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Evaluation of Anti-biofouling Progresses in Marine Application

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Abstract

Biofouling is detrimental and has been a major concern in the marine industry for several decades. This phenomenon is the accumulation, colonization and attack of organisms—which are both micro and macro, to assemblies, parts and/or structures that are submerged in freshwater and other marine environments. Even despite all the exceptional indispensable and indisputable characteristics of alloys such as steel, biofouling continues to be a major source of failures of these alloys, thereby limiting their use in service. This study presents a review of the existing means of protection against biofouling which are basically the use of paints and electrolytic deposition of anti-biofouling agents such as some nano-composite coatings. The different types of systems from the first-generational coatings such as tributyltin self-polishing copolymer paints to the novel nano-composite coatings were discussed. Ultimately, the use of nano-materials and composites consisting some anti-biofouling natural products has identified to be a promising way of combating biofouling issues in the maritime.

Keywords Biofouling · Corrosion · Electrodeposition · Marine application · Coatings · Nano-composite

1 Introduction

Globally, biofouling is an unfavourable condition being faced in the marine environment. This issue negatively affects the erected underwater structures as well as mobile watercrafts such as ships [1]. Specifically, biofouling causes severe problems like corrosion, distortion and alteration of the surfaces of structures submerged in the water, increased weight, increased drag leading to the reduction in watercrafts' speed and up to 40% fuel additional consumption [2]. The incremental fuel consumption leads to an aggravation of CO_2 emission and more water transportation cost [3]. Therefore, biofouling has posed as a huge economic stress factor.

Biofouling is a phenomenon whereby microorganisms and macroorganisms rapidly colonize the natural and synthetic structures which are submerged in an aquatic environment [4]. There are two major groups of several fouling organisms based on their size, these groups are the microorganisms and macroorganisms [4]. Microorganisms can also be regarded as slimes, biofilms or microfoulings [5].

Fouling organisms go through five major stages of progression, which are: adsorption, immobilization, consolidation, microfouling, and macrofouling. Microorganisms in the marine environment are fond of accumulating and attaching on surfaces that are not protected. They then form biofilms on the surfaces; these biofilms are quite difficult to be removed from the attached surfaces even when a high shear flow is applied [6]. Biofouling happens sequentially from initialization whereby microbes attach to a water body's surface by generating some substances which are extracellular polymeric in nature. This consequentially promotes the growth of a biofilm matrix. These growths are dependent on an interaction between the cells of the bacterial, attachment of the surface, and the content of the medium. An illustration of parts attacked with biofouling is shown in Fig. 1.

Biofouling in maritime causes environmental disasters and billions of dollars are being consumed annually in the shipping industry. Drag and surface smoothness decreases when organisms attach to the hulls of the vessel; this results to an increase in the hydrodynamic weight and subsequently lead to a reduction in top speed and loss of manoeuvrability [7]. Therefore, biofouling causes fuel consumption and stimulates harmful compound emission. Examples of such toxic compounds are SO_x and NO_x, which is the source of

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Fig. 1 Effect of biofouling on some parts of a vessel

soil damage acid rains and soil damage. Other noxious substances, such as atmospheric pollutants and CO_2, are also released to the environment, which poses serious environmental problems [8–10]. Almost 60,000 people die and about €200 billion is lost annually as a result of the toxic emissions from international marine navigation [11]. Such incremental fuel consumption which is activated by fouling can shorten the dry-docking intervals [12]. Thus, efforts of preventing biofouling prevention are highly beneficial both economically and to the environment [1].

This research aims to review the existing ways of preventing biofouling through the use of anti-biofouling coatings. This will in turn help researchers in developing a simple, non-toxic and economical way of developing material surfaces which are non-wetting, durable and with mechanical properties which are not compromised, also the surfaces are expected to repel a range of contaminants and have instant significance in applications. The scope of this work is limited to chemical and electrolytic methods used to protect against biofouling.

2 Review on Anti-biofouling Paints

2.1 First Generation of Anti-biofouling Coating

The first attempts to use an anti-biofouling system was around the seventh century BC [5]. The Carthaginians, Phoenicians, Romans and Greeks, which were ancient Mediterranean civilizations gave the written reports on the first attempts to protect structures from biofouling [13]. In seventh century BC, a timber which is lead-sheathed was originally used in Phoenician galleys. Around the fifth century BC, coatings made of a mixture of arsenic and sulphur-and-oil were used to prevent fouling and in the third century AD, the Romans and Greeks secreted lead sheathing with copper nails [14]. Then, in the thirteenth to fifteenth centuries, pitch mixed with oils or tallow was extensively used for biofouling prevention. In the sixteenth century, ships were enclosed with woods placed above film of animals' tar which is coated [5]. Although lead casings were the anti-fouling system which is most commonly used, their utilization aggravated the rusting of steel materials which led to the ban of lead sheathings [4].

