A Review on the Effect of Microstructure Surface on the Adhesion of Marine Fouling Organisms

Hongyue Yang1,*, Songling Wang2

1School of North China Electric Power University, Baoding, China
2School of North China Electric Power University, Baoding, China

*Corresponding author e-mail: annyang_ncepu@163.com

Abstract. Marine anti-pollution is a difficult issue in marine development. At present, marine anti-fouling researches mainly focus on three aspects: chemical, physical and biological. With the spread of the concept of environmental protection, the development of environmentally friendly, bio-adaptive anti-fouling technology has become a new development trend, and micro-structure surface anti-fouling technology has become a research hotspot. This paper introduces the profile and impact of biofouling, highlights the antifouling mechanism and research progress of microstructure technology, and describes the adhesion characteristics of the three main fouling organisms on the microstructure surface.

1. Introduction

At present, the development of China’s ocean is still at the initial stage with gradual progress. The research and utilization in the resources of marine water, chemicals, biology and minerals need to be deepened. The complicated marine environment has also created certain obstacles to the development and utilization of the ocean, especially the solutions to the problems caused by marine fouling organisms. In 1969, R.A.Horne pointed out in his book "Marine Chemistry” that compared with corrosion. And the fouling of marine life has been a more troublesome problem since ancient times. The reason lies in that the marine fouling organism has a more resistible vitality, thus making it an insurmountable obstacle to the ocean1.

2. The overview and impact of marine fouling organism

Marine fouling organism is a general term for animals, plants and microorganisms that grow on the surface of ships and all facilities in the sea, including bacteria, some diatoms, large algae, and a variety of species from protozoa to vertebrates, including barnacles, hydroids, ascidians, mussels, polyps, seaweeds, etc. The main impact of fouling organisms is caused by the following aspects: reduce the ship’s speed, cause the fuel loss, corrode metal facilities, etc.

It increases the ship's navigational resistance, reduce the speed, and cause a lot of fuel loss and consumption. British International Paint Company has collected statistics on more than 1,500 ships and found that when the fouling in the bottom of the ship is 5%, fuel consumption will increase by 10%. It is estimated that the global ship building industry spends nearly $10 billion on it annually. And the global fuel industry's fuel loss due to fouling organisms is up to 30%2, 3, 4
It aggravates the corrosion of various metal facilities in the ocean. The surface of the ship becomes brittle and detached due to corrosion. And the sailing cycle is reduced to shorten the service life of the ship. The annual loss of hydrogen sulfide from sulfate-reducing bacteria alone has caused the offshore oil industry to suffer losses of hundreds of millions of dollars\textsuperscript{5,6}.

It causes harm to seawater pipelines, instrumentation, hydro-meteorological buoys and other facilities and poses safety hazards for production safety. The seawater cooling system of the ship and the heat exchange equipment of the power plant that are attached by fouling organisms may cause serious accidents due to blockage of the pipeline\textsuperscript{7,8}; the moving parts of the marine monitoring instrument may be malfunctioned due to bio-fouling, and bubbles will be generated during the growth and reproduction of the fouling organisms, so that the acoustics confusion occurs in the process of instrument detection; it may increases the dynamic load effect of buoys and navigation marks in the ocean due to waves, causing them to deviate from the correct orientation and accidents due to this\textsuperscript{9}.

It is harmful to the aquaculture industry. It adheres to the surface of the net cage of aquaculture, resulting in blockage of the mesh and the inability to exchange water, and greatly reducing the yield and quality of the cultivation.

At present, marine anti-fouling research mainly starts from three aspects of: chemical, physical and biological, and the control methods are also becoming increasingly diverse. However, due to the uncontrollable elements, like varieties of fouling organisms, multiple factors contribute to the formation of fouling, and the discrepancies in different sea areas, each anti-fouling technique has its own limitations.

