The Future Trends of Sustainable Antibiofouling Research and the Development of a Naturally Derived Synergistic Antibiofouling Solution

by

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Dalhousie University is located in Mi’kma’ki, the
ancestral land and unceded territory of the Mi’kmaq.
We are all treaty people

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Abstract

Biofouling, the accumulation of organisms on submerged surfaces, presents a significant challenge to maritime industries, including navies, shipping/transport, aquaculture, and fisheries. The resulting drag on ship hulls increases fuel consumption and carbon emissions, contributing to climate change. Historically, various antibiofouling solutions have been developed, with many relying on toxic compounds such as copper and tributyltin (TBT). These substances, while effective, have raised environmental concerns, creating an urgent need for novel solutions that are both effective and environmentally benign. This study aimed to develop a sustainable, non-toxic, and cost-effective antibiofouling solution using three naturally derived chemicals: chitosan, capsaicin, and polydimethylsiloxane (PDMS). Three solutions were produced with varying concentrations of chitosan (50:1, 100:1, and 200:1) while maintaining constant levels of capsaicin and PDMS. The antibiofouling properties of these solutions were evaluated using a bacterial assay and a proteinaceous analogue test, comparing their efficacy against a control and among each other. The results indicated that these novel combinations were not effective antibiofouling agents. This outcome underscores the complexity of developing solutions that are both efficient and environmentally friendly, highlighting the need for further research into alternative substances and strategies.

List of Abbreviations and Symbols Used

CPVC - Critical pigment volume concentration

DLS - Dynamic light scattering

GHG - Greenhouse gas

IMO - International maritime organization

LCA - Life cycle analysis

PDMS - Poly(dimethylsiloxane)

ROS - Reactive oxygen species

SPC - Self-polishing copolymer

STPP- Sodium triphosphate

TBT – Tributyltin

UV - Ultraviolet

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Chapter 1: Introduction

1.0. Overview

Floating on bodies of water has been a primary means of transport for millennia. Throughout history and across civilizations, many different vessels have been used, but one persistent problem remains: biofouling. Biofouling is the attachment and accumulation of marine organisms on submerged surfaces, in both marine and freshwater environments (Qian et al., 2022). This issue affects various marine industries, such as navies, shipping, transport, and fisheries, because the attachment of these organisms creates drag on ship hulls, leading to increased fuel usage, higher transport costs, and greater greenhouse gas (GHG) emissions, which contribute to climate change (Dobretsov et al., 2019). Biofouling is also a significant problem for aquaculture, as it clogs pumps and pipes, and weighs down nets and pens leading to poor water quality and lower stock value (Garibay-Valdez et al., 2022).

Due to these persistent issues caused by biofouling across marine industries, many antibiofouling solutions have been developed throughout history (Yerba et al., 2004). An antibiofouling solution can prevent biofouling through chemical deterrents, which alter the surface chemistry to create a toxic environment (Ralston & Swain, 2009), and/or physical deterrents that change the surface structure to prevent the attachment of organisms (Abioye et al., 2019). Traditionally, biofouling was prevented using metal-based paints, such as tributyltin (TBT), copper, and mercury. However, these paints eventually break down and settle into marine sediments. TBT was one of the most popular antibiofouling solutions worldwide since the 1950s due to its low cost and availability until it was banned for being toxic to non-target species, such as mussels, periwinkles, whelks, and oysters, causing endocrine disruption and reproductive deformities (Bauer et al., 1995; Beyer et al., 2022; Carrier et al., 2023). Past and current market solutions have contributed to marine pollution, including microplastics and biocides, and the deterioration of marine ecosystems and their wildlife (Flemming et al., 1996).

Modern and experimental antibiofouling solutions also utilize chemical deterrents, such as biocides, self-polishing compounds, zwitterions, and photocatalysts, as well as physical deterrents, such as hydrophobic, hydrophilic, amphiphilic, electrochemical, and altered surface topography approaches (Amara et al., 2018; Amir et al., 2021; Pourhashem et al., 2022). However, these solutions have now been making the shift toward a focus on sustainability (Almeida et al., 2007). Sustainability meaning that materials are renewably sourced and have the potential for environmentally friendly production that can be sustained over a long period of time. Many proposed antibiofouling solutions vary in toxicity, sustainability, and cost, making them sub-optimal for commercialization and global use. Therefore, a novel, sustainable, environmentally benign, and cost-effective antibiofouling solution is needed.

1.1. Biofouling

Common biofouling organisms are bacteria, algae and larvae of many marine invertebrates, such as barnacles, lobsters and shrimp (Bixler & Bhushan., 2012). The attachment of these organisms happens in a four-stage process, which biochemically allows for the subsequent attachment of smaller to larger organisms (Dobretsov et al., 2006). The first step of this process is the absorption of dissolved organic molecules on the submerged surface; second is the colonization of bacteria on the surface; third is the colonization of microscopic eukaryotes; fourth and finally is the colonization of invertebrate larvae and algal spores (Dobretsov et al., 2006). The attachment of these organisms creates a change in the structure of the submerged surface, which can be costly for marine industries. The increased drag on ship hulls causes an overall increase in carbon emissions as more fuel is needed to move the vessel (Hadžić et al., 2022). Biofouling also increases the costs associated with aquaculture due to the need for more management during production, as nets, cages, pumps, and tubing need preventative measures in place and constant maintenance (Bannister et al., 2019). Due to these environmental and economic impacts, biofouling needs to be prevented, but historically, solutions to these issues have caused their own problems.

1.1. History of Antibiofouling Solutions

The first antibiofouling solutions were created in the 17th century BC by introducing lead and arsenic-based coatings (Abioye et al., 2019). These solutions were quickly phased out within the following centuries due to their incompatibility with steel and the cost to sustain their usage (Almeida et al., 2007). Copper-based paints were also introduced at this time but have still been commercially used in in modern times even with adverse effects to the marine environments (Gomes et al., 2020). At the start of the 20th century, mercury-based paints were introduced , which were effective and had a longer lifetime of nine months compared to the first copper-based solutions but were phased out because of their overt adverse effects on the health and safety of humans (Abioye et al., 2019). TBT was introduced in the 1960's and quickly became the number one antibiofouling solution for the next four decades until its ban in 2008 due to its toxic biocidal impacts on non-target species (Beyer et al., 2022). Since this ban, there has been a demand for the development of sustainable and environmentally friendly antibiofouling solutions (Maréchal & Hellio., 2009). Many different environmentally friendly antibiofouling solutions have been studied, such as tin-free self-polishing solutions, low-surface energy solutions, silicate solutions, nanoparticle solutions and electrolytically deposited solutions (Jin et al., 2022). These solutions are only effective against biofouling in the short term due to biocidal leaching, a lack of pungency, and/or a sensitivity to abrasion, making their application effectively unsustainable for marine industry application (Jin et al., 2022). Solutions such as low-surface energy and nanoparticle solutions have the potential for commercialization as they can be low-cost, environmentally friendly, and sustainable. However, these solutions need a further cradle-to-grave study to understand their full potential (Amir et al., 2021).

1.3. Sustainable Antibiofouling Solutions

Currently, many different researchers and entrepreneurs are developing sustainable solutions for biofouling. This research aligns with the goals of the UN SDG 14 by focusing on the reduction of marine pollution and more sustainable management, protection and conservation of marine and coastal ecosystems (Government of Canada, 2024). Currently, the main focus of sustainable antibiofouling solutions is degradable synthetic/organic compound-based biocides, photocatalytic surfaces and biocide-free physical deterrents (Carrier et al., 2023). Organic/natural compound-based biocides are environmentally friendly compounds derived from nature's naturally occurring antibiofouling solutions (e.g., chitin, capsaicin, and melanin) (Jin et al., 2022). Many of these are effective at preventing biofouling but come with challenges, such as long-term effectiveness (Carrier et al., 2023). Natural compound-based biocide solutions are typically imbedded in a matrix of other compounds that secure them to the surface and are responsible for their slow release, however, due to this intrinsic property, there is a time limit on their effectiveness (Amir et al., 2021). Photocatalytic antibiofouling solutions prevent biofouling using the power of the sun and its radiation (i.e., ultraviolet and infrared) by creating free-radical oxidants which eliminate bacteria, stopping the development of a biofilm and the subsequent stages of biofouling (Carrier et al., 2023). The major challenge with photocatalysts as antibiofouling solutions is their need for sunlight, making them less potent underwater, especially in deep areas where light cannot penetrate (Wang et al., 2022). Biocide-free physical deterrent antibiofouling solutions change the surface structure of the material they are applied to, sometimes mimicking the textures of marine mammals and sharks (Abioye et al., 2019). They are effective at preventing the attachment of microorganisms even in waters with strong currents, which the previous solutions lack: however, abrasion is an issue for these solutions as if the texture is worn off, biofouling will occur (Amir et al., 2021). The robustness of an anti-biofouling solution relies on the active ingredient against biofouling, but more importantly, the non-active constituents allow for long-term strength and effectiveness.

1.4. Components of Paint

To make an antibiofouling solution, it requires more than just its active ingredients, such as compounds that account for adhesion, strength and durability. These include pigments, solvents/thinners, binders and additives, which are often toxic to the environment (Talbert, 2008). The base pigment is used for coloration and helps protect the material intended to cover. For example, lead-based paint helps protect wood but not steel. Solvents and thinners are used to achieve the desired viscosity for application, depending on the type of paint, either water or oil-based compounds are used (e.g., turpentine and xylene). Binders help evenly spread and adhere the paint across the surface. Binders can be an oil, resin, bitumen or cellulose derivative (Ravikumar et al., 2012). Other components of paint include driers and colouring pigments, which help accelerate the drying process and provide additional coloration (Talbert, 2008). Antibiofouling paints have additional components for antibiofouling, anti-corrosion, self-healing and UV protection.

1.5. Non-toxic Alternate Components of Paint

For an antibiofouling solution to be sustainable and non-toxic to the marine environment, all the ingredients must follow suit. Typically, some components that makeup paints are innately toxic but are used due to their efficiency and cost-effectiveness (Talbert, 2008). Non-toxic alternatives exist which can replace these ingredients. The base pigment is used for protection, corrosion prevention and colouring (Talbert, 2008). One prominent non-toxic substance that is used for this purpose is henna, which chemically protects metals by donating the electron from the phenol group of the lawsone (2-hydroxy-1,4-naphthoquinone) component to the metal, which stabilizes it and prevents corrosion (Ong et al., 2021). Another corrosion inhibitor, capsaicin, is an extract of the *Capsicum* sp. Chili peppers which creates a protective layer via the protonated coating being absorbed by the negatively charged metal (i.e., mild steel), which prevents corrosion (Ong et al., 2021). Solvents and thinners are usually alcohols, esters or hydrocarbons, depending on the binder used, as they must be compatible and mixed to adjust the viscosity of the paint. However, it is common for solvents to be volatile substances (Talbert, 2008). Potential non-toxic solvent substitutes are water, 3-methoxy-

3-methyl-butanol, and propylene carbonate (Moity et al., 2012). Binders are used for adhering paints to surfaces and can commonly be non-toxic and environmentally friendly as they are derived mainly from natural sources. Environmentally friendly binders include linseed oil, safflower oil, gum arabic, kaolin, beeswax, and dextrin (Talbert, 2008). Using the preceding ingredients as alternatives to traditional toxic ones will allow for an antibiofouling solution that is non-toxic and neutral to marine environments. This can only be done in combination with active ingredients that are also non-toxic, but for the antibiofouling solution to be commercialized on a large scale the ingredients must also be sustainable using renewable materials that are cost-effective and long-lasting.

1.6. Chitosan Nanoparticles: Active Antibiofouling Ingredient

Chitosan nanoparticles are derived from chitin, the main material comprising invertebrate exoskeletons (e.g., lobster shells, crab shells, mussel shells). Chitin is the second most abundant polysaccharide, after cellulose (Zheng et al., 2011). Due to its large abundance, chitinous materials are often discarded as waste materials (e.g., overboard scallop shells after shucking, composting of lobster/crab/shrimp shells), leading to chitin being a low-cost abundant material (Perez et al., 2020). The characteristic of chitin being a waste material will help support a blue circular economy as invertebrate fishing industries can use there waste products as an antibiofouling solution (Lv et al., 2023). Chitosan has broad natural antibacterial, anti-fungal and anti-algal abilities, which help prevent the first successional stage of biofouling (Kumar et al., 2021). It is non-toxic, hydrophilic and has strong film-forming abilities under acidic conditions (optimally pH of 4) with negatively charged surfaces (e.g., steel) due to its positively charged amine group (Crini., 2022). Chitosan nanoparticles are ideal for an antibiofouling solution when combined with a hydrophobic antibiofouling counterpart, as amphiphilic solutions are superior to individual hydrophobic and hydrophilic antibiofouling solutions (Galli & Martinelli., 2017). This is due to amphiphilic substances having the properties of both hydrophobic and hydrophilic materials, creating a mosaic of chemical prevention (Galli & Martinelli., 2017). The hydrophobic component creates a physical barrier to microorganisms with hydrophilic membranes, and the hydrophilic part creates a physical barrier repelling microorganisms with hydrophobic membranes (Galli & Martinelli., 2017).

1.7. Capsaicin: Active Antibiofouling Ingredient

Capsaicin (8-Methyl-N-vanillyl-*trans*-6-nonenamide) is an extract from the chilli pepper fruit (*Capsicum annuum*) and confers chili's their distinctive flavour and pungency (Peng et al., 2011). The pungency from the capsaicin is what makes it effective as an antibiofouling ingredient, creating an inhospitable environment for microbes when applied to surfaces (Liu et al., 2022a; He et al., 2023). The antifouling ability is achieved through the release of capsaicin, which has a biocidal effect with low non-specific toxicity when released into the marine environment, effectively preventing the attachment of microorganisms (He et al., 2023). The rate of release of capsaicin from a matrix (i.e., silicone) can pose an issue for its use as an antibiofouling ingredient (Liu et al., 2022a). This happens when the capsaicin is not thoroughly incorporated into the matrix adhering to the surface and needing biofouling prevention which creates a short-lived lifespan of capsaicin as an active antibiofouling ingredient (Qin et al., 2023). This effect can be counteracted with a long-lasting binder or by combined with other antibiofouling abilities for a more robust synergistic effect (Qin et al., 2023).

1.8. Polydimethylsiloxane (PDMS): Active Antibiofouling Ingredient

Polydimethylsiloxane (PDMS) is a non-toxic silicone oil derived from abundant, and naturally occurring silica (Schmidt et al., 2006). Silica, being much of the sand on beaches, is widely available and contributes to the low price of PDMS (Schmidt et al., 2006). PDMS is hydrophobic, and due to this, it can be used as a slippery lubricant in many different applications (e.g., microfluidic devices, anti-foaming agents, and hydraulic fluid) (Miranda et al., 2021). As an active antibiofouling ingredient, PDMS prevents the attachment of fouling organisms through low surface energy and its hydrophobic quality (Seo et al., 2021). Though using PDMS alone as an anti-biofouling solution would be ineffective, combining it with another hydrophilic anti-biofouling substance, such as chitosan nanoparticles, could create a robust and effective solution (Seo et al., 2020; Seo et al., 2021). PDMS acts as a matrix to contain the chitosan nanoparticles due to their opposite polarity, which over time will degrade and slowly release the chitosan nanoparticles to prevent attachment over the long term (Mahto & Pal., 2020). Together, PDMS and chitosan nanoparticles create an ideal amphiphilic solution to biofouling that is non-toxic, sustainable and cost-effective.

1.9. Purpose and Hypothesis

The aim of this thesis was to develop a novel antibiofouling coating material that was sustainably produced (i.e., renewable material), composed using environmentally benign constituents (i.e., non-toxic, did not contain microplastics or biocides), and was cost-effective for commercialization in marine industries. The active ingredients combined and tested were chitosan, capsaicin, and polydimethylsiloxane. The study examined the coating’s properties and analyzed it's effectiveness against biofouling organisms (*Escherichia coli* and *Staphylococcus aureus*) using the paper disc diffusion method and a proteinaceous analogue test using egg whites. It was hypothesized that all replicates (with differing chitosan nanoparticle concentrations to test for efficacy: 50:1, 100:1, and 200:1 where the ratio represented chitosan: capsaicin/PDMS) would be effective at preventing biofouling of the chosen bacteria, algal spores, and diatom, with the moderate concentration being the most efficient in ability and cost as supported by the literature.

Research objectives

1. Screen for environmentally friendly constituents of the antibiofouling paints and develop a cost-effective, non-toxic paint.
2. Test combinations of the active ingredient within the antibiofouling solution to determine optimal efficiency as a paint.

Chapter 2. The World of Antibiofouling Solutions: History, Functionality and Future - Review

Abstract

The problem of biofouling has persisted throughout human history despite numerous prevention efforts. Antibiofouling solutions have been used for millennia with varying success. Traditionally, solutions embedded with heavy metal biocides were utilized. These traditional solutions have become a cause for concern as their toxic substances accumulate in the surrounding environment and negatively impact marine organisms. As a result, antibiofouling research has shifted towards sustainable alternatives to traditional toxic coatings. In this review, 100 marine antibiofouling studies were examined to understand current innovations, how environmentally benign solutions are being developed, and how solution testing can be improved. Most studies focused on the sustainability and non-toxicity of antifouling solutions by using non-toxic biocides, hydrophobicity, superhydrophobicity, low surface energy, hydrophilicity, amphiphilicity, self-polishing, and zwitterions as chemical deterrents. Additionally, altered topography, electrochemical repulsion, and photocatalysts were used as physical deterrents. The findings from the review show that an emphasis should be placed on sustainability and the utilization of synergy in future solutions by combining substances that provide multiple antibiofouling abilities to deter fouling. With the future focus on sustainable and environmentally benign solutions, it is important to understand what researchers mean by these terms and to consider the broader scope of application, as most solutions are not entirely eco-friendly as they claim (e.g., toxic base paint and microplastics). Collaborative efforts between scientists, industry stakeholders, and regulatory bodies will be essential in driving the development and adoption of innovative, sustainable antibiofouling technologies that can effectively protect marine environments and human interests alike.

2.0. Introduction

Humans have been navigating aquatic ecosystems since time immemorial. Many different modes of transport have been designed throughout history and across civilizations to transverse bodies of water. Each vessel was designed for a certain purpose, such as fishing and/or transportation, but all vessels face one common problem. Shipping vessels, along with most other human-made marine infrastructure, have faced the challenge of marine organisms colonizing their surfaces from the first submersion. This attachment and accumulation of marine organisms on submerged surfaces is known as biofouling (Dürr & Thomason, 2009; Abioye et al., 2019). Biofouling is a successional process consisting of four major stages i) the absorption of dissolved organism matter, ii) the attachment of unicellular eukaryotes and prokaryotes, iii) the secondary colonization of invertebrate larvae and algal spores; iv) and the development of macro-organisms such as kelp and barnacles (Dürr & Thomason, 2009; Bixler & Bhushan, 2012; Hadžić et al., 2022).

Biofouling occurs on most submerged surfaces and has caused significant financial burdens for marine industries throughout history (Dürr & Thomason, 2009; Schultz et al., 2010; Davidson et al., 2016; Trueba-Castaneda et al., 2021). For example, biofouling can decrease the efficiency of aquaculture systems (e.g., obstructing pumps, fouling on cages, netting, buoys and stock, increased risk for disease and decreased water quality), resulting in the need for more management and maintenance of such facilities (Schultz et al., 2010; Adams et al., 2011; Fitridge et al., 2012). It is estimated that biofouling accounts for 5-10% of aquaculture operational costs, amounting to a global expenditure of $1.5-3 billion USD annually (Fitridge et al., 2012).

Other vessel-based sectors such as the navy, shipping/transport and fishing industries also experience substantial costs due to biofouling (Townsin, 2003; Dürr & Thomason, 2009; Davidson et al., 2021; Davidson et al., 2023). The cost associated with biofouling to the US Navy alone (i.e., coating, cleaning, and fuel use) is estimated to be between $400-540 million USD annually, with comparable costs seen in other marine sectors (Jones, 2009; Schlutz et al., 2010). As the US naval fleet only represents ~0.5% of the entire world fleet (in terms of number of ships), it is suggested that the total global economic loss associated with biofouling is astronomical (Alberte et al., 1992; Schlutz et al., 2010). Overall, biofouling can increase voyage costs by up to 77% compared to a clean ship's hull (Yerba et al., 2004). Most costs are associated with biofouling, which affects ship functionality and fuel usage (Townsin, 2003; Yerba et. al., 2004; Schlutz et al., 2010; Hewitt, 2023). Attached organisms on ship hulls create roughness and frictional drag, resulting in decreased speed and increased voyage time and fuel consumption (Yerba et al., 2004; Lindholdt et al., 2015; Hakim et al., 2019; Hadžić et al., 2022). This has a dramatic economic and environmental effect due to increased GHG emissions per voyage (Amara et al., 2018; Notti et al., 2019; Farkas et al., 2021; Mosunov & Evstigneev, 2021).

Even minimal biofouling, such as a thin biofilm, can increase a vessel's fuel consumption by 8% increasing to 18% when biofilms form thicker slime with larger organisms (Notti et al., 2019). GHG (i.e., CO_2) emissions from the transport of maritime industries account for 2.6% of the total global emissions and are projected to increase to 17% by 2050 (Schnurr & Walker, 2019). This increase in emissions can be combated with more energy-efficient fuels (e.g., biofuels) and vessels equipped with effective antibiofouling solutions (Farkas et al., 2021; Hewitt, 2023). The International Maritime Organization (IMO) is enforcing this through their GHG reduction strategy, which plans on a 40% reduction of CO_2 emissions of international shipping transportation by 2030 and to reach net-zero GHG emissions by 2050 (International Maritime Organization, 2023).

An antibiofouling solution is defined as a coating, paint, surface treatment, surface or device that is used on a ship to control or prevent attachment of unwanted organisms by the International Maritime Organization (IMO) (International Maritime Organization, 2019). These solutions can act as chemical deterrents, physical deterrents, or both for a more robust solution (Almeida et al., 2007; Hellio & Yerba, 2009; Pan et al., 2022a). However, effective antibiofouling solutions pose significant challenges. Several biogeochemical and mechanical factors influence biofouling and the choice of antibiofouling solution to be used for a particular purpose. Some of the most important factors to consider when implementing antibiofouling solutions include the organismal fouling community (i.e., bacteria, fungi, algae, protists, invertebrate larvae and microscopic eukaryotes), water chemistry (i.e., salinity, hardness, pH, and light), location (i.e., temperature, seasonality, and depth), and operational movement (i.e., stationary, time remaining stagnant and travel speed) (Chambers et al., 2006; Dürr & Thomason, 2009; Bixler & Bhushan, 2012). Due to these factors and the high variability that influences biofouling, no one antibiofouling solution that can do it all.

Throughout history, there have been countless antibiofouling solutions conjured up by resolute seafaring civilizations (Readman, 2005; Chambers et al., 2006; Dafforn et al., 2011). Most implemented antibiofouling solutions have been effective in some way, but not without consequences and lessons learned that further helped inform the future generations of solutions (Readman, 2005; Dafforn et al., 2011). Building off the backs of our ancestors, researchers and entrepreneurs alike have continued working toward better, more effective antibiofouling solutions to solve the problem that is biofouling. In this literature review, the past, present and future of antibiofouling solutions is compiled, analyzed and discussed to understand what has already been developed and ways it can be further evolved. The review also examines the varying categories and mechanisms of antibiofouling solutions for a comprehensive report containing its history and functionalities. So, first, we must dip into antibiofouling's past to paint a clear picture.

