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Recent Advances in Surface-Energy-Regulated Marine Antifouling Coatings for Concrete: Focusing on Superhydrophobic Surfaces

Zhijun Liu[1],Xu Meng1,Shaochun Li[1,*],Xianpeng Wang[2],KovsharS.N[2],Tsyganov A.R[3]

[1]Qingdao University of Technology ,266520,Qingdao, China；

[2] Belarusian national technical university ,220013, Minsk, PR Belarus

[3]International Institute of Management and Business ,220086,Minsk, PR Belarus

KEYWORDS

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Durability

ABSTRACT

Concrete structures exposed to marine, hydraulic, and urban environments are susceptible to multiple fouling processes, including sediment deposition, salt crystallization, organic contamination, and biological attachment, which accelerate surface degradation and reduce durability. Surface coatings are widely applied for concrete protection due to their practical applicability. Conventional barrier-type coatings protect concrete by blocking water and aggressive ion penetration, but their long-term performance is often limited by aging, interfacial degradation, and mechanical damage. Anti-organic-fouling coatings reduce oil and organic adhesion and show effectiveness in specific scenarios, although durability and environmental adaptability remain challenging. In recent years, surface-energy-engineered superhydrophobic coatings have attracted increasing attention. By combining micro-/nanostructured surfaces with low-surface-energy materials, these coatings impart high contact angles and low adhesion, effectively suppressing liquid wetting, particulate accumulation, and early-stage biofouling. This review systematically summarizes fouling mechanisms on concrete surfaces, compares different anti-fouling coating strategies, and discusses current challenges and future directions toward durable and practical concrete protection.

INTRODUCTION

Concrete is one of the most widely used structural materials in marine engineering, hydraulic infrastructure, and coastal constructions. During long-term service in harsh environments characterized by high humidity, high salinity, strong corrosion, and intense biological activity, concrete surfaces are highly susceptible to multi-source contamination and degradation. In marine environments, the coupled effects of chloride ingress, sediment deposition, organic pollutant adsorption, and biofouling by microorganisms and macro-organisms significantly accelerate surface deterioration, resulting in reduced service life, increased maintenance costs, and potential safety risks. Therefore, the development of efficient, durable, and environmentally friendly antifouling technologies for concrete surfaces has become a critical scientific and engineering challenge in marine engineering[1-3].

Conventional antifouling strategies for concrete mainly rely on biocidal coatings or dense barrier coatings, which retard deterioration by releasing active agents or blocking the penetration of aggressive species. However, these approaches often suffer from environmental concerns, limited service life, and poor adaptability to concrete substrates, making them insufficient for long-term applications in marine environments[4]. In recent years, inspired by advances in biomimetic interface science and wetting theory, surface-energy-regulated antifouling strategies have attracted increasing attention. By reducing surface free energy and constructing micro/nanostructures,

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* Corresponding author. E-mail address:lishaochun@qut.edu.cn

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the interfacial interactions between contaminants and substrates can be significantly weakened, thereby suppressing fouling at the source[5].

In this context, superhydrophobic coatings, characterized by high water contact angles, low sliding angles, and excellent liquid repellency and self-cleaning properties, have been regarded as promising candidates for non-adhesive antifouling protection of concrete surfaces. Numerous studies have demonstrated that superhydrophobic surfaces can effectively reduce the adhesion of sediments, organic pollutants, and early-stage biofilms, while simultaneously retarding chloride penetration and reducing water absorption thus offering synergistic improvements in antifouling performance and durability. Nevertheless, their practical application is still hindered by challenges such as insufficient mechanical robustness, limited long-term durability, and difficulties in large-scale fabrication on rough concrete substrates[6-7].

Against this background, this review systematically summarizes recent advances in antifouling coatings for concrete in marine environments, with a particular focus on surface-energy-regulated and superhydrophobic coatings. Their working mechanisms, advantages, limitations, and future research directions are discussed to provide guidance for the development of durable antifouling strategies for concrete structures.

