



A review on coatings based on modified surface topography-biomimetic approach for the inhibition of marine biofouling

Lokeshwar Mahto^{*a}, Dharm Pal^{*a}, Vivek Kumar Gaba^b and Manisha Agrawal^c

^aDepartment of Chemical Engineering, ^bDepartment of Mechanical Engineering,

National Institute of Technology Raipur, Raipur-492 010, Chhattisgarh, India

^cDepartment of Chemistry, Rungta College of Engineering and Technology, Bhilai-490 020, Chhattisgarh, India

E-mail: lokeshwarmahato@gmail.com, dpsingh.che@nitrr.ac.in

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Biofouling of marine ships and structures is always detrimental to the maritime industry resulting in a reduction in speed and increased cost of operation and maintenance. Developing a non-polluting environmental-friendly and highly efficient coating towards all kind of biofouling organisms is a challenging task. Because of the recent technological advancements in nanotechnology, one of the strategies is to modify the surface topography inspired from nature. Bio-mimicking the topography of natural surfaces like animal skin and surface of leaves facilitates to repel fouling organisms from artificial surfaces effectively. In this article, the topographic properties such as surface roughness and superhydrophobicity relating to the anti-biofouling mechanism and the technologies like photolithography, laser ablation, template casting, etc. used in the fabrication of these surfaces have been critically reviewed to give a deeper insight into the latest developments and challenges associated with coating and biofouling.

Keywords: Marine coating, biofouling, topography, surface modification, bio-mimicking.

Introduction

The issue of biofouling has always been with the marine industries from the inception, where biomolecules and marine species adhere and settle over an artificial surface as it is submerged in seawater^{1,2}. This has a detrimental effect on moving vessels as it increases the weight as well as frictional resistance³. The speed of the vessel is compromised which results in increased fuel consumption for big freight vessels⁴. The effect is harmful to the environment as well as monetary losses are incurred⁵⁻⁷. The deteriorated coatings and ship hull are non-degradable which settle down over the seabed which increases chemical deposition⁸. Therefore, vast research has been carried out to develop a novel strategy to curb marine biofouling⁹.

Antifouling coating market is dominated by self-polishing copolymers (SPCs) which release biocides like zinc, copper and booster biocides from the coating surface after hydrolysis with water leaving behind a smooth surface¹⁰. Studies have shown that the dissolved copper level exceeds the per-

missible limits set by the regulatory body¹¹. Tests were done on marine organisms like *Artemia nauplii* and *Paracentrotus lividus* showing fair toxicity towards them¹²⁻¹⁴. The non-toxic coatings being developed using different strategies has proven to be effective to a large extent like foul release coatings, biomimetic materials, zwitterionic polymers, amphiphilic polymers, UV irradiation, etc.^{2,8,15-17}. But, the challenges encountered in developing these non-toxic coatings are considerable like the cost of fabrication, difficulty in applying the coating, non-facile synthesis techniques, etc.⁴.

Many natural organisms and plants defence mechanism against fouling are achieved by a micro-topography structure like the skin of a shark¹⁸, water striders (*Gerris remigis*)¹⁹, molluscs²⁰, oysters, lotus leaf²¹, etc. and upset the settlement of spores of algae, bacteria and barnacles. This also changes the wettability of the surface and it becomes hard for the foulants to adhere with the superhydrophobic surfaces formed. Mimicking the natural surfaces and developing a replica to get the desired properties of the artificial sur-

faces is called bio-mimicking. Researchers are lately investigating to incorporate the biomimetic approach to control biofouling as the technological advancements in nanotechnology has made it possible to fabricate a micro-textured and nano-textured surface facile^{8,22–25}.

In this article, the surface properties of bio-mimicking surfaces like surface roughness and super hydrophobicity responsible for antifouling behaviour and their synthesis and development methods like photolithography, template casting, electrodeposition, laser ablation, etc. are systematically reviewed.

Key topographic surface properties

Surface roughness:

The cutoff length of the surface roughness and porosity has an impact on the settlement of foulants on the submerged surface. The slopes of the rough surface are accessed by the foulants and they attach to it by secreting adhesives to form a strong bond. The bacteria and marine foulants are protected in the crevices of the surface as they are confined and become difficult to remove by external forces like water shear, abrasion, etc.²⁶.

Because of high roughness of the surface, the total adhesion sites for the foulants to colonize increases. The major microfouling organisms like spores of algae, diatoms and bacteria and the major macrofouling organisms like larvae, tubeworms, bryozoans, etc. are affected by different scales of topography as the size of microfoulants and macrofoulants vary largely from few micrometres to hundreds of micrometre. The mechanism is believed to be dependent on the number of attachment points present on a surface.