3. Study on Antifouling Mechanism of Micro-structure Surface

Wenzel and Cassie\textsuperscript{10} found that the surface roughness affected the wettability of the surface of the material, and proposed the Wenzel model and the Cassie model respectively. After that, people began to consider the effect of wetness on the bio-fouling of the microstructure surface\textsuperscript{11}. Studies have shown that the appropriate size of the microstructure (shape, height, spacing, etc.) can effectively inhibit the adhesion of fouling organisms. And the microstructure of a certain size will reduce the adhesion of specific fouling organisms, but for other types of fouling organisms, it may help them multiply.

Figure 1. Three wetting states of droplets on smooth and microstructure surfaces (a. wet smooth surface b. soaked microstructure surface c. hydrophobic microstructure surface)

Through a large number of experiments, Scardino\textsuperscript{12} found that when the feature size of the microstructure was slightly smaller than the size of the fouling organism, the antifouling effect is the best. And the "contact point theory"\textsuperscript{13} is proposed. It means that the number of sufficient contact points between the fouling organism and the attached surface is a prerequisite for the successful adhesion of fouling organisms. The relative size relationship between the surface microstructure and the attached organisms will affect the number of contact points.

Based on the contact point theory, scholars from various countries have proposed several different models for the study of biological attachment mechanisms, which are ERI, ERI II, nano-force gradient
and SEA, etc. The current mechanism research is only for the adhesion of certain organisms, and it is not universal. The antifouling mechanism of surface microstructure is still a hot topic.

Schumacher\textsuperscript{14} studied four different alignment rules of the microstructure surface and tested the antifouling performance. The ERI model was proposed to predict the adhesion of Ulva spores by comparing the experimental results with that of the smooth surface. ERI is a dimensionless ratio based on Wenzel roughness factor, concave surface fraction, and spore mobility. Studies have shown that the adhesion density of Ulva spores decreases with increasing ERI values. However, the ERI model tends to increase with the density of the adhesion of the Ulva spores. And the larger the variance of the model is, the greater the error of the prediction will be.

\begin{equation}
ERI = \frac{r \times df}{f_D}
\end{equation}

Long\textsuperscript{15} replaced D (degree of freedom of spore movement on characteristic groove) f in ERI model with n (number of different characteristic structures on surface microstructure), and established ERI II model.

\begin{equation}
ERI_{II} = \frac{r \times n}{1 - \varphi_S}
\end{equation}

The ERI II model uses the natural logarithm to represent the adhesion density of the green algae spores, which eliminates the influence of the variance in the adhesion density of the green algae spores. What’s more, it can better indicate the adhesion law. According to the model, the number of spore attachments decreases as the area fraction at the top increased or the number of different features in the design increases. The model correctly predicted spore adhesion densities on three previously untested surfaces to smooth surfaces.

Based on the ERI II model, Magin\textsuperscript{16} added the Reynolds number Re and the sensitivity factor m of the organism to the surface, which is used to describe the linear relationships between the adhesion density of marine bacteria and the ERI II value. The effective predicting formula for the adhesion of Ulva spores and marine bacteria is obtained.

Schumacher\textsuperscript{17} uses a force transfer model combined with various features of the microstructure surface to establish a nano-force gradient model (Fig. 2): when the fouling organisms adhere to the surface of the microstructure, the structures of different feature sizes will produce different stresses on the fouling organisms. The fouling organism is forced to leverage energy to adjust its contact area on each structural feature to balance the stress.

Based on the contact point theory, Decker\textsuperscript{18} built the SEA model (Fig. 3) based on two ERI models. By identifying the increasing interfacial free energy, the microstructure surface is divided into many grids of the same size. The model can better predict the adhesion of Ulva spores, diatoms, and other organisms.

The models mentioned above have their own limitations, and there is currently no universal theoretical model. Both ERI models do not consider the adhesion parameters of the substrate material, like the wettability, mechanical properties and the size of the organism. And they are only suitable for predicting the adhesion of the Ulva spores, without general antifouling function. The nano-force gradient model only considers the cuboid. The validity of other complexly shaped microstructures has not been verified. And it requires a single organism to have multiple adhesion points and can only contact the convex vertex of the groove. The nano-force gradient model and the SEA model are contradictory in the influence of the number of adhesion points on the adhesion density.
4. Research on antifouling properties of microstructure surfaces

