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Green and Sustainable Antibiofouling Coatings: A Review

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Abstract. Marine biofouling, the unwanted accumulation of living organisms on man-made structures is worsening every year. Biofouling in marine environment usually occurs in any wetted surface, submerged for a period of time in the seawater. This phenomenon happens in few stages started with conditioning and followed by attachment and colonization. Marine biofouling is a highly cost problem which requires unnecessary waste of financial source especially in fuel consumption and production time as well as safety problems. The currently used antibiofouling paint is reported to be highly toxic and threatening the marine organisms. The most commonly used antibiofouling agents are biocide-based such as the tributyl-n-tin (TBT), Sea Nine 211, Irgarol 1051, Diuron and the copper-based. The well-developed coating companies such as Hempel USA, SeaCoat Technology, Nippon Paint and many more has started their involvement in making the earths greener by introducing the non-biocide coating that is claimed to be as good as the biocide based antibiofouling coatings. Although the paints did not clearly state the antibiofouling agents used to replace biocide, the paints introduced are however, still has its own drawbacks. The secondary metabolites extracted from natural products as a defence mechanism is a potential green antibiofouling agent. Apart of its lower toxicity, natural products are also biodegradable. Further study on development of sustainable antibiofouling coating is crucial. This paper intended to review available green and sustainable antifouling compound used as paint or coating. The effectiveness and toxicity of the antifoulant will be critically reviewed.

1. Introduction

Antibiofouling coatings are special coatings applied on wetted surface to prevent the growth and attachment of merine biofouling. Common antibiofouling coatings work by diffusion of antibiofouling agent in the seawater due to chemical reaction occurs when the paint is submerged in the seawater. Marine biofouling grows on structures including deep sea oil platform, pipelines, fish nets, ships, cables as well as the bridge pillars. They basically grow on anything submerged in the seawater including on the living things such as crabs and turtles [1]. Marine biofouling became a problematic issue that needs immediate action. This is due to the significant impact left by these marine biofouling problem on daily operation of most sectors associated with marine environment. Some of serious problem arises includes a significant increase in expenses for fuel due to an increase in drag resistance and weight, cleaning the biofouling formed, harmful waste scraped from fouled surfaces into the sea, profit loss for dry docking and reduction in shelf life for the man-made structure [3]. Antifouling problems is a costly problem as it generally increases the operation cost for shipping industry for up to 50 % due to an increase in fuel consumption caused by biofouling. Besides, the need for dry docking forces the operation team to stop their operation to remove the biofouling on their structure and repaint it. Unsuprisingly, this makes the global antibiofouling coatings market published by Markets and Markets estimated a growth from USD 5.61 billion in 2015 to USD 9.92 billion in 2021. This report is projected based on the Compound Annual



Growth Rate (CAGR) which is 8.6 percent from 2016 to 2021. The market demands are projected to increase significantly each year. Rapid growth in the shipping industry is driven by the enforcement of law by the International Maritime Organization (IMO) and local authorities. The global market of antibiofouling coatings is currently dominated by the Asia-Pacific region, specifically China both for building of ship as well as ship repairs; 37.2 % China, 34.4 % South Korea and 19.2 % Japan.

The shipbuilding industry growing in India, Vietnam and the Phillipines will increase the demands for the antibiofouling coatings industry. AkzoNobel, Jotun, PPG, Hempel's Marine Paints and Chugoku Marine Paints are the main market players for antibiofouling coatings [2]. Currently used antibiofouling coating are associated with environmental concern as they are highly toxic to marine environment. The highly toxic antibiofouling coatings that leached to the seawater has increased the rate of mortality, increased the accumulation of heavy metals in bivalve and decreasing the enzymatic activity of marine organisms [9][11]. Occupational exposure during antifouling paint application is also one of the major concern as the antibiofouling paints comprises of toxic substance which are not only toxic to marine environment but to the environment as well. They usually contain high concentration of volatile organic component (VOC) that evaporated to the environment during and after the curing process of paint is completed [2].

The basic composition of the antibiofouling paint comprises of binder, solvent, antibiofouling agent, additives and pigment. Each of the main components play a major role in the performance of the antibiofouling paint. Binder acts as the agent that hold all components together on the surface of interest. In order for the paint to be possible to be applied on surface, vehicle is needed. For this case, solvent acts as the vehicle for the paint to be applied on the surface of marine structure. Solvent may be in the form of organic solvent or water but as for the antibiofouling coating, organic solvent is preferred over water as to produce a non-water based coatings. Additives are optional as it is added to improve the performance of the coating [3].

