS (left) and marine field in the Zhoushan Sea over 360 days (right). Reproduced with permission[208]. Copyright 2024, Wiley-VCH.

4.2 MOF-Based Coatings

MOFs are 3D porous crystalline materials with periodic network structures self-assembled via coordination bonds, in which metal ions or clusters serve as nodes and organic ligands as linkers[210, 211]. Owing to their highly designable structures, tunable pore environments, large specific surface areas, and diverse functional sites[212-214], MOFs have considerable application potential in numerous fields including gas storage and separation[215, 216], ion adsorption[217, 218], chemical catalysis[219], and guest molecule delivery[220]. Recently, these structural and chemical

advantages have prompted the development of MOF-based marine antifouling coatings, providing a versatile platform for constructing high-performance synergistic antifouling systems. Consequently, recent advances in MOF-based antifouling coatings can be discussed from three main aspects: (1) enabling the controlled release of guest molecules through MOF nanocontainers, (2) exploiting intrinsically active MOFs for metal-ion release or enzyme-like catalytic antifouling, and (3) constructing MOF-based composite architectures to enhance antifouling performance, mechanical robustness, and coating stability.

(1) Enabling controlled release through MOF nanocontainers. One of the most direct approaches is to exploit the intrinsic high porosity of MOFs as nanocontainers for the encapsulation of functional guest molecules, which protects the active molecules while enabling their controlled release through structural modulation of the MOFs, thus prolonging lifespan of the coating. As an example, Zhao et al.[221] fabricated a coating by blending an acrylic resin with UiO-66-NH_2 nanocontainers that had been efficiently loaded with carboxylated benzisothiazolinones (BIT-COOH) antifoulant via hydrogen bonds and electrostatic interactions (Fig. 9a). Experimental results demonstrate that the coating exhibits excellent antibacterial and antifouling properties even at a low loading level (4 wt%). Beyond a single antifouling function, this strategy can be expanded to integrate both antifouling and anticorrosion capabilities. Zhang et al.[222] reported a self-renewing zwitterionic hydrogel coating with corrosion inhibitor benzotriazole (BTA)-loaded UiO-66 (BU), which exhibits both antifouling and anticorrosion performance (Fig. 9b). The polymer is synthesized through the copolymerization of MDO and vinyl chloroacetate (VCA), followed by grafting with 2-(trimethoxysilyl) propyl dimethylamino propionate (TPD). It is capable of undergoing self-crosslinking or forming a crosslinked network with BU, while also establishing chemical bonding with the substrate to generate a stable interfacial structure. As a result, the coating displays favorable mechanical strength and strong adhesion. Owing to the continuous renewal of the zwitterionic surface during polymer degradation and hydrolysis, the coating exhibits pronounced resistance to bacteria and diatoms. Meanwhile, these degradation and hydrolysis processes facilitate sustained release of BTA, thereby effectively suppressing corrosion.

(2) Exploiting intrinsically active MOFs for ion-release and catalytic antifouling.

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Beyond serving as nanocontainers, MOFs can also function as intrinsically active components by using their metal nodes, framework chemistry, or catalytic activity to achieve active antifouling. For ion-mediated antifouling, an important design objective is not simply to increase metal-ion release, but to stabilize the active valence state and regulate its release kinetics in seawater. He et al.[223] developed a hierarchical Cu(I)-MOF/PPy composite through a redox concomitant formation strategy, in which Cu(I) sites provided sustained Cu^{+} release while the PPy phase supplied complementary photothermal activity. The resulting composite exhibits broad-spectrum activity against marine bacteria, algae, and surface-associated biofilms, reaching 100% eradication efficiency under solar-driven heating.

Moving from active-material design toward practical marine coatings, regulation of the coordination environment and metal valence becomes particularly important for achieving sustained antifouling activity with reduced metal loading. Ren et al.[224] converted HKUST-1 into mixed-valence Cu(I,II)-BTC through *in situ* reduction and subsequently incorporated it into an acrylic silicone self-polishing antifouling coating (Fig. 9c). By taking advantage of the stabilization effect of the MOF framework on Cu(I) sites and controllable hydrolysis of Cu-O bonds, the coating enables sustainable release of Cu(I) ions in seawater and enhances antibacterial efficacy through the generation of ROS. The 1.8/Cu(I,II)-BTC sample maintains a Cu(I) release rate of 37.5 μg/(mL·day) after 30 days of seawater immersion with a bactericidal efficacy of 90% and 80% against *E. coli* and *S. aureus*. Following 6 months of field seawater exposure, algal coverage of the coating remains at 6.7% with no attachment of large organisms. Notably, the total copper content of the coating is ~9.5 wt%, which is significantly lower than that of conventional commercial Cu_2O coatings (20-50 wt%), highlighting its potential for durable antifouling application with low copper loading.

Furthermore, inspired by the natural defense mechanisms of marine organisms, MOFs with enzyme-like catalytic activity, namely MOF nanozymes, have gained widespread attention in the design of marine antifouling materials. MOF nanozymes operate through catalytic cycles in which the active centers are not consumed during the reaction. This feature is expected to prolong the durability of active antifouling functions by reducing reliance on the continuous depletion of antifoulants or metal ions. Zhou et al.[225] first reported a cerium-based MOF (Ce-

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MOF-808) with natural haloperoxidase (HPO)-like activity. In the presence of hydrogen peroxide (H_2O_2) and bromide ions (Br^-), it can catalyze the oxidation-bromination reaction to produce hypobromous acid (HOBr), which directly destroys bacterial cell structures to achieve bactericidal effects. It also causes halogenation and inactivation of bacterial quorum sensing (QS) signaling molecules (AHLs), thereby interrupting bacterial communication and fundamentally inhibiting biofilm formation (Fig. 9d). However, conventional approaches to stably immobilize these nanocatalysts on a substrate typically involve simply blending the nanoenzymes into the resin matrix as fillers, which leads to significant encapsulation of the active sites by the polymer, thereby reducing the catalytic activity. Consequently, Xu et al.[226] first integrated plasma electrolytic oxidation (PEO), chemical etching, and solvothermal growth to fabricate a PEO/UiO-66(Ce) nanozyme coating in situ on titanium alloy. The hydroxyl-rich nanonetwork confines and firmly immobilizes UiO-66(Ce), thereby increasing active-site accessibility. The resulting coating shows an antibacterial efficacy of 97.8% and adhesion rate of only 3.4% against *Halamphora* sp. after three days.

(3) Constructing MOF-based composite architectures for synergistic performance enhancement. In addition to their roles as nanocontainers or intrinsically active components, MOFs can serve as functional building blocks in composite coating architectures, enabling the integration of antifouling activity, mechanical robustness, and interfacial stability. Inspired by the brick and mortar structure of pearl, Guo et al.[227] fabricated a brick-and-mortar nanocomposite coating, comprising ZIF-8 brick, modified polydimethylsiloxane (NPDI) mortar and SiO_2 surface layer (Fig. 9e). The coating combines a superhydrophobic antiadhesion barrier with active antibacterial capability through released zinc ions, demonstrating an antibacterial efficacy of >99% against *E. coli*, 85.4%-95.6% against *S. aureus*, as well as an anti-algal adhesion rate of >96%. Meanwhile, its pearl-like architecture enhances coating toughness, impact resistance and overall mechanical durability. Similarly, Xu et al.[228] developed a hydrogel-anchored MOF-based coating (HMC), as shown in Fig. 9f. The coating couples HKUST-1 bricks with a biopolymer hydrogel mortar through *in situ* biomineralization, thereby achieving universal adhesion to various substrates and excellent mechanical stability. Importantly, HMC exhibits excellent antifouling performance, significantly reducing BSA

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adsorption to only 0.13±0.01 mg·cm^{-2}, and achieving a bactericidal efficacy of 100% through the hydrolysis of HKUST-1 to release Cu^{2+}. Furthermore, the coating reduces the corrosion current density to 0.45±0.19 nA·cm^{-2} in the presence of microorganisms and exhibits pH-responsive Cu^{2+} release properties, making it suitable for intelligent protection against microbiologically influenced corrosion (MIC). In another study by the team[229], a chiral MOF-based dispense-then-kill antifouling coating was developed (Fig. 9g). The coating utilizes MPN as a universal adhesion intermediate layer to achieve stable adhesion to multiple substrates, and a copper-based MOF layer (D-Met@MOF) with D-amino acids as organic ligands is constructed *in situ* on top of it. D-amino acids induce the dispersion of biofilms, while the copper-mediated Fenton-like reaction synergizes with the mechanical bactericidal effect of the nanostructure to further eliminate the dispersed bacteria. Furthermore, its blue fluorescence properties also help inhibit microalgal adhesion. Overall, this work effectively integrates biofilm regulation, chemically driven bactericidal and structural effects, providing a new design approach for developing sustainable antifouling coatings that leverage synergistic multi-mechanism interactions in complex marine environments.

In summary, MOF-based antifouling coatings provide a versatile platform for marine antifouling by serving as controlled-release nanocontainers for functional guest molecules, intrinsically active components for metal-ion release or catalytic defense, and functional building blocks for multi-mechanism synergistic systems. Their advantages lie in integrating porous storage, tunable metal-node chemistry, catalytic activity, and interfacial design within one material platform. However, the practical performance of MOF-based coatings depends on maintaining framework stability, active-site accessibility, controlled release behavior, and strong matrix integration under long-term seawater exposure while minimizing ecological risks associated with metal ions or reactive species. Future efforts should move beyond simple MOF incorporation toward scalable, mechanically robust, and environmentally safe coating architectures that fully exploit the structural and chemical tunability of MOFs.

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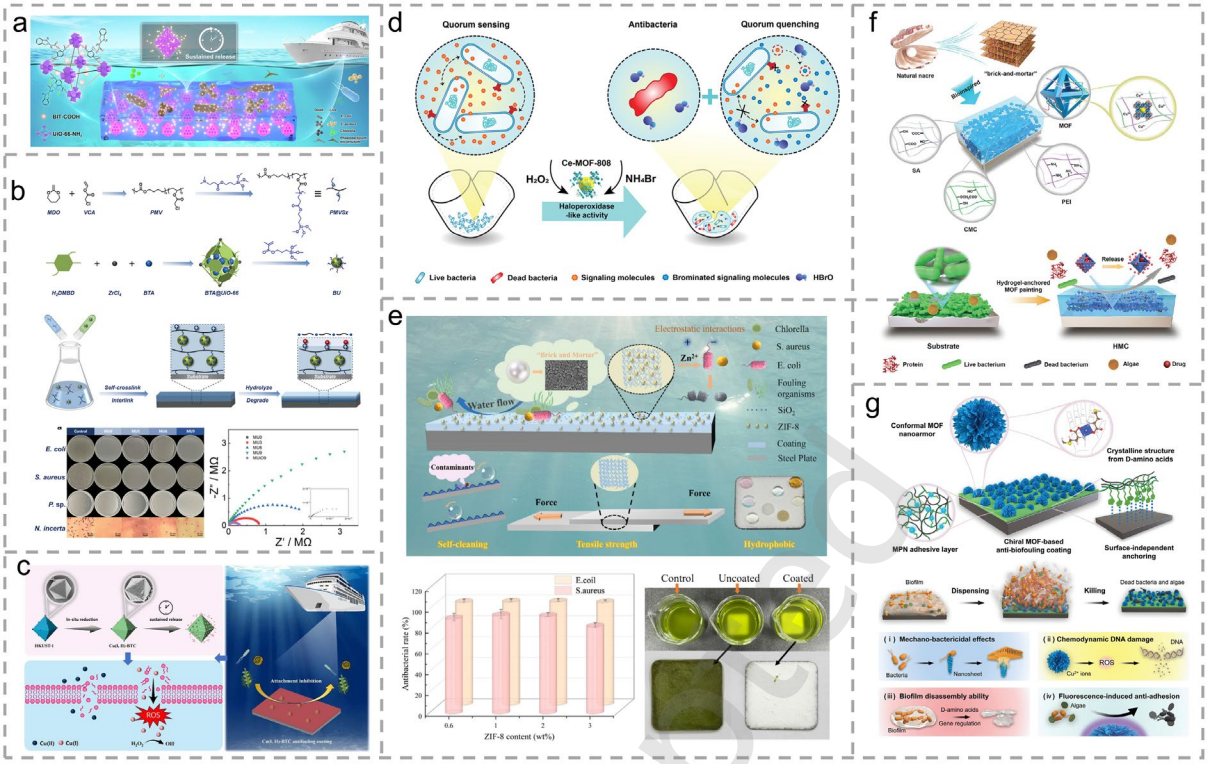

Fig. 9 Strategies of MOF-based coatings. a) Schematic illustration of the antifouling mechanism of UiO-66-NH_2@BIT-COOH incorporated acrylic coating. Reproduced with permission[221]. Copyright 2022, Elsevier. b) Illustration of preparation of the self-regenerating zwitterionic hydrogel coating with BTA-loaded MOF (top) and corresponding antifouling performance (left bottom) as well as Nyquist plots (right bottom). Reproduced with permission[222]. Copyright 2025, Wiley-VCH. c) Illustration of the antifouling mechanism for Cu(I, II)-BTC antifouling coating.. Reproduced with permission[224]. Copyright 2025, Elsevier. d) Schematic illustration of Ce-MOF-808 for antibacterial activity and inhibiting the formation of biofilms based on its HPO-like activity. Reproduced with permission[225]. Copyright 2022, Wiley-VCH. e) Schematic diagram of the antifouling mechanism of ZIF-8@SiO_2/NPDI coating (top) and corresponding antifouling performance (bottom). Reproduced with permission[227]. Copyright 2025, Elsevier. f) Schematic illustration of the HMC with anti-fouling-corrosion properties, including the organized microstructure formed by crystalline MOFs and amorphous hydrogel via multi-scale crosslinking interactions (top), as well as the corresponding antifouling and anticorrosion mechanism (bottom). Reproduced with permission[228]. Copyright 2023, Wiley-VCH. g) Schematic illustration of the chiral MOF-based anti-biofouling coating, including the organized microstructure composed of an amorphous MPN layer and crystalline chiral MOFs (top), as well as the four corresponding protection modes (bottom). Reproduced with permission[229]. Copyright 2024, Wiley-VCH.