2.1. Past: The Colorful History of Antibiofouling Solutions

Biofouling has plagued humanity since the first artificial surfaces were plunged into the water. Due to this, ancient civilizations used substances such as tar, wax and asphalt to help waterproof and prevent fouling on their wooden ship hulls (Almeida et al., 2007; Readman, 2005; Dafforn et al., 2011). The Phoenicians and Carthaginians were the first recorded societies to report the uses of antibiofouling solutions like the analogues of today (Almeida et al., 2007; Piola et al., 2009; Dafforn et al., 2011). They developed multiple rudimentary biocidal-based solutions, such as the use of pitch with a combination of copper sheathing, tallow with lead sheathing, and coatings of arsenic/sulphur in oil (Almeida et al., 2007; Dafforn et al., 2011). These biocide-based antibiofouling solutions were only the beginning of major developments for the possibilities of maritime industries.

A traditional biocide-based solution describes antibiofouling coatings that contain a biocide/toxin within a matrix (Chambers et al., 2006; Almeida et al., 2007). These solutions are classified as chemical deterrents with the use of a biocide. Biocides are any chemical substance, mixture or microorganism used to deter, or destroy harmful organisms (Konstantinou & Albanis, 2004; Thomas & Brooks, 2010). The biocide is embedded within a matrix that is either insoluble or soluble in water (Rascio et al., 1988; Almeida et al., 2007).

2.2. Insoluble and Soluble Matrix Solutions

Insoluble matrix solutions are also known as hard coatings due to the insolubility and mechanical strength of the high molecular mass substances used for the matrices (i.e., acrylic, epoxy, vinyl, and chlorinated rubber) (Yerba et al., 2004; Almeida et al., 2007).

These were implemented by the Romans and Greeks with the combined use of lead sheathing and copper nails as biocides within a metal matrix (~200 B.C. – 45 A.D.) (Marson, 1969; Yerba et al., 2004; Almeida et al., 2007). In these insoluble matrix solutions, the biocide is the only functional element acting against the fouling organisms (Kiil et al., 2003; Yerba et al., 2004). Within the matrix, biocide particles are packed densely enough to contact one another in almost all directions (Kiil et al., 2003; Yerba et al., 2004). When water encounters the particles, it starts to act on them through hydration and dissolution of the biocide. Once dissolved, the water can further penetrate within the matrix, encountering more particles over time with a gradual release (Kiil et al., 2003; Yerba et al., 2004; Almeida et al., 2007). The hydration and dissolution of the biocide prevents fouling organisms' attachment and subsequent growth as long as the insoluble matrix remains (Kiil et al., 2003; Yerba et al., 2004; Almeida et al., 2007). The effectiveness of these solutions is compromised over time because it becomes harder for the water to penetrate the pores of the matrix and reach the remaining particles, resulting in decreasing effectiveness (Marson, 1969; Kiil et al., 2003; Yerba et al., 2004; Almeida et al., 2007). Once most of the biocide is released from the matrix, a honeycomb-like structure is left behind, which contributes to drag due to its roughness and can eventually aid fouling organisms in their attachment (Kiil et al., 2003; Almeida et al., 2007). Therefore, insoluble paints are only effective for 1 to 2 years and must be renewed relatively often, limiting their application (Almeida et al., 2007). These solutions do have some advantageous qualities that other more modern solutions lack, such as oxidation and photooxidation stability due to the high inertia of the matrix substances used, which reduces problems when dry docking (i.e., degradation of coating) (Marson, 1969; Kiil et al., 2003; Yerba et al., 2004; Almeida et al., 2007).

Soluble matrix solutions consist of biocide particles and a soluble matrix (Rascio et al., 1988; Yerba et al., 2004; Almeida et al., 2007). These solutions have been used throughout history, starting with Plutarch, who used a combination of algal scrapings, slime, and pitch around 45-125 A.D. This was followed by other civilizations, such as the English in the early 19th century, who used copper, arsenic, and mercury as biocides, and linseed oil, shellac, tar, and resin/shellac varnishes as binders/matrix substances (Yerba et al., 2004; Almeida et al., 2007; Laidlaw, 2021). The solubility of the matrix ensures that two forces act against fouling organisms: the biocide and the degradation of the matrix itself (Yerba et al., 2004; Almeida et al., 2007; Banerjee et al., 2010). The degradation occurs through hydration and dissolution of the matrix and biocide (Yerba et al., 2004; Almeida et al., 2007). This process determines the rate of effectiveness based on the deterioration rate of the binder/matrix material (Yerba et al., 2004; Almeida et al., 2007; Banerjee et al., 2010). Traditionally, materials such as rosin (i.e., resin extract from pine and fir trees) have been used for this purpose (Stupak et al., 2003). Due to the fast oxidation of rosin, issues arise during dry-docking, reducing the applicability of these solutions (Yerba et al., 2004; Almeida et al., 2007). Advancements in antifouling material research have led to the discovery of novel chemicals, such as colophony for a more robust soluble matrix/binder (Yerba et al., 2004; Almeida et al., 2007; Banerjee et al., 2010). In modern times, closely related self-polishing solutions have overtaken the popularity of these more traditional soluble matrix solutions.

2.3. Self-Polishing Solutions

Self-polishing antifouling solutions can ultimately be classified as soluble matrix solutions, but there is a significant distinction between the two (Yerba et al., 2004; Almeida et al., 2007). While soluble matrix solutions degrade by hydration and dissolution, self-polishing solutions degrade through hydrolysis of the ions in the coating with the ions in the saltwater (Nwuzor et al., 2021). The matrices/binders of antifouling solutions are made up of different polymers, dictating which ablation method will be used. Hydrolysable polymers are used in self-polishing solutions and work through the hydrolysis of the side chains (e.g., copper, silyl, and zinc ester) (Löschau & Krätke, 2005; Chambers et al., 2006; Almeida et al., 2007). Many self-polishing solutions combine hydrolyzable polymers with degradable polymers (i.e., typically used in soluble matrix solutions) for a more robust solution (Almeida et al., 2007; Dafforn et al., 2011; Liu et al., 2022b). Hydrolysable polymers are best for surfaces under strong water flow, whereas degradable polymers are best suited for static conditions (Löschau & Krätke, 2005; Pan et al., 2022b). Self-polishing solutions became much more applicable with the introduction of tributyltin (TBT) as a biocide, as these solutions became the most long-lasting (i.e., >5 years) marine antifouling solutions to date (Löschau & Krätke, 2005; Chambers et al., 2006; Almeida et al., 2007).

Tributyltin Self-Polishing Solutions

Anyone who has researched antifouling solutions knows the infamous tributyltin (TBT) solutions. This can be attributed both to their success as a solution and their detriment to the marine environment (Nehring, 2001; Li et al., 2023). Tributyltin solutions have been the most universally effective antifoulants to date, having at one time covered 70% of the world’s entire fleet (Yerba et al., 2004). Tributyltin’s antifouling properties were discovered by a Dutch scientist, Van Der Kerk, who used it as a biocide to exterminate a freshwater parasitic worm (Schistosoma). Shortly after, in the late 1960s, it was implemented in marine antifouling technologies (Dafforn et al., 2011; Li et al., 2023). It was first introduced within a soluble matrix solution, having the same effectiveness as the current copper-based solutions on the market (i.e., 1–2-year longevity) (Ten Hallers-Tjabbes, 1997; Dafforn et al., 2011). Shortly after, in the 1970s, the TBT self-polishing copolymer (TBT-SPC) solutions were developed and revolutionized the field of antifouling (Dafforn et al., 2011; Li et al., 2023).

TBT is a powerful endocrine-disrupting biocide/toxin, making it a highly effective antifoulant for many marine micro- and macro-organisms (Dafforn et al., 2011; Dietert, 2014). These solutions were viewed as the global fix-all for 20 years until their first nationwide prohibition in France during the 1980s (Dafforn et al., 2011). The ban was enforced because of the negative impacts TBT was having on the oyster aquaculture industry (80-100% of oysters had developmental malformations) near locations with substantial vessel traffic (Dafforn et al., 2011). The banning was then soon followed by the UK, USA, Canada, Australia, and the EU between 1987 and 1989, as it was discovered that TBT caused the development of imposex (development of male characteristics) in female gastropods (Gibbs & Bryan, 1986; Champ, 2000; Dafforn et al., 2011). Further research also showed that TBT accumulates within the tissues of large vertebrates (crustaceans, fish, birds, mammals, and humans) (Li et al., 2023). This eventually led to the global ban on the further application of TBT in 2003, and finally, the total global ban in 2008 enforced by the IMO (International Maritime Organization, 2019).

2.4. Current Antibiofouling Solutions

With the banning of tributyltin, a significant gap was left in the world of antifouling solutions. The world's fleet had to pivot back to traditional antifouling products, causing a revival of copper-based biocidal solutions (Almeida et al., 2007). This resurgence and increased use of heavy metals are causes for concern for similar reasons as TBT (Terlizzi et al., 2001), copper biocides (i.e., cuprous oxide or copper thiocyanate) and other metals (e.g., zinc and silver) are known to bioaccumulate in filter-feeding invertebrate species (e.g., oysters). When near heavy vessel traffic, this bioaccumulation can ultimately impact coastal aquaculture facilities and the quality of fish stock intended for human consumption (Wang et al., 2014a; Wong et al., 1999; Zhang & Wang, 2005). Due to these concerns, it seems inevitable that metal-based biocidal solutions will also face a worldwide ban, as copper biocides have already been banned by several European countries for use on recreational vessels (Arai, 2009). Since the current market antifouling solutions are again facing prohibition due to environmental concerns, there is a renewed focus on the innovating novel, non-toxic, environmentally friendly solutions (Kyei et al., 2020).

2.5. Future Antibiofouling Prospects: Methods

The next section of the literature review revolves around a compilation of 100 research articles (Supplementary Material Table S1) involving marine antifouling solutions from the past decade (2015-2024). A semi-systematic review was conducted using the following search terms: "antibiofouling/antifouling, solutions/coatings/paint, novel, marine," excluding the following terms: "-membrane, -review, -biomedical, -antibiotics, -biomedical contamination, -desalination, -recovery, -biosensors, and -drug" to ensure the focus remained on novel marine antifouling solutions relevant to this paper. The first 100 papers found from multiple databases (i.e., Google Scholar, Scopus, and Novanet) meeting the requirements of the search terms were included for a robust, non-biased analysis of future trends in the field of antifouling. In this search, multiple papers came from the same journals, including *Progress in Organic Coatings* (12 papers), *ACS Applied Materials & Interfaces* (5 papers), *Chemical Engineering Journal* (5 papers), and *Langmuir* (5 papers), with all others having only 1-3 papers per journal (Supplementary Material Table S2). This is relevant for future researchers who wish to publish novel marine antifouling solutions, as these are potential avenues to pursue.

2.6. Future Antibiofouling Prospects: Results

A wide range of novel antifouling solutions is being formulated by entrepreneurs and academics alike, employing various chemical and physical methods to prevent or deter fouling organisms from attaching to various surfaces. The trend in future antifouling solutions seems to be directed toward environmentally friendly options that will not impact non-target species in the long term (Amara et al., 2018; Kyei et al., 2020). This is being attempted through many different avenues, implementing both chemical and physical deterrents such as the use of non-toxic biocides, zwitterionic forces, self-polishing capabilities, hydrophobicity/super-hydrophobicity/low surface energy, hydrophilicity, amphiphilic character, and altered surface topography (Almeida et al., 2007). Many novel solutions have also focused on combining multiple antifouling abilities to create a synergistic effect and a more robust antifouling solution (Sharmin et al., 2015). These multifaceted strategies are crucial in developing advanced coatings and materials designed to effectively combat biofouling without introducing harmful substances into marine ecosystems. Among the most promising strategies is the development of non-toxic antifouling biocides that leverage natural biochemical pathways or biomimetic approaches to deter organisms without disrupting the surrounding aquatic life (Amir et al.., 2021).

Table 2.1**.** Compilation of the antibiofouling techniques used from the literature review of 100 marine antibiofouling studies (Supplementary Material Table S1), where some solutions used multiple technique for a synergistic approach.

Type of antifouling technique	Number of solutions (number/100)
Biocidal	45
Hydrophobic/ superhydrophobic/ low surface energy	32
Self-polishing	19
Altered topography	12
Zwitterion	10
Hydrophilic	9
Amphiphilic	8
Electrochemical repulsion	3
Photocatalyst	3
Other Physical deterrent (i.e., abrasion, gas-bubbles)	2

2.6.1 Chemical Deterrents: Biocide-Based (Non-Toxic, Metals, Nanoparticles)

The most common antifouling solutions employ biocides, as previously mentioned with the historic use of TBT, copper and tin (Srinivasan & Swain, 2007). Of the solutions reviewed, 45 implemented biocides (refer to Supplementary Material Table S1). Current research shows that biocides remain a popular method for preventing biofouling, but rather than relying on the toxic chemicals of the past, researchers are exploring innovations (Lu et al., 2018; Sánchez-Lozano et al., 2019; Kumar et al., 2021; Quémener et al., 2021; Ghattavi et al., 2023). A significant body of research within the antifouling community focuses on discovering novel non-toxic biocides from the natural world for a more environmentally friendly approach (Feng et al., 2018a & b; Sánchez-Lozano et al., 2019; Tian et al., 2020; Quémener et al., 2021). Researchers are examining organisms with natural antifouling chemicals or characteristics and incorporating them into novel solutions; 12 of the 45 biocide-based studies focused on this (Noor Idora et al., 2015; Shao et al., 2015; Xin et al., 2016; Moodie et al., 2017; Zhu et al., 2017; Feng et al., 2018a & b; Sánchez-Lozano et al., 2019; Pinteus et al., 2020; Tian et al., 2020; Quémener et al., 2021; Takamura et al., 2023; Lenchours Pezzano et al., 2024). For instance, Feng et al. (2018a) examined 18 alkaloids from terrestrial plants and found five effective substances for preventing biofouling (i.e., evodiamine, camptothecin, cepharanthine, sinomenine, and strychnine). Sánchez-Lozano et al. (2019) derived non-toxic extracts from five macroalgae and two sponges (successfully from: Laurencia gardneri, Sargassum horridum (macroalgae), Haliclona caerulea, and Ircinia sp. (sponges)), and Lenchours Pezzano et al. (2024) looked at enzymatic extracts from fishery residues for antifouling potential.

There is also a large focus on nanoparticle and nanocomposite biocides, as particle size plays a significant role in the effectiveness of foul-release biocide-based antifouling solutions. Eight of the 45 biocidal-based antifouling solutions used nanoparticles for this purpose (Supplementary Material Table S1). Kumar et al. (2021) developed chitosan-based antifouling coatings with varying percentages of ZnO or ZnO–SnOx nanoparticles, with or without the glutaraldehyde crosslinking of chitosan and a photocatalyst. This solution was found to be antimicrobial, photo-stabilized, and environmentally friendly, but needed field testing. Zhang et al. (2016) created a titanium dioxide (TiO_2) nanoparticle suspension in fluorinated acrylic copolymers, which was anti-corrosive, antibacterial/algal, field-tested, and environmentally friendly, but further development for market use is needed.

Most novel antifouling solutions are being developed with environmental awareness and consideration, but this is not always true. Multiple studies released within the past decade use a metal biocide within the solution and claim to be environmentally friendly (Punitha et al., 2017; Chen et al., 2020; Hu et al., 2021a; Kumar et al., 2021). This is a misleading claim, as these metals can be non-toxic and environmentally friendly when found in low concentrations, but when used in high amounts as marine coatings, these statements no longer remain true (Katranitsas et al., 2003; Srinivasan & Swain, 2007). If these novel solutions were implemented on a large scale, it would only be a matter of time before another global ban on an antifouling substance would occur.

2.6.2 Chemical Deterrent: Self-Polishing Solutions and Zwitterions

Self-polishing solutions may be the most effective antifouling technique to date, which is why researchers are still investigating their full potential (Zhang et al., 2019; Lau & Yong, 2021). As mentioned in section 2.2, toxic biocides (e.g., tin, copper, zinc, and silver) are commonly incorporated within self-polishing solutions, making the majority toxic to off-target organisms (Chambers et al., 2006; Almeida et al., 2007). Due to this, the focus in this body of research is on developing non-toxic biocides that can replace these toxic substances within the hydrolyzable matrices (Chambers et al., 2006; Liu et al., 2021).

Of the solutions reviewed, 19 had self-polishing characteristics (Supplementary Material Table S1), four of which still incorporated the use of a metal biocide, some of which claimed to be environmentally friendly (Dai et al., 2021; Tian et al., 2021; Zhou et al., 2021a; Deng et al., 2023). For example, Tian et al. (2021) developed a Cu-Ti composite antifouling coating with a micron-sized alternating laminated structure of Cu/Ti by plasma spraying the mechanically mixed Cu/Ti powders. The researchers claimed that this coating had a controlled release rate, good adhesion, mechanical robustness, was long-lasting, and was "environmentally friendly" despite incorporating two heavy metals (i.e., copper and titanium). This claim of environmental friendliness was based on the slow and controlled release rate of the copper ions in small doses over time, which would not significantly impact the natural copper concentration found within seawater. However, this would not hold if used on a large scale (Hellio & Yebra, 2009). Zhou et al. (2021a) created a rosin-based zinc resin via a one-pot dehydration condensation reaction using non-toxic rosin acid and $Zn(OH)_2$, and Dai et al. (2021) developed self-polishing zinc-polyurethane copolymers, which contained zinc atoms in the form of a polymeric salt. Both studies used heavy metals in their solutions and claimed to be environmentally friendly. The problem is that copper and zinc concentrations would inevitably increase beyond the naturally occurring levels, potentially leading to a similar outcome as with TBT (Katranitsas et al., 2003; Srinivasan & Swain, 2007). It is important to understand how and why a solution claims to be environmentally friendly to prevent history from repeating itself (i.e., localized extinctions and future banning of copper biocides).

Zwitterionic surfaces as an antifouling technique are relatively novel in the field but are quickly becoming one of the most promising. Zwitterions make effective antibiofouling solutions due to their molecular structure and interactions, as they have both positively and negatively charged parts, resulting in an overall neutral charge (Zhang et al., 2019). This molecular characteristic prevents marine biofouling mainly through the formation of a hydration layer between the coated surface and the seawater. This layer forms because the positively charged portion of the zwitterions strongly attracts polar water molecules (Leng et al.,2016). As a result, proteins and biofouling organisms cannot attach to the slippery, soap-like surface (i.e., the hydration layer) and are repelled, creating a self-cleaning effect. Zwitterionic solutions are also long-lasting because they do not rely on the release of biocides or self-degradation like traditional insoluble/soluble matrix solutions (Zhang et al., 2019; Chen et al., 2022).

Of the studies reviewed, ten focused on zwitterions (Supplementary Material Table S1). For instance, Mei et al. (2020) developed surface-fragmenting hyper-branched copolymers with tertiary carboxybetaine ester (TCB) primary chains and poly(ε-caprolactone) bridged chains. These chains can hydrolyze and degrade in marine environments, continuously generating zwitterions, so the polymer coating has a fouling-resistant and renewable surface. Dai et al. (2021) developed a “kill–resist–renew trinity” polymeric coating using monomer-tertiary carboxybetaine ester acrylate with the antifouling group N-(2,4,6-trichlorophenyl) maleimide copolymerized with methacrylic anhydride via reversible addition-fragmentation chain transfer polymerization, yielding a degradable zwitterionic hyperbranched polymer. Another interesting study by Kim and Kang (2024) developed amphiphilic zwitterionic thin polymer brushes composed of sulfobetaine methacrylate and trifluoroethyl methacrylate synthesized on Si/SiO_2 surfaces via surface-initiated atom transfer radical polymerization. These three studies, as well as the other seven zwitterionic-based solutions, were generally found to have excellent protein resistance, antibacterial, and anti-algal abilities, preventing the first two stages of biofouling in the successional process.

Though zwitterions are very capable antifouling solutions, there are a few challenges surrounding their implementation as marine market solutions. This is mainly due to their complex synthesis requiring skilled and trained individuals, generally high cost, and low durability/mechanical strength under environmental duress (e.g., changing salinity, temperature, and pH). It should also be noted that these solutions are marked as "environmentally friendly," although there is little research into their long-term environmental impacts since this is an innovation in marine antifouling solutions. Researchers should remain aware of these assumptions.

2.6.3 Physical Deterrents: Hydrophobicity, Hydrophilicity and Amphiphilicity

Hydrophobicity

Hydrophobicity and superhydrophobicity are among the most popular trends in marine antibiofouling, being used in 32 of the solutions reviewed (Supplementary Material Table S1). Hydrophobic substances repel water, meaning they have low wettability and surface energy (Zhu et al., 2017; Yu et al., 2021; Tong et al., 2023). These characteristics give hydrophobic substances their antibiofouling ability, as fouling organisms have difficulty attaching to low surface energy/lubricant-like surfaces and find it even harder to stay attached; flowing water easily dislodges these organisms (Zhu et al., 2017; Tong et al., 2023). Hydrophobic solutions are also environmentally friendly when they avoid petroleum-based substances and the incorporation of toxic biocides, making this a very promising technique for the future (Wang et al., 2016; Zhu et al., 2017; Selim et al., 2019; Tong et al., 2023).

Wang et al. (2016) developed a slippery lubricant-infused porous surface with high underwater transmittance, fabricated on a glass sample by infusing lubricant into its porous microstructure using the hydrothermal method. This hydrophobic solution was found to be antibacterial/algal and suitable for optical equipment but was only applicable under static conditions. Yu et al. (2021) developed organic-inorganic hybrid particles (Poly@V-SiO_2) with controlled particle sizes and adjusted chemical constituents, used as building units to construct a superhydrophobic and lubricant-grafted slippery surface, which was found to be environmentally friendly and effective against fouling but needed further development to understand its full application. Selim et al. (2019) developed a superhydrophobic composite of *in situ* PDMS/β-MnO_2 nanorods coatings with a rough structured surface, which was self-cleaning, antimicrobial, and effective over a 90-day field test but needed further development for commercial use.

These studies demonstrate that hydrophobic/superhydrophobic/low surface energy substances have significant potential in antibiofouling solutions, but there must be an emphasis on the synergy of these solutions (Wang et al., 2016; Zhu et al., 2017; Selim et al., 2019; Tong et al., 2023). Only nine of the 32 hydrophobic solutions relied solely on hydrophobicity as the antifouling characteristic (see Supplementary Material Table S1). The other studies involving hydrophobic characteristics incorporated at least one other antifouling quality for a more robust solution (see Supplementary Material Table S1). Researchers interested in hydrophobic coatings as antifouling solutions should consider a synergistic strategy for more effective results.

Hydrophilicity

Hydrophilic antibiofouling solutions work like hydrophobic solutions but oppositely: while hydrophobic substances repel water molecules, hydrophilic ones attract them (Shen et al., 2019; Yang et al., 2024). This strong attraction to water molecules prevents the attachment of fouling organisms, creating a physical barrier between the coated surface and the water (Wang et al., 2016; Yao et al., 2020). Nine of the 100 marine antibiofouling solutions reviewed exhibited hydrophilicity (see Supplementary Material Table S1).

Xie et al. (2020) created a copolymer using polyacrylates, tert-butyldimethylsilyl methacrylate, eugenol methacrylate (EM), and poly(N-vinylpyrrolidone) (PVP), where EM is antibacterial, PVP causes hydrophilicity, and encapsulated eugenol gradually releases to repel marine organisms further. This solution was found to be protein-resistant, anti-bacterial/algal, and effective for an eight-month field experiment, but further development is needed for commercialization (Xie et al., 2020). Liu et al. (2019) developed a coating by incorporating 3-sulfopropyl methacrylate potassium salt on a flat hydrophilic resin surface using SSI-ATRP. It was found to be environmentally friendly, anti-diatom, and long-lasting, although its effectiveness was hindered by salt responsiveness, requiring further development. Pan et al. (2022b) synthesized degradable hyperbranched polymers with a controlled structure of methyl methacrylate, tertiary carboxybetaine ester, and divinyl monomer-methacrylic anhydride via RAFT polymerization, having superhydrophilic character. It was found to have protein resistance, anti-bacterial/diatom abilities, and a controlled degradation rate, but it was not field tested. Therefore, its application is not fully understood, and further research is needed (Pan et al., 2022b).