Size of fouling organism greater than the primary length dimension of the surface adheres weakly as it gets fewer attachment points as seen in Fig. 1(d). Similarly, fouling organisms whose size is smaller than the primary length dimension will strongly adhere to the surface because it gets more attachment points at the surface to adhere as illustrated in Figs. 1(b) and 1(c)⁵.

This theory of attachment points has been studied for several fouling organisms like spores of green fouling algae *Ulva*^{21,27}, diatoms of *Nitzschia cf. paleacea*, *Fallacia carpentariae*, *Navicula jeffreysi* and *Amphora sp.* having sizes from 1 to 14 μm ²⁸. Small motile microfoulers *U. rigida* and

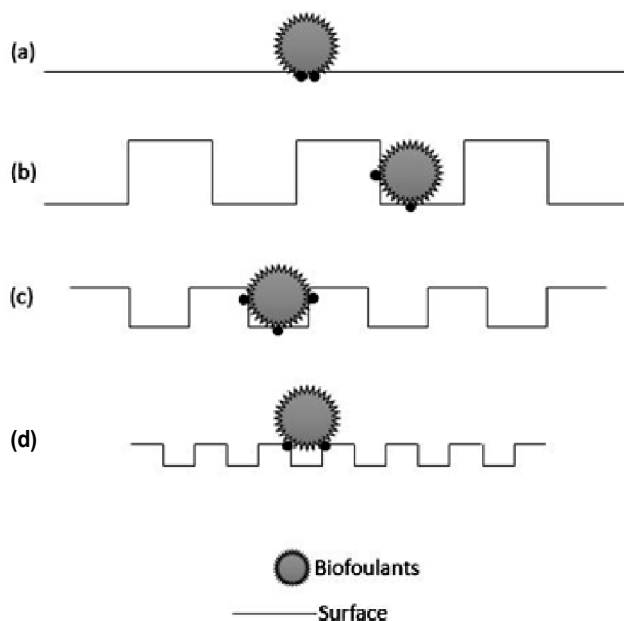


Fig. 1. Schematic of attachment point theory showing attachment sites available for microfoulants: (a) over a flat surface, (b) roughness greater than foulant size, (c) roughness equal to foulant size and (d) roughness less than the foulant size.

Amphora sp. (7 μm) showed a weak relationship to attachment points, macrofouling larvae *B. neritina* and *H. elegans* having sizes from 129–321 μm showed a good relationship with attachment points and no effect was seen for non-motile spores of algae *C. clavulatum* (37 μm). The study was conducted on nine equally segmented zones prepared on polycarbonate plates of eight different microtextured length scales between 4 and 512 μm and one unmodified region²⁹. Cyprids of *Balanus amphitrite* showed preference towards sinusoidal linear textures with aspect ratio 1:1 ranging from 0 to 32 μm ³⁰.

These studies reveal that the roughness is a dominant factor which needs to be considered when developing an antifouling surface depending on the application. For marine antifouling application, as the sizes differ greatly from nanometers to centimetres, the surface should be able to resist biofouling of all sizes of fouling organisms.

Superhydrophobicity:

Superhydrophobicity, also called a lotus effect, in nature is defined as the physicochemical phenomenon attributed by low surface energy and roughness making the surface