4.3 HEA-Based Metallic Protective Surfaces

Conventional metal materials typically face an inherent trade-off among mechanical properties, corrosion resistance, and antifouling capability under harsh marine conditions[230]. HEAs, comprising at least five principal elements, offer a new material platform[231-233]. Their characteristic high-entropy effect, sluggish diffusion, lattice distortion, and cocktail effect favor

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the formation of simple and disordered solid-solution structures including face-centered cubic (FCC), body-centered cubic (BCC), and hexagonal close-packed (HCP) structures, thereby creating favorable conditions for the design of alloys with unique properties[234-236]. Since the initial proposal by Yeh and Cantor et al. in 2004[237, 238], numerous HEAs have been explored across diverse applications, including antifouling materials[239, 240]. For instance, Ren et al.[233] proposed a new $Co_{0.4}FeCr_{0.9}Cu_{0.3}$ HEA. The as-prepared HEA achieves an antibacterial efficacy of >99.9% against both *E. coli* and *S. aureus* through the combined effects of Cu ion release and contact killing by Cu-rich phase after 24 h, significantly exceeding that of conventional antibacterial Cu-bearing stainless steel (71.50% and 80.84% antibacterial rate) while maintaining excellent corrosion resistance and mechanical properties.

Although Cu-bearing HEAs show clear antifouling potential, their antibacterial performance primarily depends on Cu ion release and direct contact with Cu-rich phase, yet excessive Cu dissolution may damage the passive film and increase MIC risks. Yang et al.[241] developed $Al_xCoCrCuFeNi$ (x = 0, 0.1, 0.3, 0.5) HEAs (Fig. 10a). Through precise control of the composition, $Al_{0.3}$HEA simultaneously improves mechanical performance, antifouling and anti-MIC properties, with a biofilm inhibition rate of 94.1% against *P. aeruginosa* and anti-algal rates of 88% and 96% against *Chlorella* and *P. tricornutum*, respectively. More importantly, its polarization resistance (R_p) is 20-fold that of Al_0HEA. This enhancement arises from the dual functionality of Al in stabilizing the Cu-rich phase to suppress excessive Cu dissolution and promoting the formation of Al- and Cr-rich oxides in passive films.

The importance of regulating Cu content and its microstructural distribution have also been examined in coating form. Xu et al.[242] fabricated a range of $CoCrFeNiCu_x$ HEA coatings by employing the high-speed laser cladding and systematically investigated effects of Cu addition on microstructural evolution, corrosion behavior and antifouling capability (Fig. 10b). The results demonstrate that the $CoCrFeNiCu_{0.3}$ coating offers the optimal compromise between corrosion resistance and antifouling performance, as minor Cu doping primarily promotes the enrichment of Cu particles near grain boundaries rather than forming distinct Cu-rich phase, thereby limiting micro-galvanic interactions while maintaining a sustained release of Cu ions.

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Building on Cu-based systems, Ag-containing HEAs have also been explored to broaden the antibacterial spectrum. Zhou et al.[243] conducted a systematic comparison among Cu-AgHEA, AgHEA, and CuHEA on a CoCrFeNi matrix, demonstrating that the Cu-Ag metastable phase in Cu-AgHEA helps reduce formation of defects in the passive film while the simultaneous release of Cu and Ag ions promotes ROS generation, achieving an antibacterial rate of 99.9% against marine bacteria.

Extending this Ag-alloying strategy to coatings, Chen et al.[244] fabricated $FeCrNiAl_{0.6}Ti_{0.6}Ag_x$ HEA coatings on 304 stainless steel via laser cladding (Fig. 10c). Owing to a dense Cr_2O_3- and TiO_2-rich passive layer, the $Ag_{0.3}$ coating exhibits the highest corrosion potential (-106.0 $mV_{vs.\ Ag/AgCl}$) and polarization resistance ($4.41 \times 10^5\ \Omega \cdot cm^2$) in 3.5 wt% NaCl solution, whereas the $Ag_{0.4}$ coating achieves an antibacterial rate of 73.74% against *P. aeruginosa* after 24 h through the synergistic effects of contact killing by Ag precipitates and ROS generation promoted by Ag ion release. However, excessive Ag addition increases the interphase potential difference and slightly weakens corrosion resistance. These studies suggest that the content, distribution, and phase state of antibacterial elements in HEA systems should be optimized within an appropriate window to balance antifouling performance with passive-film stability and long-term corrosion resistance.

4.4 TENG-Assisted Self-Powered Antifouling Systems

TENGs, as an emerging energy harvesting technology based on the principles of contact electrification and electrostatic induction, can convert mechanical energy in marine environments such as waves and wind into electrical signals while providing continuous electrical output for interface control under self-powered conditions, thereby enabling active antifouling. Recent studies[245-247] have demonstrated several routes for TENG-assisted antifouling, including direct electric-field regulation of microorganism adhesion and viability, TENG-driven seawater electrolysis, and TENG-powered ultraviolet C (UVC) sterilization, which helps to overcome the limited external energy supply of conventional active antifouling approaches.

Applying electric fields to directly hinder microorganism attachment or inactivate attached

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microorganisms serves as a fundamental approach for antifouling. Long et al.[246] proposed a wave-driven TENG comprising Kapton and copper films (Fig. 10d), which generates a low-frequency and low-intensity alternating electric field between conductive electrodes to enable self-activated antifouling. The underlying mechanism is attributed to the continuous perturbation of the double layer at the electrode and solution interface by the electric field, which impedes the formation of conditioning film during the early stage. More recently, Li et al.[248] developed a dual-electric-field self-powered antifouling system by harvesting wind and wave energy. The wind-driven high-voltage output generated enhanced local electric fields at Cu_7S_4 nanowire tips, inducing electroporation and ROS production. At a flow rate of 1500 mL min^{-1}, the system achieved a bactericidal rate of 99.99% against both *E. coli* and *S. aureus* within 10 min. Meanwhile, the wave-driven low-frequency alternating electric field continuously disturbed the interfacial electrical environment and reduced bacterial adhesion.

TENG output can also be directly coupled with seawater electrolysis for microorganism inactivation. Liu et al.[247] developed a wind-driven rotational direct-current TENG for the self-powered inactivation of seawater microorganisms. At a wind speed of 10 m s^{-1}, the device generated an open-circuit voltage of 450 V and a short-circuit current of 11 μA, driving seawater electrolysis to produce approximately 0.8 mg L^{-1} chlorine within 60 min for the inactivation of marine microorganisms.

Besides, TENG-generated electricity can further power UVC sources for non-contact sterilization. Chen et al.[245] proposed a self-powered UVC sterilization system based on a modified freestanding rotating TENG (MFR-TENG) with discharge excitation capability (Fig. 10e). The breakdown discharge effect enables an output voltage of 6.9 kV, approximately three times that generated without excitation, providing sufficient energy for UVC emission. Following a ten-minute sterilization period, the system achieves an antibacterial rate of ~75% against mixed marine bacterial communities, significantly surpassing the 44% observed without discharge.

Notably, harsh marine conditions, including high humidity, biofouling, and repeated mechanical friction, can significantly degrade the output performance of TENGs and restrict their application potential in marine environments. Therefore, Sun et al.[249] addressed these

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limitations by incorporating MXene into PVA to enhance dielectric properties, wear resistance, and moisture tolerance, while a biomimetic hydrophobic polyvinylidene fluoride (PVDF) layer further improved moisture resistance. The resulting TENG maintained an open-circuit voltage of 1056 V at 95% relative humidity.

In summary, HEA-based protective surfaces and TENG-related self-powered systems represent two emerging coating-related directions beyond conventional polymer-based antifouling coatings. HEAs can integrate mechanical robustness, corrosion resistance, and antifouling performance through multi-component alloy design, phase regulation, and antibacterial element control, but their practical application still depends on scalable and cost-effective fabrication and long-term stability in seawater. TENG-related systems, although not coatings in a strict sense, harvest marine mechanical energy to drive electric-field interference, electrochemical generation of reactive species, or UVC sterilization, yet maintaining stable output, moisture resistance, and long-term durability remains a significant challenge. Integrating HEA-based structural stability with TENG-assisted energy harvesting and active actuation, such as using antifouling HEAs as corrosion-resistant electrodes or structural substrates, may provide a promising route toward mechanically robust and self-powered marine protection systems.

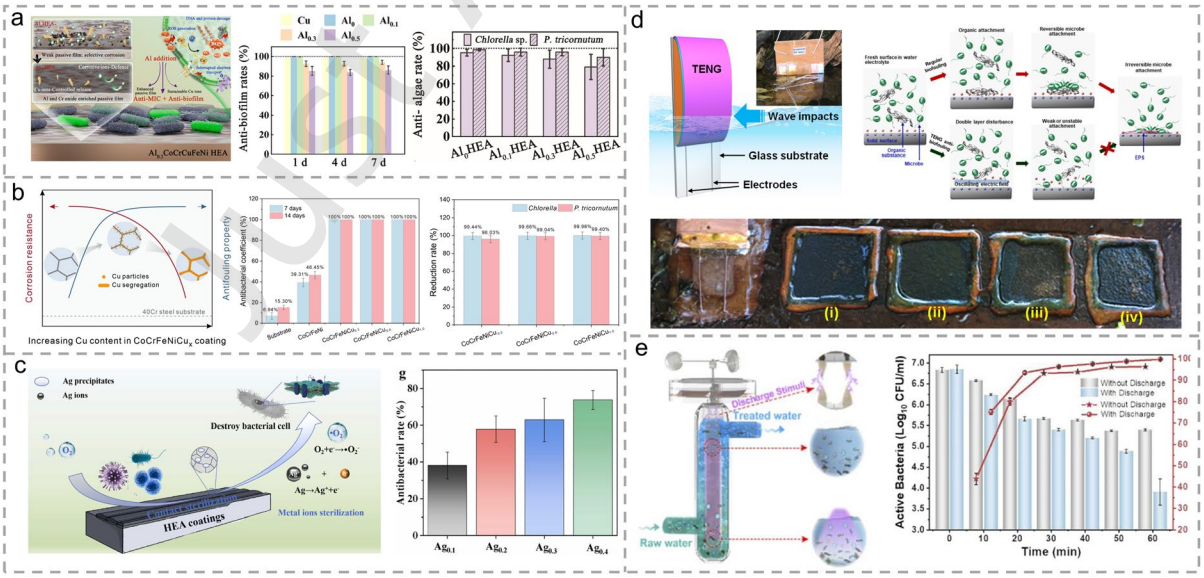

Fig. 10 Emerging coating-related antifouling materials and self-powered systems. a) Schematic diagram of anti-biofilm and anti-MIC mechanisms of $Al_{0.3}$CoCrCuFeNi HEA (left) and corresponding antifouling performance (right). Reproduced with permission[241]. Copyright 2025, Wiley-VCH. b) Schematic illustration of Cu-regulated antifouling-corrosion resistance balance in CoCrFeNiCu$_x$ HEA coatings (left) and

corresponding antifouling performance (right). Reproduced with permission[242]. Copyright 2025, Elsevier. c) Schematic diagram of the antibacterial mechanism of the $FeCrNiAl_{0.6}Ti0.6Ag_x$ HEA coatings (left) and corresponding antibacterial performance (right). Reproduced with permission[244]. Copyright 2025, Elsevier. d) Schematic illustration of the setup, mechanism, and on-site performance of water-driven TENG antifouling. Reproduced with permission[246]. Copyright 2019, Elsevier. e) Schematic diagram of the antifouling mechanism of a self-powered antifouling UVC pipeline sterilizer based on a modified freestanding rotary triboelectric nanogenerator (left) and corresponding antibacterial performance (right). Reproduced with permission[245]. Copyright 2022, Elsevier.