1.Types and Formation Mechanisms of Surface Fouling on Concrete

Concrete surfaces are highly susceptible to multiple pollutants during service, and different fouling types often act synergistically to accelerate deterioration. Based on contaminant characteristics and mechanisms, surface fouling can be classified into four categories: inorganic salt fouling, organic fouling, sediment and particulate pollution, and biological fouling.

1.1.Inorganic Salt Fouling

Inorganic salt fouling arises from the migration, crystallization, and deposition of soluble salts (e.g., Cl^-, SO_4^{2-}) on concrete surfaces [8]. In humid or saline conditions, salts accumulate via capillary transport and precipitate during wet-dry cycles or evaporation [9], as shown in Table1. Although its effect on structural capacity is limited, salt crystallization markedly reduces durability. It can increase surface roughness by about 60%, while crystal growth and pore blockage diminish freeze-thaw resistance, abrasion resistance, and resistance to moisture and ion transport [10]. Liang et al. [11] further reported that banded deposits formed under flowing water reduce surface compactness and enhance adhesion, promoting particulate accumulation, deeper carbonation, microcrack propagation, and up to a 40% drop in impermeability.

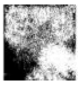
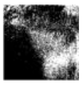
Specimen	MK-0	MK-5	MK-10	MK-15	MK-20
Efflorescence image					
Efflorescence area image					
Area(%)	60.16	50.91	42.24	34.12	30.27

Table.1. Relative efflorescence area of concrete with different mix proportions

* Source: Study on the efflorescence behavior of concrete by adding metakaolin [10]

1.2.Organic Fouling

Organic fouling occurs because the porous concrete matrix readily adsorbs and retains oils and hydrocarbons, forming sticky contamination layers [12]. Their oxidation products and associated microbes reduce alkalinity and induce chalking, as shown in Fig1. Prolonged grease exposure may cause tackiness, discoloration, mold, and compressive strength losses exceeding 60% [13]. Low-viscosity fuels such as gasoline and diesel, though weakly corrosive, can dissolve loose surface phases and weaken steel-concrete bonding, causing an 18-22% strength reduction after 150 days [14]. In heavily oil-contaminated industrial environments, oxidized mineral oils combine with dust to form sludge, producing dark staining and reducing the elastic modulus (~18%) and friction coefficient (~37%) [15].

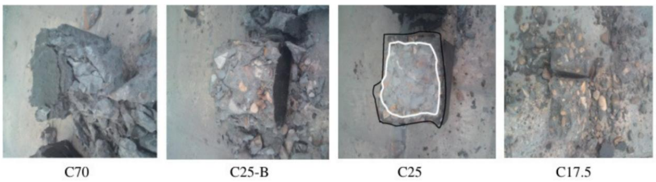


Fig.1. Penetration of petroleum through different types of concrete

* Source: Compressive strength performance of low-and high-strength concrete soaked in mineral oil [15]

1.3.Sediment and Particulate Fouling

Sediment and particulate fouling commonly occurs in environments with flowing water or airborne dust. Fine particles increase surface roughness, promoting the retention of dust, organics, and microbes and raising the risk of secondary contamination [16]. Particle impact under water or wind erosion further induces abrasion and microcrack propagation, reducing compactness as well as impermeability and abrasion resistance [17]. Fernandez [18] reported that airborne particulates not only alter surface appearance but also adsorb moisture and acidic species, accelerating weathering and alkalinity loss. Notably, the increasing proportion of carbonaceous and nitrogenous particles enhances SO_2 oxidation and gypsum formation, triggering “black crust” development and advancing calcite sulfate conversion, thereby compromising surface integrity [19].

1.4.Biological Fouling

Biological fouling is a major deterioration process in moist, nutrient-rich, and mild-temperature environments[21], as shown in Fig2. It is driven by the adhesion, growth, and metabolism of bacteria, fungi, algae, and macrofoulers, typically progressing through initial attachment, biofilm formation, metabolite accumulation, and microbial corrosion.