2.2 Paints from Copper Compounds

In 1860, copper compounds which are embedded in rosin was developed using hot-plastic paint. This paint was utilized in combating biofouling. Usually, a layer of anti-corrosive substances is applied on the substrate to protect it from corrosion and this copper compound paint can then be applied as an additional layer afterwards. However, the copper compound coatings are too expensive, their performances are relatively inefficient, and they are not durable [5].

2.3 Mercury-Based Coating

Early in the twentieth century, there was a development of spirit varnish paint. After this, the Americans developed the ships' bottom part paint in 1908. Most of these paint advances were based on the dispersion of mercury oxide in graded turpentine and grain alcohol. These paints, protected their substrates from fouling for their lifetime which was about 9 months [15]. An investigation by the US Navy in the early twentieth century confirmed that rosin exhibited some desirable properties such as its availability, affordability, and effective replacement for the costly and rare shellac. Therefore, rosin was mixed with toxic compositions of mercury or copper so as to enhance their performance; an example is the case of rosin combined with resins derived artificially, which improved their mechanical characteristics but has led to apprehensions on the health and safety [4].

2.4 Paints with Tributyltin (TBT)

In the 1990s, the marine industry started to use paints from the exacerbated poisoning of organotin compounds [6]. These paints actively released a broad-spectrum biocide, TBT, which presented outstanding anti-fouling properties [5] by killing a large amount of the prospective fouling organisms [1]. The TBT self-polishing coatings (TBT–SPC) composed of TBT acrylate esters as anti-biofouling coatings were developed in 1958. Regrettably, the TBT-bearing biocides' rate of degradation was slow in the sea water column; as a result of this, the toxic lipophilic compounds then became readily bio-available to aquatic species which are not the targets [15]. Later, the long-term release of toxic TBT was realised to cause severe destruction to aquatic life [16]. At significantly low concentrations, TBT causes some sub-lethal effects, such as the disruption of the endocrine which leads to sexual disorders [17]. In addition, the International Maritime Organization (IMO) gave a report that TBT accrues in mammals and weakens the immunological defences in sea lives such as fishes [5]. These aquatic lives provide above 70% of global oxygen production, so a harm to them is a huge risk to humanity. Thus, due to the toxicity, the TBT was banned globally in 2008 by the international convention [1].

2.5 Coating with Tin-Free Self-Polishing Anti-fouling Materials

It is acrylic based and has an extent of anti-fouling characteristics. The toxicity of improved paint is far lesser, and it could meet the environmental protection requirements and anti-fouling properties. It, therefore, became the conventional products in the anti-fouling paint for a number of years [18]. The coating contains copper and some other metals that have the tendency leading to "the black pollution" and tend to be harmful to the marine environment, thus, a restriction was placed on it by the European Union.

2.6 Coatings with Materials That Have Low Surface Energy Anti-biofouling Abilities

The very low surface-energy makes it challenging for any attachment of marine fouling organisms. They are of either silicone based or fluoride based [19]. Marine lives are unable to attach on silicon coating because surface tension is low. Silicone does not have the ability to stop marine biofouling, however, it is able to reduce how fouler will adhere to the material. Although silicone's surface energy is quite low, its adhesion on the substrate material is very weak [18]. On the other hand, Fluorocarbon resin has strong resistance to water and oil, weather, stain, chemical, solvent and many more, it, therefore, seemed to be a kind of excellent material for anti-biofouling coating [20]. Fluoride has excellent resistance to chemicals and durability but it is too expensive. Fluorine paint has demonstrated good performance in the low surface energy anti-fouling coatings, however, it obtained widespread concerns especially as fluoride-based chemicals do not easily dissolve in common organic solvent.

2.7 Marine Creatures Anti-fouling Paints

Marine animals such as dolphin, seaweed, whale, shellfish and others have the natural anti-fouling ability because marine organisms mostly do not attack them despite living with them. It was found by some researchers that coral and other marine animals can produce anti-fouling properties on its own. The extracts from coral extracts were studied, four kinds of steroids compounds with open loop were gotten and confirmed to be able to effectively prevent fouling [21]. Although some achievements were made with these extracts, they are too expensive, only effective within a shortterm, also the anti-fouling effect is not an ideal one and was unable to meet the prerequisite.