5 Summary and Outlook

In this review, recent advances are summarized in terms of dynamic surface antifouling, static surface antifouling, and emerging antifouling approaches (**Table 1**). Specifically, chemical dynamic coatings consisting of degradable polymers achieve antifouling performance through self-renewal mechanisms. However, these coatings are inherently consumable, and the lifetime is closely related to their thickness and degradation rate. Incorporating eco-friendly antifoulants may assist in prolonging their effective antifouling lifetime. Physical dynamic coatings avoid material consumption by generating deformable or unstable interfaces under flow, electric or magnetic fields, thereby disrupting foulant settlement and promoting detachment. However, they still face challenges associated with energy supply, actuation stability, as well as long-term durability. Combining them with other antifouling strategies, such as the incorporation of fouling-resistant groups, is expected to enhance their overall performance under conditions where external energy supply is unavailable. FRCs weaken interfacial adhesion through low surface energy, low elastic modulus, and smooth interfaces, allowing attached organisms to be removed by hydrodynamic shear. Nevertheless, poor mechanical strength, weak substrate adhesion, and limited antifouling performance remain challenging. Developing polymer-ceramic hybrid coatings, further endowed with dynamic-bond-mediated self-healing provides practical routes to improve antifouling performance and durability. Fouling-resistant coatings based on PEG, zwitterions and hydrogels utilize a dense hydration layer as a physical barrier to suppress protein adsorption and microbial attachment. However, the trade-off among high hydrophilicity, low swelling ratio and high mechanical stability remains challenging. Integrating hydrophobic crosslinked networks with surface self-enrichment strategies may help preserve hydration-mediated resistance while improving

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stability. Biomimetic micro-/nanostructured coatings demonstrate unique antifouling potential by mimicking natural topographies such as lotus leaves, shark skin and cicada wings. Yet their limited scalability and high fabrication cost still hinder their practical implementation. Therefore, there is an urgent need to develop simple, reproducible and mechanically durable structuring methods compatible with industrial-scale fabrication. In addition to conventional dynamic and static antifouling coatings, emerging approaches have also been discussed, including stimuli-responsive coatings, MOF-based coatings, HEA-based protective surfaces, and TENG-assisted self-powered systems. These methods expand antifouling design by introducing environment-adaptive interfacial regulation, controlled release, catalytic or ion-mediated defense, corrosion resistance, and energy-assisted protection. Designing optimal coating structures in conjunction with these methods may improve antifouling efficiency, extend service life, and promote the development of multifunctional marine protection systems.

Although significant progress has been achieved, the practical application of marine antifouling coatings remains constrained by several unresolved issues. Real marine environments are harsh and complex, with temperature fluctuation, salinity variation, pressure, UV exposure and abrasion collectively influencing coating performance. In addition, the diversity of fouling organisms requires coatings capable of resisting multiple species and multiple fouling stages rather than a single model organism under simplified laboratory conditions. Long-term material stability is equally important because degradation, wear, swelling, lubricant depletion, and changes in interfacial structure can progressively weaken the mechanisms responsible for antifouling during service. These processes may also increase the release of coating-derived species into the surrounding seawater, thereby linking coating durability directly with environmental sustainability. This issue is particularly relevant to systems containing Cu-, Ag-, or Zn-based components, DCOIT, or nanomaterials, in which antifouling activity can involve or be accompanied by the release of metal ions, biocides, nanomaterial particles, or transformation products, potentially causing adverse effects on non-target marine organisms when their release is not adequately controlled[250-253]. Degradable and lubricant-infused coatings also require consideration of the environmental fate of degradation products and lubricant residues. Consequently, long-term sustainability should be assessed

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together with release kinetics, environmental fate, and ecotoxicological effects rather than inferred from antifouling efficacy alone. The persistence of the polymer matrix is another relevant factor, since non-degradable fragments generated during coating erosion and maintenance can contribute to marine microplastic emissions. Therefore, reducing coating-derived microplastic generation through degradable, recyclable, or erosion-resistant coating designs should be considered an important component of sustainable antifouling development.

The antifouling strategies reviewed here, fouling release, fouling resistance, and biocidal action, play an important role in marine antifouling. However, dependence on a single mechanism is often inadequate for complex marine environments. Therefore, synergistic coatings that integrate multiple antifouling mechanisms are needed to achieve more durable and broad-spectrum performance. Specifically, combinations of fouling-release and fouling-resistant functions may offer immediate practical potential by reducing initial adhesion while facilitating the detachment of already attached organisms. Moreover, integrating emerging strategies such as stimuli-responsive coatings, MOF-based coatings, HEA-based protective surfaces, and TENG-assisted self-powered systems, which require stable responsiveness, controllable functional output, substrate adhesion and long-term durability, with mechanically robust, self-healing and environmentally compatible coatings may improve antifouling efficiency, extend service life, and promote the development of multifunctional marine protection systems.

Notably, a clear mismatch remains between laboratory evaluation and real marine validation. Coatings that exhibit good performance under controlled testing conditions may fail to maintain comparable effectiveness in real marine environments. In addition to field validation, comparison with commercial products provides an important reference for judging whether improvements observed in laboratory studies translate into practically meaningful performance. In this review, the fluorine-free amphiphilic coating showed higher N. incerta removal than the commercial 1100SR coating[101], while the barnacle removal strength of the zwitterionic silicone coating remained within the <0.4 MPa range reported for commercial FRCs[107]. The Cu(I,II)-BTC coating achieved six-month marine protection with a total Cu content of ~9.5 wt%, compared with 20-50 wt% for conventional commercial Cu_2O

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coatings[224]. These examples provide useful practical reference points, but comparable commercial benchmarks remain limited for many of the emerging approaches discussed in this review, for which the available evidence is still dominated by laboratory testing or field demonstrations. Moreover, the availability of scalable manufacturing routes, reasonable production costs, and compatibility with existing coating application processes are also critical for translating emerging antifouling technologies into commercial use. Recent progress in characterization techniques makes their incorporation into marine antifouling coating evaluation increasingly necessary, as these methods can help bridge the gap between laboratory studies and real-world applications. In parallel, introducing computer simulations and artificial intelligence into coating development may accelerate the establishment of correlations among coating composition, interfacial properties, environmental variables, and long-term field performance. Such data-assisted approaches can enable more efficient multi-objective optimization of antifouling efficacy, durability, environmental safety, cost, and processability, providing a promising avenue for the rational design of novel marine antifouling coatings.

Table 1. Summary of different marine antifouling strategies.

Strategy	Mechanism	Antifouling performance	Limitations	Refs
Chemical dynamic coating	Degradation/hydrolysis-driven renewal	Adhesion inhibition: ~0 *N. incerta* cells mm^{-2} for degradable h-LLA_x coating	Lifetime limited by coating thickness and degradation rate	[67]
Physical dynamic coating	Field-induced deformation	Dynamic removal: 99.5% removal by a single sweeper; >90% bacterial removal within 1 h by DIVA	Dependence on external power	[77, 80]
Fouling-release coating	Low adhesion shear-assisted release	Adhesion inhibition: >90% (*P. aeruginosa*) reduction; Fouling release: 99.7% (*N. incerta*) removal after water jetting	Limited static antifouling; trade-off in modulus, adhesion, and strength	[101]
Fouling-resistant coating	Hydration layer-mediated resistance	Adhesion inhibition: 100% (protein and algal) inhibition; Killing: 99.97% (*E. coli* and *S. aureus*) antibacterial rate	Difficulty in balancing high hydrophilicity and mechanical stability	[126]

Just Accepted

Biomimetic micro-/nanostructured surface	Topographical resistance/killing	Killing: >99% (*E. coli)* antibacterial rate; Adhesion inhibition: >94% (*algal)* inhibition	High fabrication cost; limited durability and large-scale application	[182]
Stimuli-responsive coating	Triggered release/resistance/killing	Killing: 99.99% (*E. coli* and *S. aureus*) antibacterial rate; Fouling release: 99.99% (*E. coli* and *S. aureus*) release; Adhesion inhibition: >99% (protein and algal) inhibition	High system complexity; uncertain long-term responsiveness in seawater	[186]
MOF-based coating	controlled release/catalytic activity/coating reinforcement	Killing: >99% (*E. coli*) and 85.4%-95.6% (*S. aureus*) antibacterial rate; Adhesion inhibition: >96% (algal) inhibition	High scale-up cost; unresolved aqueous stability and environmental compatibility	[227]
HEA-based protective surface	Antibacterial alloying and corrosion protection	Killing: >99.9% (*E. coli* and *S. aureus*) antibacterial rate with $Co_{0.4}FeCr_{0.9}Cu_{0.3}$ HEA after 24 h;	Complex alloy fabrication; high cost; trade-off in ion release/passivation	[233]
TENG-assisted system	Self-powered active antifouling	Killing: 99.99% (*E. coli* and *S. aureus*) antibacterial rate within 10 min; Adhesion inhibition: 76.45% (*E. coli*) and 81.61% (*S. aureus*) inhibition after 7 days	Humidity-, fouling-, and wear-induced output decay; large-scale integration challenge	[248]

Just Accepted

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Author contributions

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Competing interests

The authors have no competing interests to declare that are relevant to the content of this article.

Just Accepted

6 References

[1] H. Shu, J. Cui, Y. Liu, X. Bai, C. Yuan, et al. Map-inspired dual crosslinked pvp-phenol sprayable hydrogel coating for stable marine antifouling applications. *Friction* **13**(11): 9440946 (2025)

[2] H. Yang, L. Qin, W. Zhao, F. J. Mawignon, H. Guo, et al. Eco-friendly polysaccharide coatings for antifouling and drag-reduction and potential application for marine devices. *Friction* **12**(4): 726-744 (2024)

[3] E. A. K. Murphy, J. M. Barros, M. P. Schultz, K. A. Flack, C. N. Steppe, et al. Roughness effects of diatomaceous slime fouling on turbulent boundary layer hydrodynamics. *Biofouling* **34**(9): 976-988 (2018)

[4] M. P. Schultz. Effects of coating roughness and biofouling on ship resistance and powering. *Biofouling* **23**(5): 331-341 (2007)

[5] M. Zhu, L. Ma. A review of recent advances in the effects of surface and interface properties on marine propellers. *Friction* **12**(2): 185-214 (2024)

[6] J. Xie, H. Wu, J. Tao, Z. Lu, X. Liu, et al. Meniscus-inspired segmented network

intercalation strategy: Stiffened yet toughened toward healable antifouling materials. *Adv. Funct. Mater.* **35**(39): 2503647 (2025)

[7] S. M. A. Mousavi, R. Pitchumani. Temperature-dependent dynamic fouling on superhydrophobic and slippery nonwetting copper surfaces. *Chem. Eng. J.* **431**: 133960 (2022)

[8] S. Li, K. Feng, J. Li, Y. Li, Z. Li, et al. Marine antifouling strategies: Emerging opportunities for seawater resource utilization. *Chem. Eng. J.* **486**: 149859 (2024)

[9] H. H. Tuson, D. B. Weibel. Bacteria–surface interactions. *Soft Matter* **9**(17): 4368-4380 (2013)

[10] A. G. Nurioglu, A. C. C. Esteves, G. de With. Non-toxic, non-biocide-release antifouling coatings based on molecular structure design for marine applications. *J. Mater. Chem. B* **3**(32): 6547-6570 (2015)

[11] S. Lin, H. Bi, C. E. Weinell, K. Dam-Johansen. Submerged surfaces exposed to marine biofouling – experimental investigation of cleaning parameters effectiveness. *Prog. Org. Coat.* **172**: 107097 (2022)