Hydrophilic surfaces as antifouling solutions seem to be gaining traction, as two of the nine hydrophilic-based studies were from the current year (2024) (Liu et al., 2024; Yang et al., 2024). The future trend in hydrophilic solutions appears similar to hydrophobic ones, emphasizing synergy, as none of the nine studies solely relied on hydrophilic quality to prevent biofouling (see Supplementary Material Table S1). Therefore, hydrophilic solutions are a promising avenue for antifouling coatings, seemingly only when combined with other antifouling techniques.

Amphiphilicity

Amphiphilic antifouling solutions prevent organismal attachment by having both hydrophobic and hydrophilic parts (Guazzelli et al., 2020; Jiang et al., 2024). The antifouling ability is achieved by combining the hydrophobic portion repelling water molecules. In contrast, the hydrophilic portion simultaneously attracts the water molecules, creating a physical barrier that makes it difficult for fouling organisms to penetrate or invade (Su et al., 2021; Sven et al., 2021; Jiang et al., 2024). Among the solutions reviewed, eight exhibited amphiphilic character (see Supplementary Material Table S1).

Guazzelli et al. (2020) developed environmentally friendly amphiphilic methacrylate copolymers (Si-co-EF) containing polysiloxane (Si) and mixed poly(oxyethylene)–perfluorohexyl (EF) side chains synthesized with different compositions. They are combined with polysiloxane-functionalized nanoparticles as additives to condensation-cured nanocomposite poly(siloxane) films. They found that the solutions had anti-diatom and anti-polychaeta abilities, but biofouling occurred during a 10-month field test (Guazzelli et al., 2020). Guo et al. (2020) developed a novel amphiphilic block copolymer that combined hydrophilic polyvinylpyrrolidone (PVP) with hydrophobic poly(1-(1H,1H,2H,2H-perfluorodecyloxy)-3-(3,6,9-trioxadecyloxy)-propan-2-yl acrylate) and PDMS. It was found that the solution had antifouling abilities against bacteria, diatoms, and a macro-fouler (i.e., pseudobarnacle), but field testing was needed to understand its application fully. Su et al. (2021) created a polyurethane coating with amphiphilic micro/nanodomains structure triggered by phase segregation. It was found to be protein resistant, have anti-algal/macro fouler abilities, and be mechanically robust, but field testing was also needed to understand its real-world application.

These studies are good representations of the future trends in amphiphilic antibiofouling solutions. Amphiphilic-based solutions are intrinsically synergistic, combining both hydrophobic and hydrophilic abilities, which underlines the importance and significance of synergistic interactions within an antifouling solution. This holds especially true when pursuing environmentally friendly solutions with non-toxic characteristics.

2.6.4. Physical Deterrent: Altered Topography

Altered surface topography antibiofouling solutions are becoming a favourable option for environmentally friendly coatings. Achieving altered topography can be done through a variety of different methods and chemicals, but ultimately, the goal of these solutions is to change the surface topography so that fouling organisms can no longer attach (Duan et al., 2023; Yang et al., 2023; Richards et al., 2024). Typically, this is accomplished using microparticles and/or nanoparticles/nanocomposites, as the altered surface topography must prevent the attachment of organisms while also not increasing drag force (Selim et al., 2022a; Yang et al., 2023; Richards et al., 2024). For this reason, and due to the generally low abrasion resistance, altered surface topography solutions are best used under static conditions (Fu et al., 2017; Yang et al., 2023; Richards et al., 2024).

Of the solutions reviewed, 12 involved altered surface topography (see Supplementary Material Table S1). Richards et al. (2024) engineered six surfaces inspired by Brill fish scales fabricated through a 2-photon polymerization process. They discovered that the biomimetic antifouling surfaces had anti-diatom abilities, although further development would be needed to understand mechanical strength and real-world applications. Fu et al. (2017) developed an ethylene-vinyl acetate copolymer composite coating prepared with N-(2,4,6-trichlorophenyl) maleimide (TCPM) using halloysite clay as tubule nanocontainers to encapsulate the TCPM. The coating had anti-bacterial abilities lasting through a 60-day field test with claims of being environmentally benign. Although this may not be the case, as the coating may contribute to microplastic pollution as it degrades, further testing is needed to understand these implications. Another study by Zhang et al. (2017) developed polymer brushes on a wrinkled silicone elastomer using surface-initiated atom transfer radical polymerization, resulting in an altered surface topography coating that was environmentally friendly and had anti-algal abilities. The application of this coating is not fully understood, as no field testing was performed, which is needed to understand a coating's real-world effectiveness and market potential. There is potential for altered surface topography antibiofouling solutions to become the next market solution, but there are a few issues to work out first. Altered surface topography coatings can easily be environmentally friendly if the substances used for their construction do not degrade over time or alternatively do not accumulate in the surrounding ecosystem and/or organisms (Yang et al., 2023; Richards et al., 2024; Selim et al., 2024). This seems to be the biggest issue with altered surface topography solutions. As previously mentioned, many of these solutions claim to be environmentally friendly but are plastic-based (Fu et al., 2017; Zhang et al., 2017; Richards et al., 2019; Su et al., 2021). Although most of these plastics are plant-based rather than petroleum-based, it is not fully understood how these "eco-friendly plastics" will impact marine ecosystems in the long term (Spierling et al., 2018; Zimmermann et al., 2020). This must be a consideration when formulating these coatings for large-scale commercial use if environmental conservation is to be a priority.

2.6.5. Physical Deterrent: Other

As biofouling remains a problem, some innovative researchers have come up with unique ways to combat the challenge of biofouling. Three of the solutions reviewed incorporated photocatalysts for an antifouling effect (Zhang et al., 2020b; Li et al., 2022b; Xiong et al., 2023). Photocatalysts (e.g., titanium dioxide and zinc oxide) deter fouling organisms through the creation of reactive oxygen species (ROS) after encountering photons (Zhang et al., 2020b; Li et al., 2022b; Xiong et al., 2023). The ROS are highly reactive and can easily disturb the settlement of dissolved organic matter and proteins needed for succession, as well as damage the cells of fouling organisms (Zhang et al., 2020b; Li et al., 2022b; Xiong et al., 2023). Photocatalysts as antifouling additives are generally viewed as environmentally friendly (although the use of heavy metals must be considered when making such claims). They have self-cleaning abilities if light is present, are typically long-lasting, and do not degrade over time (Zhang et al., 2020b; Li et al., 2022b; Xiong et al., 2023). Though these antifouling additives are promising for the future, researchers must consider a few issues in their implementation. As photocatalysts only work in the presence of light and often need UV activation, which could be an issue in their effectiveness over time, seasonality, and location (Zhang et al., 2020b; Szeto et al., 2021; Li et al., 2022b; Xiong et al., 2023).

The use of electrochemical repulsion was also implemented in three of the solutions reviewed (Jia et al., 2017; Elmas et al., 2020; Zhang et al., 2023). Electrochemical antifouling works by generating an electric field, which prevents the attachment of fouling organisms (Jia et al., 2017; Elmas et al., 2020; Zhou et al., 2021b; Zhang et al., 2023). This is typically done by installing conductive materials such as electrodes upon the surface where fouling is to be prevented and then passing an electrical current through it. This generates the electric field, changing the surface chemistry and ultimately ROS, which deter fouling organisms by creating an inhospitable environment (Zhou et al., 2021b; Zhang et al., 2023). Generally, electrochemical repulsion antifouling is environmentally friendly, versatile with adjustable intensity, low maintenance, and long-lasting if equipment persists (Jia et al., 2017; Elmas et al., 2020; Zhou et al., 2021b; Zhang et al., 2023). However, there are some disadvantages, mainly due to the need for a constant supply of electrical power and the potential for increased corrosion with incompatible materials (Elmas et al., 2020; Zhou et al., 2021b; Zhang et al., 2023). Electrochemical repulsion may be the solution to biofouling, but much more research is needed before this can ever come to fruition.

Two studies of those solutions reviewed used unique antibiofouling techniques to prevent fouling. Mo et al. (2022) developed a novel spherical cavity microstructure array constructed on a conventional silicone coating (PDMS). The bioinspired (i.e., based on the concave structure of springtail and the hairy structure of *Salvinia*) coating prevents biofouling through the gas-liquid biphase interface generated by the air bubble arrays, which are produced by the microcavity structure under water pressure (Mo et al., 2022). This antibiofouling solution had anti-bacterial and anti-diatom abilities but only applies under static conditions (Mo et al., 2022). The other unique study, Zanje et al. (2017), developed a scalable floating body antifouling device consisting of a set of durable components forming a movable chain-like apparatus around offshore structures. The device was eco-friendly and cost-effective but only applies under static conditions (i.e., specifically designed for offshore oil facilities).

2.7. Conclusion: Research Gaps and Next Steps

The historical and ongoing challenges of biofouling have driven significant advancements in antibiofouling technologies, yet there remain crucial research gaps and opportunities for innovation. The resurgence of metal-based biocidal solutions in response to the banning of tributyltin highlights the need for environmentally friendly alternatives. Given the environmental concerns associated with metal biocides, there is an urgent need for the continued exploration and development of non-toxic, environmentally friendly antibiofouling solutions. Major considerations for what environmentally friendly means are paying attention to the use of heavy metals and materials that may contribute to pollution.

This literature review underscores the importance of synergy in developing antibiofouling technologies. Combining multiple antifouling properties, such as biocides, self-polishing substances, zwitterions, hydrophobicity, hydrophobicity, amphiphilic character, and altered surface topography, creates more robust and effective solutions. The future of antibiofouling will likely depend on integrating such multifaceted strategies, leveraging the strengths of different approaches to achieve optimal performance without adverse environmental impacts.

The future of antibiofouling research should be advancing non-toxic biocides derived from natural renewable sources, developing novel materials with robust antifouling capabilities, and enhancing the mechanical robustness and long-term efficacy of future coatings. There is also a promising potential in using photocatalysts and electrochemical repulsion techniques, although these require further research to address limitations related to environmental conditions and power requirements. Ultimately, the following for antibiofouling research should prioritize field testing and real-world application assessments to ensure the practicality and environmental safety of novel solutions. Collaborative efforts between scientists, industry stakeholders, and regulatory bodies will be essential in driving the development and adoption of innovative, sustainable antibiofouling technologies that can effectively protect marine environments and human interests alike.

Chapter 3: Development of a Naturally Derived Synergistic Antibiofouling Solution based on Incorporating Chitosan, Capsaicin and Polydimethylsiloxane

Abstract

Biofouling, the accumulation of organisms on submerged surfaces, presents a significant challenge to maritime industries like navies, shipping, aquaculture, and fisheries, costing these sectors hundreds of millions of dollars annually. This is primarily due to increased fuel consumption caused by the drag biofouling creates on ship hulls, which also leads to higher carbon emissions and contributes to climate change. Historically, various antibiofouling solutions have been developed, many of which contain toxic compounds such as copper, mercury, and tributyltin (TBT). While effective, these solutions pose environmental risks, prompting the need for novel, eco-friendly alternatives. This study aimed to create a sustainable, environmentally benign, and cost-effective antibiofouling solution using naturally derived chemicals: chitosan, capsaicin, and polydimethylsiloxane (PDMS). Three solutions with different chitosan concentrations were tested using bacterial assays and proteinaceous analogue tests to evaluate their efficacy. Unfortunately, the results indicated that these combinations were not effective as antibiofouling agents. The study highlights the challenges in developing solutions that are both effective and environmentally safe, stressing the need for further research into alternative substances and strategies. The findings emphasize the complexity of balancing efficacy with environmental considerations in antibiofouling solutions. The study calls for continued exploration of innovative, sustainable approaches that not only address biofouling effectively but also minimize long-term impacts on marine ecosystems. The pursuit of an optimal solution remains ongoing, and the importance of developing ecologically responsible methods is as critical as ever.

3.0. Introduction

Throughout history and across civilizations, many different water-faring vessels have been used, but one persistent problem remains: biofouling. Biofouling is the attachment and accumulation of marine organisms on submerged aquatic surfaces, affecting marine industries such as navies, shipping, transport, and fisheries (Dobretsov et al., 2019; Qian et al., 2022). This is due to drag caused by biofouling on ship hulls, leading to increased fuel usage, higher transport costs, and greater greenhouse gas (GHG) emissions, contributing to climate change (Dobretsov et al., 2019). Biofouling also impacts aquaculture, clogging pumps and pipes, leading to poor water quality and lower stock value (Fitridge et al.,2012).

Due to these persistent issues, many antibiofouling solutions have been developed throughout history. These solutions have prevented biofouling through chemical deterrents, which alter surface chemistry, and physical deterrents, which change surface structure (Yerba et al., 2004; Abioye et al., 2019). Traditionally, biofouling was prevented using metal-based paints like tributyltin (TBT), copper, and mercury (Almeida et al., 2007). However, these paints eventually break down and settle into marine sediments, causing environmental harm (Bauer et al., 1995; Carrier et al., 2023). Current antibiofouling research focuses on antibiofouling solutions, including biocides, self-polishing compounds, zwitterions, photocatalysts, and various surface treatments, focusing on sustainability (Qiu et al., 2024). However, many solutions vary in toxicity, sustainability, and cost, making them suboptimal for commercialization and global use (Almeida et al., 2007). Therefore, a novel, sustainable, environmentally benign, and cost-effective antibiofouling solution is needed.

Researchers and entrepreneurs are developing sustainable solutions for biofouling, aligning with the goals of UN SDG 14, which focuses on reducing marine pollution and promoting sustainable management and conservation of marine ecosystems (Government of Canada, 2024). Key approaches include degradable synthetic and organic compound-based biocides, hydrophobicity/hydrophilicity/amphiphilicity, self-polishing surfaces, low surface energy and biocide-free physical deterrents (Carrier et al., 2023). Organic biocides, derived from natural substances like chitin and capsaicin, are effective but face challenges in long-term effectiveness (Jin et al., 2022).

Hydrophobic/hydrophilic/amphiphilic chemicals prevent fouling organisms from attaching to a surface through its water-repelling/water-attracting/both abilities, respectively, but often must be combined with other antifouling substances for a more robust solution (Qiu et al., 2024). Self-polishing solution, effectively prevent organisms from attaching to surfaces through hydrolysis. However, they are too often combined with heavy metals, which can leach into the marine environment (Yerba et al., 2004). Low surface energy solutions work through their almost fluid-like bonds, makeing surface attachment extremely difficult for fouling organisms (Almeida et al., 2007). Biocide-free physical deterrents alter surface structure topography to prevent organisms from attaching, but these solutions are sensitive to abrasion and can be easily damaged (Amir et al., 2021). All these antibiofouling solutions have advantages and disadvantages regarding effectiveness and longevity. For this reason, it is important to continue research into these environmentally benign solutions that are non-toxic to off target species and do not accumulate in the environment and determine what combination of antifouling mechanisms will prevent organismal attachment while simultaneously having no adverse effects on the surrounding marine environment.

All its ingredients must adhere to these principles for an antibiofouling solution to be sustainable and non-toxic to the marine environment. Creating an antibiofouling solution requires more than just active ingredients; it also needs compounds for adhesion, strength, and durability. These include pigments, solvents, binders, and additives, which are often toxic but chosen for their efficiency and cost-effectiveness (Talbert., 2008). Base pigments provide colouration and material protection. Solvents and thinners, such as turpentine and xylene, adjust the paint's viscosity (Ravikumar et al., 2012). Binders, like oil, resin, bitumen, or cellulose derivatives, ensure even spreading and adhesion. Additional components like driers and colouring pigments accelerate drying and enhance colour. Antibiofouling paints also include specific additives for antibiofouling, anti-corrosion, self-healing, and UV protection (Talbert., 2008). Non-toxic alternatives exist, such as henna for corrosion protection and capsaicin for creating protective layers on metals (Stupak et al., 2003). Solvents and thinners can be replaced with non-toxic substitutes like water and propylene carbonate. Environmentally friendly binders include linseed oil, gum Arabic, and beeswax (Torres & De-la-Torre., 2021). Combining these non-toxic ingredients with sustainable and cost-effective antibiofouling materials will enable the development of an eco-friendly antibiofouling solution suitable for large-scale commercialization.

This study focuses on the synergistic combination of foul-release, non-toxic naturally derived biocides, amphiphilicity and low surface energy to prevent biofouling. Chitosan nanoparticles, derived from abundant chitin in invertebrate exoskeletons, possess natural antibacterial, antifungal, and anti-algal properties, making them effective in preventing biofouling (Zheng et al., 2011; Perez et al., 2020). Their use supports a blue circular economy by repurposing fishing industry waste (Lv et al., 2023). Chitosan is non-toxic, hydrophilic, and forms strong films under acidic conditions. When combined with hydrophobic substances, such as polydimethylsiloxane (PDMS), they create superior amphiphilic antibiofouling solutions (Galli & Martinelli., 2017; Kumar et al., 2021). PDMS, derived from silica, is cost-effective and prevents organism attachment through its hydrophobic qualities (Schmidt et al., 2006; Mahto & Pal., 2020). Capsaicin, an extract from chilli peppers, adds a biocidal effect, creating an inhospitable environment for microbes (Liu et al., 2022a; He et al., 2023). Combining these elements can produce a robust, non-toxic, and sustainable antibiofouling solution that benefits marine environments and supports sustainable maritime practices (Qin et al., 2023).

The aim of this research is to develop a novel antibiofouling coating material that is sustainable, environmentally benign, and cost-effective for commercialization. The study will test combinations of chitosan nanoparticles, capsaicin, and PDMS for effectiveness against biofouling organisms with a paper disc diffusion test against two bacterial strains (*E. coli* and *S. aureus*) and a proteinaceous analogue test using egg whites. It is hypothesized that different concentrations of chitosan nanoparticles (50:1, 100:1, and 200:1, where the ratio is chitosan: capsaicin/PDMS) will effectively prevent biofouling, with moderate concentrations being the most efficient in ability and cost.

3.1. Methods and Materials

Active ingredients were obtained from the following: Chitosan (Sigma-Aldrich, SKU: 448869-250G, low molecular weight), poly(dimethylsiloxane) (Sigma-Aldrich, SKU: 469319-50mL, Viscosity 1.0 cSt (25°C)), and capsaicin (Sigma-Aldrich, SKU: 360376-250MG, natural). The non-active ingredients were obtained from the following: propylene carbonate (Sigma-Aldrich, SKU: P52652-500G, ReagentPlus® 99%), henna/2-Hydroxy-1,4-naphthoquinone/lawsone (Sigma-Aldrich, SKU: H46805-100G, 97%), and linseed oil (Sigma Aldrich, SKU: 430021-250mL, liquid). The following reagents and solvents were supplied by Sigma-Aldrich: Sodium triphosphate (STPP), acetic acid (ethanoic acid, 1% *v/v*), and isopropyl alcohol.

3.1.1. Ingredient Selection

Active Biofouling Ingredients

A literature review was performed before laboratory experiments to determine optimal active and non-active ingredients to assess sustainable, non-toxic and cost-effective antibiofouling solutions. The literature review also informed how to synthesize chitosan nanoparticles from chitin, synthesize antibiofouling solutions, determine preferred biofouling organisms for laboratory assays, and the experimental design and analysis of the effectiveness of antibiofouling solutions. Thus, active ingredients were selected based on the literature review.

Chitosan nanoparticles were used as a non-toxic biocide to deter biofouling organisms through their inherent anti-bacterial, anti-fungal, anti-algal and hydrophilic abilities (Al-Naamani et al., 2017; Elshaarawy et al., 2017; Abdelsalam et al., 2022; Crini, 2022; Ramasamy et al., 2023). Chitosan nanoparticles were chosen as it is a common waste product of marine invertebrate fishing industries in the Western North Atlantic in the form of chitin (El-saied & Ibrahim, 2020; Santos et al., 2020; Abdelsalam et al., 2022). Using chitin in the form of chitosan nanoparticles will be much more effective at deterring biofouling organisms as the particle size remains within the matrix of the antibiofouling solution longer with a slow-release rate (Ju et al., 2020; Mahto & Pal., 2020). PDMS helps create the matrix in the antibiofouling solution, which controls the slow release of the chitosan nanoparticles (Al-Naamani et al., 2017; Elshaarawy et al., 2017; Atthi et al., 2020; Abdelsalam et al., 2022). PDMS was chosen as an active ingredient due to its film-forming and lubricant-like properties, which create a hydrophobic low surface energy surface that biofouling organisms have difficulty attaching to (Nendza, 2007; Atthi et al., 2020; Chungprempree et al., 2022). The combination of these two ingredients together creates an amphiphilic solution that is ideal for preventing biofouling by creating a physical and chemical barrier (Galli & Martinelli., 2017; Zhang et al., 2017; Soleimani et al., 2023). To create an even more robust antibiofouling solution, the chemical capsaicin was used for its anti-bacterial, anti-algal and anti-corrosion abilities for added protection as a biocide (Xu et al., 2005; Lu et al., 2018; Hao et al., 2020). These three active ingredients will, in theory, form an amphiphilic, non-toxic, slow-release (i.e., low solubility binder and nanoparticle incorporation) biocide soluble matrix solution with low surface energy (Lu et al., 2018; Mahto & Pal, 2020; Liu et al., 2022a; Soleimani et al., 2023).

Non-Active Biofouling Ingredients

The non-active ingredients were selected based on the literature review. The antibiofouling solution needs all three main components of paint (i.e., base pigment, binders, and solvents/thinners), but these ingredients must also be non-toxic and sustainable (Bentley & Turner, 1998; Lambourne & Strivens, 2004; Talbert, 2008; Ravikumar, 2012). Henna was used as a base pigment and anticorrosion element due to its properties described in *section 1.6.* Linseed oil was used for as a non-toxic binder due to its historical use, low cost, and compatibility with the chosen solvent (Phillip, 1973; Drisko, 1977; Sharmin et al., 2015; Hu et al., 2021b). Propylene carbonate is one of the best non-toxic solvents because it is biodegradable, non-corrosive, low vapour pressure and an affinity to oil-based binders (Moity et al., 2012; Yao et al., 2020).

3.1.2. Synthesising the Base Paint

Determining the Critical Pigment Volume Concentration (CPVC)

To develop the paint base 1 g of henna was combined with drops of linseed oil (the binder) to determine the critical pigment volume concentration (CPVC). This provided the ratio of pigment to binder needed in the case of henna and linseed oil (using the spatula rub-out oil absorption (OA) method (i.e., place on glass plate/marble slab mix ingredients together until a stiff putty-like paste forms) creating a millbase (American Society for Testing and Materials, 2021). This ratio (1 g henna: 0.1 g linseed oil) was scaled up depending on the needed materials. To the millbase, the paint thinner/solvent, propylene carbonate, was added 1 drop (~0.01 mL) at a time to achieve the desired consistency (i.e., a thin but opaque layer formed when applied to glass/mild steel surface), resulting in a total of 0.15 g propylene carbonate added. This experiment was performed under a chemical fume hood for safety.