Barthlot, director of the Institute of Botany at the University of Bonn, Germany, discovered the self-cleaning properties of the lotus leaf when observing the leaves of the plant. Studies have shown that there are micron-sized mastoid structures on the surface of the lotus leaf, and the nanostructures are finely distributed. This secondary micro-nano composite structure imparts super-hydrophobic properties to the surface of the lotus leaf\(^{19,20}\). The skin of a fast-moving shark is distributed with finely spaced scale and ridge to prevent the attachment of micro-organisms such as bacteria. In addition,
other sea lives like crabs, starfish, shells, etc. all have very precise microstructures, and their
drophobic and self-cleaning properties have a great inspiration in marine antifouling research.

Ball\textsuperscript{21} found that the skin of the shark is closely packed with a large number of micron-sized scaly
structures. Studies have shown that the skin can self-clean because the V-shaped and U-shaped
grooves are cross-combined on the shark scales. The research team guided by Brennan modeled the
Sharklet AF with microstructure based on the shark skin. The results show that the micro-structured
material can reduce the adhesion rate of algae, barnacle and other fouling organisms by about 85%\textsuperscript{22}.

Schumacher\textsuperscript{14} conducted a comparative study of the Sharklet AF surface and first proposed the
concept of ERI (engineering roughness index, Fig.4). Four different alignment microstructures with a
space between 2 mm and a height of 3 mm were studied. These surfaces all have reduced sporulation.
The ERI model calculation formula is proposed, and the ERI values of the four structures in the
following figure are calculated respectively. Figure A shows the Sharklet with the AF surface with a
77% reduction in spore settlement.; Figure B shows a new multi-feature topography consisting of a 2
mm diameter cylinder and a 10 mm equilateral triangle and a 58% reduction in spore settlement;
Figure C, D is about the 2 mm diameter cylinder and the 20 um high groove and the sedimentation
reduction by 36% and 31%, respectively.

Carman ET al.\textsuperscript{11} modeled the surface microstructure of shark skin and designed three kinds of
surfaces with cylindrical, ridge and bionic appearance. It was found that the surface with bionic
microstructure can reduce the attachment of ulva by 85%.

Halder ET al.\textsuperscript{23} fabricated micropores with a depth of 5 μm with different distances and diameters
on the surface of PDMS. The adhesion of colibacillus on the surface of PDMS under static and flow
conditions were tested. The characteristics of flow field in the channel were simulated in the
experiments. The surface of the microporous structure of 2 μm in distance and 10 μm in diameter can
significantly reduce bacterial adhesion.

Scardino ET al.\textsuperscript{24} studied the effects of microstructure of shell surface on the adhesion of fouling
organisms. Four different samples of mussels and pearl shells were tested in the same sea area. It was
found that there were fewer microorganisms attached to the surface of the mussels, indicating that the
surface microstructure of the shells was influential to adhesion. At the same time, the rule between surface texture and antifouling performance was studied. It was found that with the decrease of skewness of surface profile, the surface roughness became higher and the surface antifouling performance became weaker.

Zheng ET al. studied the micron-scale protrusions on the surface of the starfish, ranging 100-250 μm. Under the scanning electron microscope, there is a finer structure on the protrusion. The surface microstructure of the starfish was simulated with polydimethylsiloxane (PDMS). The regular hexagon was formed by 19 small cylindrical protrusions, with diameter of 2.5μm, and spacing between of 6μm. Through the test of the laboratory dynamic anti-fouling device, the surface of the PDMS with starfish microstructure has better antifouling performance when compared with the smooth PDMS surface.

The research team of Wuhan University of Technology led by Bai Xiuqin 26-28 duplicated the surface of the three shells of dosinia japonica, chlamys nobilis and gafarium pectinatum. The antifouling test showed that the number of diatoms attached to the surface of the simulated microstructure was significantly reduced. The effect of the rib spacing was studied by modelling the microstructure of the dosinia surface to the ribbed surface with adjacent rib spacing w. The results showed that the rib spacing w has a non-monotonic effect on the reduction performance. Wang et al. studied the arrangement of surface microstructures of different kinds of shellfish, such as radiation and concentric circles: the surface of the mussel is distributed with many protrusions with a diameter of 10 to 25 μm; the surface of the dosinia has a ribbed structure; the surface of the perna viridis is arranged with a groove structure.