[12] S. Lin, H. Bi, C. E. Weinell, K. Dam-Johansen. Mapping the biofouling activities of aged fouling release coating surfaces undergoing underwater cleaning. *Appl. Ocean Res.* **144**: 103860 (2024)

[13] J. Zhang, L. Pan, H. Yan, J. Zhang, H. Zhu, et al. Mechanistic insights into ultrasonic vibration-induced avoidance behavior of fouling organisms on ti alloy surfaces. *Ultrasonics* **165**: 108085 (2026)

[14] X. Zhong, J. Dong, M. Liu, R. Meng, S. Li, et al. Experimental study on ship fouling cleaning by ultrasonic-enhanced submerged cavitation jet: A preliminary study. *Ocean Eng.* **258**: 111844 (2022)

[15] I. Kviatkovski, H. Mamane, A. Lakretz, I. Sherman, D. Beno-Moualem, et al. Resistance of a multiple-isolate marine culture to ultraviolet c irradiation: Inactivation vs biofilm formation. *Lett. Appl. Microbiol.* **67**(3): 278-284 (2018)

[16] E. Ryan, S. Turkmen, S. Benson. An investigation into the application and practical use of (uv) ultraviolet light technology for marine antifouling. *Ocean Eng.* **216**: 107690 (2020)

[17] T. Matos, V. C. Pinto, P. J. Sousa, M. S. Martins, E. Fernández, et al. Exploring local chlorine generation through seawater electrolysis to extend optical sensor lifespan in marine environments. *Chem. Eng. J.* **500**: 156836 (2024)

[18] S. Lin, H. Bi, C. E. Weinell, K. Dam-Johansen. Scratched surface: Quantifying the impact and evaluating underwater cleaning efficacy on fouling release coatings. *Ocean Eng.* **316**: 120013 (2025)

[19] J. Liu, Z. Tong, F. Gao, J. Wang, J. Hu, et al. Pearl-inspired intelligent marine hetero nanocomposite coating based on “brick&mortar” strategy: Anticorrosion durability and switchable antifouling. *Adv. Mater.* **36**(26): 2401982 (2024)

[20] Z. Tong, H. Guo, Z. Di, S. Chen, L. Song, et al. P. Pavoninus-inspired smart slips marine antifouling coating based on coumarin: Antifouling durability and adaptive adjustability of lubrication. *Adv. Funct. Mater.* **34**(8): 2310702 (2024)

[21] Z. Zhang, R. Chen, J. Yu, G. Sun, Q. Liu, et al. Fluorocarbon-based self-layering interpenetrating polymer-network coatings with anti-fouling and anti-icing properties.

Just Accepted

Chem. Eng. J. **474**: 145540 (2023)

[22] Q. Cheng, A. B. Asha, Y. Liu, Y.-Y. Peng, D. Diaz-Dussan, et al. Antifouling and antibacterial polymer-coated surfaces based on the combined effect of zwitterions and the natural borneol. *ACS Appl. Mater. Interfaces* **13**(7): 9006-9014 (2021)

[23] T.-N. Le, C.-K. Lee. Surface functionalization of poly(n-vinylpyrrolidone) onto poly(dimethylsiloxane) for anti-biofilm application. *Appl. Biochem. Biotechnol.* **191**(1): 29-44 (2020)

[24] K. Yu, A. Alzahrani, S. Khoddami, J. T. J. Cheng, Y. Mei, et al. Rapid assembly of infection-resistant coatings: Screening and identification of antimicrobial peptides works in cooperation with an antifouling background. *ACS Appl. Mater. Interfaces* **13**(31): 36784-36799 (2021)

[25] M. R. L. Jones, P. M. Ross. Recovery of the new zealand muricid dogwhelk haustrum scobina from tbt-induced imposex. *Mar. Pollut. Bull.* **126**: 396-401 (2018)

[26] C. C. Ten Hallers-Tjabbes. Tributyltin and policies for antifouling. *Environ. Technol.* **18**(12): 1265-1268 (1997)

[27] J. Beyer, Y. Song, K. E. Tollefsen, J. A. Berge, L. Tveiten, et al. The ecotoxicology of marine tributyltin (tbt) hotspots: A review. *Mar. Environ. Res.* **179**: 105689 (2022)

[28] L. A. R. Simões, É. L. Vogt, C. S. da Costa, M. de Amaral, M. L. M. Hoff, et al. Effects of tributyltin (tbt) on the intermediate metabolism of the crab callinectes sapidus. *Mar. Pollut. Bull.* **182**: 114004 (2022)

[29] D. Vanavermaete, K. Hostens, G. Everaert, K. Parmentier, C. Janssen, et al. Assessing the risk of booster biocides for the marine environment: A case study at the belgian part of the north sea. *Mar. Pollut. Bull.* **197**: 115774 (2023)

[30] C. Ma, W. Wang, D. Kong, S. Wei, W. Li, et al. A novel core/shell cuprous oxide-based structure with improved microwave absorbing and antibacterial performance. *J. Clean. Prod.* **378**: 134419 (2022)

[31] T. Mao, G. Lu, C. Xu, H. Yu, J. Yu. Preparation and properties of polyvinylpyrrolidone-cuprous oxide microcapsule antifouling coating. *Prog. Org. Coat.* **141**: 105317 (2020)

[32] W. Zhou, Y. Wang, C. Ni, L. Yu. Preparation and evaluation of natural rosin-based zinc resins for marine antifouling. *Prog. Org. Coat.* **157**: 106270 (2021)

[33] J. Pan, X. Ai, C. Ma, G. Zhang. Degradable vinyl polymers for combating marine biofouling. *Acc. Chem. Res.* **55**(11): 1586-1598 (2022)

[34] T. Kim, S. Eo, W. J. Shim, M. Kim. Qualitative and quantitative assessment of microplastics derived from antifouling paint in effluent from ship hull hydroblasting and their emission into the marine environment. *J. Hazard. Mater.* **477**: 135258 (2024)

[35] M. Sharifinia, Z. A. Bahmanbeigloo, M. Keshavarzifard, M. H. Khanjani, B. P. Lyons. Microplastic pollution as a grand challenge in marine research: A closer look at their adverse impacts on the immune and reproductive systems. *Ecotoxicol. Environ. Saf.* **204**: 111109 (2020)

[36] L. Zhao, R. Chen, J. Liu, Q. Liu, J. Yu, et al. Layer-by-layer-assembled biomimetic microstructure surface with multiple synergistic antifouling performance. *Prog. Org. Coat.* **183**: 107812 (2023)

[37] G. He, W. Liu, Y. Liu, S. Wei, Y. Yue, et al. Antifouling hydrogel with different mechanisms:Antifouling mechanisms, materials, preparations and applications. *Adv.*

Just Accepted

Colloid Interface Sci. **335**: 103359 (2025)
[38] X. Shi, H. Wei, W. Zhou, P. E. D. Soto Rodriguez, C. Lin, et al. Advanced strategies for marine antifouling based on nanomaterial-enhanced functional PDMS coatings. *Nano Mater. Sci.* **6**(4): 375-395 (2024)
[39] H. Jin, L. Tian, W. Bing, J. Zhao, L. Ren. Bioinspired marine antifouling coatings: Status, prospects, and future. *Prog. Mater. Sci.* **124**: 100889 (2022)
[40] J. Sun, W. Yang, X. Yang, T. Guo, J. Duan, et al. Recent advances in fouling-resistant silicone-based marine antifouling coatings: Mechanisms, fabrication and performances. *Nano Mater. Sci.*: (2026)
[41] H. Qiu, K. Feng, A. Gapeeva, K. Meurisch, S. Kaps, et al. Functional polymer materials for modern marine biofouling control. *Prog. Polym. Sci.* **127**: 101516 (2022)
[42] X. Zang, Y. Ni, Q. Wang, Y. Cheng, J. Huang, et al. Non-toxic evolution: Advances in multifunctional antifouling coatings. *Mater. Today* **75**: 210-243 (2024)
[43] Q. Xie, J. Pan, C. Ma, G. Zhang. Dynamic surface antifouling: Mechanism and systems. *Soft Matter* **15**(6): 1087-1107 (2019)
[44] J. Tang, R. Chen, X. Huo, G. Sun, J. Yu, et al. Constructing micro-dynamic surface of boron-polyurethane with endurable anti-fouling properties. *Prog. Org. Coat.* **174**: 107243 (2023)
[45] T. Masaki, D. Fujita, N. T. Hagen. The surface ultrastructure and epithallium shedding of crustose coralline algae in an 'isoyake' area of southwestern hokkaido, japan. *Hydrobiologia* **116**(1): 218-223 (1984)
[46] K. Yamamoto, H. Endo, S. Yoshikawa, K. Ohki, M. Kamiya. Various defense ability of four sargassacean algae against the red algal epiphyte neosiphonia harveyi in wakasa bay, japan. *Aquat. Bot.* **105**: 11-17 (2013)
[47] G. M. Nylund, H. Pavia. Chemical versus mechanical inhibition of fouling in the red alga dilsea carnosa. *Mar. Ecol. Prog. Ser.* **299**: 111-121 (2005)
[48] L. Tian, Y. Yin, H. Jin, W. Bing, E. Jin, et al. Novel marine antifouling coatings inspired by corals. *Mater. Today Chem.* **17**: 100294 (2020)
[49] H. Nagamine, K. Yamahata, Y. Hagiwara, R. Matsubara. Turbulence modification by compliant skin and strata-corneas desquamation of a swimming dolphin. *J. Turbul.* **5**(null): N18 (2004)
[50] R. Ganguli, V. Mehrotra, B. Dunn. Bioinspired living skins for fouling mitigation. *Smart Mater. Struct.* **18**(10): 104027 (2009)
[51] M. Qi, Q. Song, J. Zhao, C. Ma, G. Zhang, et al. Three-dimensional bacterial behavior near dynamic surfaces formed by degradable polymers. *Langmuir* **33**(45): 13098-13104 (2017)
[52] W. Xu, C. Ma, J. Ma, T. Gan, G. Zhang. Marine biofouling resistance of polyurethane with biodegradation and hydrolyzation. *ACS Appl. Mater. Interfaces* **6**(6): 4017-4024 (2014)
[53] J. Yao, S. Chen, C. Ma, G. Zhang. Marine anti-biofouling system with poly(ε-caprolactone)/clay composite as carrier of organic antifoulant. *J. Mater. Chem. B* **2**(31): 5100-5106 (2014)
[54] Z. Ma, Y. Hong, D. M. Nelson, J. E. Pichamuthu, C. E. Leeson, et al. Biodegradable polyurethane ureas with variable polyester or polycarbonate soft segments: Effects of
Just Accepted

crystallinity, molecular weight, and composition on mechanical properties. *Biomacromolecules* **12**(9): 3265-3274 (2011)

[55] I. Manavitehrani, A. Fathi, H. Badr, S. Daly, A. Negahi Shirazi, et al. Biomedical applications of biodegradable polyesters. *Polymers* **8**(1): 20 (2016)

[56] C. Ma, L. Xu, W. Xu, G. Zhang. Degradable polyurethane for marine anti-biofouling. *J. Mater. Chem. B* **1**(24): 3099-3106 (2013)

[57] H. Y. Chiang, J. Pan, C. Ma, P.-Y. Qian. Combining a bio-based polymer and a natural antifoulant into an eco-friendly antifouling coating. *Biofouling* **36**(2): 200-209 (2020)

[58] C. Ma, W. Zhang, G. Zhang, P.-Y. Qian. Environmentally friendly antifouling coatings based on biodegradable polymer and natural antifoulant. *ACS Sustain. Chem. Eng.* **5**(7): 6304-6309 (2017)

[59] Q. Liu, C. Xu, L. Pang, X. Xiong, H. Zhou, et al. Cuo_2 and tannic acid mediated synergistic antimicrobial and antifouling coatings. *Chem. Eng. J.* **522**: 167589 (2025)

[60] H. Xu, Y. Wang, C. Ni, G. He, X. Li, et al. Degradable polylactic acid-reinforced acrylate polymer coating for marine antifouling. *Prog. Org. Coat.* **197**: 108824 (2024)

[61] Q. Xie, X. Zhou, C. Ma, G. Zhang. Self-cross-linking degradable polymers for antifouling coatings. *Ind. Eng. Chem. Res.* **56**(18): 5318-5324 (2017)