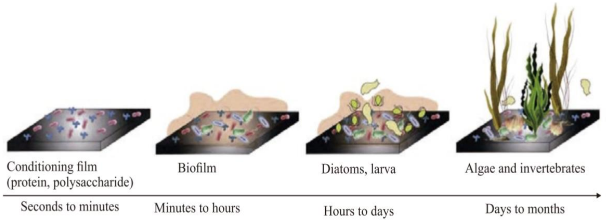


Fig.2. Typical growth processes of algal organisms

* Source: Latest Research Progress of Marine Microbiological Corrosion and Bio-fouling, and New Approaches of Marine Anti-corrosion and Anti-fouling [20]

It is characterized by concealed onset, cumulative effects, and complex mechanisms. Existing studies generally categorize it into three types[22]:

(1) Non-structural fouling

Caused by the deposition of microbial pigments (e.g., melanin, carotenoids, phycobilins), producing black, yellow, or green staining. Although it does not directly damage the material, it degrades appearance and alters optical and moisture-related surface properties, facilitating further microbial colonization [23].

(2) Functional fouling

Originates from biofilm formed by extracellular polymeric substances (EPS). Biofilms increase roughness and enhance adhesion of dust, particulates, and organics, and may clog capillary pores, impeding moisture and gas transport. This leads to scaling, chalking, and surface softening, reducing impermeability and self-cleaning ability and fostering composite fouling layers [24].

(3) Structural fouling

Driven by corrosive microbial metabolites such as organic acids, sulfides, and ammonia, causing dissolution, alkalinity loss, pore structure alteration, and elevated moisture content, as shown in Fig3. These changes accelerate carbonation, decalcification, and sulfate reactions [23]. In marine settings, microbial-induced corrosion (MIC), combined with the localized stress from barnacles and mussels, disrupts surface uniformity and promotes microcrack initiation, significantly reducing durability [25-26].

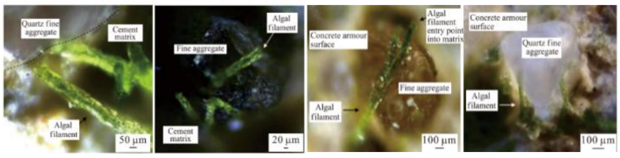


Fig.3. Algae filaments grow inside the concrete

* Source: Microscopic Study into Biodeterioration of Marine Concrete [26]

2.Progress in Anti-Fouling Coatings for Concrete

2.1.Barrier-Type Coatings

Barrier-type coatings form dense, continuous layers on concrete surfaces to block moisture, Cl^-, SO_4^{2-}, and acidic media, delaying degradation and enhancing durability. Common materials include epoxy, polyurethane, acrylic, and inorganic silicate coatings.

Epoxy coatings feature high crosslink density and low porosity; for instance, kaolin/epoxy composites still exhibit ~80% lower Rp than uncoated samples after three wet-dry cycles [27], as shown in Fig4. Polyurethane coatings, valued for flexibility, suit microcracked or impact-prone concrete; asphalt-polyurethane (As/PU) composites maintain ~0.9× compressive strength after 28-day immersion in 3% H_2SO_4 [28]. Bio-based polyurethanes show improved dynamic performance, with bending strain rate rising from 0.00033 to

0.067 s^{-1} and impact strain energy density increasing ~11.3× under dynamic loading [29], as shown in Fig5(a). Acrylic coatings can penetrate 20-30 mm into concrete to fill microcracks; after fluorosilane modification, contact angle increased from 82.0° to 93.7°, and surface energy decreased from 33.8 to 24.0 mJ/m^2 [30], as shown in Fig5(b).

However, long-term weathering and environmental stability remain concerns. Epoxy loses interfacial adhesion under heat, moisture, and UV [31]; TDI monomer release during polyurethane curing may reach 0.3 mg/m^3, accelerating aging at crack tips [32]; acrylics often retain residual monomer, a potential carcinogen, though modification can reduce it below 0.05%. Moreover, acrylics have poor water compatibility and relatively lower hardness and abrasion resistance.