Drying the base paint

It had to dry once the paint base was developed with maximum efficacy (critical pigment volume concentration). Multiple drying strategies were employed to determine the quickest method for drying. The trials consisted of ambient room temperature drying (drying conditions under a chemical fume hood, 20-22°C), heat shock drying (where the paint was heated to 40°C and subsequently allowed to dry after in ambient room temperatures), increased airflow drying (air blown directly onto the base paint), heat curing (constantly heated to 30-35°C and allowed to dry) and a heating/cooling trial (where the paint was heated to 30-35°C for a day and then left at ambient room temperature for the next day on repeat.).

3.1.3. Synthesizing the Active Antibiofouling Solution

Chitosan to chitosan nanoparticles (ionotropic gelatin method)

Based on the literature review, the methods and materials used to synthesize chitosan nanoparticles were chosen (Triwulandari et al., 2018; Mahto & Pal., 2020). Chitosan was dissolved in 1% acetic acid *v/v* (1:100, 1% solid to solvent) at 80°C with a mixing speed of 200 rpm for 2 h. After dissolution, the solution was filtered through a glass wool filter to remove high molecular weight chitosan and other impurities that may have been present. Deionized water was added to the solution to create a 0.10% *w/v* concentration with a pH of 5. A 0.10% w/v solution of STPP was created by mixing 1g of STPP with 1000 mL of deionized water while consistently stirring for 30 minutes. The 0.10% w/v chitosan solution was mixed with the 0.10% w/v STPP solution in a 4:1 ratio, respectively, at a mixing speed of 800 rpm for 1 h. This mixture was centrifuged at 11,000 rpm for 40 min. Following centrifugation, the suspension was rinsed and filtered with deionized water until a neutral pH was achieved. UV-vis and DLS particle analysis was utilized to determine whether the formation of chitosan nanoparticles occurred. If successful, any resulting chitosan nanoparticles could be used to develop the antibiofouling solutions.

Active ingredient paint preparation

Chitosan (200mg) was dissolved in 20 mL of 1% acetic acid using a magnetic stirring device set at 350 rpm for 4 h at room temperature, as indicated in various studies (Kim et al., 2002; Rutnakornpituk et al., 2006; Akyuz et al., 2018). Then, 0.002 g of powder capsaicin was incorporated into the chitosan solution by centrifuging at 21,000 rpm for 15 min. Subsequently, the chitosan-capsaicin aqueous solution was combined with 100 μL of PDMS, previously dissolved in 10 mL of isopropyl alcohol. This mixture was stirred for 2 h at 80°C to facilitate the reaction. Sonication was then used for 4 h to eliminate any bubbles, as suggested in the literature (Rutnakornpituk et al., 2006; Akyuz et al., 2018). This process resulted in the formation of a hydrogel that eventually dried into a film.

This experiment was repeated three times using varying concentrations of the chitosan-capsaicin mixture (ratios of 200:1, 100:1, and 50:1 chitosan: capsaicin/PDMS, respectively) to determine efficacy. Additionally, multiple controls were utilized, including a solvent control and a chitosan-PDMS control (i.e., to evaluate if capsaicin was needed for effectiveness). Finally, the hydrogel film (chitosan-capsaicin-PDMS film) was applied on top of the prepared mild steel plates or stored in a sealed test tube for further bacterial testing.

3.1.4. Bacterial Assay: Paper Disc Diffusion Test

Bacterial Fouling Organisms

Based on the literature review, *Escherichia coli* and *Staphylococcus* aureus were chosen to test the antibiofouling solution against common bacteria. *E. coli* is ideal as a general fouling organism as it is a gram-negative bacterium which is more common in nature and harder to kill due to its hard cell wall. It is common in many environments, making it relevant to seawater, freshwater and waste-water systems (Seo et al., 2021). *S. aureus* was chosen as a gram-positive bacterium to understand the effectiveness of the solution against a broader range of organisms and the solutions full applicability. *S. aureus* is also used in many other studies on antifouling solutions as it is a common fouling species (Dobretsov et al., 2006; Papa et al., 2015; Long et al., 2021)

Bacterial assay: using agar plate paper disc diffusion assay.

The bacterial paper disc diffusion test was based on the standard proceedings from the literature review (Andrews, 2001). Sterile filter paper and agar plates were obtained, with three plates inoculated with *E. coli* and another three with *S. aureus* according to standard procedure (Andrew, 2001; Schwalbe et al., 2007). Six individual filter paper discs were applied to each agar plate to conduct a maximum of six testing trials per plate, ensuring no contact between effective zones. The controls and test trial antibiofouling solutions were then applied to the filter papers, including the three active efficacy test solutions (200:1, 100:1, and 50:1, the ratio being chitosan: PDMS/capsaicin), two controls **(**a solvent and chitosan control) and an antibiotic standard. Plates were then inverted and incubated at 37°C for approximately 10 h. Following incubation, the inhibition zone of each test filter paper was measured in millimetres.

Visual and statistical analysis

Visual analysis was performed using photographs of the surfaces with the antibiofouling solutions, followed by measurement of the inhibition zone. Statistical analysis was done using an ANOVA in Rstudio to test for significant differences in the inhibition zones of the three efficacy tests (200:1, 100:1, and 50:1, the ratio being chitosan: PDMS/capsaicin), two controls **(**a solvent and chitosan control) and the antibiotic standard.

3.1.5. Proteinaceous Analogue of Amphiphilic Fouling Organisms: Egg White Test

Experimental surface preparation

Mild steel metal plates measuring 5 cm by 10 cm were obtained. Plates were first cleaned with sandpaper, using 120 and 180 grit, to remove particles and increase the surface area for better coating attachment. Subsequently, the plates were cleaned with a gentle dish soap solution (i.e., Dawn™ dish soap) mixed with deionized water on a clean cloth, ensuring the removal of any grease or dust particles that remained from the sandpaper cleaning.

The plates were then cleaned with acetone and then rinsed with deionized water to eliminate any remaining particles and impurities. These steps were repeated a total of five times. After these cleaning steps, the plates were ready for paint application.

Application of antibiofouling coating

A non-active ingredient paint base was applied to the previously prepared mild steel plates using a metal spatula to spread a thin but opaque layer. This was allowed to dry, with the drying time recorded. Subsequently, one of the active ingredient efficacy test trials consisting of the Chitosan-capsaicin-PDMS mixtures (200:1, 100:1, and 50:1) was applied over the top of the base paint in a thin layer. (If drying of base paint does not occur, proceed without it and continue using only the active ingredient portion). This layer was left to dry (~24 h).

Testing against amphiphilic fouling organisms (egg white test)

Egg white (1 mL) was applied to each test trial surface and allowed to dry. Once dry, the adhesion of the egg white was tested, and each test trial was given a grade based on its antibiofouling ability. The literature review informed the grading scale and is as follows (Mahto & Pal, 2020; Barletta et al., 2018; Vesco et al., 2018): Grade 1 indicated that the egg white came off after shaking the test substrate for 30 s. Grade 2 was assigned when the egg white completely came off after brushing for 30 s; Grade 3 was given when less than 25% of the egg white (approximately determined visually) remained on the solution after 30 sec of brushing; and Grade 4 was noted when greater than 25% of the egg white remained after 30 s of brushing (Table 3.1). After grading, results were recorded to determine which concentration of chitosan nanoparticles exhibited the best efficacy.

Table 3.1. Evaluation scale of fouling due to egg white. Grade 1 is assigned to solutions with the most effective anti-adhesion ability, and grade 4 being the least effective. Adapted from: Mahto & Pal (2020); Barletta et al. (2018); Vesco et al. (2018).

Grade	Antifouling ability
1	Clean after shaking
2	Clean after brushing
3	<25% remaining after brushing
4	>25% remaining after brushing

3.2 Results

3.2.1. Base Paint Synthesis

The critical pigment volume concentration (CPVC) of linseed oil and henna was determined to be 0.1 g linseed oil to 1 g powdered henna using the spatula rub-out oil absorption method (Figure 3.1). To reach the desired consistency, 0.15 g of propylene carbonate was added. The henna slowly dissolve out of the linseed oil/propylene carbonate mixture, never fully reaching a smooth consistency without small particle aggregations. Further trials revealed that excluding propylene carbonate from the base paint significantly reduced the dry time in ambient conditions (three months compared to one month), although neither result is ideal for commercial market solutions.

Figure 3.1**.** Millbase (Binder/linseed oil and pigment/henna) paste was created to be combined with a solvent (propylene carbonate) for the desired consistency.

Once the desired consistency was reached, the dry time of the paint was determined through five different drying trials (Table 3.2). The ambient room temperature trial had the longest dry time of three months, followed by the increased airflow trial with a dry time of two months, the heat curing trial with a dry time of one month, and the heating/cooling method with the fastest dry time of two weeks (Figure 3.2). The heat shock trial melted after exposure to a hot plate and had no dry time, as the paint never dried (Table 3.2). After the trials dried, they were tested for abrasion and scratch resistance with a metal spatula. Upon drying, the scratch/abrasion resistance of each trial was virtually the same. The trials resisted flaking or peeling off with moderate to hard force from the constant pressure of the metal spatula. When applying force hard enough to leave marks on the glass slide with the metal spatula, the base paint would peel or flake off from glass and steel substrates.

Table 3.2. Methods for drying the base paint (i.e., linseed oil, henna and propylene carbonate) of antibiofouling solution and the amount of time it took to dry.

Drying Trial	Description	Time result
Trial 1: Ambient room temperature	Drying conditions under a chemical fume hood, 20-22°C	3 months
Trial 2: Heat shock drying	Heated to 40°C and subsequently allowed to dry after in ambient room temperatures.	N/A (melted)
Trial 3: Increased air flow drying	Ambient room temperature air blown directly onto the base paint.	2 months
Trial 4: Heat curing	Constantly heated to 30-35°C and allowed to dry.	1 month
Trial 5: Heating/cooling	Heated to 30-35°C for a day and then left at ambient room temperature for the next day on repeat.	2 weeks

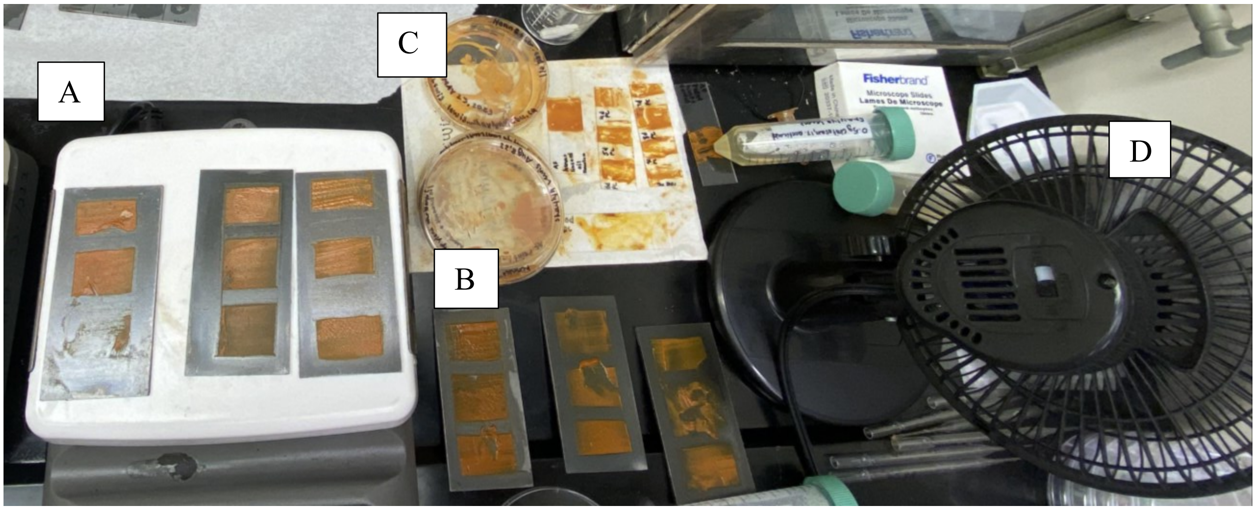

Figure 3.2. Paint drying trials: A- heat/cooling method (heat phase), B- Heat/cooling method (cooling phase), C- Ambient conditions, D- Increased airflow method (out of view).

3.2.2. Synthesizing the Active Antibiofouling Solution

Chitosan to Chitosan Nanoparticles

The synthesis of chitosan nanoparticles was not achieved through the ionic gelation method. Five test trials were performed using this method, all with similar results. Particle size results from dynamic light scattering (DLS) showed that the particle size was in the 10,000 nm range, or in other words, the microparticle range (10 µm). The first trial was burned due to the variable heat from the available hot plate (Figure 3.3). In the following trials, the temperature was consistently monitored throughout the experiment to ensure it did not exceed 80°C (as determined through literature review). The second trial was further adjusted by decreasing the amount of chitosan added to the solution (i.e., from 1 g to 0.5 g), as suggested by further literature review, to test if the high concentration caused the burning. Although the experiment was completed, DLS particle results showed that the size range was still in the micrometer range, indicating further adjustments were needed. In the third trial, 1g of chitosan was used with constant monitoring to see if the concentration from the second trial was insufficient for nanoparticle formation under the ionic gelation method. This trial was completed but had the same results as the second trial, with the particle size range still in the micrometer category rather than the nanometer category. The fourth trial was similar to the third trial, but instead of creating a 1% chitosan nanoparticle solution using 999 mL of deionized-water, a 1% solution was created using 99 mL of di-water. These steps were also followed for the fifth and final trial. Both the fourth and fifth trials were unsuccessful in creating chitosan nanoparticles. Due to the unavailability of advanced equipment capable of performing the ionic gelation method within a short time frame, the nanoparticles were not included in the final active solution test trials and were replaced with the original purchased form from Sigma-Aldrich.

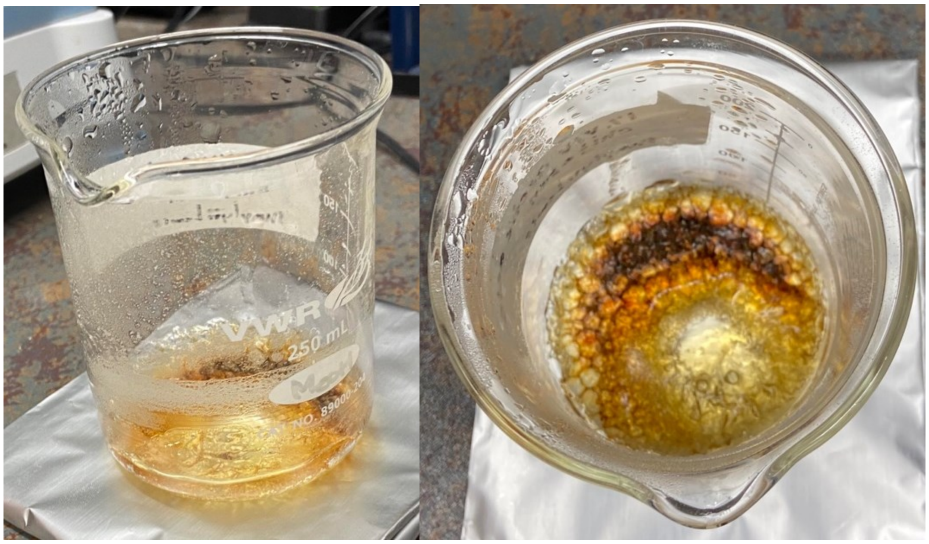

Figure 3.3. First trial synthesising chitosan nanoparticles from chitosan. Trial was over heated and burned, sticking to the glass beaker resulting in an unusable test trial methods were adjusted following this trial. Side (left) and aerial view (right)

Combining Active Ingredients

The active ingredients (i.e., chitosan, capsaicin, and PDMS) were combined to create a hydrogel that could be applied in a layer, forming a film once dry. Each of the three test trials and the chitosan control were prepared using the same method, with varying amounts of chitosan depending on the desired result. This resulted in the formation of a hydrogel film when applied to mild steel plates in each trial (chitosan control, 50:1, 100:1, and 200:1) (Figure 3.4). The chitosan control and the 200:1 test trial displayed the best results. The chitosan control showed the best performance overall, forming a smooth surface once dried. The 200:1 test trial, on the other hand, had a rough surface once the film dried, which is inefficient for moving structures or vessels due to the drag it may create. The other two test trials (50:1 and 100:1) had concerning results, as they appeared to have corroded the mild steel plates after application (Figure 3.4).

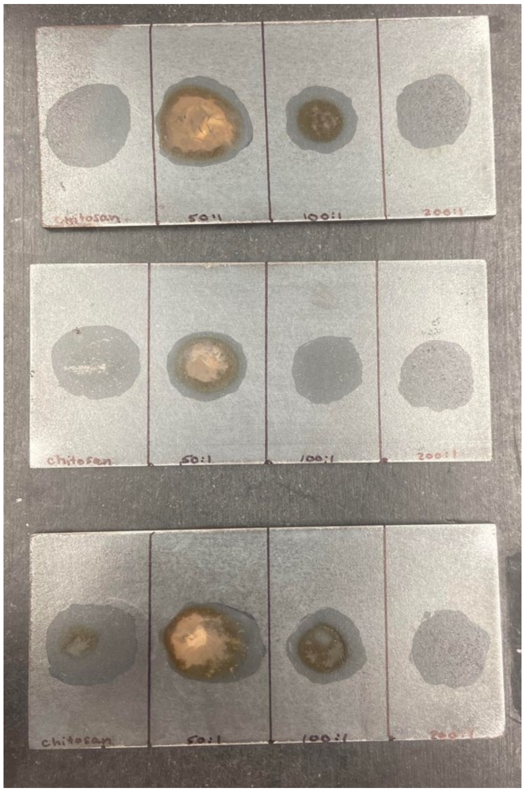

Figure 3.4. First trial application (x3 for each mild steel plate) of active ingredient solution to mild steel test substrate. From left to right (on each of the three plates) chitosan control, test trials 50:1, 100:1 and 200:1.

3.2.3. Paper Disc Diffusion Test

The paper disc diffusion results showed that the three test trial solutions (50:1, 100:1, and 200:1) were not effective at preventing the bacterial growth of *E. coli* or *S. aureus*. This was determined by the lack of inhibition zones surrounding the test trial paper discs (Figure 3.5). The chitosan control (i.e., chitosan/PDMS) and solvent control (i.e., 2-propanol and 1% *v/v* acetic acid) also did not form inhibition zones around their respective paper discs (Figure 3.5). The antibiotic standard (used to determine the effectiveness of the test solutions) was the only one to have an inhibition zone, averaging ~1 cm on *S. aureus* plates and ~6 mm on *E. coli* plates. This result was consistent across the different tests on various plates and bacterial strains (Figure 3.5).

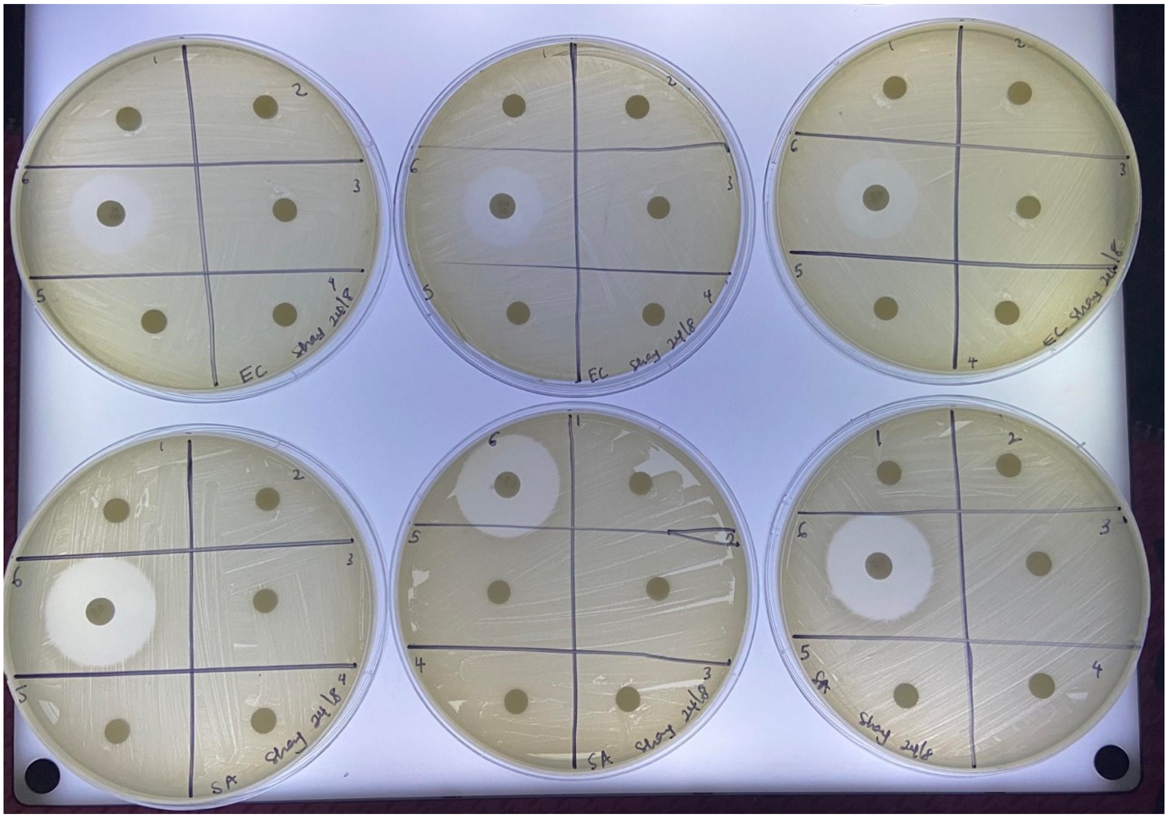

Figure 3.5. Paper disc diffusion results against *S. aureus* (bottom) and *E. coli* (top) of the three chitosan/capsaicin/PDMS test trials (i.e., (1) 50:1, (2)100:1, and (3)200:1), (4) chitosan control, (5) solvent control (i.e., 2-propanyl and 1% *v/v* acetic

3.2.4. Proteinaceous Analogue Test (Egg White Test)

The results from the proteinaceous analogue test, or the egg white (albumin) test, showed that some test trials had better anti-adhesive abilities compared to others (Table 3.3). The chitosan control without the incorporation of capsaicin achieved the best grade out of all the test trials (Table 3.3). The egg white started cracking and flaking off before shaking (Figure 3.6). After 30 sec of shaking, 85% of the egg white was removed, and after 30 sec of brushing, 100% of the egg white was removed, giving this trial Grade 2. The 100:1 test trial had the second-best anti-adhesive results, as it was slightly cracking and flaking off after drying (although more flaking was observed on the untreated surface of the mild steel plate) (Figure 3.6). After 30 sec of shaking, 15% of the egg white was removed, and after 30 sec of brushing, 85% of the egg whites were removed, giving this trial a Grade 3. The last two test trials (50:1 and 200:1) had similar results, with the 200:1 test trial showing slightly less anti-adhesive power. The 50:1 test trial was cracking and slightly flaking after the egg whites had dried (Figure 3.6). After 30 sec of shaking, 35% of the egg white was removed, and after 30 sec of brushing, 65% of the egg whites were removed. The 200:1 test trial result showed that the egg whites were flaking around the perimeter of the solution but not within its limits (Figure 3.6). Less than 10% of the egg whites were removed after 30 sec of shaking, and 65% was removed after 30 sec of brushing.

Table 3.3. Proteinaceous analogue test (egg white test) results of active ingredient solutions (50:1, 100:1 and 200:1) and the chitosan control including test result description.