[62] G. Dai, Q. Xie, X. Ai, C. Ma, G. Zhang. Self-generating and self-renewing zwitterionic polymer surfaces for marine anti-biofouling. *ACS Appl. Mater. Interfaces* **11**(44): 41750-41757 (2019)

[63] G. Dai, Q. Xie, C. Ma, G. Zhang. Biodegradable poly(ester-co-acrylate) with antifoulant pendant groups for marine anti-biofouling. *ACS Appl. Mater. Interfaces* **11**(12): 11947-11953 (2019)

[64] X. Zhou, Q. Xie, C. Ma, Z. Chen, G. Zhang. Inhibition of marine biofouling by use of degradable and hydrolyzable silyl acrylate copolymer. *Ind. Eng. Chem. Res.* **54**(39): 9559-9565 (2015)

[65] C. Aydogan, G. Yilmaz, Y. Yagci. Synthesis of hyperbranched polymers by photoinduced metal-free atrp. *Macromolecules* **50**(23): 9115-9120 (2017)

[66] M. S. Selim, M. A. Shenashen, S. A. El-Safty, S. A. Higazy, M. M. Selim, et al. Recent progress in marine foul-release polymeric nanocomposite coatings. *Prog. Mater. Sci.* **87**: 1-32 (2017)

[67] C. Si, G. Hu, W. Jiang, P. Sun, J. Cao, et al. Hydrophobic biodegradable hyperbranched copolymers with excellent marine diatom resistance. *Biomacromolecules* **23**(10): 4327-4338 (2022)

[68] X. Ai, Q. Xie, C. Ma, G. Zhang. Fouling release coating consisting of hyperbranched poly(ε-caprolactone)/siloxane elastomer. *ACS Appl. Polym. Mater.* **2**(4): 1429-1437 (2020)

[69] X. Zhang, Z. Yin, S. Xiang, H. Yan, H. Tian. Degradation of polymer materials in the environment and its impact on the health of experimental animals: A review. *Polymers* **16**(19): 2807 (2024)

[70] M. Suzuki, Y. Tachibana, K.-i. Kasuya. Biodegradability of poly(3-hydroxyalkanoate) and poly(ε-caprolactone) via biological carbon cycles in marine environments. *Polym. J.* **53**(1): 47-66 (2021)

[71] H. Ko, H.-H. Park, H. Byeon, M. Kang, J. Ryu, et al. Undulatory topographical waves

Just Accepted

for flow-induced foulant sweeping. *Sci. Adv.* **5**(11): eaax8935 (2019)

[72] P. Shivapooja, Q. Wang, B. Orihuela, D. Rittschof, G. P. López, et al. Bioinspired surfaces with dynamic topography for active control of biofouling. *Adv. Mater.* **25**(10): 1430-1434 (2013)

[73] S. Zhang, P. Zuo, Y. Wang, P. Onck, J. M. J. d. Toonder. Anti-biofouling and self-cleaning surfaces featured with magnetic artificial cilia. *ACS Appl. Mater. Interfaces* **12**(24): 27726-27736 (2020)

[74] J. Wang, H. Jin, W. Bing, L. Tian. Dynamic antifouling strategies of soft corals: Disrupting the flow field by tentacles. *Colloids Surf., A* **701**: 134771 (2024)

[75] H. Jin, T. Zhang, W. Bing, S. Dong, L. Tian. Antifouling performance and mechanism of elastic graphene–silicone rubber composite membranes. *J. Mater. Chem. B* **7**(3): 488-497 (2019)

[76] W. Bing, L. Tian, Y. Wang, H. Jin, L. Ren, et al. Bio-inspired non-bactericidal coating used for antibiofouling. *Adv. Mater. Technol.* **4**(2): 1800480 (2019)

[77] Q. Wu, H. Yan, L. Chen, S. Qi, T. Zhao, et al. Bio-inspired active self-cleaning surfaces via filament-like sweepers array. *Adv. Mater.* **35**(25): 2212246 (2023)

[78] F. Qian, Z. Wang, M. Xia, J. Yin, X. Jia, et al. The micro-motion nanostructure surface induced by the electric field for efficient inhibition of biofilm. *Chem. Eng. J.* **527**: 171984 (2026)

[79] M. Gao, W. Bing, Z. Wang, J. Fu, J. Li. Bioinspired magnetic-responsive microcolumn arrays for active/passive antifouling and droplet manipulation. *ACS Appl. Mater. Interfaces* **17**(50): 68627-68638 (2025)

[80] Z. Wang, Y. Chen, D. Cheng, B. Wang, S. Qi, et al. Dynamic interfacial vortex arrays for advanced antifouling. *Cell Rep. Phys. Sci.* **6**(8): 102727 (2025)

[81] P. Hu, Q. Xie, C. Ma, G. Zhang. Silicone-based fouling-release coatings for marine antifouling. *Langmuir* **36**(9): 2170-2183 (2020)

[82] A. K. Leonardi, C. K. Ober. Polymer-based marine antifouling and fouling release surfaces: Strategies for synthesis and modification. *Annu. Rev. Chem. Biomol. Eng.* **10**(Volume 10, 2019): 241-264 (2019)

[83] R. F. Brady Jr, I. L. Singer. Mechanical factors favoring release from fouling release coatings. *Biofouling* **15**(1-3): 73-81 (2000)

[84] L. Cavas, P. G. Yildiz, P. Mimigianni, A. Sapalidis, S. Nitodas. Reinforcement effects of multiwall carbon nanotubes and graphene oxide on PDMS marine coatings. *J. Coat. Technol. Res.* **15**(1): 105-120 (2018)

[85] M. S. Selim, S. A. El-Safty, N. A. Fatthallah, M. A. Shenashen. Silicone/graphene oxide sheet-alumina nanorod ternary composite for superhydrophobic antifouling coating. *Prog. Org. Coat.* **121**: 160-172 (2018)

[86] M. S. Selim, N. A. Fatthallah, S. A. Higazy, Z. Hao, P. Jing Mo. A comparative study between two novel silicone/graphene-based nanostructured surfaces for maritime antifouling. *J. Colloid Interface Sci.* **606**: 367-383 (2022)

[87] Z. Zhou, A. Seif, S. Pourhashem, J. Duan, A. Rashidi, et al. Multi-treatments based on polydimethylsiloxane and metal-organic framework wrapped with graphene oxide for achieving long-term corrosion and fouling protection: Experimental and density functional theory aspects. *Constr. Build. Mater.* **384**: 131229 (2023)

Just Accepted

[88] I. O. Arukalam, E. E. Oguzie, Y. Li. Fabrication of fdts-modified PDMS-zno nanocomposite hydrophobic coating with anti-fouling capability for corrosion protection of q235 steel. *J. Colloid Interface Sci.* **484**: 220-228 (2016)

[89] M. S. Selim, A. Elmarakbi, A. M. Azzam, M. A. Shenashen, A. M. El-Saeed, et al. Eco-friendly design of superhydrophobic nano-magnetite/silicone composites for marine foul-release paints. *Prog. Org. Coat.* **116**: 21-34 (2018)

[90] H. Qiu, I. Hölken, A. Gapeeva, V. Filiz, R. Adelung, et al. Development and characterization of mechanically durable silicone-polythiourethane composites modified with tetrapodal shaped zno particles for the potential application as fouling-release coating in the marine sector. *Materials* **11**(12): 2413 (2018)

[91] M. S. Selim, H. Yang, S. A. El-Safty, N. A. Fatthallah, M. A. Shenashen, et al. Superhydrophobic coating of silicone/*β*–mno_2 nanorod composite for marine antifouling. *Colloids Surf., A* **570**: 518-530 (2019)

[92] C. Liu, Q. Xie, C. Ma, G. Zhang. Fouling release property of polydimethylsiloxane-based polyurea with improved adhesion to substrate. *Ind. Eng. Chem. Res.* **55**(23): 6671-6676 (2016)

[93] S. K. Rath, J. G. Chavan, S. Sasane, Jagannath, M. Patri, et al. Two component silicone modified epoxy foul release coatings: Effect of modulus, surface energy and surface restructuring on pseudobarnacle and macrofouling behavior. *Appl. Surf. Sci.* **256**(8): 2440-2446 (2010)

[94] S. K. Rath, J. G. Chavan, S. Sasane, A. Srivastava, M. Patri, et al. Coatings of PDMS-modified epoxy via urethane linkage: Segmental correlation length, phase morphology, thermomechanical and surface behavior. *Prog. Org. Coat.* **65**(3): 366-374 (2009)

[95] Q. Xie, C. Liu, X. Lin, C. Ma, G. Zhang. Nanodiamond reinforced poly(dimethylsiloxane)-based polyurea with self-healing ability for fouling release coating. *ACS Appl. Polym. Mater.* **2**(8): 3181-3188 (2020)

[96] E. Martinelli, D. Gunes, B. M. Wenning, C. K. Ober, J. A. Finlay, et al. Effects of surface-active block copolymers with oxyethylene and fluoroalkyl side chains on the antifouling performance of silicone-based films. *Biofouling* **32**(1): 81-93 (2016)

[97] S. Zhao, D. Zhang, Y. Jiang, C. Gao, L. Liu, et al. A silicone coating containing natural borneol fluorinated side chains with excellent static antifouling properties. *Eur. Polym. J.* **193**: 112064 (2023)

[98] Y. Qin, J. Xue, S. Wang, Y. Fan, L. Wang, et al. Capsaicin-based silicone antifouling coating with enhanced interlocking adhesion via sipn. *Colloids Surf., A* **677**: 132346 (2023)

[99] Z. Lu, Z. Chen, Y. Guo, Y. Ju, Y. Liu, et al. Flexible hydrophobic antifouling coating with oriented nanotopography and nonleaking capsaicin. *ACS Appl. Mater. Interfaces* **10**(11): 9718-9726 (2018)

[100] H. Guo, C. Wen, S. Tian, X. Zhang, Y. Ma, et al. Universal intraductal surface antifouling coating based on an amphiphilic copolymer. *ACS Appl. Mater. Interfaces* **13**(18): 21051-21059 (2021)

[101] H. Shu, A. Khlyustova, K.-W. Park, S. Stafslien, G. Kang, et al. Fluorine-free amphiphilic copolymers for broad-spectrum marine biofouling deterrence. *Adv. Funct. Mater.* **35**(38): 2502065 (2025)

Just Accepted

[102] Z. Zhang, Q. Xie, G. Zhang, C. Ma, G. Zhang. Surface-enriched amphiphilic polysiloxane coating with superior antifouling ability and strong substrate adhesion. *ACS Appl. Polym. Mater.* **5**(5): 3524-3533 (2023)
[103] G. Galli, E. Martinelli. Amphiphilic polymer platforms: Surface engineering of films for marine antibiofouling. *Macromol. Rapid Commun.* **38**(8): 1600704 (2017)
[104] H. Guo, J. Yang, W. Zhao, T. Xu, C. Lin, et al. Direct formation of amphiphilic crosslinked networks based on pvp as a marine anti-biofouling coating. *Chem. Eng. J.* **374**: 1353-1363 (2019)
[105] J. Sun, J. Duan, Y. Zhang, X. Zhai, Y. Zhu, et al. Transparent hybrid coatings for marine antifouling: Synergizing amphiphilicity, nanocellulose lubrication, and electrostatic repulsion. *Chem. Eng. J.* **521**: 166869 (2025)
[106] R. B. Bodkhe, S. J. Stafslien, J. Daniels, N. Cilz, A. J. Muelhberg, et al. Zwitterionic siloxane-polyurethane fouling-release coatings. *Prog. Org. Coat.* **78**: 369-380 (2015)
[107] J. Tan, A. C. C. Esteves, J. Yang, S. Zhou. Zwitterion-bearing silica sol for enhancing antifouling performance and mechanical strength of transparent PDMS coatings. *Prog. Org. Coat.* **191**: 108415 (2024)
[108] Y.-Y. Cui, M.-Q. Sheng, Y. Liu, Y. Feng, H.-J. Li, et al. Antifouling and self-healing performance of marine coatings based on hydrogen-bond interactions. *ACS Appl. Mater. Interfaces* **15**(50): 58967-58975 (2023)
[109] J. Sun, J. Duan, X. Liu, X. Dong, Y. Zhang, et al. Environmentally benign smart self-healing silicone-based coating with dual antifouling and anti-corrosion properties. *Appl. Mater. Today* **28**: 101551 (2022)
[110] P. Hu, Q. Xie, C. Ma, G. Zhang. Fouling resistant silicone coating with self-healing induced by metal coordination. *Chem. Eng. J.* **406**: 126870 (2021)
[111] Y. Liu, S. Zhao, Y. Jiang, Q. Liu, Z. Liu, et al. Agnp hybrid organosilicon antifouling coating with spontaneous self-healing and long-term antifouling properties. *Ind. Eng. Chem. Res.* **62**(33): 12999-13008 (2023)
[112] Y. Hou, G. Zhu, J. Cui, N. Wu, B. Zhao, et al. Superior hard but quickly reversible si–o–si network enables scalable fabrication of transparent, self-healing, robust, and programmable multifunctional nanocomposite coatings. *J. Am. Chem. Soc.* **144**(1): 436-445 (2022)
[113] Z. Zhang, Y. Huang, Q. Xie, G. Liu, C. Ma, et al. Functional polymer–ceramic hybrid coatings: Status, progress, and trend. *Prog. Polym. Sci.* **154**: 101840 (2024)
[114] W. Zheng, J. Huang, X. Zang, X. Xu, W. Cai, et al. Judicious design and rapid manufacturing of a flexible, mechanically resistant liquid-like coating with strong bonding and antifouling abilities. *Adv. Mater.* **34**(42): 2204581 (2022)
[115] W. Zheng, H. Wang, Q. Huang, Y. Li, J. Huang, et al. Ultra-antifouling liquid-like surfaces for sustainable viscous water-in-oil emulsions separation and oil recovery. *Adv. Mater.* **37**(5): 2413751 (2025)
[116] M. Wang, Q. Xie, J. Pan, C. Ma, G. Zhang. Strong yet tough cross-linked oligosiloxane elastomer with impact-resistant and antifouling properties. *Chem. Eng. J.* **464**: 142769 (2023)
[117] Z. Zhang, B. Zhang, Z. Zhang, Q. Xie, G. Zhang, et al. Self-reinforcing gradient antifouling coating with high adhesion, on-demand peelability, and superior recyclability.