2. Review of Research on Development of Antibiofouling Coatings

The recent antibiofouling coatings mainly used in industry which are tabulated in Table 1 are the copper-based, biocide-based and non-biocide based antibiofouling coatings. Numbers of research has been conducted however unable to compete the performance of the highly toxic coating. The biocide based and metal based antibiofouling coatings are still preferred over the non- biocide antibiofouling coating mainly due to the price of both products. The problems with the biocide based antibiofouling coatings is that it also kills the non-target organism which are referring to the marine life. Moreover, the paint scraped during dry docking and repainting process that are not properly disposed will do more harm than good. The scraped paint from biocide-based coating are still functioning and when it is being washed away in the sea, marine life will be affected as biocide also kills the non-target organisms too [4].

2.1. Biocide based Antibiofouling Coatings

Biocide has been widely used as antibiofouling agents due to its efficiency in preventing biofouling problem. Diuron has been reported to perform well for at least 2 years. However, the toxicity concentration may also provide a toxic environment to the marine life living nearby the ships or painted surface. Due to environmental concern, the usage of these antibiofouling coating has been restricted only for ships with a minimum of 25 m in length. For the antibiofouling coatings made from Poly (ϵ -caprolactone) diol and butenolide, the polymer degrades in seawater as it is a soluble matrix paint. However, the rate of dissolution in water depends on the composition of the antifouling paint itself. A faster rate of dissolution may reduce the longevity of the coatings and increase the accumulation of antibiofouling agent in the seawater [34]. Enzyme and marine organisms may increase the rate of degradation of the coating and reduce its lifetime. This type of coating is highly dependent on environmental parameter as the rate of degradation of butenolide are highly affected by temperature too. Addition of rosin as binder may help in increasing the self-polishing rate and improve the late release of butenolide [14]. The longest lifetime of antibiofouling coatings are the one that incorporating the TBT. TBT works by providing a highly toxic environment that not only marine fouling organisms but

other marine organisms too with toxicity reported for as low as 1.87 µg/L (LC50) which is extremely toxic in low concentration. The trends are presented in Table 1.

Table 1. Toxicity and Longevity of Biocide based Antibiofouling Coatings [8][10].

No.	Composition of Antibiofouling Coating		Toxicity (LC50)	Lifetime (Years)	Reference
	Antibiofouling agent	Pigment			
1	Tributyl-n-tin (TBT)	Zinc oxide (ZnO), talc, Barium sulphate (BaSO4)	1.87 ppb (Schmidtea mediterranea)	5	[7]
2	Copper (I) Thiocyanate	Copper metal oxide or sulphide	5 ppm (Rat, inhalation)	2	[9]
3	Irgarol 1051	-	1.62 ppm (Brine shrimp)	1-2	[10]
4	Diuron	Copper oxide (Cu2O)	6.00 ppm (Zebrafish)	3	[11]
5	Sea Nine 211	Copper oxide (Cu2O)	2.70 ppb (Rainbow trout)	>1	[12]
6	Chlorothalonil	-	76.00 ppm (Rainbow trout)	2	[13]

2.2. Metal based antibiofouling coatings

One of the most widely used metal based antibiofouling agent is copper oxide and zinc oxide. Copper and zinc oxide has been widely used in antibiofouling coatings as it has the antibiofouling properties as well as imparting colours to the coating produced. Its usage has been phased out in the USA in January 2018. Copper and zinc oxide has shown a great performance for at least 3 years. However, the toxicity of copper oxide against zebrafish is as high as 242.40 ppb and zinc oxide against rainbow trout is 1.10 ppm which is very toxic to the marine organisms.

2.3. Non-biocide based antibiofouling coatings

The non-biocide based antibiofouling coatings has also been slowly introduced to the market since the copper based and biocide based antibiofouling coatings has been restricted. The non-biocide based antibiofouling coatings are able to serve as long as the biocide based antibiofouling coatings. One of the main problem with the non-biocide antibiofouling coating is that its efficiency is not as good as the biocide based [31]. Although it can serve longer than the biocide based, the amount of fouling observed on the non-biocide antibiofouling coatings are more severe and this makes the cleaning process harder. Most of the non-biocide antibiofouling coatings required mechanical and physical removal rather than cleaning by movement of water. Thinning has also been observed in most of the non-biocide based antibiofouling coatings which is also caused by the composition of the coatings itself. The longevity of non-biocide antibiofouling coatings recently available in market is able to compete the biocide based antibiofouling coatings which are in the range between 2 to 7.5 years. Furthermore, the toxicity of the non-biocide based antibiofouling coatings are significantly lower than the biocide based antibiofouling coatings as the LC50 less than 1000 ppm is considered as non-toxic. Details of the products are tabulated in Table 2.

Table 2. Non-biocide Antibiofouling Coatings [25][16].