2.8 Silicate Anti-biofouling Paints

Silicate anti-fouling paint is a kind of soluble alkali silicates anti-fouling agent, it tends to form a strong basic region between the sea areas of the paint in contact and the surface of the hull. Normally, fouling organisms survive suitably in an environment with pH 7.5–8.0 in slightly basic marine water, and it is not fit for them in an acidic environment. Silicate possibly will become the anti-fouling agent (examples of such are zeolites, crystallization alumina silicate with water), and its molecular sieve or ion exchange effect tend to form the anti-fouling mechanism. In the course of the procedure, ion exchange occurs in-between H^+ and silicate coming from the marine. Then, an anti-biofouling agent is then released and anti-biofouling function can be achieved. This method characterised as being economical and effective, but can affect the climate conditions and the aquatic environment adversely [22].

2.9 Capsaicin Anti-fouling Paints

Capsaicin can be gotten from pepper fruit. Capsaicin is the key substance of chilli's spicy flavour and it is the alkaloid which is quite stable. In addition, it has antibacterial properties and it tends to prevent marine organisms from growing. Until now, local and international researchers have done a plethora of researches for the natural anti-fouling agent which are spicy [23]. Anti-biofouling paint containing capsaicin attains the purpose of anti-biofouling by driving effect. Its effective components could be gradually and gently released through the release control technology and long-term and the non-toxic anti-fouling effect will be achieved. Fischer used pepper derivative for the anti-biofouling agents, such as oleoresin capsaicin and red capsaicin. The derivative was added to an ordinary paint which is waterproof so as to synthesize the anti-biofouling paint that can inhibit marine organisms biofouling. Three acrylamide compounds were then derived from capsaicin and all the three compounds were found to possess antibacterial activities which are noteworthy. The compounds are 3-hydroxy-4-methoxybenzylamine (HMOBA), butylmethacrylate (BMA) and *N*-[4-hydroxy-2-methyl-5-(methylthio)benzyl]benzamide (HMMBA), with their formulas presented in Fig. 2. These

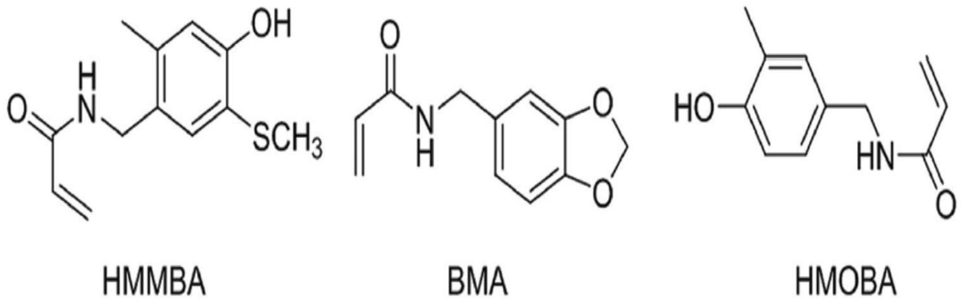

Fig. 2 Formulas for the three acrylamide compounds [24]

compounds were also studied as an anti-biofouling agent in the anti-biofouling paint, and test panels exposed in seawater for 186 days was almost free of organisms from the marine. The result thus showed that it could prevent the adhesion of marine organisms [24]. However, this means of protection have a harmful effect risk on other non-target marine animals due to its hotness.

2.10 Nano-Anti-fouling Paints

The introduction of nanoparticles has the ability to improve and enhance the rheology, adhesion of the coating, the hardness of the coating, smoothness and patients' anti-ageing properties. Therefore, it is a vital course to be considered in anti-biofouling agent development. An effective anti-biofouling nanotechnology is the microencapsulation technology. This technique uses a type resin material which is water-soluble in making the anti-biofouling agent, in nanopowder form, to produce the particulates and then formulate them in the paint. In micro-encapsulation technology, droplets or tiny particles are enclosed by a coating so as to produce small capsules of a lot of properties which are useful. Generally, this is used in the incorporation of enzymes, ingredients for food, cells or some other materials which are on a micrometric scale. It is also possible to use microencapsulation in the enclosure of gases, liquids and solids inside a micrometric wall which is made of either soft or high-soluble films, to lessen the seawater contact and achieve a reduction in the rate of release to achieve the stability of the anti-biofouling effect. The anti-biofouling agent which is micro-encapsulated can obtain a more lasting and much better anti-biofouling effect, also the environmental impact can be reduced. Practically, it is possible to steadily dissolve the microcapsules by sea water, and the anti-biofouling agent is gradually and effectively released to achieve an anti-biofouling effect with high stability. The microencapsulated anti-biofouling agent is more durable in that its anti-biofouling efficiency period is longer, it also has a good anti-biofouling effect and the ability to reduce the impact on the environmental. The size of the polymer particles decreases whereas the reaction temperature reduces or there was a rise in the amount of dispersing agent and stirring speed [25].