Just Accepted

Adv. Funct. Mater. **35**(46): 2508400 (2025)
[118] J. Sun, J. Duan, C. Liu, X. Liu, Y. Zhu, et al. Transparent and mechanically durable silicone/zro2 sol hybrid coating with enhanced antifouling properties. *Chem. Eng. J.* **490**: 151567 (2024)
[119] Y.-H. Cheng, C.-T. Wu, L.-H. Hu. Dual functional low surface energy coating of anti-corrosion / fouling via crosslinking polysilazane preceramic precursor incorporated with fluorine. *Prog. Org. Coat.* **177**: 107409 (2023)
[120] R. Chen, Y. Zhang, Q. Xie, Z. Chen, C. Ma, et al. Transparent polymer-ceramic hybrid antifouling coating with superior mechanical properties. *Adv. Funct. Mater.* **31**(19): 2011145 (2021)
[121] Y. Huang, Z. Zhang, B. Zhang, C. Ma, G. Zhang. Self-adaptive zwitterionic polysilazane coatings with mechanical robustness, high transparency, and broad-spectrum antiadhesion properties. *Adv. Mater.* **37**(6): 2413035 (2025)
[122] M. Zhang, W. Choi, M. Kim, J. Choi, X. Zang, et al. Recent advances in environmentally friendly dual-crosslinking polymer networks. *Angew. Chem. Int. Ed.* **63**(24): e202318035 (2024)
[123] A. Jain, N. B. Bhosle. Biochemical composition of the marine conditioning film: Implications for bacterial adhesion. *Biofouling* **25**(1): 13-19 (2009)
[124] P. Cao, S. Tian, J. Dai, J. Li, G. Lu, et al. A catfish skin-inspired dual-antifouling epoxy thermoset as coatings for marine antifouling applications. *Chem. Eng. J.* **504**: 159057 (2025)
[125] X. Song, J. Man, X. Zhang, J. Wang, Y. Zhang, et al. Atomistic insights into the ionic response and mechanism of antifouling zwitterionic polymer brushes. *Small* **21**(6): 2406233 (2025)
[126] Y. Xiong, Z. Fang, P. Tang, Y. Li, H. Jiang, et al. Mxene and cellulose nanofibers reinforced hydrogel with high strength and photothermal self-healing performances for marine antifouling. *Carbohydr. Polym.* **348**: 122879 (2025)
[127] X. Xu, J. Liu, J. Weng, W. Wen, D. Yu, et al. Underwater antiadhesion hydrogel coatings with spontaneous segment orientation. *Chem. Eng. Sci.* **301**: 120700 (2025)
[128] C. Zhang, Y. Qi, Y. Guo, S. Zhang, G. Xiong, et al. Anti-marine biofouling adhesion performance and mechanism of PDMS fouling-release coating containing ps-peg hydrogel. *Mar. Pollut. Bull.* **194**: 115345 (2023)
[129] C. Ma, H. Yang, X. Zhou, B. Wu, G. Zhang. Polymeric material for anti-biofouling. *Colloids Surf., B* **100**: 31-35 (2012)
[130] Y. Yang, H. Luo, J. Yang, D. Huang, S. Zhou. Facile uv-curing technique to establish a 3D-grafted poly(ethylene glycol) layer on an epoxy resin base for underwater applications. *J. Appl. Polym. Sci.* **133**(39): 43972 (2016)
[131] I. Jiménez-Pardo, L. G. J. Van der Ven, R. A. T. M. Van Benthem, G. De With, A. C. C. Esteves. Hydrophilic self-replenishing coatings with long-term water stability for anti-fouling applications. *Coatings* **8**(5): 184 (2018)
[132] S. Jiang, Z. Cao. Ultralow-fouling, functionalizable, and hydrolyzable zwitterionic materials and their derivatives for biological applications. *Adv. Mater.* **22**(9): 920-932 (2010)
[133] L. Zheng, H. S. Sundaram, Z. Wei, C. Li, Z. Yuan. Applications of zwitterionic polymers.
JBE Accepted

React. Funct. Polym. **118**: 51-61 (2017)
[134] Y. He, J. Hower, S. Chen, M. T. Bernards, Y. Chang, et al. Molecular simulation studies of protein interactions with zwitterionic phosphorylcholine self-assembled monolayers in the presence of water. *Langmuir* **24**(18): 10358-10364 (2008)
[135] J. Wu, C. Zhao, R. Hu, W. Lin, Q. Wang, et al. Probing the weak interaction of proteins with neutral and zwitterionic antifouling polymers. *Acta Biomater.* **10**(2): 751-760 (2014)
[136] X. Li, C. Tang, D. Liu, Z. Yuan, H.-C. Hung, et al. High-strength and nonfouling zwitterionic triple-network hydrogel in saline environments. *Adv. Mater.* **33**(39): 2102479 (2021)
[137] Y. Chang, S. Chen, Q. Yu, Z. Zhang, M. Bernards, et al. Development of biocompatible interpenetrating polymer networks containing a sulfobetaine-based polymer and a segmented polyurethane for protein resistance. *Biomacromolecules* **8**(1): 122-127 (2007)
[138] Y. Chang, S. Chen, Z. Zhang, S. Jiang. Highly protein-resistant coatings from well-defined diblock copolymers containing sulfobetaines. *Langmuir* **22**(5): 2222-2226 (2006)
[139] C. Ma, H. Zhou, B. Wu, G. Zhang. Preparation of polyurethane with zwitterionic side chains and their protein resistance. *ACS Appl. Mater. Interfaces* **3**(2): 455-461 (2011)
[140] Z. Liu, C. He. Fabrication of amphiphilic self-healing coatings with high mechanical properties and their antifouling performance. *Prog. Org. Coat.* **175**: 107371 (2023)
[141] J. Niu, Y. Li, Y. Shen, S. Yuan, D. Wang, et al. Hydrogel-anchored caffeic acid-copper ions coating: Integrated defensive and offensive strategy for synergistic antifouling. *Chem. Eng. J.* **515**: 163766 (2025)
[142] M. Zou, Y. Zeng, X. Wang, S. Liu, Q. Ye, et al. Preparation of nanocomposite hydrogels based on zwitterionic polymer functionalized mxene nanosheets for antifouling application. *Carbon* **243**: 120590 (2025)
[143] H. Sun, X. He, C. Yuan, X. Bai. Research progress and performance enhancement strategies of marine antifouling hydrogel coatings. *Prog. Org. Coat.* **206**: 109365 (2025)
[144] Y. Xiong, D. Hu, L. Huang, Z. Fang, H. Jiang, et al. Ultra-high strength sodium alginate/pva/phmb double-network hydrogels for marine antifouling. *Prog. Org. Coat.* **187**: 108175 (2024)
[145] J. P. Gong, Y. Katsuyama, T. Kurokawa, Y. Osada. Double-network hydrogels with extremely high mechanical strength. *Adv. Mater.* **15**(14): 1155-1158 (2003)
[146] H. Chen, Q. Chen, R. Hu, H. Wang, B.-m. Z. Newby, et al. Mechanically strong hybrid double network hydrogels with antifouling properties. *J. Mater. Chem. B* **3**(27): 5426-5435 (2015)
[147] X. Li, J. Gou, H. Feng, X. Sun, X. Qin, et al. Improved antifouling ability for double-network hydrogel coatings with excellent elastic and toughness under marine tidal environment. *Adv. Eng. Mater.* **25**(10): 2201801 (2023)
[148] J. Zhang, L. Chen, L. Chen, S. Qian, X. Mou, et al. Highly antifouling, biocompatible and tough double network hydrogel based on carboxybetaine-type zwitterionic polymer and alginate. *Carbohydr. Polym.* **257**: 117627 (2021)
[149] J. Zhang, S. Qian, L. Chen, L. Chen, L. Zhao, et al. Highly antifouling double network hydrogel based on poly(sulfobetaine methacrylate) and sodium alginate with great toughness. *J. Mater. Sci. Technol.* **85**: 235-244 (2021)
[150] J. Ren, Y. Wang, Z. Liu, K. Liu, X. Xiang. Balancing stretchability and conductivity:
JUST Accepted

Carbon nanotube layer-enhanced non-ionic conductive hydrogels with a sandwich structure. *Chem. Eng. J.* **500**: 156641 (2024)
[151] F. Wang, J. Wang, W. Li, Y. Wu, J. Liu. High performance hydrogel sensor based on rgo integrated p(aa-hema) and starch for motion and temperature monitoring. *Carbohydr. Polym.* **371**: 124497 (2026)
[152] Y. Wu, C. Zhou, J. Xie, A. Mo, D. Lee, et al. Functional iron oxide nanoparticles cross-linked hydrogel for craniofacial bone regeneration. *Biomaterials* **327**: 123782 (2026)
[153] J. Yang, R. Bai, B. Chen, Z. Suo. Hydrogel adhesion: A supramolecular synergy of chemistry, topology, and mechanics. *Adv. Funct. Mater.* **30**(2): 1901693 (2020)
[154] J. Liu, S. Qu, Z. Suo, W. Yang. Functional hydrogel coatings. *Natl. Sci. Rev.* **8**(2): nwaa254 (2021)
[155] J. Shen, M. Du, Z. Wu, Y. Song, Q. Zheng. Strategy to construct polyzwitterionic hydrogel coating with antifouling, drag-reducing and weak swelling performance. *RSC Adv.* **9**(4): 2081-2091 (2019)
[156] X. Jiang, X. Xu, Z. Xia, D. Lin, Y. Chen, et al. Simultaneous segment orientation and anchoring for robust hydrogel coating with underwater superoleophobicity. *Adv. Funct. Mater.* **34**(24): 2314589 (2024)
[157] Y. Liu, S. Miao, H. Ren, L. Tian, J. Zhao, et al. Synthesis and functionalization of scalable and versatile 2D protein films via amyloid-like aggregation. *Nat. Protoc.* **19**(2): 539-564 (2024)
[158] A. Kamada, M. Rodriguez-Garcia, F. S. Ruggeri, Y. Shen, A. Levin, et al. Controlled self-assembly of plant proteins into high-performance multifunctional nanostructured films. *Nat. Commun.* **12**(1): 3529 (2021)
[159] M. Rabe, D. Verdes, S. Seeger. Understanding protein adsorption phenomena at solid surfaces. *Adv. Colloid Interface Sci.* **162**(1): 87-106 (2011)
[160] Z. Zhao, M. Pan, C. Qiao, L. Xiang, X. Liu, et al. Bionic engineered protein coating boosting anti-biofouling in complex biological fluids. *Adv. Mater.* **35**(6): 2208824 (2023)
[161] Z. Song, R. Li, X. Yang, A. Ambrosi, X. Luo. Ultralow fouling electrochemical detection of uric acid directly in serum based on phase-transited bovine serum albumin and conducting polymer. *Chin. Chem. Lett.* **34**(12): 108314 (2023)
[162] N. Zhang, Z. Deng, Q. Wang, M. Zhou, P. Wang, et al. Phase-transited lysozyme with secondary reactivity for moisture-permeable antibacterial wool fabric. *Chem. Eng. J.* **432**: 134198 (2022)
[163] C. Fu, Z. Wang, Y. Gao, J. Zhao, Y. Liu, et al. Sustainable polymer coating for stainproof fabrics. *Nat. Sustain.* **6**(8): 984-994 (2023)
[164] Y. Han, Z. Lin, J. Zhou, G. Yun, R. Guo, et al. Polyphenol-mediated assembly of proteins for engineering functional materials. *Angew. Chem. Int. Ed.* **59**(36): 15618-15625 (2020)
[165] X.-T. Wang, L.-Y. Liu, H. Liang, W.-Y. Ge, L.-L. Chen, et al. Super stable coating based on ovalbumin and tannic acid for hydrophilic and antibacterial functionalization of polymer materials. *ACS Appl. Mater. Interfaces* **17**(10): 16040-16056 (2025)
[166] A. Ali, D. Culliton, S. Fahad, Z. Ali, E.-T. Kang, et al. Nature-inspired anti-fouling strategies for combating marine biofouling. *Prog. Org. Coat.* **189**: 108349 (2024)
[167] Y. Liu, X. He, C. Yuan, P. Cao, X. Bai. Antifouling applications and fabrications of biomimetic micro-structured surfaces: A review. *J. Adv. Res.* **59**: 201-221 (2024)