No.	Company	Paint Name	Toxicity (LC50)	Lifetime (Years)	Reference
1	Ecological Coatings	EC-4300	1283 ppm ^a	3	[16]
2	Hempel USA	Hempasil X3	2447 ppm ^a	7.5	[17]
3	Oceanic Surfaces International	ECO-5	2212 ppm ^a	5	[18]

4	KISS Polymers	MegaGuard Ultra LiquiCote	2311 ppm ^c	3	[19]
5	International Paint	VC Performance Epoxy	2142 ppm ^b	5	[20]
6	Xurex Nano-Coating	ProGlide Plus	1947 ppm ^a	5	[21]
7	Seacoat Technology	Sea Speed GC V4	-	5	[22]
8	Petit Paint	Klean N' Klean	2309 ppm ^c	5	[23]
19	Sound Specialty Coatings	AQUAPLY M	1281 ppm ^c	5	[24]
10	Specialty Products	Polyshield HT		3	[25]
11	International Paint	Intersleek 900	2102 ppm ^b	5	[26]
12	Ram Protective Coatings	Ceram-Kote 99M	1117 ppm ^c	3	[27]
13	Jotun	SeaForce 30	-	3	[28]
14	Nippon Paint	AquaTerras	-	5	[29]

^a Brine shrimp

^b Rainbow trout

^c Zebrafish

Non-biocide based antibiofouling coatings was studied to decide whether or not it is suitable to replace biocide. The performance of poly (ϵ -caprolactone) diol and butenolide [14], polyelectrolyte chitosan Schiff [6], sodium benzoate has been studied as non-biocide antibiofouling agent. However, the longevity is not as good as biocide based antibiofouling agent. These agent can only last for 2 years for poly (ϵ -caprolactone) diol and butenolide, 1 year for polyelectrolyte chitosan Schiff and 2 years for sodium benzoate. The toxicity of both poly(ϵ -caprolactone) diol and butenolide and polyelectrolyte chitosan Schiff were reported to be non toxic which are 2400 ppm and 3738.47 pm respectively [14][6]. Sodium benzoate however, is toxic with toxicity of 484 ppm [32]. Besides the drawbacks that the non-biocide antibiofouling coatings has, there are also much more on the advantages side. One of it is it does not need to be reapplied as frequent as the copper- based coatings. The copper-based coatings need to be reapplied for at least once in 2 years while the non-biocide coatings may last for up to 7.5 years [34]. This helps in reducing the operational cost too. Next is some of the non-biocide coating can be easily applied by using roller to achieve the smooth and standard condition of coated surface but some of the biocide-based coating need to be applied only by spraying method to achieve the desired surface condition. This reduce the amount of time needed for repainting process [3]. Stripping is also only required during initial application of non- biocide antibiofouling paint only [3]. A lesser surface preparation process leads to a lesser production cost [30]. Although it has been considered non-toxic, dry docking of the coated surface should be done properly by collecting all the scraped coating and dispose it accordingly as it may still be harmful to the environment especially if it is swallowed by marine life due to its colour or properties that might mimic their food source. However, in terms of production cost, the manufacturing cost for the non- biocide antibiofouling coatings are higher than the biocide based antibiofouling coatings due to the complex production process and low percentage of yield if natural sources is used [22]. Delamination of coating are also reported for the non-biocide based antibiofouling coatings [30].

2.4. Antifouling agent from natural sources

Moving towards the greener approach, one of the best candidate to replace the current antifoulant is the one from natural sources. In truth, numbers of research have been conducted to study the potential of the natural antifouling agent in combating the marine biofouling problems. The biofouling agent can be extracted from living things including plants, animals and microorganisms. These natural sources came with plentiful advantages. Natural antifoulant are commonly known to be safe-to-use. Since natural antifoulant are commonly come from animal, plants, fungi and bacteria, most of it are proven to be highly secured to be. Besides, natural antifoulants are also not harmful to the environment as well in compared to biocide-based antifoulant. They are biodegradable and disposing the antifoulant back to the environment will not cause harm or pollution since it is the origin of the antifoulant itself. In addition, natural antifoulants are obtained from a renewable sources and it can be utilized and reproduce without harming the environment [30]. In addition, most of the extracted antifouling agent from natural sources

are pigmented, hence, no additional metal-based pigment is needed as to give colour to the antibiofouling paint. The pigment incorporated in the antibiofouling paint is also one of the major source of metal accumulation in seawater due to high leaching rate in the environment [31]. The potential natural antibiofouling agent are as tabulated in Table 3.

Table 3. Potential antifouling agent from natural sources [33][31].