Trial	Grade	Description
Chitosan solution	2	Egg white cracking and flaking off before shaking, 85% of the egg white was removed after 30s of shaking, 100% of the egg white was removed after 30s of brushing.
50:1	4	Cracked and slightly flaking (less than chitosan solution. 35% of the egg white removed after 30s shaking, 65% of egg whites removed after 30s of brushing.
100:1	3	Slightly cracking (although more flaking was observed on the untreated surface of the mild steel plate), 15% of the egg white was removed after 30s of shaking, 85% of egg whites were removed after 30s of brushing.
200:1	4	Egg whites flaking around the perimeter of solution but not within its limits, <10% of the egg whites were removed after 30s of shaking, 65% was removed after 30s of brushing

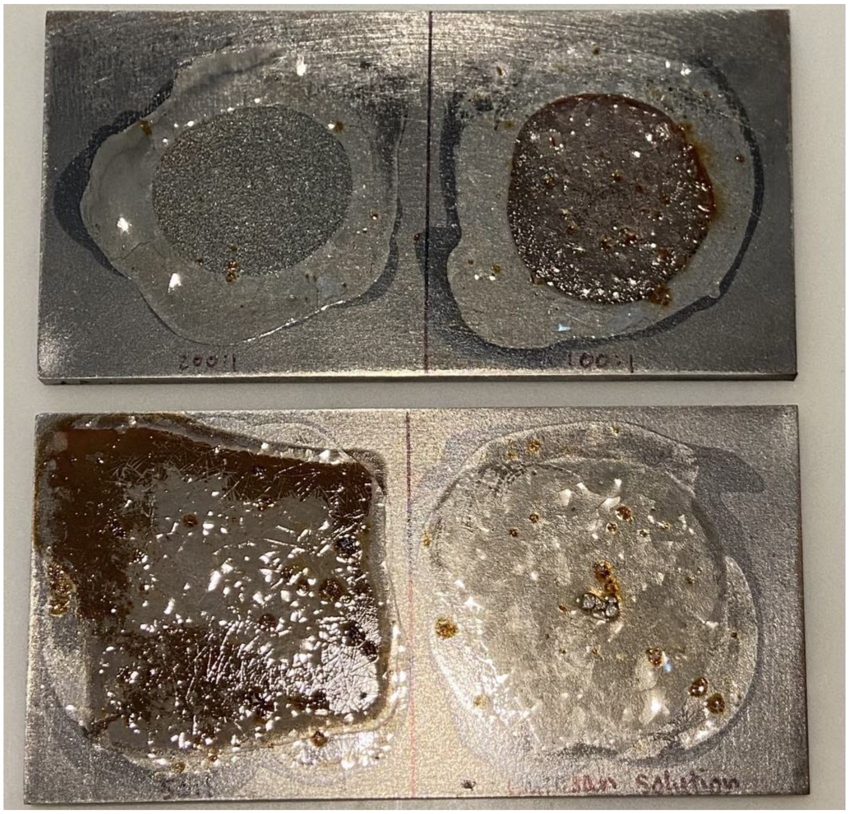

Figure 3.6. Second proteinaceous analogue test (egg white test) with test trials 200:1 (top left), 100:1 (top right), 50:1 (bottom left) and the chitosan control (bottom right) with egg white applied and dried on top after 24h.

3.3. Discussion

The goal of this study was to investigate sustainable substances that could be utilized as active ingredients within an antibiofouling solution. Chitosan, capsaicin, and PDMS were chosen for this purpose. Using these ingredients, an antibiofouling solution was developed to create a sustainably produced, eco-friendly, cost-effective antibiofouling coating suitable for the commercial market. The antibiofouling coating (i.e., chitosan/capsaicin/PDMS) was further tested for antibiofouling characteristics through abrasion, anti-bacterial (paper disc diffusion test), and anti-adhesion testing (egg whites test). To develop a fully environmentally benign antibiofouling solution, a base paint of henna, linseed oil, and propylene carbonate was first developed. The first issue discovered when using this base paint was drying time: the shortest dry time was two weeks (under conditions of being repeatedly heated and cooled) to a total of three months (under ambient room conditions). This is impractical for commercial use, especially on ship hulls that would need to be dry-docked for that time (Almeida et al., 2007). When dry, the paint was not smooth throughout the coated surface. This is not ideal for dynamic/moving systems (e.g., ship hulls), as this "bumpy" surface would most likely increase roughness and ultimately increase drag force and increase fuel consumption, which is the opposite of the intended purpose of an antibiofouling solution (Abioye et al., 2019). Due to the long dry time and the increased surface roughness induced by the developed base paint, it was not included in the final test trials. The base paint could have been improved with additional ingredients such as drying agents and surfactants, but these ingredients would also have to be environmentally friendly to not compromise the integrity of the solution (Gooch, 2002).

Most novel marine antibiofouling studies do not include the development of a base paint, but rather opt for a commercial marine paint (i.e., a paint intended for submerged surfaces without antibiofouling properties), incorporating this with their novel solution or opting out entirely (Kyei et al., 2020; Liu et al., 2022b). This is convenient for researchers as they can focus on the antifouling solution itself, but it becomes a problem when papers claim that their novel solution is environmentally friendly while using a toxic base paint as part of the solution (Armelin et al., 2007; Amara et al., 2018). This raises concerns within sustainable antibiofouling research: the meaning of "environmentally benign." Many marine antibiofouling solutions may be falsely represented as environmentally benign when the full life cycle of the coating is not fully understood (Torres & De-la-Torre, 2021; Turner, 2021). As biocides are public enemy number one in these solutions, they are less of a concern due to the focus on their non-toxic counterparts. The bigger issue is the trend of incorporating naturally derived and/or eco-friendly plastics within antibiofouling solutions (Almeida et al., 2007; Turner, 2021). Plastic pollution is a major environmental concern and has been for many years, but recent research has highlighted the negative impacts of microplastics on the marine environment especially those from marine paints (Turner, 2021; Yang et al., 2021). Therefore, some "environmentally friendly" solutions may not be as friendly as they seem and may require further research to fully understand the environmental impact, using environmental management tools, such testing as a Life Cycle Analysis (LCA).

The active ingredients selected for this study's potential novel antibiofouling solution were chitosan (nanoparticles), capsaicin, and PDMS, which were combined to create a lubricant-like hydrogel film intended to prevent biofouling. Results indicated that the three active ingredient solution test trials (200:1, 100:1, and 50:1 ratio of chitosan to capsaicin) were not effective antibiofouling coatings. The results of the three active solutions were comparable in both the paper disc diffusion test and the proteinaceous analogue test. The paper disc diffusion test results showed that the trials were virtually identical, as none of the test trials were successful in preventing the growth of the two chosen bacteria (*E. coli* and *S. aureus*) (Chapter 4 Figure 3.5). This could be due to the concentrations of the three solutions being too low to prevent the growth of these organisms, necessitating increased concentrations in future studies. This test may also not provide enough evidence as to whether the novel hydrogel has antibacterial properties, as the purpose of the hydrogel is to prevent growth in the form of a dried film, but the test only shows the prevention of growth of its surroundings, not within its boundaries. A similar study by Qin et al. (2023) developed a novel epoxy poly(dimethylsiloxane)/capsaicin antifouling coating with antibacterial abilities, which was tested using a submersion simulation. They submerged the coating into a bacterial (*Paracoccus pantotrophus*) suspension and determined the cell count using a LIVE/DEAD staining kit. This test would help determine more conclusively if the coating developed in the present study has antibacterial abilities better than the paper disc diffusion test. The bacteria used for the paper disc diffusion test may not have been the ideal candidates for testing an antibiofouling solution. Previous studies that have developed novel antibiofouling coatings have used *E. coli* and *S. aureus* as their test bacteria due to their cosmopolitan distribution (Zhao et al., 2019; Seo et al., 2021). However, these may not be appropriate generalist bacteria to use, as they may not represent the first colonizers in a marine biofouling succession scenario (Rampadarath et al., 2017; Khanzada et al., 2020). Marine diatom species could be more representative of the first settling organisms and should be involved in future antifouling testing.

This issue highlights one of the main problems in antibiofouling research, which is the lack of standardized testing for the effectiveness of novel antifouling solutions (Rittschof, 2009; Gu et al., 2020). Although biofouling is so diverse that standardized methods may not represent the entire global ocean, there still could be select tests included in novel antibiofouling studies as a starting point for current researchers and for future comparability. The recommended tests would include submersion suspension testing with bacteria and algae (diatoms) in phosphate-buffered saline (e.g., providing a protein source for the bacteria's survival as well as further protein absorption testing), followed by LIVE/DEAD staining, abrasion/adhesion tests (i.e., to test the robustness and mechanical strength of the novel coating), field testing within conditions of the intended environment, and toxicity testing (e.g., filter-feeding bivalves) to understand how the novel antibiofouling coating may impact the surrounding environment and ecosystem (Briand, 2009). Including all the previously mentioned tests would give a robust understanding of the coating's antifouling ability and performance under real-world conditions (Gu et al., 2020). However, these tests can be expensive and time-consuming (i.e., field testing >2 months minimum) (Rittscof, 2009). This may be why many studies on novel marine antibiofouling solutions do not include all the suggested tests, as it is not within the means of every individual research group. Nevertheless, if these tests were implemented as a standard, the quality of these novel solutions would be better monitored and reviewed for commercial potential (Rittscof, 2009).

The proteinaceous analogue test showed slightly differing results between the trials. The 100:1 test trial achieved Grade 3 (i.e., <25% remaining after brushing), which was the highest of the test trials (i.e., 200:1 and 50:1 both achieved Grade 4). This suggests that the moderate concentration of chitosan has the best anti-adhesion ability of the three test trials (i.e., it is best at preventing the attachment of fouling organisms). However, it must be noted that the chitosan control (i.e., chitosan/PDMS minus capsaicin) performed the best, achieving Grade 2 (i.e., clean after 30 sec of brushing) in the test. This is noteworthy as it may suggest that the addition of capsaicin within a silicone-based antifouling solution could decrease its anti-adhesive ability, but further research with increased concentrations is needed for more comprehensive and conclusive results. Previous studies have found that incorporating capsaicin within their antibiofouling solutions results in excellent antifouling abilities (i.e., anti-bacterial, anti-diatom, and biodegradable) at low concentrations. However, they also found that the pungency of capsaicin may only be effective at the microorganism scale, as the settlement of macroorganisms (e.g., barnacle and mussel larvae) has been observed on capsaicin-containing coatings under real-world conditions (Peng et al., 2011; Wang et al., 2014b). This suggests that this study's solution (chitosan/capsaicin/PDMS hydrogel) should have prevented the growth of bacteria, and field testing of the solution may have shown the true antibacterial behavior of the hydrogel. The antibacterial test (i.e., paper disc diffusion) selected for this study only displayed the surrounding effects of the solution rather than the settlement of bacterial organisms directly on the coating's surface. The anti-adhesive/lubricant-like abilities of PDMS are also not displayed during the paper disc diffusion test, as the settlement of microorganisms could not be observed. Future research involving field testing is suggested to address this issue.

The ineffectiveness of these solutions in the antibacterial and anti-adhesion tests may be due to the form of chitosan used. Initially, chitosan nanoparticles were intended to be incorporated within the active solutions, but they were replaced with microscale particles of chitosan due to the unavailability of the nanoparticle form. Chitosan nanoparticle synthesis was attempted in this study using the ionic gelation method but was unsuccessful. This suggests that more trials are needed with high-performing equipment in a timely manner, as the methods and materials used were variable and time-consuming (e.g., inconsistent hot plate temperatures and equipment availability). The ionic gelation method requires an experienced person with proper laboratory training, which may have contributed to the lack of formation due to inexperience; this can be addressed with further trials. Previous studies suggest that chitosan, regardless of size, should have been successful in preventing bacterial growth (Kumar et al., 2019; Liu et al., 2021). This places limitations on the interpretation of the results of this study but suggests that the concentrations used were overall insufficient in preventing bacterial growth and would need to be increased in further research (Ghiggi et al., 2017). Many studies that use chitosan as an active ingredient in their antibiofouling solutions are for biomedical materials and membrane surfaces (e.g., water treatment systems) rather than robust protective coatings for moving surfaces (e.g., ship hulls) or dynamic surfaces (e.g., surfaces in tidal conditions) (Roux et al., 2005; Kumari et al., 2021; Teixeira-Santos et al., 2021). When chitosan is used in the latter, it is typically combined with a biocide (e.g., ZnO, TiO_2, and Ag) for a more effective solution (Al-Naamani et al., 2017; Natarajan et al., 2018; Pourhashem et al., 2022). This indicates that chitosan must be combined with a more effective antifouling substance to deter or prevent the attachment of biofouling organisms. Future antibiofouling research interested in using chitosan as an active ingredient should focus on incorporating it within solutions intended for biomedical applications (e.g., implants) and/or filtering systems (e.g., ultrathin membranes) rather than dynamic/moving systems (Ghiggi et al., 2017; Pourhashem et al., 2022). Chitosan should still be pursued as an antifouling ingredient for these instances, as it is a large untapped resource from the waste produced by invertebrate fishing industries and would help support a circular economy with its utilization as an antifouling substance (Junceda-Mena et al., 2023).

3.4. Conclusion

This study explored the potential of chitosan, capsaicin, and PDMS as active ingredients in an environmentally friendly antibiofouling solution. Despite the development of a hydrogel film intended to prevent biofouling, the tests— anti-abrasion, antibacterial (paper disc diffusion), and anti-adhesion (egg whites test)—revealed the solution's limitations. The primary issues included inadequate concentrations of active ingredients and the inappropriate choice of bacterial testing, highlighting the necessity for standardized tests in the field of antibiofouling research in future studies. The development of the base paint using henna, linseed oil, and propylene carbonate proved impractical due to long drying times and an uneven surface, making it unsuitable for commercial use. This underscores the broader challenge in antibiofouling research: the need to ensure that all components of the coating, including the base paint, are truly environmentally benign and effective. This study also raises critical questions about the current definitions of "environmentally friendly" solutions, suggesting that many novel coatings may not be as benign as claimed, necessitating comprehensive LCA to assess their true environmental impact. The findings also suggest that while chitosan, capsaicin, and PDMS show promise, their combination in the tested ratios did not produce effective antibiofouling properties. The use of chitosan in its microscale form, instead of nanoparticles, may have compromised the results, emphasizing the need for further refinement in synthesis methods and concentration levels. Additionally, future research should consider alternative testing protocols, including submersion tests with bacteria and diatom species, abrasion/adhesion tests, toxicity tests and field trials, to better simulate real-world conditions and evaluate the long-term effectiveness of these coatings.

Given the challenges encountered, future efforts should focus on refining the formulation of chitosan-based coatings for biomedical and filtration applications, where its properties can be better utilized. Moreover, integrating chitosan with more potent antifouling agents, while maintaining environmental integrity, could enhance its efficacy. This approach aligns with the principles of a circular economy, leveraging waste products from invertebrate fishing industries to develop sustainable antifouling solutions, although scaling up the use of chitosan may reduce its sustainability due to production processes. In conclusion, while the current study did not achieve its goals, it lays a foundation for future research aimed at developing effective, environmentally friendly antibiofouling technologies. Continued exploration, innovative formulations, and rigorous testing are essential to advancing this field and achieving sustainable solutions for marine biofouling control.

Chapter 4: Conclusion

4.1 Knowledge Gaps and the Future of Antibiofouling Research

A key theme emerging from recent literature is the importance of synergy in the development of antibiofouling technologies. Combining multiple antifouling properties—such as biocides, self-polishing substances, zwitterions, hydrophobic and hydrophilic characteristics, amphiphilic behavior, and altered surface topography—can create more robust and effective solutions. The future of antibiofouling will likely depend on integrating these multifaceted strategies, leveraging the strengths of different approaches to achieve optimal performance without adverse environmental impacts.

Future research should focus on advancing non-toxic biocides derived from natural renewable sources, developing novel materials with robust antifouling capabilities, and enhancing the mechanical robustness and long-term efficacy of future coatings. The literature review revealed that photocatalysts and electrochemical repulsion techniques also hold promise, though they require further research to address limitations related to environmental conditions and power requirements. Ultimately, the next steps in antibiofouling research should prioritize field testing and real-world application assessments to ensure the practicality and environmental safety of novel solutions. Collaborative efforts between scientists, industry stakeholders, and regulatory bodies will be essential in driving the development and adoption of innovative, sustainable antibiofouling technologies that effectively protect marine environments and human interests alike.

4.2 Addressing the Gaps

The experimental study aimed to investigate sustainable substances for use as active ingredients in an antibiofouling solution. Chitosan, capsaicin, and polydimethylsiloxane (PDMS) were chosen for their potential antibiofouling properties. The developed solution was intended to be sustainable, eco-friendly, and cost-effective, suitable for commercial market applications. Various tests, including abrasion, anti-bacterial (paper disc diffusion), and anti-adhesion tests (egg white test), were conducted to evaluate the solution's efficacy. However, the novel chitosan/capsaicin/PDMS combination was found ineffective as an antibiofouling coating.

One significant challenge identified was the development of a suitable base paint. Initial attempts using a base paint of henna, linseed oil, and propylene carbonate resulted in impractical drying times and surface roughness, rendering it unsuitable for commercial use. Most novel marine antibiofouling studies do not include the development of a base paint, often opting for commercial marine paint instead. This approach can be problematic when novel solutions are claimed to be environmentally friendly but use toxic base paints. This issue raises concerns about the definition of "environmentally benign" in sustainable antibiofouling research. Many novel solutions may be misrepresented as environmentally friendly when the full life cycle of the coating is not fully understood. The use of plastics, even those labeled as eco-friendly, contributes to plastic pollution, particularly microplastics from marine paints. Therefore, further research is needed to fully understand the environmental impact of these solutions, potentially utilizing tools such as Life Cycle Analysis (LCA).

The lack of standardized testing for the effectiveness of novel antifouling solutions is a significant issue in antibiofouling research. While standardized methods may not represent the entire global ocean, including certain tests as a starting point could improve comparability and understanding of novel solutions. Recommended tests include submersion suspension testing with bacteria and algae, LIVE/DEAD staining, abrasion/adhesion tests, field testing in the intended environment, and toxicity testing. Although these tests can be expensive and time-consuming, they would provide a robust understanding of the coating's antifouling ability and performance under real-world conditions. In conclusion, the journey towards developing effective, environmentally friendly antibiofouling solutions is complex and ongoing. The integration of multifaceted strategies, collaborative efforts, and standardized testing will be crucial in advancing this field. Continued exploration of sustainable substances and innovative approaches will help protect marine environments and support sustainable maritime practices.

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Appendix 1: Supplementary Material

Supplementary Table S1. The 100 marine antibiofouling studies reviewed for the semi-systematic literature review.

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Ali, A., Xiao, Y., Song, L., Hu, J., Rao, Q., Shoaib, M., . . . Zhang, Q. (2021). Biodegradable polyurethane-based clay composite and their anti-biofouling properties. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 625 doi:10.1016/j.colsurfa.2021.126946	Colloids and Surfaces A: Physicochemical and Engineering Aspects	CL-PU/DCOIT/clay composite	biodegradable polyurethane, ε-CL and 4,4′-methylenebis(cyclohexyl isocyanate) (H12MDI), 1,4 butanediol (1,4 BD), 4, 5-dicholoro-2-octyl-isothiazolone (DCOIT)	Soluble matrix solution with non-toxic biocide	Anti-bacterial, anti-diatom, degrades in artificial seawater (clay determines rate of release)	Needs more field testing
Atthi, N., Sripumkhai, W., Pattamang, P., Thongsook, O., Srihapat, A., Meananeatra, R., Supadech, J., Klunngien, N., & Jeamsaksiri, W. (2020). Fabrication of robust PDMS micro-structure with hydrophobic and antifouling properties. Microelectronic Engineering, 224, 111255. https://doi.org/10.1016/j.mee.2020.111255	Microelectronic Engineering	circular rings with eight stripe supporters (C-RESS) PDMS	PDMS with C-RESS pattern	Hydrophobicity low surface energy	Anti-algal, anti-larval	Needs biocide for functionality
Barletta, M., Aversa, C., Pizzi, E., Puopolo, M., & Vesco, S. (2018). Design, manufacturing and testing of anti-fouling/foul-release (AF/FR) amphiphilic coatings. Progress in Organic Coatings, 123, 267–281. https://doi.org/10.1016/j.porgcoat.2018.07.016	Progress in Organic Coatings	Silanes and polyeurethanes using sol-gel reactions	fluorinate-alkyl bearing silanes (FTSi), poly-siloxane backbones, PEG-ilatechain silanes, isocyanates	Amphiphilic antifoul/foul release	successful antifoul/ foul release system, amphiphilic	Field testing needed, longevity unknown

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Elmas, S., Gedefaw, D. A., Larsson, M., Ying, Y., Cavallaro, A., Andersson, G. G., . . . Andersson, M. R. (2020). Porous PEI coating for copper ion storage and its controlled electrochemical release. Advanced Sustainable Systems, 4(3) doi:10.1002/adsu.201900123	Advanced Sustainable Systems	cross-linked polyethylene imine (PEI) coated on conducting carbon cloth electrodes with copper biocide	copper, cross-linked PEI	soluble matrix solution with biocide and controlled electrochemical release	controlled rate of release of biocide, effective antifoulant	Non-environmentally friendly use of copper
Ghattavi, S., Homaei, A., Kamrani, E., Saberi, D., & Daliri, M. (2023). Fabrication of antifouling coating based on chitosan-melanin hybrid nanoparticles as sustainable and antimicrobial surface. Progress in Organic Coatings, 174, 107327. https://doi.org/10.1016/j.porgcoat.2022.107327	Progress in Organic Coatings	Chitosan-melanin hybrid nanoparticle complex	chitosan nanoparticles, melanin nanoparticles	Nanoparticle biocides	Anti-bacterial, anti-algal, anti-invertebrate, cost-effective, biocompatible and long-lasting	Further field testing needed to understand application
Kumar, S., Ye, F., Mazinani, B., Dobretsov, S., & Dutta, J. (2021). Chitosan nanocomposite coatings containing chemically resistant zno–snox core shell nanoparticles for photocatalytic antifouling. International Journal of Molecular Sciences, 22(9), 4513. https://doi.org/10.3390/ijms22094513	International Journal of Molecular Sciences	Chitosan-based antifouling coatings with varying percentages of ZnO or ZnO–SnOx nanoparticles, with or without the glutaraldehyde (GA) crosslinking of chitosan and a photocatalyst	biopolymer chitosan, ZnO-SnOx (core-shell) nanoparticles, glutaraldehyde	Soluble matrix solution Nanoparticle biocides	Anti-microbial, anti-diatom photo stabilized	use of metal as biocide (toxic), field testing needed

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Liu, J., Sun, J., Duan, J., Dong, X., Wang, X., Liu, C., & Hou, B. (2022). Capsaicin-modified Fluorosilicone-based acrylate coating for marine anti-biofouling. Coatings, 12(7), 988. https://doi.org/10.3390/coatings12070988	Coatings	capsaicin-modified marine antifouling organofluorosilicone which is based on silicone and fluorine acrylate monomers covalently bound to an organic antimicrobial monomer, HMBA (N-(4-hydroxy-3-methoxybenzyl)-acrylamide) on a polymer network.	HMBA-containing polyacrylic resin, chitosan, Vanillylamine hydrochloride, PDMS, acrylates	low surface energy, soluble matrix with non-toxic biocide	good adhesion, long-lasting, anti-bacterial, anti-algal	Field testing needed
Lu, Z., Chen, Z., Guo, Y., Ju, Y., Liu, Y., Feng, R., Xiong, C., Ober, C. K., & Dong, L. (2018). Flexible hydrophobic antifouling coating with oriented nanotopography and nonleaking capsaicin. ACS Applied Materials & Interfaces, 10(11), 9718–9726. https://doi.org/10.1021/acsami.7b19436	ACS Applied Materials & Interfaces	capsaicin bonded to CoFe2O4/gelatin magnetic nanoparticles was mixed with a polydimethylsiloxane (PDMS)-based block copolymer	Capsaicin, PDMS, CoFe2O4/gelatin nanospheres, polystyrene-block-poly(dimethylsiloxane-stat-vinylmethylsiloxane) (PS-b-P(DMS-stat-VMS) or PSDV)	Nanoparticle biocide, low surface energy, nanorough topography	easy spray application, anti-bacterial,	Field testing needed
Periyasamy, Raorane, C. J., Haldhar, R., Asrafali, S. P., & Kim, S.-C. (2022). Development of arbutin-based sustainable polybenzoxazine resin for antifouling and anticorrosion of low-carbon steel. Progress in Organic Coatings, 170. https://doi.org/10.1016/j.porgcoat.2022.106968	Progress in Organic Coatings	polybenzoxazine/co-polymer coatings (PAB/BX) containing arbutin and silane	bio-based benzoxazine (arbutin, [3-(2-aminoethylamino) propyl]trimethoxysilane (AEAPTMS) and paraformaldehyde), hydrophilic co-polymer, i.e., PEG-PPG-PEG,	Hydrophobic (creating a barrier through the cross-linking)	Anti-corrosive, good adhesion, anti-bacterial	Field testing needed