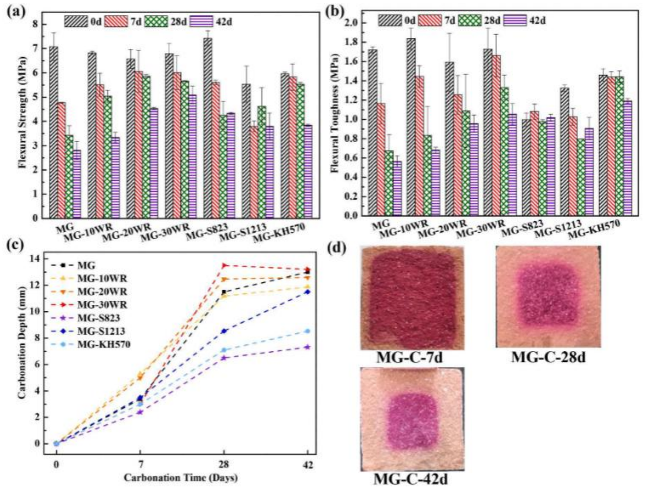


Fig.4. Increase of Rp of the cylinders coated with MG,MG-WR and MG-SCA compared to uncoated samples cured for different ages and conditioned for (a)0,(b)2 and (c)3 cycles of seawater wet-dry conditioning, and(d)the decrease of Rp after 2 and 3 wet-dry conditioning cycles

* Source: Coating performance, durability and anti-corrosion mechanism of organic modified geopolymer composite for marine concrete protection [27]

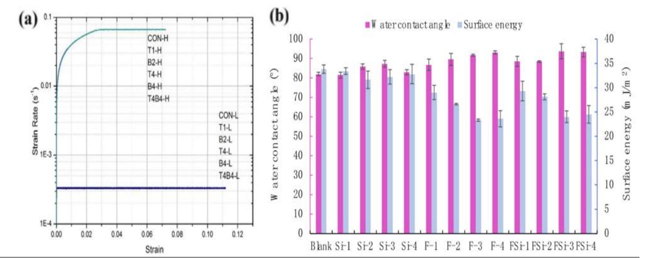


Fig.5.(a) The actual strain rate-strain curves of test specimens at varying strain rates,(b) Contact angle and surface energy of the hardened modified resins

* Source: Behaviour of concrete specimens retrofitted with bio-based polyurethane coatings under dynamic loads [29] Multiscale modification on acrylic resin coating for concrete with silicon/fluorine and graphene oxide (GO) nanosheets [30]

2.2.Anti-Organic-Fouling Coatings

Concrete in traffic, food-processing, and marine environments is prone to oil and organic contamination, which affects appearance and surface performance. Superhydrophilic hydration layer coatings have recently attracted attention [33]. Their mechanism involves introducing hydrophilic polymer chains on the surface, which rapidly absorb water to form a stable hydration layer.

This layer exhibits low oil adhesion and excellent self-cleaning. PSBMA significantly enhances surface hydrophilicity and reduces protein adsorption; Cu-modified PDA-based coatings show >90% antibacterial efficiency and 85% reduction in protein adsorption. PEG coatings rely on ether-water hydrogen bonding to resist adsorption of diverse organics; TEOS-modified PEG emulsions reduce capillary water absorption by ~84.1% (C40) and 83.1% (C50) [34], as shown in Fig6. Superhydrophilic coatings from TiO_2 nanoparticles and siloxane oligomers reduce water uptake by >85%, display a 152° contact angle with chloroform, and allow easy removal of oil and dust without detergents [35].

Compared with traditional barrier coatings, hydration-layer coatings offer environmental friendliness and high biocompatibility, showing potential for marine oil spill protection and nearshore anti-fouling applications [36]. However, chemical, mechanical, and long-term stability remain limited; in 3.5 wt.% NaCl, pH 5, and pH 9 solutions, performance declines. Molecular design, interfacial engineering, and fabrication optimization are required to achieve durable, high-efficiency anti-fouling [37].