Source	Species (Local Name)	Colour	MIC	Reference
Plant	<i>Morus rubra</i> (Mulberry fruit)	Dark purple	8.00 ppb ^a	[32]
	<i>Beta vulgaris</i> (Beetroot)	Red	39.10 ppb ^b	[33]
	<i>Oryza sativa</i> (black rice)	Dark brown	6.25 ppm ^b	[34]
	<i>Curcuma longa</i> (turmeric)	Yellow-orange	800.00 ppb ^b	[35]
	<i>Capsicum annuum</i> (capsicum)	Red	256.00 ppb ^c	[36]
	<i>Rhodophyta sp.</i> (Red algae)	Light blue	10.00 ppb ^b	[37]
	<i>Callistemon citrinus</i> (Bottle brush tree)	Orange	2.50 ppb ^b	[38]
Bacteria	<i>Vogesella indigofera</i>	Violet	125.00 ppm ^b	[39]
	<i>Hahella chejuensis</i>	Red	0.40 ppm ^b	[40]
	<i>Micromonospora lupine</i>	Orange-red	0.31 ppm ^b	[41]
	<i>Chromobacterium violacein</i>	Violet	1.25 ppb ^b	[42]
	<i>Chryseobacterium indologenes</i>	Yellow	4.00 ppm ^b	[43]
	<i>Paracoccus zeaxanthinifaciens</i>	Yellow	15.9 ppm ^b	[44]
	<i>Agrobacterium aurantiacum</i>	Pink-Red	0.80 ppm ^b	[40]
	<i>Paracoccus carotinifaciens</i>	Pink-Red	2.50 ppb ^b	[45]
	<i>Pseudomonas argentinesis</i>	Yellow	625.00 ppb ^b	[46]
<i>Streptomyces shaanxiensis</i>	Blue	-	[47]	
Fungi	<i>Aspergillus versicolor</i>	Yellow	0.62 ppm ^b	[48]
	<i>Stemphyllum lycopersici</i>	Red	3.13 ppb ^b	[49]
	<i>Talaromyces verruculosus</i>	Red	-	[50]
	<i>Penicillium oxalicum</i>	Red	2.12 ppm ^b	[42]

^a*Streptococcus mutans*

^b*Staphylococcus aureus*

^c*Pseudomonas aeruginosa*

Table 3 summarizes the potential natural antifouling agent extracted from natural sources. Based on Table 3, the minimum inhibitory concentration (MIC) which is the minimum concentration at which the compound will act as the antibacterial agent. The MIC of the natural antifouling candidate is low, hence, only a low concentration of compound is needed for the compound to be effective. This is an advantage as referring to the drawbacks of natural antifoulants regarding the high production cost and low percentage of yield. When only a low concentration of antifoulant is needed to inhibit the targeted microorganisms from producing the extracellular polymeric substance (EPS) that cause biofilm, a lower production is sufficient as to cater the needs to be incorporated in the antibiofouling paint [31].

Apart from its advantages comes the drawbacks of natural antifoulants. One of the major drawbacks is in terms of cost. Natural antifoulant extracted usually are low in percentage of yield in compared to the biocide-based antifouling agent. Hence, the production cost is higher in compared to the biocide-based antifoulant. Besides, solubility of the extracted antifoulant are also very low which makes the process to incorporate the antifoulant in the antifouling paint to be harder. In addition, although the antifoulants extracted comes together with colour, the colour of the antifoulant are highly unstable and further process such as stabilization needs to be done to stabilize the antifoulant and the physiochemical properties that it carries together [31]. In spite of all the cons, this potential antibiofouling agent will save the marine life is a sustainable option to adopt. A comprehensive research should be carried out to make this natural sources feasible to be commercially used as antibiofouling agent to combat biofouling problem and at the same time, save the world. Current research such as hydrogel based antibiofouling

coating has not yet able to promise an efficiency as good as the biocide-based antibiofouling coatings [14].

3. Conclusions and Future Perspectives

After years of neglect, environment fates need to be given its right. The development of sustainable antibiofouling coatings need to be prioritize as its impacts is severe in all stages of life and a lot of living things especially the one associated with marine environment. There are numbers of available antibiofouling coatings in the market and endless number of antibiofouling agent candidates in the world. Generally, the toxicity of the antibiofouling agent and the final products and its performance are the two factors that differ one coating with another. EC50 and LC50 values are often used as a standard in measuring the toxicity of the antibiofouling coatings. Development of a sustainable antibiofouling coatings are crucial as to reduce the footprint of current antibiofouling coatings. The main challenge in development of sustainable antibiofouling coatings has been highlighted previously [15]. Natural products are one of a suitable candidate to replace the toxic antibiofouling agents. However, due to inadequate and thorough research as well as the production cost, it has not widely been studied especially in a long term effect. Hence, as to achieve the Sustainable Development Goals, specifically goal number 9, the life below the water, an immediate action needs to be taken into account. One of it is by developing a sustainable antibiofouling coatings.

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