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Quémener, M., Kikionis, S., Fauchon, M., Toueix, Y., Aulanier, F., Makris, A. M., Roussis, V., Ioannou, E., & Hellio, C. (2021). Antifouling activity of halogenated compounds derived from the red alga Sphaerococcus coronopifolius: Potential for developing Environmentally Friendly Solutions. Marine Drugs, 20(1), 32. https://doi.org/10.3390/md20010032	Marine Drugs	red alga (Sphaerococcus coronopifolius) 15 metbolites (sphaerococcinol A and 14R-hydroxy-13,14-dihydro-sphaerococcinol A)	Specimens of S. coronopifolius	non-toxic biocides	Anti-larval, anti-bacterial Environmentally friendly, renewable	Needs further incorporation within an antibiofouling solution matrix
Seo, E., Seong, M. R., Lee, J. W., Lim, H., Park, J., Kim, H., Hwang, H., Lee, D., Kim, J., Kim, G. H., Hwang, D. S., & Lee, S. J. (2020). Anti-biofouling features of eco-friendly oleamide–PDMS copolymers. ACS Omega, 5(20), 11515–11521. https://doi.org/10.1021/acsomega.0c00633	ACS Omega	oleamide–PDMS copolymer (OPC)	PDMS, Oleamide (fatty acid)	low surface energy, hydrophobic	Anti-algal, anti-invertebrate, drag-reduction	needs other antibiofouling mechanisms incorporated for robustness
Seo, E., Lee, J. W., Lee, D., Seong, M. R., Kim, G. H., Hwang, D. S., & Lee, S. J. (2021). Eco-friendly erucamide–polydimethylsiloxane coatings for marine anti-biofouling. Colloids and Surfaces B: Biointerfaces, 207, 112003. https://doi.org/10.1016/j.colsurfb.2021.112003	Colloids and Surfaces B: Biointerfaces	erucamide-polydimethylsiloxane (EP) coating	PDMS, Erucamide (fatty acid)	low surface energy, hydrophobic	Anti-bacterial, anti-algal, field-tested longevity (5.5 months), drag reduction	Best application for vessel needing drag reduction, further field testing needed to understand the appropriate application

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Sun, L., Wang, W. -., Li, M., Zhang, J., Chai, T., Cao, W., . . . Zhou, F. (2022). Natural product zanthoxylum bungeanum based multi-functionalized self-polishing interface for sustainable marine antifouling. Advanced Materials Interfaces, doi:10.1002/admi.202201195	Advanced Materials Interfaces	triclosan was co-polymerized with general acrylic molecules to fabricate a self-polishing resin by free radical polymerization	Acrylic resin, pulverized Z. bungeanum seeds	Self-polishing with non-toxic biocide, hydrophobic	Anti-algal, anti-diatom, tough mechanical properties	Field research needed, longevity unknown
Wang, D., Chen, Y., Huang, Y., Bai, H., Tan, Y., Gao, P., . . . Jiang, L. (2022a). Universal and stable slippery coatings: Chemical combination induced adhesive-lubricant cooperation. Small, 18(32) doi:10.1002/smll.202203057	Small	bio-inspired adhesive polydopamine (PDA) adhered to substrates and chemically bonded with liquid-like lubricant PDMS	polydopamine, PDMS	Low surface energy	highly versatile and suitable for most applications (beyond marine environment),	needs other antibiofouling mechanisms incorporated for robustness, further research on marine antibiofouling needed

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Zhang, H., Li, Y., Tian, S., Qi, X., Yang, J., Li, Q., Lin, C., Zhang, J., & Zhang, L. (2022). A switchable zwitterionic ester and capsaicin copolymer for Multifunctional Marine antibiofouling coating. Chemical Engineering Journal, 436, 135072. https://doi.org/10.1016/j.cej.2022.135072	Chemical Engineering Journal	PDMS-based coatings grafted with different contents of hydrolyzable EB and capsaicin copolymer	Dibutyltin dilaurate (DBTDL), 2-mercaptoethanol (98%), (3-Isocyanatopropyl)-triethoxysilane (IPTS), methyltriacetoxysilane (METES), dihydroxyl-terminated polydimethylsiloxane (HO-PDMS-OH, MW = 4200 g mol−1), 2,2′-Azoisobutyronitrile (AIBN), N-2-(Dimethylamino) ethyl methacrylamide (DMA, 99%, stabilized with 4-methoxyphenol)	Self-polishing with non-toxic biocide, Low surface energy, switchable zwitterionic esters	Anti-bacterial, anti-algal, long-lasting (261 day field test)	Further research needed to understand marine applications, use of metals
Sánchez-Lozano, I., Hernández-Guerrero, C. J., Muñoz-Ochoa, M., & Hellio, C. (2019). Biomimetic approaches for the development of New Antifouling Solutions: Study of incorporation of macroalgae and sponge extracts for the development of new environmentally friendly coatings. International Journal of Molecular Sciences, 20(19), 4863. https://doi.org/10.3390/ijms20194863	International Journal of Molecular Sciences	naturally derived non-toxic extracts from 5 macroalgae and 2 sponges (successfully from: Laurencia gardneri, Sargassum horridum (macroalgae), Haliclona caerulea and Ircinia sp. (sponges))	LgEt (Laurencia gardneri ethanol), LgDC (Laurencia gardneri CH2Cl2), UlEt (Ulva lactuca ethanol), UlDC (Ulva lactuca CH2Cl2), CfEt (Codium fragile ethanol), CfDC (Codium fragile CH2Cl2), ShEt (Sargassum horridum ethanol), ShDC (Sargassum horridum CH2Cl2), GvEt (Gracilaria vermiculophylla ethanol), GvDC (Gracilaria	non-toxic biocide	Anti-bacterial, environmentally friendly	may not be economically feasible, needs other antibiofouling mechanisms incorporated for robustness

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
			vermiculophylla CH2Cl2), Hc (Haliclona caerulea) and Isp (Ircinia sp.)			
Pan, J., Ai, X., Ma, C., & Zhang, G. (2022). Degradable vinyl polymers for combating marine biofouling. *Accounts of Chemical Research*, *55*(11), 1586–1598. https://doi.org/10.1021/acs.accounts.2c00187	Accounts of Chemical Research	hybrid copolymerization of degradable vinyl polymers	degradeable vinyl polymers, polyurethane	self-polishing copolymer	Long-lasting, environmentally friendly, broad spectrum usage, no microplastics	needs other antibiofouling mechanisms incorporated for robustness

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Feng, D. Q., He, J., Chen, S. Y., Su, P., Ke, C. H., & Wang, W. (2018). The plant alkaloid camptothecin as a novel antifouling compound for marine paints: Laboratory bioassays and field trials. Marine Biotechnology, 20(5), 623–638. https://doi.org/10.1007/s10126-018-9834-4	Marine Biotechnology	18 alkaloids frm terrestrial plants examined, 5 being effective, with camptothecin (CPT) being the most effective	evodiamine, CPT, cepharanthine, sinomenine, and strychnine	non-toxic biocide	Anti-algal, anti-invertebrate	may accumulate in the environment (CPT) harmful to off-target species
Tian, L., Yin, Y., Jin, H., Bing, W., Jin, E., Zhao, J., & Ren, L. (2020). Novel marine antifouling coatings inspired by corals. Materials Today Chemistry, 17, 100294. https://doi.org/10.1016/j.mtchem.2020.100294	Materials Today Chemistry	AF developed from corals using their 5 strategies against biofouling: natural antifoulants, foul release effect, sloughing effect, soft tentacles and fluorescent effect	sesquiterpenes, steroids, alkaloids, and diterpenes	non-toxic biocide soluble matrix	environmentally friendly, broad spectrum	Further development is needed
Dai, Z., Cao, M., Li, S., Yao, J., Wu, B., Wang, Y., Wang, H., Dong, J., & Yi, J. (2021). A novel marine antifouling coating based on a self-polishing zinc-polyurethane copolymer. Journal of Coatings Technology and Research, 18(5), 1333–1343. https://doi.org/10.1007/s11998-021-00496-8	Journal of Coatings Technology and Research	zinc-polyurethane copolymers	self-polishing zinc-polyurethane copolymers, which contained zinc atom in the form of a polymeric salt	Self-polishing	Long-lasting, controlled release	Further field testing needed to understand the application
Xie, C., Guo, H., Zhao, W., & Zhang, L. (2020). Environmentally friendly marine antifouling coating based on a synergistic strategy. Langmuir, 36(9), 2396–2402. https://doi.org/10.1021/acs.langmuir.9b03764	Langmuir	Antifouling based on synergistic strategy:	copolymer polyacrylates, tert-butyldimethylsilyl methacrylate (TBSM), eugenol methacrylate (EM), and poly(N-vinylpyrrolidone) (PVP)	self-polishing, hydrophilic, non-toxic, biocide	Anti-bacterial, anti-diatom long-lasting (8 months in the field), prevents protein absorption	Further field testing needed to understand application

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Mo, Y., Xue, P., Xing, J., Liu, H., Wang, J., & Liu, J. (2022). Conventional silicone coating improved by novel bioinspired microcavity arrays: Generating stable air bubble arrays for gas-liquid interface antifouling. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 644, 128887. https://doi.org/10.1016/j.colsurfa.2022.128887	Colloids and Surfaces A: Physicochemical and Engineering Aspects	Bioinspired biofouling based on the concave structure of springtail and the hairy structure of Salvinia	PDMS, polystyrene	gas-liquid biphase interface generated by the air bubble arrays (physical deterrent), microcavity structure under water pressure	Made of conditions of static water, anti-bacterial, anti-diatom, hydrophobic	Not applicable to moving vehicles, further research needed to understand full application ability
Zhang, Y., Hu, H., Pei, X., Liu, Y., Ye, Q., & Zhou, F. (2017). Polymer brushes on structural surfaces: A novel synergistic strategy for perfectly resisting algae settlement. Biomaterials Science, 5(12), 2493–2500. https://doi.org/10.1039/c7bm00842b	Biomaterials Science	polymer brushes on a wrinkled silicone elastomer. (via surface-initiated atom transfer radical polymerization)	Polymer brushes (POEGMA and PSPMA), PDMS	low surface energy, altered surface topography	Anti-algal, environmentally friendly	Further field testing is needed for longevity
Feng, K., Li, X., & Yu, L. (2018). Synthesis, antibacterial activity, and application in the antifouling marine coatings of novel acylamino compounds containing gramine groups. Progress in Organic Coatings, 118, 141–147. https://doi.org/10.1016/j.porgcoat.2017.10.027	Progress in Organic Coatings	acylamino compounds containing gramine groups	Allylamine, n-octylamine and 5-bromo-1H-indole,	non-toxic biocide	Anti-bacterial, environmentally friendly	Further development and field testing are needed

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Hölken, I., Hoppe, M., Mishra, Y. K., Gorb, S. N., Adelung, R., & Baum, M. J. (2016). Complex shaped zno nano- and microstructure based polymer composites: Mechanically stable and environmentally friendly coatings for potential antifouling applications. Phys. Chem. Chem. Phys., 18(10), 7114–7123. https://doi.org/10.1039/c5cp07451g	Phys. Chem. Chem. Phys.	Solvent-free polymer/particle-composite coating based on two-component polythiourethane (PTU) and tetrapodal-shaped ZnO (t-ZnO) nano- and microstructures	polyurethane, sulphur-containing polyol (polythiourethane, PTU), t-ZnO	Nanoparticle biocides	cost-effective, strong mechanical solid properties, spray coating, environmentally friendly, long-lasting	Further development and field research is needed
Lenchours Pezzano, J., Rodriguez, Y. E., Fernández-Gimenez, A. V., & Laitano, M. V. (2024). Exploring fishery waste potential as an antifouling component. Environmental Science and Pollution Research, 31(13), 20159–20171. https://doi.org/10.1007/s11356-024-32491-y	Environmental Science and Pollution Research	enzymatic extracts from fishery residues for antifouling potential	Extracts from Pleoticus muelleri shrimp, Illex argentinus squid, and Lithodes santolla king crab	non-toxic biocides	environmentally friendly, cost-effective, circular economy	Further development needed
Zanje, S. R., Jain, I., & Dhar, S. (2017). Design and analysis of novel antifouling device (AFD) by FEM. SSRN Electronic Journal. https:// doi.org/10.2139/ssrn.3101370	SSRN Electronic Journal.	A scalable device consisting of a durable set of components forming a movable chain-like structure around offshore structures	floating body antifouling device (AFD)	abrasion (rubbing device)	environmentally friendly, cost-effective	Further development needed, only for static objects

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Shao, C.-L., Xu, R.-F., Wang, C.-Y., Qian, P.-Y., Wang, K.-L., & Wei, M.-Y. (2015). Potent antifouling marine dihydroquinolin-2(1h)-one-containing alkaloids from the gorgonian coral-derived fungus scopulariopsis sp.. Marine Biotechnology, 17(4), 408–415. https://doi.org/10.1007/s10126-015-9628-x	Marine Biotechnology	marine natural alkaloids (1–6) were isolated from the gorgonian coral-derived fungus Scopulariopsis sp	six dihydroquinolin-2-one-containing alkaloids, three monoterpenoids combined with a 4-phenyl-3,4-dihydroquinolin-2(1H)-one (1–3) and three 4-phenyl-3,4-dihydroquinolin-2(1H)-one alkaloids (4–6), were isolated from the gorgonian coral-derived fungus Scopulariopsis sp.	non-toxic biocides	Anti-larval, anti-Environmentally friend	Further development needed
Mei, L., Ai, X., Ma, C., & Zhang, G. (2020). Surface-fragmenting hyperbranched copolymers with hydrolysis-generating zwitterions for antifouling coatings. Journal of Materials Chemistry B, 8(25), 5434–5440. https://doi.org/10.1039/d0tb00886a	Journal of Materials Chemistry B	surface-fragmenting hyperbranched copolymer with tertiary carboxybetaine ester (TCB) primary chains and poly(ε-caprolactone) (PCL) bridged chains, where the former and the latter can hydrolyze and degrade in marine environments, continuously generating zwitterions, so the polymer coating has a fouling resistant and renewable surface.	hyperbranched copolymer via RAFT polymerization of divinyl-functionalized poly(ε-caprolactone) (PCL-V2), tertiary carboxybetaine ester (TCB), and methyl methacrylate (MMA)	self-polishing (zwitterions)	protein resistant anti-bacterial, controlled degradation rate	Field testing needed

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Yu, Y., Xu, N., Zhu, S., Qiao, Z., Zhang, J., Yang, J., & Liu, W. (2021). A novel Cu-doped high entropy alloy with excellent comprehensive performances for Marine Application. Journal of Materials Science & Technology, 69, 48–59. https://doi.org/10.1016/j.jmst.2020.08.016	Journal of Materials Science & Technology	Cu-doped high entropy alloy for marine application	AlCoCrFeNiCu0.5 alloy, AlCoCrFeNi HEA powders	Biocide (copper)	Anti-bacterial, anti-algal, anti-diatom, superior to market copper paints	toxic at high concentrations
Zhang, Z., Liu, Y., Feng, M., Wang, N., Du, C., Peng, S., Guo, Y., Liu, Y., Liu, Y., & Wang, D. (2023). Charge storage coating based triboelectric nanogenerator and its applications in self-powered anticorrosion and antifouling. Frontiers of Materials Science, 17(1). https://doi.org/10.1007/s11706-023-0635-y	Frontiers of Materials Science	TENG coating with charge-storage properties acrylic resin, a friction material, with nano-BaTiO3 particles and gas phase fluorination	CSC-TENG, acrylic resin, F/BT coating, nano-BaTiO3 particles	electrochemical repulsion	Anti-corrosive, environmentally friendly, anti-bacterial, anti-algal protein resistant	further development is needed for application, and field testing is needed
Xu, Y., Wang, G., Xie, Z., & Wang, A. (2019). Preparation and evaluation of degradable polyurethane with low surface energy for marine antifouling coating. Journal of Coatings Technology and Research, 16(4), 1055–1064. https://doi.org/10.1007/s11998-018-00180-4	Journal of Coatings Technology and Research	new polyurethane modified by dodecafluoroheptyl methacrylate (DFHMA)	Alcoholates (PLA and DFHMA), polyurethane (PLIPFI and PLI&PFI)	self-polishing, low surface energy (antiadhesion)	environmentally friendly, anti-bacterial, antialgal	for moving conditions, further field testing needed to understand the application

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Liu, H., Ma, Z., Yang, W., Pei, X., & Zhou, F. (2019). Facile preparation of structured zwitterionic polymer substrate via sub-surface initiated atom transfer radical polymerization and its synergistic marine antifouling investigation. European Polymer Journal, 112, 146–152. https://doi.org/10.1016/j.eurpolymj.2018.07.025	European Polymer Journal	3-sulfopropyl methacrylate potassium salt (SPMA) on the flat hydrophilic resin surface using SSI-ATRP	SPMA polymer brushes, zwitterionic SPMA, acrylate, resin	Altered surface chemistry and topography, super-hydrophilic	long-term, environmentally friendly, anti-diatom	Further development is needed Hindered by salt responsiveness, further field testing is needed to understand the application.
Yang, M., Gu, L., Yang, B., Wang, L., Sun, Z., Zheng, J., Zhang, J., Hou, J., & Lin, C. (2017). Antifouling composites with self-adaptive controlled release based on an active compound intercalated into layered double hydroxides. Applied Surface Science, 426, 185–193. https://doi.org/10.1016/j.apsusc.2017.07.207	Applied Surface Science	intercalation of sodium paeonolsilate (PAS) into MgAl and ZnAl layered double hydroxide (LDH)	PAS, Zn(NO3)2·6H2O, Al(NO3)3·9H2O, Mg(NO3)2·6H2O	Soluble matrix with biocide	Anti-algal, anti-bacterial, anti-diatom, self-adaptive, long-acting	May be toxic due to metals (further research is needed); field testing is needed
Takamura, H., Kinoshita, Y., Yorisue, T., & Kadota, I. (2023). Chemical synthesis and antifouling activity of monoterpene–furan hybrid molecules. Organic & Biomolecular Chemistry, 21(3), 632–638. https://doi.org/10.1039/d2ob02203f	Organic & Biomolecular Chemistry	monoterpene–furan hybrid molecules with introduced structural variety (monoterpenes	geraniol, nerol, (R)-citronellal, (s)-citronellal (monoterpenes and furan)	non-toxic biocide	environmentally friendly, anti-larval	Further development is needed and field testing is needed

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Dai, G., Ai, X., Mei, L., Ma, C., & Zhang, G. (2021). Kill–resist–renew trinity: Hyperbranched polymer with self-regenerating attack and defense for antifouling coatings. ACS Applied Materials & Interfaces, 13(11), 13735–13743. https://doi.org/10.1021/acsami.1c02273	ACS Applied Materials & Interfaces	"kill–resist–renew trinity" polymeric coating	monomer-tertiary carboxybetaine ester acrylate with the antifouling group N-(2,4,6-trichlorophenyl)maleimide (TCB-TCPM) copolymerized with methacrylic anhydride via reversible addition-fragmentation chain transfer polymerization yielding a degradable hyperbranched polymer.	Self-polishing (zwitterion), degradable, biocide, hydrophobic	Controlled degradation rate, anti-bacterial, synergy ("attacking" and "defending"), protein resistant	Field testing needed
Zhu, P., Meng, W., & Huang, Y. (2017). Synthesis and antibiofouling properties of crosslinkable copolymers grafted with fluorinated aromatic side chains. RSC Advances, 7(6), 3179–3189. https://doi.org/10.1039/c6ra26409c	RSC Advances	functional ternary copolymers grafted with short fluoroalkyl or perfluoropolyether-modified fluorinated aromatic side chains and cross-linkable functional groups were prepared via radical polymerization	Fluoropolymers (2,3,4,5,6-Pentafluorostyrene, 1H,1H-Heptadecafluoro(2,5-dimethyl-3,6-dioxanonan-1-ol) and 1H,1H,2H,2H-perfluorooctanol), arcylates	hydrophobic, low surface energy	environmentally friendly, anti-bacterial, antialgal, thermo-stable	Further development needed, field testing needed
Wei, M.-Y., Wang, C.-F., Wang, K.-L., Qian, P.-Y., Wang, C.-Y., & Shao, C.-L. (2017). Preparation, structure, and potent antifouling activity of sclerotioramine derivatives. *Marine Biotechnology*, *19*(4), 372–378. https://doi.org/10.1007/s10126-017-9760-x	Marine Biotechnology	new sclerotia in amine derivatives (30) by the one-step semisynthetic method	sclerotia in amine derivatives (unnamed)	non-toxic biocide	Environmentally friendly,	Further development needed

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Tian, J., Xu, K., Hu, J., Zhang, S., Cao, G., & Shao, G. (2021). Durable self-polishing antifouling cu-ti coating by a micron-scale Cu/Ti laminated microstructure design. Journal of Materials Science & Technology, 79, 62–74. https://doi.org/10.1016/j.jmst.2020.11.038	Journal of Materials Science & Technology	Cu-Ti composite antifouling coating with micron-sized alternating laminated structure of Cu/Ti by plasma spraying of mechanically mixed Cu/Ti powders	Cu and Ti powders	Self-polishing, biocide	Controlled release rate, environmentally friendly, good adhesion, mechanically strong, long-lasting	toxic at high concentrations
Shen, J., Du, M., Wu, Z., Song, Y., & Zheng, Q. (2019). Strategy to construct polyzwitterionic hydrogel coating with antifouling, drag-reducing and weak swelling performance. RSC Advances, 9(4), 2081–2091. https://doi.org/10.1039/c8ra09358j	RSC Advances	soft and wet coating composed of zwitterionic polymer	combination of the anti-polyelectrolyte effect of poly-N-(3-sulfopropyl)-N-(methacryloxyethyl)-N, N-dimethylammonium betaine (PSBMA) and the typical polyelectrolyte effect of polyacrylic acid (PAA), a bicomponent hydrogel coating with weak swelling in saline solution, silanes	Zwitterion, hydrophilic	mechanically strong, anti-bacterial, protein resistant	not good for drag, field testing is needed
Pan, J., Mei, L., Zhou, H., Zhang, C., Xie, Q., & Ma, C. (2022). Self-regenerating zwitterionic hyperbranched polymer with tunable degradation for anti-biofouling coatings. Progress in Organic Coatings, 163, 106674. https://doi.org/10.1016/j.porgcoat.2021.106674	Progress in Organic Coatings	synthesized degradable HBPs with the controlled structure of methyl methacrylate (MMA), tertiary carboxybetaine ester (TCB), and divinyl monomer-methacrylic anhydride (MAAH) via RAFT polymerization	degradable hyperbranched anti-biofouling polymers of methyl methacrylate, tertiary carboxybetaine ester (TCB), and divinyl monomers via reversible addition-fragmentation chain transfer (RAFT) polymerization	Zwitterion self-polishing, Superhydrophilicity, MMA degradation (not self-polishing)	Controlled degradation rate, anti-diatom, anti-bacterial, protein resistant	Field testing needed