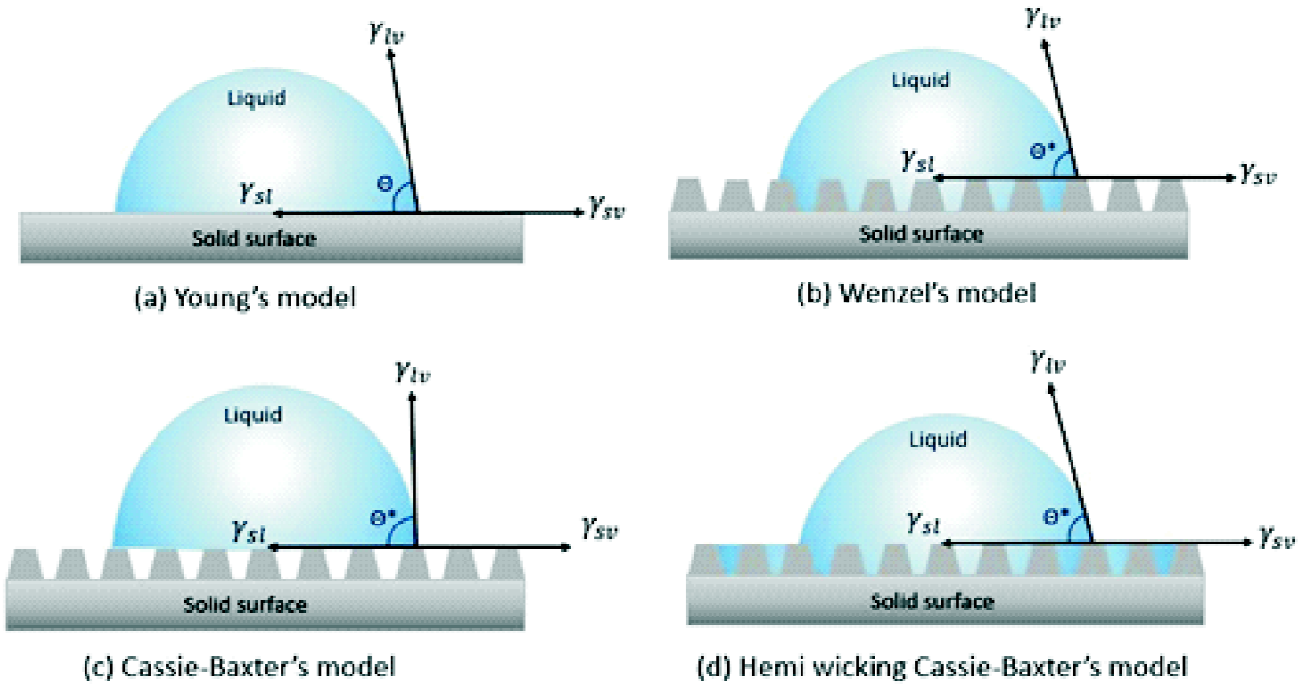


Fig. 2. Schematic of different models proposed to show the wettability: (a) Young's model for a smooth surface, (b) Wenzel, (c) Cassie-Baxter and (d) hemi wicking Cassie-Baxter model for rough surfaces.

extremely difficult to wet. The water droplets formed are nearly spherical and is easily rolled off the surface taking contaminants and dust particles with it making the surface clean³¹.

A surface is said to be superhydrophobic when the angle formed between the water droplet and surface is more than 150° . Contact angle (θ) is calculated theoretically by Young's equation³² giving us the wettability characteristics of a smooth surface, eq. (1):

$$\cos \theta = \frac{\gamma_{sa} - \gamma_{sl}}{\gamma_{la}} \quad (1)$$

where, γ_{sa} is the surface tension between air and solid, γ_{sl} is the surface tension between liquid and solid and γ_{la} is the surface tension between liquid and air, respectively.

For heterogeneous surfaces, the wetting behaviour is followed by a more complicated route given by Wenzel and Cassie^{33,34}. Wenzel model assumes rough surface and water droplets penetrate the troughs filling all the protrusions. The apparent contact angle by Wenzel model (θ^*) corresponding to the stable equilibrium state for rough surface related to smooth surface contact angle (θ) is given as

$$\cos \theta^* = r \cos \theta \quad (2)$$

where, r , is the roughness factor and is given by

$$r = \frac{\text{Actual rough surface area}}{\text{Smooth surface area}} > 1 \quad (3)$$

Solid fraction ϕ_s is a characteristics parameter of the roughness effect given by the ratio of surface cross-sectional area to substrate area. For smooth surfaces, $r = 1$ and $\phi_s = 0$. It is inferred from the Wenzel equation that a hydrophilic surface renders more hydrophilicity and a hydrophobic surface renders more hydrophobicity³². On a rough surface, air pockets are created in the protrusions generating a solid-liquid-gas interface below the liquid, the Cassie-Baxter model is suitable for this surface. The hemi-wicking state is said to form when a precursor liquid film is developed on the rough surface. The Cassie-Baxter model equation is given by³⁵:

$$\cos \theta^* = \phi_s \cos \theta - (1 - \phi_s) \quad (4)$$

and Hemi-wicking Cassie-Baxter equation model is given by

$$\cos \theta^* = \phi_s \cos \theta + (1 - \phi_s) \quad (5)$$

Natural surfaces like insect wings which are super-

hydrophobic repel dirt and fouling organisms as they are easily rolled off. The contact angle of cicada wings and dragonfly wings when examined were between 149–164°. This gives researchers the idea to incorporate superhydrophobicity to the artificial surfaces to make it repel fouling organisms as a natural surface would do. This was studied by Gangadoo *et al.* where they made use of template replication technique which gave fair antifouling results to the artificial surfaces prepared³⁶. It is also seen that the superhydrophobicity along with surface topography, roughness level and amount of air trapped in the groove plays an important role in developing antifouling surfaces³⁷.