5. Study on surface adhesion of several major fouling biological microstructures

Different biofouling organisms have different adhering characteristics, and scholars from various countries have also conducted targeted researches on several common fouling organisms.

5.1. Attachment of ulva spores

Ulva spores are about 5 μm in diameter, and the spores with a feature size of 5 μm have the most indirect contacts and the strongest adhesion. This characteristic is in accordance with the contact point theory. When the microstructure feature size is greater than 5 μm, the spore will increase the contact point with the surface by "tilting"; when the microstructure feature size is too large, the spores will adhere together to the surface; when the microstructure feature size is less than 5 μm, it can effectively decrease spore adhesion.

Callow found that the adhesive of the ulva spores has a hydrophilic nature, making it less adherent on the hydrophobic structure. Decker found that different landforms have a certain effect on the location of individual zoospores and the screening distance of zoospores, which may contribute to antifouling. Schumacher found that the ulva spores were preferred to adhere to the bottom of the groove of the feature structure, rather than the sidewall or protrusion of it, and the larger the aspect ratio of the microstructure, the stronger the ability of the host to inhibit the adhesion of the ulva. Rajab35 believed that most of the spores were attached to the grooves due to the large contact area of the grooves and the gravity of the bacteria itself. At the same time, the bacteria in the grooves are protected by the side walls from the washing of the water. Hoipkemeier-Wilson et al. studied the effects of microstructures of 5 μm height and 5 μm width with intervals of 5, 10 and 20 μm on the adhesion and shedding of Ulva spores. The experiment found that the microstructure with width and depth of 5 μm significantly reduced the spore adhesion density, and the intervals over 5 μm were more likely to adhere to the spores.

5.2. Diatom adhesion

Marine diatoms are diverse in variety and have different attachment features. There are great differences in the size of different diatoms (2 μm for Nitzschia; 4 μm for Navicula; 7 μm for Amphora.). Microstructure surface has a single antifouling ability and can only target one or several
diatoms. Unlike the adhesion of the ulva spores, the diatom will crawl along the wall after the base is initially attached, thus achieving deep attachment. Studies have shown that diatoms can climb to the beach during the day and climb back into the bottom at night. In an aquatic environment, diatoms have the ability to float upward and migrate vertically. Ákess committed to the study of the adhesion of some diatoms have a lower adhesion density on the nano-peak structure than on the nano-convex structure. The reason may be the adhesives secreted by diatoms are hydrophobic, which are less likely to adhere to the surface of the hydrophilic structure.

5.3. Barnacle adhesion
Studies in the 1960s abroad have shown that the initial adhesion of barnacles relies on the mechanical attachment of the Venus larvae tentacle sucker, which can withstand a water flow impact of 3.6 knots (close to 2 meters per second). Petronis compared V-grooves and bulges of the same structural height (69 μm) and size (97 μm) and found that the grooves were more effective than bulges. Bers found that the same kind of sinusoidal micro-scale groove with the relatively smaller surface roughness is better in the antifouling effect.

Xiao et al. designed and manufactured different sized honeycomb microstructures like cone and spike to investigate the effects of surface topography on zoospore adhesion of marine microbes such as Dendrobium and diatom.

Aldred observed that Venus larvae preferred to adhere to smaller sized microstructures because the adhesion of Venus larvae (the tip of the small antenna) is about 20-30 μm.

6. Conclusion
Microstructure antifouling technology still needs to be improved with further research and discussion. First, the research on the mechanism of microstructure antifouling remains to be confirmed. Although the "attachment point theory" has high recognition, it has not been verified by a large number of experiments. Second, there are many kinds of fouling organisms, but a single type of microstructure can only be effective for some fouling organisms. The surface with composite structure becomes the development trend of microstructure research. Third, the current preparation method cannot meet the requirements of large-scale processing technology, and the preparation conditions require much. So the cost-effective surface processing of microstructures has gained momentum.

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