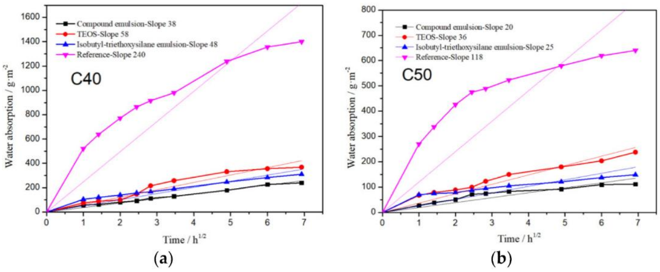


Fig.6. Effects of capillary water absorption of concrete for different coating: (a) C40; (b) C50

* Source: Preparation of Modified Silane Composite Emulsion and Its Effect on Surface Properties of

Cement-Based Materials [34]

2.3.Antimicrobial / Anti-Biofouling Coatings

Concrete structures in marine, hydraulic, and high-humidity environments are prone to biofouling by bacteria, fungi, algae, and larger organisms, which increase surface roughness and promote biofilm formation, accelerating microbiologically influenced corrosion (MIC) and compromising durability [38-39]. Antimicrobial/anti-biofouling coatings inhibit microbial attachment and growth via chemical, photocatalytic, or contact-killing mechanisms [40].

Chemical killing relies on active metal ions (e.g., Ag^+, Cu_2O, ZnO) to disrupt microbial metabolism. ZnO-NPs combinations can suppress biofilm formation by 65–85% [41], while Ag-NPs reduce E. coli by ~5.8 log CFU/cm² and S. aureus by ~4.11 log CFU/cm² [42]. However, these coatings mainly act in early fouling stages, and sustained ion release poses environmental risks; for example, organotin compounds (TBT) persist in marine environments, bioaccumulate, and disrupt endocrine systems [43]. In addition, metal ion agents may be incompatible with coating matrices, e.g., Cu_2O in epoxy or fluorocarbon systems can weaken crosslinking, reducing adhesion and tensile strength [44].

Photocatalytic killing, exemplified by TiO_2, generates OH^- and O_2^- under UV or visible light, oxidizing microbial membranes and DNA, as shown in Table2. Fe-doped TiO_2 (1 g/L) achieved 3.57 ± 0.21 log (E. coli) and 3.49 ± 0.42 log (S. aureus) reduction under 240 min visible light, reaching 99.97% killing efficiency [45]. Effectiveness declines under low light or underwater conditions [46]; element doping, graphene composites, or noble metal modifications expand light response but increase cost and environmental concerns [47].

Contact-killing coatings introduce cationic groups (e.g., quaternary ammonium salts, guanidine polymers) that disrupt microbial membranes electrostatically. QAC-functionalized coatings reduce E. coli, drug-resistant A. baumannii, and Listeria monocytogenes by >5.0 log CFU/cm² (~99.999%) within seconds [48]. However, covalently grafted functional polymers lose activity if the coating suffers mechanical damage, chemical cleaning, or aging, limiting long-term efficacy [49].

Overall, antimicrobial/anti-biofouling coatings effectively suppress microbial adhesion and biofilm formation, offering a key strategy against concrete bio-corrosion. Yet, chemical toxicity, photocatalytic light dependence, and contact-killing durability remain challenges, necessitating further optimization in material design, interface engineering, and ecological safety.

Table.2. Photocatalytic efficiency of cementitious materials with titanium dioxide incorporation

Material	Application Process	Quantity	Efficiency	Environmental Conditions	Observations
Anatase I	Mixing technique	10% by weight	$44.1\ mg \times h^{-1} \times m^{-2}$	Flow rate 1.0 L × min^{-1}; UV-A radiation 10 ± 2 W × m^{-2}, relative atmospheric humidity 50 ± 5% and pollutant concentration (NO_x) 20 ppmv.	The efficiency is in terms of removal of NO_x.
Anatase II			$11.71\ mg \times h^{-1} \times m^{-2}$		
Rutile			$37.24\ mg \times h^{-1} \times m^{-2}$		
Nano-TiO_2-SiO_2	Dip coating	-	87% of MG, 80% MB and 65% MO	UV irradiation, room temperature.	The photocatalytic material was added to WPC blocks. The efficiency is in terms of percent of decomposit