3 Review on Electrolytic Deposited Coatings

In addition to modifying a surface with a coating chemically (chemical paints), the characteristics of material surface can also be altered physically for example, through electrodeposition or by creating micropatterns. This is stimulated by the marine creatures such as whales and sharks. Thus, micropatterns can be produced to imitate marine creatures' skin's surface topography [26]. It is realistic to anticipate that the surface enhancement technology combined with biofouling-resist or biofouling-release coating materials will further enhance and improve the anti-biofouling function. The electrolytic deposition has successfully enhanced the physical surface properties of different materials for corrosion protection [27–31]. Electroplated zinc coatings, for instance, is one of the major ways of protecting steels from corrosion [32]. The potentials of the electrolytic coating have been leveraged by a number of researchers for biofouling prevention.

3.1 Electrodeposition of Nanoporous Tungstite Films

Metallic surfaces are exceptionally durable and some tend to be resistant to biofouling. Surfaces based on electrodeposition of nanoporous tungsten oxide (TO) films on steel substrate were developed [33]. The TO-modified steels are comparably mechanically durable with bare steel, they are also extremely tolerating to tensile and compressive stresses as a result of chemical bonding to the steel and their morphology which is island-like. Although the coatings were impinged with zirconia particles stabilized with yttria, and sometimes exposed to ultraviolet (UV) light and extremely high/low temperatures, they were non-wetting as the TO coatings which are inherently superhydrophilic were converted to superhydrophobic. These surfaces exhibited omniphobicity properties when lubricated and thus retained hidden mechanical durability. This coating was confirmed to considerably reduce marine *Escherichia coli* attachment and algal film adhesion [33]. However, the coating is not effective as expected to biofouling because after 8 days, biofilm firmly attached to the surface coated leaving about 88 ± 4% fouled surfaces.

3.2 Electrodeposition of Nickel Matrix–Silver Nanoparticle Composite Coatings

Nickel matrix–silver nanoparticles were produced and characterized for anti-biofouling and anti-corrosive applications in the aquatic environment. Electrodeposition of two Ni–Ag composite coatings of compositions, Ni–0.25 at.% Ag and Ni–0.75 at.% Ag was done on copper. The highlights of the effects of silver nanoparticles on the biofouling, Ni passivation and electrochemical corrosion of these coatings made of the composite were done. In the course of anodic polarisation in about 3.5% sodium chloride, silver nanoparticles which are embedded into a matrix of nickel-induced galvanic passivation, which enriched the possibility of the use of coatings of composites for passivation. When these composite coatings were exposed to sulphate-reducing bacteria, they showed a superior resistance to biofouling by inhibiting the formation of biofilms. The level of bio-film reduced with the quantity of silver nanoparticles incorporated in the composite coatings. Bio-film led to a reduction in corrosion resistance of the coatings. Thus, the anti-microbial nanoparticles when embedded in a suitable metal matrix can protect substrates from biofouling [34]. However, the use of silver in the huge structures in the marine environment is quite expensive.

3.3 Electrodeposition with Lanosterol

The anti-fouling ability of lanosterol-coated PVC sheets was studied. The sheets were immersed in natural seawater for almost 1 year. Remarkably, up to 1 year of exposure, there was no substantial fouling organisms or growth on the coupons coated with lanosterol [35]. Consequently, the bactericidal chemical compounds can serve as alternatives for anti-biofouling compounds because they are friendly to the environment [35, 36]. Lanosterol displayed anti-biofilm activity and anti-quorum sensing activity against the leading micro-fouling bacterial groups and their receptor proteins such as *N*-acyl-homoserine lactone and oligopeptide. The chemical compounds which was produced by the biofilm bacteria interrupted the biofilm formation [37, 38]. The original biofilm forming bacterial colonies in the marine environment has the ability to produce allelopathic substances that tend to act in contrast to the neighbouring cell [39]. Results from the research showed that early stage micro-fouling bacterial colonies produced an allelopathic substance which look like bacteriocin or antimicrobials for biofouling control. The research recommended the screening of natural products as a promising approach to discover innovative biofilm inhibitors or anti-fouling compounds.