Just Accepted

[168] Y. Zhang, C. Su, Z. Zou, J. Li, M. Ji, et al. Underwater biomimetic structures and surface technologies: A frontier review from natural inspiration to engineering implementation. *Ocean Eng.* **334**: 121591 (2025)
[169] R. Jiang, L. Hao, L. Song, L. Tian, Y. Fan, et al. Lotus-leaf-inspired hierarchical structured surface with non-fouling and mechanical bactericidal performances. *Chem. Eng. J.* **398**: 125609 (2020)
[170] W. Jo, H. S. Kang, J. Choi, J. Jung, J. Hyun, et al. Light-designed shark skin-mimetic surfaces. *Nano Lett.* **21**(13): 5500-5507 (2021)
[171] L.-C. Pan, S.-Y. Hsieh, W.-C. Chen, F.-T. Lin, C.-H. Lu, et al. Self-assembly of shark scale-patterned tunable superhydrophobic/antifouling structures with visual color response. *ACS Appl. Mater. Interfaces* **15**(29): 35311-35320 (2023)
[172] K. Yang, X. Yu, X. Cui, D. Chen, T. Shen, et al. Surface modification of 3D biomimetic shark denticle structures for drag reduction. *Adv. Mater.* **37**(9): 2417337 (2025)
[173] T. Vladkova. Surface engineering for non-toxic biofouling control. *J. Univ. Chem. Technol. Metall.* **42**: 239-256 (2007)
[174] D. Howell, B. Behrends. A review of surface roughness in antifouling coatings illustrating the importance of cutoff length. *Biofouling* **22**(6): 401-410 (2006)
[175] A. J. Scardino, E. Harvey, R. De Nys. Testing attachment point theory: Diatom attachment on microtextured polyimide biomimics. *Biofouling* **22**(1): 55-60 (2006)
[176] Y. Liu, K. Zhang, W. Li, B. He, C. Lu, et al. Dual-bioinspired superhydrophobic coating with physicochemical synergy for enhanced antifouling performance. *J. Bionic Eng.* **22**(6): 3108-3117 (2025)
[177] L. Zhao, R. Chen, L. Lou, X. Jing, Q. Liu, et al. Layer-by-layer-assembled antifouling films with surface microtopography inspired by laminaria japonica. *Appl. Surf. Sci.* **511**: 145564 (2020)
[178] F. J. Mawignon, L. Qin, A. N. Kouediatouka, S. Lu, H. Yang, et al. Highly strong bio-inspired zno/PDMS superhydrophobic surface with drag reduction and antibacterial properties. *Tribology International* **189**: 109003 (2023)
[179] H.-W. Hsueh, S.-H. Lin, Z.-X. Chen, H.-Y. Yang, Y.-Y. Wu, et al. Self-assembly of antifouling wavy taco shell arrays inspired by hierarchically wrinkled shark scales. *ACS Appl. Mater. Interfaces* **18**(21): 30329-30339 (2026)
[180] E. P. Ivanova, J. Hasan, H. K. Webb, G. Gervinskas, S. Juodkazis, et al. Bactericidal activity of black silicon. *Nat. Commun.* **4**(1): 2838 (2013)
[181] E. P. Ivanova, J. Hasan, H. K. Webb, V. K. Truong, G. S. Watson, et al. Natural bactericidal surfaces: Mechanical rupture of pseudomonas aeruginosa cells by cicada wings. *Small* **8**(16): 2489-2494 (2012)
[182] S. Qi, J. Wang, D. Cheng, T. Pan, R. Tan, et al. Rational design of microbicidal inorganic nano-architectures. *Small* **21**(30): 2502663 (2025)
[183] J. Wang, B. Macdonald, T. H. Cho, T. Repetto, K. Sun, et al. Bioinspired zwitterionic nanowires with simultaneous biofouling reduction and release. *Small* **20**(40): 2400784 (2024)
[184] S. Burkert, E. Bittrich, M. Kuntzsch, M. Müller, K.-J. Eichhorn, et al. Protein resistance of pnipaam brushes: Application to switchable protein adsorption. *Langmuir* **26**(3): 1786-1795 (2010)

Just Accepted

[185] Q. Yu, J. Cho, P. Shivapooja, L. K. Ista, G. P. López. Nanopatterned smart polymer surfaces for controlled attachment, killing, and release of bacteria. *ACS Appl. Mater. Interfaces* **5**(19): 9295-9304 (2013)

[186] Y. Liu, G. I. N. Waterhouse, X. Jiang, Z. Zhang, L. Yu. Smart antibiofouling composite coatings based on photothermally-active polyaniline and thermo-responsive poly(n-isopropylacrylamide). *Chem. Eng. J.* **490**: 151669 (2024)

[187] M. Rahimi, M. Nasiri. Polymer brushes prepared by surface-initiated atom transfer radical polymerization of poly (n-isopropyl acrylamide) and their antifouling properties. *Eur. Polym. J.* **125**: 109536 (2020)

[188] M. Divandari, J. Pollard, E. Dehghani, N. Bruns, E. M. Benetti. Controlling enzymatic polymerization from surfaces with switchable bioaffinity. *Biomacromolecules* **18**(12): 4261-4270 (2017)

[189] Y. Ni, D. Zhang, Y. Wang, X. He, J. He, et al. Host–guest interaction-mediated photo/temperature dual-controlled antibacterial surfaces. *ACS Appl. Mater. Interfaces* **13**(12): 14543-14551 (2021)

[190] F. Zhang, J. Gao, Z. Zhao, M. Xia, X. Li, et al. Dynamic responsive liquid-like smooth coatings developed via multi-mechanism synergistic strategy for intelligent antibiofouling. *Prog. Org. Coat.* **213**: 109950 (2026)

[191] Q. Tan, G. Zhang, X. Xu, T. Hu, Y. Zhang, et al. Stimuli-responsive composite coating based on dynamic metal-ligand interactions for sustainable marine antifouling. *Sustain. Mater. Technol.* **45**: e01521 (2025)

[192] C. Wei, X. Zhang, Y. Deng, B. Wu, J. Wu. Antifouling behaviour of pH responsive amphiphilic smart coatings on the basis of charge conversion in the marine environments. *Prog. Org. Coat.* **210**: 109701 (2026)

[193] D.-H. Qu, Q.-C. Wang, Q.-W. Zhang, X. Ma, H. Tian. Photoresponsive host–guest functional systems. *Chem. Rev.* **115**(15): 7543-7588 (2015)

[194] W. Szymański, J. M. Beierle, H. A. V. Kistemaker, W. A. Velema, B. L. Feringa. Reversible photocontrol of biological systems by the incorporation of molecular photoswitches. *Chem. Rev.* **113**(8): 6114-6178 (2013)

[195] Y. Qu, X. Li, H. Zhang, R. Huang, W. Qi, et al. Controllable synthesis of a sponge-like z-scheme n,s-cqds/bi2moo6@TiO2 film with enhanced photocatalytic and antimicrobial activity under visible/nir light irradiation. *J. Hazard. Mater.* **429**: 128310 (2022)

[196] G. Wang, H. Feng, L. Hu, W. Jin, Q. Hao, et al. An antibacterial platform based on capacitive carbon-doped TiO_2 nanotubes after direct or alternating current charging. *Nat. Commun.* **9**(1): 2055 (2018)

[197] M. Xiao, L. Zhang, B. Luo, M. Lyu, Z. Wang, et al. Molten-salt-mediated synthesis of an atomic nickel co-catalyst on TiO_2 for improved photocatalytic h2 evolution. *Angew. Chem. Int. Ed.* **59**(18): 7230-7234 (2020)

[198] W. Szeto, M. K. H. Leung, D. Y. C. Leung. Recent developments of titanium dioxide materials for aquatic antifouling application. *J. Mar. Sci. Technol.* **26**(1): 301-321 (2021)

[199] A. S. Vijayan, A. Joseph, B. G. Nair, S. Vandana. MoS_2/ag-TiO_2/polyurethane nanocomposite as a photocatalytic coating for antibiofouling applications. *ACS Appl. Nano Mater.* **7**(16): 19024-19042 (2024)

[200] A. Zhou, S. Li, W. Sand, X. Wang, Y. Geng, et al. Self-growing composite photocatalytic

Just Accepted

materials for surface self-renewal of marine antifouling coatings. *Sep. Purif. Technol.* **354**: 129387 (2025)

[201] J. Lin, J. Wu, Y. Fan, Y. Su, K. Chen, et al. Awakenable smart-responsive hydrogel armor for adaptive surface protection in marine environments. *ACS Sustain. Chem. Eng.* **13**(33): 13642-13655 (2025)

[202] M. Villegas, Y. Zhang, N. Abu Jarad, L. Soleymani, T. F. Didar. Liquid-infused surfaces: A review of theory, design, and applications. *ACS Nano* **13**(8): 8517-8536 (2019)

[203] R. Li, R. Liu, V. Abetz, L. Zhao, Z. Zhang, et al. Nepenthes-inspired liquid-infused membranes with capillary-enhanced thermal efficiency anti-scaling, and anti-fouling for direct contact membrane distillation. *Chem. Eng. J.* **518**: 164726 (2025)

[204] S. Amini, S. Kolle, L. Petrone, O. Ahanotu, S. Sunny, et al. Preventing mussel adhesion using lubricant-infused materials. *Science* **357**(6352): 668-673 (2017)

[205] T.-S. Wong, S. H. Kang, S. K. Y. Tang, E. J. Smythe, B. D. Hatton, et al. Bioinspired self-repairing slippery surfaces with pressure-stable omniphobicity. *Nature* **477**(7365): 443-447 (2011)

[206] J. Wang, L. Wang, N. Sun, R. Tierney, H. Li, et al. Viscoelastic solid-repellent coatings for extreme water saving and global sanitation. *Nat. Sustain.* **2**(12): 1097-1105 (2019)

[207] Z. Tong, L. Song, S. Chen, J. Hu, Y. Hou, et al. Hagfish-inspired smart slips marine antifouling coating based on supramolecular: Lubrication modes responsively switching and self-healing properties. *Adv. Funct. Mater.* **32**(27): 2201290 (2022)

[208] Z. Tong, F. Gao, S. Chen, L. Song, J. Hu, et al. Slippery porous-liquid-infused porous surface (spips) with on-demand responsive switching between “defensive” and “offensive” antifouling modes. *Adv. Mater.* **36**(9): 2308972 (2024)

[209] Z. Tong, S. Zhang, S. Chen, R. Sun, P. He, et al. Sea anemone-inspired slippery liquid-infused porous surface (slips) with bionic cilia for responsive 4d antifouling. *Small* **20**(36): 2401658 (2024)

[210] H. Li, M. Eddaoudi, M. O'Keeffe, O. M. Yaghi. Design and synthesis of an exceptionally stable and highly porous metal-organic framework. *Nature* **402**(6759): 276-279 (1999)