					ion of dyes.
P25 (75 % anatase and 25% rutile)	Mixing technique	2% by weight of binders	NOx removal rate: 28 μmol $m^{-2}\times h^{-1}$. NO_x removal ratio: 5.8%	24 h UV-A irradiation.	The TiO_2 was incorporated in self-compacting glass mortar (SCGM). WPC and metakaolin were used as cementitious materials.
Nano-TiO_2	Dip coating and vacuum saturation	TiO_2 ethanol suspension of 0.05 g × mL^{-1}	Toluene removal efficiency: 95 % elimination rate: 60–70 mg × $m^{-2}\times h^{-1}$	24 °C, 52% relative humidity.	Initial toluene concentration: 15 ppmv gas residence time: 3 min.

* Source: Challenges and Opportunities of Using Titanium Dioxide Photocatalysis on Cement-Based Materials [46]

3.Surface-Energy-Controlled Coatings

Surface-energy-controlled coatings reduce the free energy of concrete surfaces, hindering wetting or adhesion of dust, sediments, oils, and biofouling proteins, thereby achieving anti-fouling and self-cleaning. Inspired by the lotus effect (static water contact angle >150°, roll angle ~2°) [50-51], as shown in Fig7.superhydrophobic coatings are created by combining micro/nano surface structures with low-surface-energy materials, forming a solid-gas-liquid interface. Droplets rest on trapped air within the microstructures (Cassie state) [52].

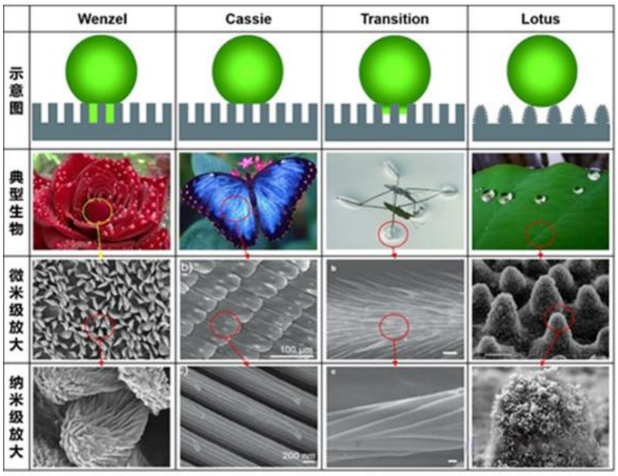


Fig.7. Superhydrophobic Phenomenon in Nature

Yang et al. [53] developed a sprayable superhydrophobic antibacterial coating exploiting concrete hydrophilicity and capillarity, reaching 162.4° contact angle, 5° roll angle, >70% water repellency improvement, and 84.6% (E. coli) / 90.4% (S. epidermidis) antibacterial rates. Song et al. [54] produced low-cost, fluorine-free, mechanically robust coatings (160 ± 1° contact, 6.5 ± 0.5° roll) maintaining superhydrophobicity after blade scratches and sandpaper abrasion, with excellent anti-icing, corrosion resistance, and low ice adhesion. Gu et al. [55] reported multi-layer surfaces retaining 154.4° contact and 4.3° roll after 115 days outdoor exposure. Wu et al. [56] used perfluorinated copolymers with SiO_2 nanoparticles to delay ice formation 1765 s at -20 ℃ and reduce ice adhesion 22-fold, as shown in Fig8. Kong et al. [57] fabricated hierarchical DE/Al_2O_3@STA-geopolymer surfaces, achieving 159.1 contact angle and 4.5° roll angle, with excellent water repellency, self-cleaning, and resistance to Cl^- corrosion and mechanical abrasion.

However, the Cassie state is thermodynamically metastable and vulnerable: static pressure or impacts exceeding the Laplace limit, prolonged immersion, or droplet evaporation can collapse the air layer; mechanical wear or oil contamination damages microstructures; its energy barrier is lower than the Wenzel state, allowing irreversible transition to full wetting [58-60]. These intrinsic limitations critically restrict long-term stability and anti-fouling performance of superhydrophobic coatings in real-world applications.