Fabrication techniques to produce biomimetic surface

Microtopographical surfaces are produced by laser ablation^{27,28}, photolithography³⁸, castings, etc. They are generally applied to polymeric substrates like polydimethylsiloxane (PDMS), polyvinyl chloride, polycarbonate, etc. The adhesion of spores of *Ulva* is studied to be greater in the hydrophilic surface as compared to hydrophobic surfaces indicating surfaces with low surface energy being prone to fouling by *Ulva*³⁹. Engineered microtopographic surfaces show a reduction in the growth of *Ulva* spores by 58% on a pillar of diameter 2 μm and pyramid of 10 μm . About 77% of the surface imitated shark skin⁵. Fabricating a surface having a single length dimension will not be fruitful as biofoulants come in all kind of shapes and sizes⁴⁰.

Efimenko *et al.*⁴¹ developed hierarchically wrinkled surfaces with dimensions ranging from nanometer to millimeter where the larger wrinkled structure was placed below the small wrinkles in a nested pattern. The roughness was achieved of variable length scale by wrinkling process in which the surface was stretched by applying uniaxial strain then the surface was treated with UV light and ozone after which the strain was released. They reported fouling of flat topographies within a few weeks (4–15 weeks) while hierarchically wrinkled surface topography coatings with same chemistries showed relatively negligible fouling even after a prolonged period of 18 months. Also, barnacles were not seen to settle in. This indicates that surface topography plays an important role against adhesion of barnacles without disturbing the chemical structure of the coating⁴¹.

Amphiphilic surface with nano-roughness is also seen to resist biofouling effectively as demonstrated by Wang *et al.*⁴² where they made polydimethylsiloxane based amphiphilic coating incorporating polyethylene glycol and dopamine methacrylamide. Heterogeneous nano-scale roughness was seen in the range of 4.54 nm to 13.8 nm.

Table 1 gives detailed information about the recent researches carried out in fabricating non-fouling surfaces using biomimetic approaches. It can be seen that the surface roughness itself affects the biofouling adhesion and settlement. But, it still does not prohibit attachment completely and also the mechanical strength of the fabricating surfaces is a factor to ponder upon. Bio-mimicking the lubricating behaviour of natural surfaces like fish skin is also studied recently. It is difficult to make a robust surface with artificial lubricating behaviour which lasts for a long period because it gets depleted quickly and the lubricant attachment strength to the substrate is non-lasting⁵⁵.

Liu *et al.* proposed a different approach centred around water jets spraying outward for marine antifouling surfaces where a flowing field layer is developed which produce forces greater than the adhesion force based on attachment point theory⁵⁴.

Making a robust coating with excellent mechanical strength and antifouling properties requires the incorporation of multiple techniques and strategies like making the surface rough and lubricating. The chemistry of the coating material is an important factor because wettability is dependent on the interface molecules. So, the material used in the preparation needs to be looked upon.

Economic analysis

The cost of antifouling coating mainly depends on the material used, the synthesis method adopted and coating application technique.

As the prices differ according to the variation in average molecular weight and viscosity of the polymers, the cost of raw material also depends on the specification required. For the generally used materials encountered in this review, silicone resins are comparatively expensive as compared to epoxy resins but the antifouling results achieved by silicone resins are much better than achieved by epoxy resins which

Table 1. Different biomimetic surfaces produced using various techniques to counter biofouling