[211] H. C. Zhou, J. R. Long, O. M. Yaghi. Introduction to metal-organic frameworks. *Chem. Rev.* **112**(2): 673-674 (2012)

[212] S. Canossa, Z. Ji, C. Gropp, Z. Rong, E. Ploetz, et al. System of sequences in multivariate reticular structures. *Nat. Rev. Mater.* **8**(5): 331-340 (2023)

[213] D. Han, X. Liu, S. Wu. Metal organic framework-based antibacterial agents and their underlying mechanisms. *Chem. Soc. Rev.* **51**(16): 7138-7169 (2022)

[214] L.-H. Xu, S.-H. Li, H. Mao, Y. Li, A.-S. Zhang, et al. Highly flexible and superhydrophobic mof nanosheet membrane for ultrafast alcohol-water separation. *Science* **378**(6617): 308-313 (2022)

[215] S. Zheng, H. Zhou, H. Xue, P. Braunstein, H. Pang. Pillared-layer ni-mof nanosheets anchored on ti_3c_2 mxene for enhanced electrochemical energy storage. *J. Colloid Interface Sci.* **614**: 130-137 (2022)

[216] H. Raza, I. Yildiz, F. Yasmeen, K. S. Munawar, M. Ashfaq, et al. Synthesis of a 2D copper(ii)-carboxylate framework having ultrafast adsorption of organic dyes. *J. Colloid Interface Sci.* **602**: 43-54 (2021)

[217] X. Li, K. Chen, R. Guo, Z. Wei. Ionic liquids functionalized mofs for adsorption. *Chem.*

Just Accepted

Rev. **123**(16): 10432-10467 (2023)

[218] L. Niu, L. Zeng, D. Yu, S. Cheng, G. Feng. Overscreening-driven modulation of ion adsorption and desorption in conductive mof electrodes by charging rates. *ACS Nano* **19**(2): 2581-2590 (2025)

[219] T. Li, Z. Jin. Unique ternary ni-mof-74/ni_2p/mos_x composite for efficient photocatalytic hydrogen production: Role of ni_2p for accelerating separation of photogenerated carriers. *J. Colloid Interface Sci.* **605**: 385-397 (2022)

[220] D. Zhou, Y. Chen, W. Bu, L. Meng, C. Wang, et al. Modification of Metal-Organic Framework nanoparticles using dental pulp mesenchymal stem cell membranes to target oral squamous cell carcinoma. *J. Colloid Interface Sci.* **601**: 650-660 (2021)

[221] X. Zhang, X. Hao, S. Qiu, G. Lu, W. Liu, et al. Efficient capture and release of carboxylated benzisothiazolinone from uio-66-nh_2 for antibacterial and antifouling applications. *J. Colloid Interface Sci.* **623**: 710-722 (2022)

[222] Y. Zhang, J. Pan, Q. Xie, C. Ma, G. Zhang. Self-regenerating zwitterionic hydrogel coating modified by Metal-Organic Framework for integrated anti-biofouling and anti-corrosion. *Adv. Funct. Mater.* **35**(25): 2424994 (2025)

[223] W. He, X. Li, X. Dai, L. Shao, Y. Fu, et al. Redox concomitant formation method for fabrication of Cu(I)-mof/polymer composites with antifouling properties. *Angew. Chem. Int. Ed.* **63**(44): e202411539 (2024)

[224] X. Ren, L. Wang, Z. Xie, C. Huang, J. Wang, et al. Stable Cu(I) release from multivalent Cu-MOFs: Achieving long-lasting antifouling performance in marine coatings. *Chem. Eng. J.* **524**: 169242 (2025)

[225] Z. Zhou, S. Li, G. Wei, W. Liu, Y. Zhang, et al. Cerium-based metal–organic framework with intrinsic haloperoxidase-like activity for antibiofilm formation. *Adv. Funct. Mater.* **32**(39): 2206294 (2022)

[226] Y. Xu, Z. Yao, X. He, J. Wang, M. Zheng, et al. In-situ growth of peo/uio-66(ce) nanozyme coatings on titanium alloy for antibacterial applications. *Surf. Coat. Technol.* **515**: 132627 (2025)

[227] R. Guo, M. Du, W. Wu, P. Guo, H. Shi, et al. Metal organic frameworks reinforced triple protective high-strength coating inspired by pearl structure for marine anti-fouling. *Chem. Eng. J.* **519**: 165046 (2025)

[228] Z. Yu, X. Li, X. Li, B. Zheng, D. Li, et al. Nacre-inspired Metal-Organic Framework coatings reinforced by multiscale hierarchical cross-linking for integrated antifouling and anti-microbial corrosion. *Adv. Funct. Mater.* **33**(47): 2305995 (2023)

[229] Z. Yu, X. Li, Z. Wang, Y. Fan, W. Zhao, et al. Robust chiral Metal-Organic Framework coatings for self-activating and sustainable biofouling mitigation. *Adv. Mater.* **36**(45): e2407409 (2024)

[230] Z. Wang, J. Fu, Y. Feng, Y. Wan, Q. Tian, et al. Corrosion behavior and antifouling performance of cocrfenicu$_x$ high-entropy alloys in 3.5 wt% NaCl solution. *Electrochim. Acta* **553**: 148303 (2026)

[231] J. Gao, Y. Jin, Y. Fan, D. Xu, L. Meng, et al. Fabricating antibacterial cocrcufeni high-entropy alloy via selective laser melting and in-situ alloying. *J. Mater. Sci. Technol.* **102**: 159-165 (2022)

[232] Y. Li, L. Yang, Y. Liao, R. Zhao, L. Ji, et al. Photothermal heating-assisted superior

Just Accepted

antibacterial and antibiofilm activity of high-entropy-alloy nanoparticles. *Adv. Funct. Mater.* **33**(35): 2302712 (2023)

[233] G. Ren, L. Huang, K. Hu, T. Li, Y. Lu, et al. Enhanced antibacterial behavior of a novel cu-bearing high-entropy alloy. *J. Mater. Sci. Technol.* **117**: 158-166 (2022)

[234] H. Shahmir, T. Mousavi, J. He, Z. Lu, M. Kawasaki, et al. Microstructure and properties of a cocrfenimn high-entropy alloy processed by equal-channel angular pressing. *Mater. Sci. Eng., A* **705**: 411-419 (2017)

[235] W.-R. Wang, W.-L. Wang, J.-W. Yeh. Phases, microstructure and mechanical properties of al$_x$cocrfeni high-entropy alloys at elevated temperatures. *J. Alloys Compd.* **589**: 143-152 (2014)

[236] I. S. Wani, T. Bhattacharjee, S. Sheikh, Y. P. Lu, S. Chatterjee, et al. Ultrafine-grained alcocrfeni$_{2.1}$ eutectic high-entropy alloy. *Mater. Res. Lett.* **4**(3): 174-179 (2016)

[237] B. Cantor, I. T. H. Chang, P. Knight, A. J. B. Vincent. Microstructural development in equiatomic multicomponent alloys. *Mater. Sci. Eng., A* **375-377**: 213-218 (2004)

[238] J. W. Yeh, S. K. Chen, S. J. Lin, J. Y. Gan, T. S. Chin, et al. Nanostructured high-entropy alloys with multiple principal elements: Novel alloy design concepts and outcomes. *Adv. Eng. Mater.* **6**(5): 299-303 (2004)

[239] A. Burla, M. Khandelwal, M. Vaidya. Antibacterial properties of cu containing complex concentrated alloys. *Mater. Today Commun.* **33**: 104915 (2022)

[240] X. Zhang, Y. Yu, B. Ren, Z. Liu, T. Li, et al. Design of a novel cocrfenicu$_{0.3}$ high entropy alloy with desirable mechanical, corrosion and anti-bacterial properties via adjusting cu distribution. *Mater. Today Commun.* **35**: 105946 (2023)

[241] L. Yang, E. Zhou, Y. Dong, J. He, Z. Tian, et al. Al/cu enhancement in marine anti-biofouling and anti-biocorrosion performance of high-entropy alloys. *Adv. Funct. Mater.* **35**(39): 2502816 (2025)

[242] K. Xu, S. Li, S. Xie, H. Cao, C. Chen, et al. Microstructural evolution, corrosion resistance and antifouling performance of cocrfenicu$_x$ high-entropy alloy coatings fabricated by high-speed laser cladding. *Surf. Coat. Technol.* **514**: 132565 (2025)

[243] E. Zhou, Y. Yang, J. Li, G. Ren, L. Li, et al. A novel cu and ag bearing high-entropy alloy with remarkable antibacterial, antiviral and anti-microbial corrosion performances. *Adv. Funct. Mater.* **35**(15): 2419467 (2025)

[244] H. Chen, S. Han, H. Zhang, Z. Wang, Q. Du, et al. Corrosion resistance and antibacterial performances of a novel ag-bearing fecrnial$_{0.6}$ti$_{0.6}$ag$_x$ high entropy alloy coatings. *Corros. Sci.* **256**: 113224 (2025)

[245] J. Chen, P. Wang, J. Li, C. Wang, J. Wang, et al. Self-powered antifouling uvc pipeline sterilizer driven by the discharge stimuli based on the modified freestanding rotary triboelectric nanogenerator. *Nano Energy* **95**: 106969 (2022)

[246] Y. Long, Y. Yu, X. Yin, J. Li, C. Carlos, et al. Effective anti-biofouling enabled by surface electric disturbance from water wave-driven nanogenerator. *Nano Energy* **57**: 558-565 (2019)

[247] C. Liu, J. Liu, J. Liu, C. Zhao, B. Shan, et al. A wind-driven rotational direct current triboelectric nanogenerator for self-powered inactivation of seawater microorganisms. *Mater. Today Energy* **26**: 100991 (2022)

[248] Y. Li, L. Dong, X. Sun, X. Xu, W. Li, et al. Tuning charge donation and trapping abilities

Just Accepted

in triboelectric nanogenerators via amino-modified zif-8 mof for self-powered dual-electric-field antifouling. *Nano Energy* **156**: 112152 (2026)
[249] X. Sun, L. Dong, Y. Liu, X. Li, J. Liu, et al. Biomimetic pva-pvdf-based triboelectric nanogenerator with mxene doping for self-powered water sterilization. *Mater. Today Nano* **24**: 100410 (2023)
[250] A. Calisi, C. Lorusso, J. A. Gallego-Urrea, M. Hassellöv, F. Dondero. Ecotoxicological effects of silver nanoparticles in marine mussels. *Sci. Total Environ.* **851**: 158113 (2022)
[251] B. G. Campos, L. B. Moreira, P. G.F.E, A. C. F. Cruz, F. C. Perina, et al. Water and sediment toxicity and hazard assessment of dcoit towards neotropical marine organisms. *Environ. Pollut.* **330**: 121797 (2023)
[252] M. Lagerström, F. Le Bihanic, L. Veensalu, R. Boisserie-Gimenez, L. Racofier, et al. Sustainability ranking of antifouling coatings for leisure boats – balancing efficacy and environmental impact. *J. Hazard. Mater.* **506**: 141600 (2026)
[253] J.-y. Xu, P. U. Xin-Ming, D.-c. Lu, Y.-y. Xing, C. Liu, et al. Seawater quality criteria and ecotoxicity risk assessment of zinc oxide nanoparticles based on data of resident marine organisms in china. *Sci. Total Environ.* **905**: 166690 (2023)



Zhen Zhang earned his master's degree in Mechanical Engineering from Southeast University in 2025 and is currently pursuing a Ph.D. in the Department of Mechanical Engineering at Tsinghua University. His research focuses on marine antifouling coatings and functional polymer coatings.



Yuhong Liu received her bachelor's degree in Chemistry from Tsinghua University in 2000 and her Ph.D. in Physical Chemistry from the Institute of Chemistry, Chinese Academy of Sciences in 2005. She is currently a tenured professor in the Department of Mechanical Engineering at Tsinghua University. Her research focuses on water-based lubrication, superlubricity, nanotribology, and surface and interface science.

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Jianbin Luo received his bachelor's degree in Metal Working from Northeastern University in 1982, his master's degree in Metallurgy from the University of Science and Technology Beijing in 1988, and his Ph.D. in Mechanical Design and Theory from Tsinghua University in 1994. He is currently a professor at Tsinghua University and a Member of the Chinese Academy of Sciences. His research focuses on thin-film lubrication, superlubricity, and nanomanufacturing.

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