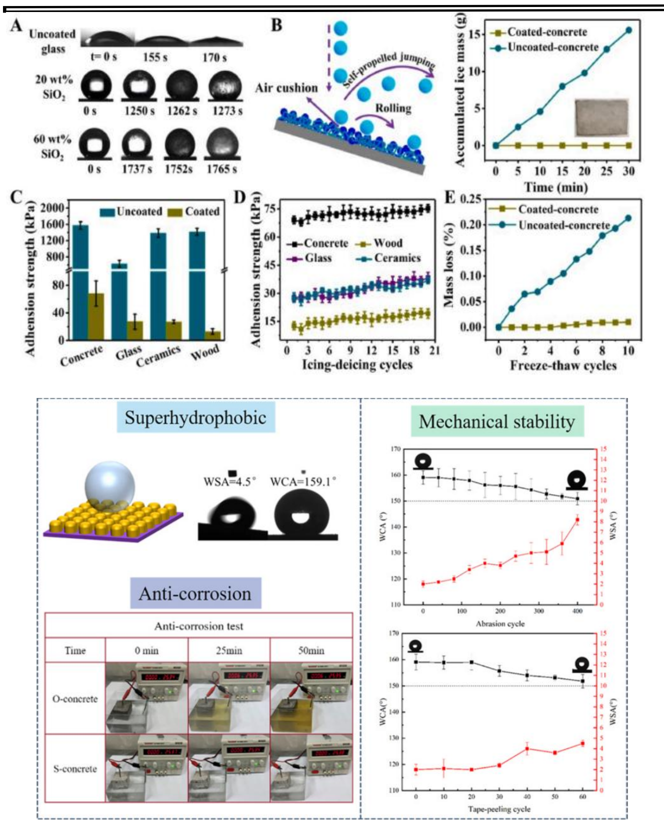


Fig.8. Anti-icing performance of the FSC$_4$/SiO$_2$F composite coating.(A)Photographs showed dynamic icing process of 2 uL water droplets on the untreated and FSC/SiO$_2$-F coated surface at -20 "C.(B)Schematic illustration of the cold-rain simulated experiments, and the accumulated ice mass as a function of the treating time.(C)Ice adhesion strength of various FSC$_4$/SiO$_2$-F coated substrates.(D)Ice adhesion strength as a function of icing-deicing cycles.(E)Mass loss of coated and uncoated concrete as a function of freeze-thaw cycles. (F) The surface contact angle and rolling angle of superhydrophobic concrete

* Source: An extremely chemical and mechanically durable siloxane bearing copolymer coating with self-crosslinkable and anti-icing properties.[56] Superhydrophobic concrete coating with excellent mechanical robustness and anti-corrosion performances.[57]

Conclusion

Surface fouling and bioadhesion are critical factors that limit the long-term durability of concrete structures in marine environments. Compared with conventional antifouling strategies based on the release of active agents, surface-energy-regulated coatings can fundamentally reduce fouling by weakening the interfacial interactions between contaminants and substrates, offering clear advantages in terms of environmental friendliness and long-term stability. Among them, superhydrophobic coatings, featuring low surface energy, micro/nanostructured roughness, and excellent self-cleaning behavior, have demonstrated considerable potential in suppressing sediment deposition, organic contamination, and early-stage biofouling.

Nevertheless, the engineering application of superhydrophobic coatings on concrete still faces several challenges, including insufficient mechanical robustness of micro/nanostructures under abrasion and erosion, limited adhesion to concrete substrates, and difficulties in large-area and complex-surface fabrication. In addition, issues related to material cost, process complexity, and long-term performance degradation require further investigation.

Future efforts should focus on enhancing mechanical durability and interfacial bonding, developing multifunctional protective systems, and promoting scalable fabrication techniques suitable for practical engineering applications. With continuous advances in surface engineering, materials science, and construction technologies, surface-energy-regulated superhydrophobic coatings are expected to play an increasingly important role in antifouling protection and durability enhancement of marine concrete structures.

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