3.4 Electrodeposition of Ag–PPy Composite Coating

A conductive PPy/resin coating which has worthy electrochemical anti-fouling effects was developed in the cathodic polarization anti-fouling application. A synthetic conductive Ag–PPy/resin composite was done. The anti-fouling effect under significantly less cathodic voltage was improved, leading to an increase in its anti-fouling effectiveness. However, its conductivity was negatively affected and this needs to be improved so as to save electrical energy [40].

3.5 Coating with Silver–Titania Nanotube Composites

Silver is a famous antimicrobial agent, it is eco-friendly and can be tolerated well by mammals. Titania nanotubes have improved properties because of a superior specific surface area on the tubular structure's inner and outer surfaces. An innovative 2-step hydrothermal synthesis of a silver–titania nanotube (Ag/TNT) composite material was produced [41]. The inhibitory properties of the biofilm produced was studied. This showed that the Ag/TNT which has the lowest silver content (0.95 wt% Ag) and is decorated with Ag nanoparticles has the lowest formation of the biofilm known as bacterium *Halomonas pacifica*. In addition, an inhibition of marine microalgae (*Isochrysis* sp. and *Dunaliella tertiolecta*) growths were detected [41].

3.6 Electrodeposition to Produce Manganese Stearate Superhydrophobic Surface

Manganese stearate superhydrophobic (SHPB) surface was produced using a versatile one-step method. A *Chlorella vulgaris*-inoculated culture medium immersion test was done which recommends that the SHPB surface prepared can efficiently suppress algae-induced biofouling. The synthesized SHPB surface demonstrates pronounced enriched corrosion resistance and outstanding mitigation against biological adhesion [42]. Despite these favourable outcomes, the SHPB surfaces maintain superhydrophobicity in harsh environments ranging from strongly acidic to alkaline environments [42, 43].

4 Conclusion

Biofouling and corrosion induced by microbes significantly lessen the robustness and usefulness of materials such as steel components [44–46]. Most major disasters in the naval industry are usually a result of biofouling and this aggravates fuel drain, resistance to drag, costs of maintenance, and the negative environmental impacts [47–51]. Thus, the predominant use of biocidal

Table 1 Some key anti-biofouling painting systems [61]

Anti-fouling system	Life time	Rate of erosion	Cost (USD/m^2)	Limitations
(TBT) Self-polishing copolymer paints	4–5 years	< 3 µm/month	$680.88	Banned 2008
(Tin-free) Self-polishing	5 years	N/A	$1,382,670. 00	Lifespan is shorter than TBT-based paint systems; thus increasing the overall maintenance cost of the ship
Conventional paint (tin free)	12–18 months	N/A	N/A	A non-polishing ability which is hard and leads to build-up on the coatings. Performance is only appropriate for environments which are very low in fouling
Control depletion Polymers (copper paint)	3 years	Matrix erodes because of coating binder's dissolution	$1,357,786. 00	The release of biocides varies, self-smoothing is poor, expensive because of the need to recoat Drying time is slow
Foul release	2–5 years	N/A	N/A	In-water cleaning problematic as silicone can be damaged by the brushes, foul release coatings has abrasion damage tendencies

anti-biofouling coatings has led to several environmental consequences [1] and this has given rise to strict legislation in the shipping industry [52, 53]. This has led to the use of an eco-friendly means of protection against biofouling through coating which is either painted or deposited through techniques such as electrolytic deposition [54–59].

On one hand, coating with chemicals such as nano-antifouling paints has proven to successfully mitigate biofouling on substrates over some time [25, 60]. However, the durability of these paints still remains a challenge as re-painting will often be required which makes this solution quite expensive and not sustainable in the long-run. Some key anti-biofouling painting systems are summarised in Table 1.

On the other hand, electrolytic deposition of nano-composite coating seems to be a more sustainable solution especially if the composite has natural products as part of its constituents. The use of “nanomaterials” is very effective in inhibiting the adhesion of bacteria and formation of biofilm because they possess effective antimicrobial properties and due to the fact that their specific surface area is large, this specific surface area is inversely proportional to the nanomaterials' particle size [62, 63]. In addition, some natural products exhibit a broad spectrum of anti-fouling actions against algal spore adherence, microbes, barnacles and mussel which can form marine biofilm to substratum which is synthesized [64, 65]; making them good constituents of the anti-biofouling composite coating.

Currently, little work has been done using electrolytically deposited nanocomposites from anti-biofouling natural products in biofouling protection. This will be a novel and promising way of combating biofouling issues in the marine application.

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Compliance with Ethical Standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

Ethical Approval This review work has no ethical issues.

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