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Punitha, N., Saravanan, P., Mohan, R., & Ramesh, P. S. (2017). Antifouling activities of β-cyclodextrin stabilized peg based silver nanocomposites. Applied Surface Science, 392, 126–134. https://doi.org/10.1016/j.apsusc.2016.07.114	Applied Surface Science	β-cyclodextrin (β-CD) stabilized silver nanocomposites (SNCs) in the presence of Polyethylene glycol (PEG)	Biocompatible polymer β-CD and adhesive resistance polymer PEG were used to functionalize the silver nanoparticles (SNPs) and the synthesized SNCs exhibit excellent micro fouling activities	Hydrophobic-hydrophobic interaction, metal nanocomposite biocide	Anti-bacterial, "environmentally friendly" biocide	Uses metal (potential environmental contamination), toxicity invertebrate testing needed
Overturf, C. L., Wormington, A. M., Blythe, K. N., Gohad, N. V., Mount, A. S., & Roberts, A. P. (2015). Toxicity of noradrenaline, a novel anti-biofouling component, to two non-target zooplankton species, daphnia magna and Ceriodaphnia dubia. Comparative Biochemistry and Physiology Part C: Toxicology & Pharmacology, 171, 49–54. https://doi.org/10.1016/j.cbpc.2015.01.006	Comparative Biochemistry and Physiology Part C: Toxicology & Pharmacology	examine the toxicity of Noradrenaline NA to the non-target zooplankton D. magna and C. dubia	Noradrenaline	biocide	Anti-bacterial, anti-larval, less toxic than previously used antibiofouling chemicals	Toxic to off-target zooplankton species
Hu, P., Xie, Q., Ma, C., & Zhang, G. (2021). Fouling-resistant silicone coating with self-healing induced by metal coordination. Chemical Engineering Journal, 406, 126870. https://doi.org/10.1016/j.cej.2020.126870	Chemical Engineering Journal	poly(dimethylsiloxane) (PDMS) coating cross-linked via coordination bonds between 2-(2-benzimidazolyl)ethanethiol (BET) and zinc ions	PDMS, zinc ions, BET	low surface energy, hydrophobic, biocide (non-leaching?), synergistic	Anti-bacterial, anti-diatom, environmentally friendly, self-healing, good adhesion	Use of metal biocide (potential environmental contaminant), field testing needed

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Liu, L., Li, Z. J., Sun, S., You, T., Qi, X. T., & Song, K. (2017). Preparation and characterization of a novel hydrogel-based fouling release coating. Materials Science Forum, 898, 1539–1544. https://doi.org/10.4028/www.scientific.net/msf.898.1539	Materials Science Forum	hydrogel-based fouling release coating	PDMS resin	soluble matrix biocide (foul release), low surface energy	Anti-diatom, good adhesion, me	only for stationary situations
Bi, S., Xu, K., Shao, G., Yang, K., & Tian, J. (2023). Mechanically robust antifouling coating with dual-functional antifouling strategy by infiltrating PDMS into plasma-sprayed porous al2o3-cu coating. Journal of Materials Science & Technology, 159, 125–137. https://doi.org/10.1016/j.jmst.2023.02.034	Journal of Materials Science & Technology	Al2O3-PDMS-Cu composite coating	PDMS, Al2O3, Cu powders	soluble matrix solution with metal biocide, low surface energy	Mechanically strong, good adhesion, anti-bacterial, anti-algal, long lasting	toxic in high concentrations (copper), field testing needed
Moodie, L. W., Trepos, R., Cervin, G., Bråthen, K. A., Lindgård, B., Reiersen, R., Cahill, P., Pavia, H., Hellio, C., & Svenson, J. (2017). Prevention of marine biofouling using the natural allelopathic compound batatasin-III and synthetic analogues. Journal of Natural Products, 80(7), 2001–2011. https://doi.org/10.1021/acs.jnatprod.7b00129	Journal of Natural Products	22 synthetic allelopathic terrestrial natural dihydrostilbenes (Batatasin-III and Synthetic Analogues) with varying substitution patterns	3,5-dimethoxybibenzyl, 3,4-dimethoxybibenzyl, and 3-hydroxy-3′,4,5′-trimethoxybibenzyl	non-toxic biocides	Anti-bacterial, anti-diatom, anti-larval	Further development needed

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Tan, J., Xu, J., Wang, D., Yang, J., & Zhou, S. (2020). Seawater-responsive sio2 nanoparticles for in situ generation of zwitterionic polydimethylsiloxane antifouling coatings with underwater superoleophobicity. Journal of Materials Chemistry A, 8(45), 24086–24097. https://doi.org/10.1039/d0ta07617a	Journal of Materials Chemistry A	zwitterionic groups were endowed with PDMS coatings by blending SiO2 nanoparticles that were modified by using two new analogues of zwitterion precursors (MAPS and BMPAS) and subsequently, the in situ generation of zwitterions was conducted	PDMS, silanes, (N-methoxyacylethyl)-3-aminopropyltriethoxysilane (MAPS) and bis(N-methoxyacylethyl)-3-aminopropyltriethoxysilane (BMAPS)	zwitterion, low surface energy, hydrophobic, foul release	Anti-bacterial, anti-diatom, anti-algal, cost-effective, super wettable applications	For static conditions, Further field testing needed
Li, H., Xin, L., Zhang, K., Yin, X., & Yu, S. (2022). Fluorine-free fabrication of robust self-cleaning and anti-corrosion superhydrophobic coating with photocatalytic function for enhanced anti-biofouling Property. Surface and Coatings Technology, 438, 128406. https://doi.org/10.1016/j.surfcoat.2022.128406	Surface and Coatings Technology	fluorine-free robust superhydrophobic coating was fabricated on the Al alloy substrate	the adhesive of epoxy resin (EP) was firstly dripped on the Al alloy substrate, and then TiO2 nanoparticles modified with stearic acid were quickly sprayed onto the semi-cured adhesive layer to prepare the fluorine-free superhydrophobic coating	Superhydrophobic, low surface energy, photocatalyst	Anti-corrosive, anti-bacterial, self-cleaning	Needs further development and field testing
Zhang, K., Zhou, Z., Deng, Y., Wang, G., Wang, Z., Wu, L., & Yang, G. (2022). Protection performance of plasma sprayed AL2O3-13 wt%tio2 coating sealed with an organic-inorganic hybrid agent. Journal of Wuhan University of Technology-Mater. Sci. Ed., 37(3), 331–335. https://doi.org/10.1007/s11595-022-2535-x	Journal of Wuhan University of Technology-Mater. Sci. Ed.	organic-inorganic hybrid sealing agent was fabricated and used in the plasma sprayed Al2O3-13 wt%TiO2 coating	Al2O3-13 wt%TiO2 coating was plasma sprayed on the steel substrate. An organic-inorganic hybrid sealing agent was fabricated and used successfully in the sprayed coating	Plasma spray soluble matrix, metal biocide, superhydrophobic	anti-corrosive, antibiofouling (no specifics), sealing treatment	use of metal biocide (potential environmental contaminant),

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Xin, X., Huang, G., Zhou, X., Sun, W., Jin, C., Jiang, W., & Zhao, S. (2016). Potential antifouling compounds with antidiatom adhesion activities from the sponge-associated bacteria,bacillus pumilus. Journal of Adhesion Science and Technology, 31(9), 1028–1043. https://doi.org/10.1080/01694243.2016.1242219	Journal of Adhesion Science and Technology	antidiatom adhesion compounds from marine microbes, fatty acids from this sponge-associated bacterium (B. pumilus)	UST050418-683 was isolated from 120 strains sponge-associated bacteria (B. pumilus)	Non-toxic biocide	Anti-diatom, environmentally friendly	Further development needed
Yuan, C.,, Cao, P., & Xiuqin B. (2017). Study on Antimicrobial Surface Design Using Biological Modification Attempting for Marine Antifouling. The 27th International Ocean and Polar Engineering Conference, San Francisco, California, USA.	The 27th International Ocean and Polar Engineering Conference	new bioorganic material (Bs prepared by a new chemical reaction between 304 stainless steel and a short-chain peptide (Bp)	Peptide (Bp), stainless steel	Altered surface topography, low surface energy, hydrophobic	Anti-bacterial,	Field testing needed
Guazzelli, E., Perondi, F., Criscitiello, F., Pretti, C., Oliva, M., Casu, V., Maniero, F., Gazzera, L., Galli, G., & Martinelli, E. (2020). New Amphiphilic Copolymers for PDMS-based nanocomposite films with long-term marine antifouling performance. Journal of Materials Chemistry B, 8(42), 9764–9776. https://doi.org/10.1039/d0tb01905d	Journal of Materials Chemistry B	Amphiphilic methacrylate copolymers (Si-co-EF) containing polysiloxane (Si) and mixed poly(oxyethylene)–perfluorohexyl (EF) side chains were synthesized with different compositions and used together with polysiloxane-functionalized nanoparticles as additives of condensation cured nanocomposite poly(siloxane) films	PDMS methacrylate (Si), poly(oxyethylene)-perfluorohexyl methacrylate (EF)	low surface energy, amphiphilic, nanoparticles	Anti-polychaeta, anti-diatom, environmentally friendly	further development needed, Fouling occurred in field test

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Noor Idora, M. S., Ferry, M., Wan Nik, W. B., & Jasnizat, S. (2015). Evaluation of tannin from Rhizophora apiculata as natural antifouling agents in epoxy paint for marine application. Progress in Organic Coatings, 81, 125–131. https://doi.org/10.1016/j.porgcoat.2014.12.012	Progress in Organic Coatings	the mangrove, Rhizophora apiculata tannin derivatives (zinc tannate) in epoxy paint	Zinc tannate, epoxy	non-toxic biocide	environmentally friendly, anti-bacterial	Further development needed, high concentration needed, solubility problems in seawater, Further field testing needed
Yao, J., Dai, Z., Yi, J., Yu, H., Wu, B., & Dai, L. (2020). Degradable polyurethane based on triblock polyols composed of polypropylene glycol and ε-caprolactone for marine antifouling applications. Journal of Coatings Technology and Research, 17(4), 865–874. https://doi.org/10.1007/s11998-019-00313-3	Journal of Coatings Technology and Research	Degradable PU films were prepared by mixing triblock polyols [composed of polyether (polypropylene glycol or polyethylene glycol) and ε-caprolactone] and crosslinker agent (tolylene diisocyanate trimer)	degradable polyurethane (PU), polyether (PPGx or PEG800) and CL	self-polishing, hydrophilic,	long-lasting (8months in field), controlled release rate, mechanically strong	Microplastics potential (polyurethane)
Wang, P., Zhang, D., Sun, S., Li, T., & Sun, Y. (2016). Fabrication of slippery lubricant-infused porous surface with high underwater transparency for the control of marine biofouling. ACS Applied Materials & Interfaces, 9(1), 972–982. https://doi.org/10.1021/acsami.6b09117	ACS Applied Materials & Interfaces	slippery lubricant-infused porous surface (SLIPS) with high underwater-transparency	slippery lubricant-infused porous surface with high underwater-transmittance was fabricated on glass sample via infusing lubricant into its porous microstructure fabricated with hydrothermal method	low surface energy, amphiphilic (superhydrophobic and hydrophilic), nanotextured	anti-bacterial, anti-algal, good for optical equipment	only for texture hydrophilic surfaces and superhydrophobic, static conditions, Further development needed

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Baldissera, A. F., Miranda, K. L., Bressy, C., Martin, C., Margaillan, A., & Ferreira, C. A. (2015). Using conducting polymers as active agents for marine antifouling paints. Materials Research, 18(6), 1129–1139. https://doi.org/10.1590/1516-1439.261414	Materials Research	paints containing polyaniline (PAni) and derivatives as active pigment	doped polyaniline (PAni-ES and PAni/DBSA) and its sulphonated form (SPAN), epoxy	soluble matrix, biocide (zinc pyrithione (PyZn),	Patented, long-lasting (12 months in field), reduce the use of Cu2O	use of metal biocide (potential environmental contaminant)
Jia, M.-Y., Zhang, Z.-M., Yu, L.-M., & Wang, J. (2017). Pani-PMMA as cathodic electrode material and its application in cathodic polarization antifouling. Electrochemistry Communications, 84, 57–60. https://doi.org/10.1016/j.elecom.2017.09.021	Electrochemistry Communications	The electrochemical properties of PANI-PMMA as a cathode material and the application of this material to cathodic polarization antifouling were investigated	Camphor sulfonic acid doped polyaniline (PANI-CSA) solution in m-cresol, Dodecyl-benzene sulfonic acid doped PANI (PANI-DBSA) solution in chloroform, glass carbon electrode (GCE), polymethyl methacrylate (PMMA)	electrochemical repulsion	Environmentally friendly, anti-bacterial,	Field testing needed, static conditions only
Hu, P., Zeng, H., Zhou, H., Zhang, C., Xie, Q., Ma, C., & Zhang, G. (2021). Silicone elastomer with self-generating zwitterions for antifouling coatings. Langmuir, 37(27), 8253–8260. https://doi.org/10.1021/acs.langmuir.1c00984	Langmuir	a silicone elastomer with zwitterionic pendant chains has been prepared by grafting a telomer of tertiary carboxybetaine dodecafluoroheptyl ester ethyl acrylate (TCBF) and 3-mercaptopropyltriethoxysilane to the bis-silanol-terminated poly(dimethylsiloxane) (PDMS)	3-Mercaptopropyltriethoxysilane (MPTS), methyltriacetoxysilane (MeAc), methylaminoethanol, acryloyl chloride, acetic acid (AA), and triethylamine (TEA), Dodecafluoroheptyl acrylate (DFA), Bis-silanol-terminated PDMS	Self-polishing, zwitterion, low surface energy	Anti-bacterial, anti-algal	Field testing needed

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Chen, C., Wang, Y., & Zhang, D. (2020). Bifunctional nanozyme activities of layered double hydroxide derived co-al-ce mixed metal oxides for antibacterial application. *Journal of Oceanology and Limnology*, *38*(4), 1233–1245. https://doi.org/10.1007/s00343-020-0041-6	Journal of Oceanology and Limnology	LDH derived Co-Al-Ce MMOs composed of CeO 2 compounding with Co 3 O 4 and CoAl 2 O 4	3,3',5,5'-tetramethylbenzidine (TMB), 2',7'-dichlorodihydrofluorescein diacetate (DCFHDA), propidium iodide (PI), and SYTO 9, Hydroxyphenyl fluorescein (HPF) and dihydroethidium (DHE)	Biocide (metal), nanozyme	Anti-bacterial, "green",	use of metal biocide (potential environmental contaminant), field testing needed
Zhang, H., Ma, Y., Tan, J., Fan, X., Liu, Y., Gu, J., Zhang, B., Zhang, H., & Zhang, Q. (2016). Robust, self-healing, superhydrophobic coatings highlighted by a novel branched thiol-ene fluorinated siloxane nanocomposites. Composites Science and Technology, 137, 78–86. https://doi.org/10.1016/j.compscitech.2016.10.023	Composites Science and Technology	superhydrophobic fabric after being dip-coated with polydimethylsiloxane (PDMS), novel branched fluorinated siloxane (T-FAS) and hydrophobic fumed silica nanoparticles (SiO2 NPs)	Perfluorinated (N-methyl-perfluorohexane-1-sulfonamide) ethyl acrylate, Pentaerythritol tetra (3-mercaptopropionate) (PETMP), γ-methacryloxypropyltrimethoxysilane (MPS), dimethyl phenyl phosphine (DMPPH)	Self-polishing, low surface energy, superhydrophobic	Mechanically strong,	Field testing needed, further development needed
Fu, Y., Cai, M., Zhang, E., Cao, S., & Ji, P. (2016). A novel hybrid polymer network for efficient anticorrosive and antibacterial coatings. Industrial & Engineering Chemistry Research, 55(16), 4482–4489. https://doi.org/10.1021/acs.iecr.5b04818	Industrial & Engineering Chemistry Research	polydopamine and silica hybrid polymer (PDSHP) coatings	hybrid polymer network coating has been formed by in situ simultaneous polymerization of dopamine and hydrolytic polycondensation of 3-aminopropyltriethoxysilane in alkaline solutions,	biocide, hydrophobic	anti-bacterial, anti-corrosive	toxicity testing needed, field testing needed

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			polyethylenimine (PEI) to form a PEI			
Guo, X., Guo, R., Fang, M., Wang, N., Liu, W., Pei, H., Liu, N., & Mo, Z. (2022). A novel composite protective coating with UV and corrosion resistance: Load floating and self-cleaning performance. Ceramics International, 48(12), 17308–17318. https://doi.org/10.1016/j.ceramint.2022.02.293	Ceramics International	superhydrophobic and oleophobic composite coating of PPS-COOH, FAS-ZnO with ATP	FAS-ZnO and ATP were mixed with PPS-COOH to fabricate a stable hydrophobic and oleophobic composite coating by a one-step spraying process	Superhydrophobic, oleophobic	mechanically strong, self cleaning, anti-corrosive, UV stability	Only for static conditions, field testing needed
Yu, M., Liu, M., Zhang, D., & Fu, S. (2021). Lubricant-grafted omniphobic surfaces with anti-biofouling and drag-reduction performances constructed by Reactive Organic–Inorganic hybrid microspheres. Chemical Engineering Journal, 422, 130113. https://doi.org/10.1016/j.cej.2021.130113	Chemical Engineering Journal	slippery liquid-infused surfaces (SLIPSs)	organic–inorganic hybrid particles (Poly@V-SiO2) with controlled particle sizes and adjusted chemical constituents were used as building units to construct superhydrophobic and lubricant-grafted slippery surface (LGSS) surface	Super hydrophobicity, low surface energy	"anti-fouling", dar reduction	Further development needed, field testing needed

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Zhang, J., Wang, Y., Zhou, S., Wang, Y., Wang, C., Guo, W., Lu, X., & Wang, L. (2020). Tailoring self-lubricating, wear-resistance, anticorrosion and antifouling properties of ti/(cu, MOS2)-DLC coating in marine environment by controlling the content of Cu Dopant. Tribology International, 143, 106029. https://doi.org/10.1016/j.triboint.2019.106029	Tribology International	Multi-functional Ti/(Cu, MoS2)-DLC coatings with different Cu content were deposited via RF/DC magnetron sputtering technique.	The Ti/(Cu, MoS2)-DLC (diamond-like carbon (DLC)) coatings with various Cu contents were successfully deposited on 304 stainless steel substrate via magnetron sputtering technique	biocide	mechanically strong, anti-corrosive,	use of metal biocide (potential environmental contaminant), field testing needed
Choi, S., Jepperson, J., Jarabek, L., Thomas, J., Chisholm, B., & Boudjouk, P. (2007). Novel approach to anti-fouling and fouling-release Marine Coatings based on dual-functional siloxanes. Macromolecular Symposia, 249–250(1), 660–667. https://doi.org/10.1002/masy.200750452	Macromolecular Symposia	biocide incorporated silicone coatings	triclosan, Derivatives of thebiocide, Triclosan (5-chloro-2-(2, 4-dichlorophenoxy) phenol), were used to covalentlyattach the biocide moiety to a silicone backbone.	biocide, hydrophobic, low surface energy, foul-release	prevented macrofouling, mechanically strong	further development needed to prevent microfouling, further field testing needed
Jiang, X., Xu, X., Xia, Z., Lin, D., Chen, Y., Wang, Y., Yu, D., Wu, X., & Zeng, H. (2024). Simultaneous segment orientation and anchoring for robust hydrogel coating with underwater Superoleophobicity. Advanced Functional Materials. https://doi.org/10.1002/adfm.202314589	Advanced Functional Materials.	synergistic segment orientation/covalent anchoring and locking strategy, a superoleophobic hydrogel coating	PEG, PTMG, epxoy resin, DMAc	Amphiphilic	long-lasting, good adhesion, self-cleaning,	Further development needed

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Tong, Z., Guo, H., Di, Z., Chen, S., Song, L., Hu, J., Hou, Y., Zhan, X., & Zhang, Q. (2023). P. pavoninus-inspired smart slips marine antifouling coating based on coumarin: Antifouling durability and adaptive adjustability of lubrication. Advanced Functional Materials, 34(8). https://doi.org/10.1002/adfm.202310702	Advanced Functional Materials	bionic CmSLIPS based on coumarin	SLIPS were modified with coumarin to prepare a “smart” SLIPS antifouling coating (CmSLIPS), which has adaptive adjustability of lubrication and antifouling durability	Low surface energy, hydrophobic, biocide	Anti-bacterial, anti-algal, surface stability/durability, adaptive lubricity, self-cleaning, protein resistant, self-healing, long lasting (150day field test), harsh marine envr.'s with many biofoulers	static conditions, Further field testing needed
Selim, M. S., Fatthallah, N. A., Higazy, S. A., Hao, Z., & Jing Mo, P. (2022). A comparative study between two novel silicone/graphene-based nanostructured surfaces for maritime antifouling. Journal of Colloid and Interface Science, 606, 367–383. https://doi.org/10.1016/j.jcis.2021.08.026	Journal of Colloid and Interface Science	superhydrophobic PDMS series decorated with RGO and GO-γ-AlOOH nanorod hybrid composites as novel FR nanocomposite coating	PDMS/GO-γ-AlOOH (3 wt%) nanocomposite, graphene-based nanocomposites	Superhydrophobic, low surface energy, nanostructure	(both)anti-bacterial, anti-fungal, environmentally friendly, field test (45days)	Further development needed

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Hu, K., Li, M., You, C., Zhang, Y., Xu, Z., Xu, Y., Fan, L., Yi, Y., & Chu, Y. (2024). Novel indole-based self-polishing environmentally friendly acrylic antifouling coatings. Progress in Organic Coatings, 190, 108384. https://doi.org/10.1016/j.porgcoat.2024.108384	Progress in Organic Coatings	indole derivatives containing carbon-carbon double bond (specifically allyl 5-Cl/NO2-indole-carboxylate, abbreviated as ACIC/ANIC) were synthesized from 5-Cl/NO2-indole and incorporated into zinc acrylate resins via N-acylation reaction	indole derivatives containing vinyl groups were synthesized through N-acylation reaction utilizing 5-chloro-1H-indole, 5-nitro-1H-indole, and allyl 1,2,4-triazole-carboxylate , these indole derivatives were introduced into zinc acrylate resin. A composite material was then prepared by blending zinc acrylate resin and PCL with acrylic urethane lacquer	Self-polishing, biocide	Anti-bacterial, antialgal, environmentally friendly	Further development and field testing needed
Svenson, johan, Grant, T. M., Rennison, D., Cervin, G., Pavia, H., Hellio, C., Foulon, V., Brimble, M. A., & Cahill, P. (2021). Towards eco-friendly marine antifouling biocides – nature inspired tetrasubstituted 2,5-diketopiperazines. SSRN Electronic Journal. https://doi.org/10.2139/ssrn.3967417	SSRN Electronic Journal	the pharmacophore derived from amphiphilic micropeptides into a 2,5-diketopiperazine (DKP) scaffold	four distinct libraries of 2,5-DKPs, an established amphiphilic antimicrobial peptide pharmacophore (2 + 2 pharmacophore),	non-toxic biocide, amphiphilic	Environmentally friendly, anti-microbial	field testing needed
Ou, B., Chen, M., Guo, Y., Kang, Y., Guo, Y., Zhang, S., Yan, J., Liu, Q., & Li, D. (2018). Preparation of novel marine antifouling polyurethane coating materials. Polymer Bulletin, 75(11), 5143–5162. https://doi.org/10.1007/s00289-018-2302-5	Polymer Bulletin	novel PLLA-grafted graphene was synthesized via ring-opening reaction	polyurethane containing PLLA,hydroxylation-terminated poly(l-lactide)-functionalized graphene (G-g-PLLA) PDMS, graphene	low surface energy, hydrophobic,	environmentally friendly, anti bacterial	further development needed