Fabricating material	Bio-mimicking surface	Synthesis technique	Maximum contact angle	Surface roughness	Biofouling testing environment	Ref.
Silicon wafers	Swimming crabs	Reactive ion etching	–	Dia.: 3 µm	70% reduction in <i>Phaeodactylum</i>	43
Silicone resin modified with polyvinyl alcohol and polyacrylamide hydrogel powder	Fish slime-like surface	Blending, spray coating	–	Spacing: 4 µm and 7 µm Dia.: 40 µm	<i>tricornutum</i> settlement No barnacle settlement for 12 weeks in field trials	44
Silicone modified acrylic resin	Lotus leaf, crab shell	Template replication	113.5°	Dia.: 6 to 8 µm Spacing: 9 µm and 10 µm	73% reduction in <i>Closterium</i> and 70% reduction in <i>Navicula</i> after 7 days	45
Epoxy resin	Cicadas wing, dragonfly wing	Template replication	–	Random	Dragonfly replicated surface was the most efficient as compared to commercial coatings	36
Manganese stearate modified aluminium plate	Lotus leaf	Electro-deposition, constant voltage of 30 V for 10 min	169.7°	500 nm to 4 µm	<i>Chlorella vulgaris</i> settlement on the modified surface was 0.02% whereas bare Al surface was 12.8%	46
Silicone modified acrylic resin	Taro leaf, rose petal, shark skin	Template replication	Taro leaf - 136.7°, Rose petal - 123.7°, Shark skin - 112.2°	Taro leaf diameter - 7 to 20 µm, rose petal width - 800 nm, shark skin length - 168 µm and width 180 µm	The overlapping and V-shaped boundary structure of the shark skin showed best results against <i>N. closterium</i> , <i>P. tricornutum</i> and <i>Chlorella</i>	47
Polyethylene oxide and fluorinated chain segments grafted on a substrate	Lubricating protein lubricin	Click coupling reaction	–	Roughness - 1 nm, Sidechain length - 1.5 to 40 nm	91% protein resistance and 84% reduction in cell adhesion	48
Perfluoropoly-ether lubricant infused in the aluminium surface	Lubricant infused surfaces	Pouring onto surface and heating for 120°C	124.1°	30 to 100 nm thick and 400 to 1000 nm long	Stable lubricant infused micro/nano-structured surface resisted adhesion against <i>C. pyrenoidosa</i> and <i>P. tricornutum</i>	49
PDMS elastomer	Placoid scales	Spin coating over moulds created using KOH and reactive ion etching technique	109°	1.5 to 2 µm spacing	75% reduction in <i>E. coli</i> attachment	50

Table-1 (contd.)

Paint matrix prepared with macroalgae and sponge extracts painted on acrylic tiles	Marine macroalgae and sponge extracts	Regular painting	-	Random	51
Polystyrene (PS) coat on silicon wafers	-	Electro-hydrodynamics to prepare PS fibres, spin coating Laser ablation	162.1°	Random	52
Poly(methyl methacrylate)	-		68-78°	Dia: 550 µm, 600 µm and 700 µm Spacing: 1 mm, 1.5 mm and 2 mm	53
Water jet micropores	Kelp	Jet holes	-	Pore dia.: 0.6 mm Pore spacing: 4.5 mm Pore length: 3 mm	54

H. Caerulea and *S. horridum* incorporated paint showed good results

The greater hole size of 720 µm and smaller spacing of 1000 µm gave the least cell attachment of *C. pagurus*

P. tricornutum attachment reduced significantly in jet treated pieces

gives silicone paints better lifespan and becomes a favourable choice. However, the majority of the current market uses copper biocides which presents an economical coating but due to its increasing level of concern towards environment, the market of copper is seen to be diminishing with stringent laws and regulations being enforced in future. Thus, giving impetus to alternatives like silicone and epoxy resins.

Simple synthesis techniques like direct blending of additives and template casting are easy to operate and bear less cost as compared to reactive ion etching and laser ablation technique which require the use of sophisticated machines. But, the cost of application of coating bears a fair share which is not an issue for reactive ion etching and laser ablation technique. As we got closer to the nanoscale, the cost of operation also increases as it requires higher energy but the product gives great antifouling results. The cost of application to big marine structures and ships is equivalent as the coating needs to be uniform and consistent throughout the surface, it requires sophisticated coating techniques like spray coating.

Challenges

As we have seen that modifying the surface topography with chemical changes gives us one of the best results but the challenges associated with it are significant. Super-hydrophobic coatings show excellent anti-corrosion properties with good antifouling properties as well. But, marine fouling organisms come with all kind of adhesives and adhesion mechanism like the bryozoan is said to be hydrophobic in nature and prefers to settle on hydrophobic surfaces. So, the incorporation of nano-scale roughness imitating natural non-fouling surfaces with superhydrophobicity can have a highly efficient non-fouling surface². Non-fouling method solely based on physical attributes are not long-lasting, also they act on only specific foulants^{55,56}. Its applicability and fabrication technique is still not economical for a large scale commercial application⁵⁷.

Conclusion and future perspective

Environmental legislations are becoming more regulated nowadays because of rising environmental concerns. So, the antifouling coatings being developed need to be environmentally-benign and non-toxic. Modifying the surface topogra-

phy of coatings is a viable strategy to inhibit biofouling. Developing a high performance modified surface which inhibits the settlement of fouling organisms efficiently will require multiple factors to be accounted for like chemistry, modulus, topography, etc. This area of research is gaining wide relevance with the recent advancements in nanotechnology but still, it is not economically feasible to employ it on a large scale. In future with more sophisticated, economical fabrication techniques and incorporating multiple mechanisms, it could be possible to see commercial marine structures with micro-textured or nano-textured topography.

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