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Selim, M. S., Yang, H., El-Safty, S. A., Fatthallah, N. A., Shenashen, M. A., Wang, F. Q., & Huang, Y. (2019). Superhydrophobic coating of silicone/β–MNO2 nanorod composite for Marine Antifouling. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 570, 518–530. https://doi.org/10.1016/j.colsurfa.2019.03.026	Colloids and Surfaces A: Physicochemical and Engineering Aspects	superhydrophobic series of 1D β-MnO2-silicone hybrid composites for FR coating	superhydrophobic composite of in situ PDMS/β-MnO2 nanorods coatings with the rough structured surface	superhydrophobic, low surface energy, altered surface texture	long-lasting (90day field test), self-cleaning, antimicrobial (bacteria, fungi yeast), environmentally friendly	further development needed
Richards, C., Briciu-Burghina, C., Jacobs, M. R., & Barrett, A. (2019). Assessment of antifouling potential of novel transparent Sol Gel Coatings for application in the marine environment. Molecules, 24(16), 2983. https://doi.org/10.3390/molecules24162983	Molecules	several novel transparent sol-gel materials	diethoxydimethylsilane (DMDEOS, tetraethyl orthosilicate (HC006)	Altered surface chemistry and topography, hydrophobic	long lasting (9-13month field test), anti-macrofouling, anti-diatom	for sensors and devices with windows (clear coating), further development needed
Liu, X., Gu, X., Zhou, Y., Pan, W., Liu, J., & Song, J. (2023). Antifouling slippery surface against marine biofouling. Langmuir, 39(38), 13441–13448. https://doi.org/10.1021/acs.langmuir.3c00986	Langmuir	antifouling slippery surface with good slippery performance and marine antifouling properties	nontoxic antifouling slippery surface (AFSS) using silicone oil, silane coupling agent, nanosilica, nanoceramic coating, epoxy resin, and capsaicin	Hydrophobic	anti-bacterial, anti-algal, protein resistant	for protecting titanium alloy piping systems

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Guo, H., Chen, P., Tian, S., Ma, Y., Li, Q., Wen, C., Yang, J., & Zhang, L. (2020). Amphiphilic Marine antifouling coatings based on a hydrophilic polyvinylpyrrolidone and hydrophobic fluorine–silicon-containing block copolymer. Langmuir, 36(48), 14573–14581. https://doi.org/10.1021/acs.langmuir.0c02329	Langmuir	PVP–PFA–PDMS block copolymer	novel amphiphilic block copolymer that combined hydrophilic polyvinylpyrrolidone (PVP) with hydrophobic poly(1-(1H,1H,2H,2H-perfluorodecyloxy)-3-(3,6,9-trioxadecyloxy)-propan-2-yl acrylate) (PFA) and polydimethylsiloxane (PDMS)	amphiphilic, foul release, low surface energy	environmentally friendly, anti-bacterial, anti-diatom, anti-macro fouler (pseudobarnicle)	field testing needed, further development needed
Su, X., Yang, M., Hao, D., Guo, X., & Jiang, L. (2021). Marine antifouling coatings with surface topographies triggered by phase segregation. Journal of Colloid and Interface Science, 598, 104–112. https://doi.org/10.1016/j.jcis.2021.04.031	Journal of Colloid and Interface Science	surface structures (chemical heterogeneity) triggered by phase segregation	polyurethane coating with amphiphilic micro/nanodomains structure triggered by phase segregation	amphiphilic, altered surface topography	mechanically robust, anti-algal, anti-macro fouler (barnacles), protein resistant	field testing needed
Jakobi, V., Schwarze, J., Finlay, J. A., Nolte, K. A., Spöllmann, S., Becker, H.-W., Clare, A. S., & Rosenhahn, A. (2018). Amphiphilic alginates for marine antifouling applications. Biomacromolecules, 19(2), 402–408. https://doi.org/10.1021/acs.biomac.7b01498	Biomacromolecules	amphiphilic alginates with fluorinated side chains	derivatization of hydrophilic alginates with fluorinated substituents provides a route to amphiphilic polysaccharides	amphiphilic	protein resistant, anti-algal, anti-diatom, renewable, environmentally friendly	Further development needed

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Pinteus, S., Lemos, M. F. L., Freitas, R., Duarte, I. M., Alves, C., Silva, J., Marques, S. C., & Pedrosa, R. (2020). Medusa polyps adherence inhibition: A novel experimental model for antifouling assays. Science of The Total Environment, 715, 136796. https://doi.org/10.1016/j.scitotenv.2020.136796	Science of The Total Environment	the seaweeds A. armata and S. muticum were evaluated for their ability to produce antifouling substances	Asparagopsis armata and Sargassum muticum, extracts	biomimetic, non-toxic biocide	environmentally friendly	further development needed
Yang, W., Lin, P., Cheng, D., Zhang, L., Wu, Y., Liu, Y., Pei, X., & Zhou, F. (2017). Contribution of charges in Polyvinyl Alcohol Networks to Marine Antifouling. ACS Applied Materials & Interfaces, 9(21), 18295–18304. https://doi.org/10.1021/acsami.7b04079	ACS Applied Materials & Interfaces	Semi-interpenetrated polyvinyl alcohol polymer networks (SIPNs) were prepared by integrating various charged components into polyvinyl alcohol polymer	cationic, anionic, zwitterionic, and neutral precursors, were integrated into SIPNs	Zwitterion/ anion, hydrophilic	anti-algal, anti-diatom, field tested, environmentally friendly, cheap, tunable mechanical properties	Further development needed.
Kim, I., & Kang, S. M. (2024). Formation of amphiphilic zwitterionic thin poly(sbma-co-tfema) brushes on solid surfaces for marine antifouling applications. Langmuir, 40(6), 3213–3221. https://doi.org/10.1021/acs.langmuir.3c03687	Langmuir	amphiphilic zwitterionic polymers using zwitterionic and hydrophobic monomers	amphiphilic zwitterionic thin polymer brushes composed of sulfobetaine methacrylate (SBMA) and trifluoroethyl methacrylate (TFEMA) were synthesized on Si/SiO2 surfaces via surface-initiated atom transfer radical polymerization	amphiphilic, zwitterion,	anti-diatom, sediment resistance	field testing needed, further development needed

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Guo, H., Liu, X., Zhao, W., Xie, C., Zhu, Y., Wen, C., Li, Q., Sui, X., Yang, J., & Zhang, L. (2021). A polyvinylpyrrolidone-based surface-active copolymer for an effective marine antifouling coating. Progress in Organic Coatings, 150, 105975. https://doi.org/10.1016/j.porgcoat.2020.105975	Progress in Organic Coatings	surface-active PVP-PDMS-PVP triblock copolymer	surface-active copolymer basing on hydrophilic polyvinylpyrrolidone (PVP) and hydrophobic poly(dimethylsiloxane) (PDMS) was designed and incorporated into a crosslinked PDMS matrix to form a surface-renewable antifouling coating	low surface energy, amphiphilic, fouling release	protein resistant, anti-diatom, anti-adhesion, field tested	Further development needed
Richards, Chloe, Ollero, A. D., Daly, P., Delauré, Y., & Regan, F. (2024). Disruption of diatom attachment on marine bioinspired antifouling materials based on Brill (Scophthalmus Rhombus). Science of The Total Environment, 912, 169348. https://doi.org/10.1016/j.scitotenv.2023.169348	Science of The Total Environment	six engineered surfaces, inspired by Brill fish scales, fabricated through a 2-photon polymerization (2PP) process, for their potential as antifouling solutions	microtextured surfaces inspired by the Brill fish scales	altered surface topography,	environmentally friendly, anti-diatom	Further development needed
Zhang, J., Pan, M., Luo, C., Chen, X., Kong, J., & Zhou, T. (2016). A novel composite paint (Tio2/fluorinated acrylic nanocomposite) for antifouling application in Marine Environments. Journal of Environmental Chemical Engineering, 4(2), 2545–2555. https://doi.org/10.1016/j.jece.2016.05.002	Journal of Environmental Chemical Engineering	TiO2/fluorinated acrylic nanocomposite paint with functional surfaces	titanium dioxide (TiO2) nanoparticle suspension into the fluorinated acrylic copolymers	hydrophobic, low surface energy	environmentally friendly, anti-corrosive, field tested, anti-bacterial, anti-diatom	uses mental (titanium although lightweight), further development needed

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Duan, Y., Wu, J., Qi, W., & Su, R. (2023). Eco-friendly marine antifouling coating consisting of cellulose nanocrystals with bioinspired micromorphology. Carbohydrate Polymers, 304, 120504. https://doi.org/10.1016/j.carbpol.2022.120504	Carbohydrate Polymers	natural sourced nanomaterials (cellulose nanocrystals, CNCs) as nanofillers, a nanocomposite superhydrophobic coating	green nanomaterials CNCs as nanofillers with micro/nanoscale hierarchical structures via a simple sol-gel method	superhydrophobic, altered surface topography, biomimicry	environmentally friendly, self-cleaning, anti-adhesion, cost-effective	Further development needed
Zhou, W., Wang, Y., Ni, C., & Yu, L. (2021). Preparation and evaluation of natural rosin-based zinc resins for marine antifouling. Progress in Organic Coatings, 157, 106270. https://doi.org/10.1016/j.porgcoat.2021.106270	Progress in Organic Coatings	rosin metal salt resin was prepared mainly by rosin and zinc hydroxide (Zn(OH)2)	rosin-based zinc resins via a one-pot dehydration condensation reaction using nontoxic rosin acid and Zn(OH)2	self-polishing, hydrophobic	long-lasting field tested (6 months), anti-bacterial, protein resistant, anti-algal, environmentally friendly, cost effective	heavy metal (zinc) used, further development needed
Yang, Z., He, X., Lou, T., Su, D., Bai, X., & Yuan, C. (2024). Fabrication of dual functional marine antifouling coatings by infusing epoxy silicone oil modification of quaternary ammonium. European Polymer Journal, 210, 112947. https://doi.org/10.1016/j.eurpolymj.2024.112947	European Polymer Journal	SLIPS endowed with both resist and contact-killing antimicrobial performances silicone oil containing quaternary ammonium salt groups infused into specially-designed AAO, forming a QAS-SLIPS coating	anodic aluminum oxides (AAO) with composite structures of the pores and pyramids were prepared and modified with siloxane a silicone oil containing quaternary ammonium salt (QAS-SO) groups was synthesized and infused into specially designed AAO, forming a QAS-SLIPS	contact killing (biocidal adjacent), hydrophilic, isolation of lubricant	anti-bacterial, anti-algal,	Field testing needed, further development needed

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Alarif, W. M., Shaban, Y. A., Orif, M. I., Ghandourah, M. A., Turki, A. J., Alorfi, H. S., & Tadros, H. R. (2023). Green synthesis of tio2 nanoparticles using natural marine extracts for antifouling activity. Marine Drugs, 21(2), 62. https://doi.org/10.3390/md21020062	Marine Drugs	TiO2 Nanoparticles Using Natural Marine Extracts	Titanium dioxide (TiO2) nanoparticles were synthesized via a novel eco-friendly green chemistry approach using marine natural extracts of two red algae (Bostrychia tenella and Laurencia obtusa), a green alga (Halimeda tuna), and a brown alga (Sargassum filipendula) along with a marine sponge sample identified as Carteriospongia foliascens	nanoparticle biocide	Field tested (108days, anti-bacterial, anti-algal, environmentally friendly	use of metal (titanium), did not prevent macrofouling
Yang, Z., He, X., Lou, T., Bai, X., & Yuan, C. (2023). Infusing paraffin-based lubricant into micro-/nanostructures for constructing slippery marine antifouling coatings. Progress in Organic Coatings, 185, 107919. https://doi.org/10.1016/j.porgcoat.2023.107919	Progress in Organic Coatings	paraffin-based SLIPS (1) pyramids-pores structures provided the storage for the lubricant; (2) The paraffin-based framework reduced the rheological properties of the lubricant	anodized 5083 Al surfaces of different scales were prepared, the porous structure with pyramid-like clusters in the upper layer was most suitable for the preparation of SLIPS	altered surface topography, hydrophobic	cost-effective, strong mechanical properties, spray coating, environmentally friendly, long lasting	field testing needed

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Liu, J., Li, Q., Gui, T., Guo, H., Zhang, K., Meng, F., Zhan, X., Liu, Q., & Zhang, Q. (2024). Carp-inspired self-regulating marine antifouling coating: Featuring robust controlled release of eugenol and high-efficiency self-healing performance. Chemical Engineering Journal, 486, 149929. https://doi.org/10.1016/j.cej.2024.149929	Chemical Engineering Journal	PDMS-Pun-x	fluorinated eugenol copolymer was synthesized and covalently grafted onto polyurea-modified PDMS to achieve the synthesis of a self-regulating coating (PDMS-Pun-x) with robust controlled release of eugenol	self-polishing, low surface energy, hydrophilic	Anti-corrosive, environmentally friendly, mechanically strong	field testing needed
Selim, M. S., Azzam, A. M., Higazy, S. A., El-Safty, S. A., & Shenashen, M. A. (2022). Novel graphene-based ternary nanocomposite coatings as ecofriendly antifouling brush surfaces. Progress in Organic Coatings, 167, 106803. https://doi.org/10.1016/j.porgcoat.2022.106803	Progress in Organic Coatings	PDMS/GO/TiO2 nanocomposites	ternary nanocoatings comprising PDMS–GO/TiO2 nanofiller as a durable marine FR top coat	Superhydrophobic, altered surface topography.	cost-effective, long-lasting (45 day field test), mechanically robust, anti-microbial, anti-fungal	Further development needed
Zhang, L., Sha, J., Chen, R., Liu, Q., Liu, J., Yu, J., Zhang, H., Lin, C., Zhou, W., & Wang, J. (2020). Surface Plasma AG-decorated Bi5o7i microspheres uniformly distributed on a zwitterionic fluorinated polymer with superfunctional antifouling property. Applied Catalysis B: Environmental, 271, 118920. https://doi.org/10.1016/j.apcatb.2020.118920	Applied Catalysis B: Environmental	surface plasma Ag-decorated three-dimensional Bi5O7I flower-like microspheres incorporated in a zwitterionic fluorinated polymer (ZFP) fabricated	Bi5O7I/Ag/zwitterionic fluorinated polymer (ZFP) (ABZFP) composite films consisting of a flower-like Bi5O7I structure, plasma-enhanced Ag co-catalyst, and ZFP	zwitterionic, photothermal catalytic	anti-diatom, anti-adhesion, anti bacterial, environmentally friendly, long lasting	field testing needed, use of heavy metal

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Fu, Y., Gong, C., Wang, W., Zhang, L., Ivanov, E., & Lvov, Y. (2017). Antifouling thermoplastic composites with Maleimide encapsulated in clay nanotubes. ACS Applied Materials & Interfaces, 9(35), 30083–30091. https://doi.org/10.1021/acsami.7b09677	ACS Applied Materials & Interface	ethylene-vinyl acetate copolymer [EVA, (C2H4)x(C4H6O2)y] with trichlorophenyl maleimide (TCPM) loaded inside of halloysite clay nanotubes	ethylene-vinyl acetate (EVA) copolymer composite was prepared with the N-(2,4,6-trichlorophenyl) maleimide (TCPM) using halloysite clay as tubule nanocontainers to encapsulate TCPM	foul-release, nanocomposite surface (altered surface topography)	anti-bacterial, field tested (field tested 60days), environmentally friendly, cost-effective	further development needed
Yee, M. S.-L., Khiew, P.-S., Chiu, W. S., Tan, Y. F., Kok, Y.-Y., & Leong, C.-O. (2016). Green synthesis of graphene-silver nanocomposites and its application as a potent marine antifouling agent. Colloids and Surfaces B: Biointerfaces, 148, 392–401. https://doi.org/10.1016/j.colsurfb.2016.09.011	Colloids and Surfaces B: Biointerfaces	flexible sheets of graphene-Ag nanomaterial	graphene-silver nanomaterials using a hydrothermal reduction of AgNPs involving the citrate method on pristine few layer graphene	biocide (silver) nanoparticles	Anti-bacterial, "green' process, anti-algal	use of heavy metal (silver), further development needed
Zhang, H., Liang, T., Liu, Y., Misra, R. D. K., & Zhao, Y. (2021). Low-surface-free-energy go/FSIAC coating with self-healing function for anticorrosion and antifouling applications. Surface and Coatings Technology, 425, 127690. https://doi.org/10.1016/j.surfcoat.2021.127690	Surface and Coatings Technology	fluorine-silicon copolymer (FSiAC) and 0.1 wt% graphene oxide (GO) was prepared by free radical copolymerization	FSiAC and GO/FSiAC coating with low surface free energy	low surface free energy, self-polishing	anti-corrosive, anti-algal, anti-bacterial	Field testing needed

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Deng, Z., Wang, Y., Zhang, D., & Chen, C. (2023). 3D Printing Technology Meets Marine Biofouling: A Study on Antifouling Resin for Protecting Marine Sensors. SSRN Electronic Journal. https://doi.org/10.2139/ssrn.4428663	SSRN Electronic Journal	3D printing photosensitive resin	acrylic photosensitive resin with controllable hydrolysis as a base material to develop a novel 3D printing photosensitive resin with excellent dispersion properties of Cu2O	self-polishing, biocide (copper)	field tested (1month), anti-bacterial, anti-algal, mechanically robust	developed for marine sensors, uses heavy metal
Selim, M. S., Fatthallah, N. A., Higazy, S. A., Zhuorui, W., & Hao, Z. (2024). Superhydrophobic silicone/graphene oxide-silver-titania nanocomposites as eco-friendly and durable maritime antifouling coatings. Ceramics International, 50(1), 452–463. https://doi.org/10.1016/j.ceramint.2023.10.121	Ceramics International	nonstick and superhydrophobic PDMS-PMMA/Ag–TiO2@GO nanocomposites	polydimethylsiloxane-polymethylmethacrylate (PDMS-PMMA) blend filled with silver nanospheres and titanium dioxide nanorods grafted on the graphene oxide surface (Ag–TiO2@GO)	Altered surface topography (silver nanorods), superhydrophobic, low surface free energy, foul release	"environmentally friendly", mechanically strong, anti-bacterial, non-toxic to off-target species	uses heavy metal
Xiong, G., Zhang, Z., Hao, S., Chen, Q., Zhang, C., Zhang, S., Wang, K., & Qi, Y. (2023). Construction of novel fluorescent synergistic photocatalytic double Z-scheme photocatalyst for efficient antifouling of Polydimethylsiloxane coatings. Progress in Organic Coatings, 181, 107575. https://doi.org/10.1016/j.porgcoat.2023.107575	Progress in Organic Coatings	g-C3N4/TNTs/SLAP/PDMS (P-CTS) biomimetic fluorescent synergistic photocatalytic AF coating	novel g-C3N4/TNTs/SLAP (CTS) fluorescent photocatalytic composite powder by thermal polymerization and liquid phase method. We added it into PDMS to prepare g-C3N4/TNTs/SLAP/PDMS (P-CTS)	biomimetic, photocatalytic, low surface energy,	anti-bacterial, anti-diatom, environmentally friendly	field testing needed

References	Journal title	Antibiofouling technology	Antifouling properties	Functionality	Advantages	Disadvantages
Guo, Hongyu, Song, L., Hu, J., Lin, T., Li, X., Yu, H., Cheng, D., Hou, Y., Zhan, X., & Zhang, Q. (2021). Enhanced antifouling strategy with a strong synergistic effect of fluorescent antifouling and contact bacteriostasis using 7-amino-4-methylcoumarin. Chemical Engineering Journal, 420, 127676. https://doi.org/10.1016/j.cej.2020.127676	Chemical Engineering Journal	DMS-based polyurethane coating containing 7-amino-4-methylcoumarin	PDMS-based polyurethane with 7-amino-4-methylcoumarin was synthesized by a simple condensation polymerization method	foul-release, low surface energy, hydrophobic, non-toxic biocide	Environmentally friendly, synergistic effect, anti-bacterial, good adhesion, anti-corrosive, anti-icing, flame-resistant, self-cleaning, intelligent fluorescent response topcoat	field testing, further development needed
Lakhan, M. N., Chen, R., Liu, F., Shar, A. H., Soomro, I. A., Chand, K., Ahmed, M., Hanan, A., Khan, A., Maitlo, A. A., & Wang, J. (2023). Construction of antifouling marine coatings via layer-by-layer assembly of Chitosan and acid siloxane resin. Journal of Polymer Research, 30(4). https://doi.org/10.1007/s10965-023-03518-8	Journal of Polymer Research	A Layer-by-layer (LBL) assembly approach of alternative charged CHT/ASR multilayer films	LBL technique to build the PEM films using Chitosan (CHT) and acid siloxane resin (ASR), utilizing a dipping method between positively charged (CHT) and negatively charged (ASR) solutions for the deposition of various numbers of layers	self-polishing, foul release	Environmentally friendly, anti-bacterial, anti-adhesion	field testing needed

Supplementary Table S2. Journal title and the amount papers associated from the 100 marine antifouling studies reviewed (Table S1).

Name of Journal	# of papers from journal (#/100)
Accounts of Chemical Research	1
ACS Applied Materials & Interface	5
ACS Omega	1
Advanced Functional Materials	2
Advanced Materials Interfaces	1
Advanced Sustainable Systems	1
Applied Catalysis B: Environmental	1
Applied Surface Science	2
Biomacromolecules	1
Biomaterials Science	1
Carbohydrate Polymers	1
Ceramics International	2
Chemical Engineering Journal	5
Coatings	1
Colloids and Surfaces A: Physicochemical and Engineering Aspects	3
Colloids and Surfaces B: Biointerfaces	2
Comparative Biochemistry and Physiology Part C: Toxicology & Pharmacology	1
Composites Science and Technology	1
Electrochemistry Communications	1
Environmental Science and Pollution Research	1
European Polymer Journal	2
Frontiers of Materials Science	1
Industrial & Engineering Chemistry Research	1
International Journal of Molecular Sciences	2
Journal of Adhesion Science and Technology	1
Journal of Coatings Technology and Research	3
Journal of Colloid and Interface Science	2
Journal of Environmental Chemical Engineering	1
Journal of Materials Chemistry A	1
Journal of Materials Chemistry B	2
Journal of Materials Science & Technology	3
Journal of Natural Products	1
Journal of Oceanology and Limnology	1
Journal of Polymer Research	1
Journal of Wuhan University of Technology-Mater. Sci. Ed.	1
Langmuir	5
Macromolecular Symposia	1
Marine Biotechnology	3
Marine Drugs	2

Name of Journal	# of papers from journal (#/100)
Materials Research	1
Materials Science Forum	1
Materials Today Chemistry	1
Microelectronic Engineering	1
Molecules	1
Organic & Biomolecular Chemistry	1
Phys. Chem. Chem. Phys.	1
Polymer Bulletin	1
Progress in Organic Coatings	12
RSC Advances	2
Science of The Total Environment	2
Small	1
SSRN Electronic Journal	3
Surface and Coatings Technology	2
The 27th International Ocean and Polar Engineering Conference	1
Tribology International	1