

ANTIFOULING STUDY OF ZINC TANNATE (TZn) FOR MILD STEEL IN TROPICAL SEAWATER

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Abstract

This study was coordinated to investigate the potential of tannin extracted from mangrove bark (*Rhizophora apiculata* sp.) as an antifouling agent. Using natural resources as antifouling agents is an environmentally friendly alternative in industrial applications. Extracted tannin was formulated in a readily available epoxy coating and then coated on the mild steel samples. The samples were immersed in seawater for 60 days in Chendering Port and Marang Jetty. A screening test was performed via the anti-biofilm assay method to characterise the antimicrobial abilities. The IC₅₀ of zinc tannate extract against *Bacillus Cereus* and *Salmonella* sp was 2.6 mg/ml and 28.75 mg/ml, respectively. It was discovered that fouling attachment is more noticeable in Chendering Port due to the physical parameters of the surrounding waters. Furthermore, the percentage of the weight acquired at Chendering port is higher than at Marang jetty. Surface inspection using digital photographs and scanning electron microscopy (SEM) reveals a denser tubeworm and mollusc attachment on the substrate without TZn. Higher tannin concentration has resulted in greater antifouling potential. Overall, the findings implicate that TZn has great potential as an antifouling additive in commercial marine coating.

Keywords: Antifouling; Mangrove Bark; Mild Steel; Seawater; Zinc Tannate

Introduction

Fouling and corrosion are two frequent phenomena when structures are exposed to seawater. Due to the complex marine environment and marine creatures, researchers discovered that microbiologically influenced biofouling and marine corrosion are two major processes (Nall *et al.*, 2017). Biofouling is the colonisation and proliferation of organisms on manufactured objects submerged in seawater which can build up on the surface of the engine cooling tube, as well

as the water supply and turbine systems. Biofilms and macrofoulers are two types of biofouling (Flemming, 2011). Biofouling control is costly, with lots of money spent on biocides, cleansers, and antifouling products worldwide (Winfield *et al.*, 2018). Biofilms substantially influence vessels, raising service costs and increasing greenhouse gas and other pollutants due to excessive fuel use (Soroldoni *et al.*, 2017; Vidales and López, 2020).

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Antifouling is defined as the prevention of fouling organisms from accumulating. Antifouling coatings work by poisoning any marine life that tries to attach itself to the ship's submerged surface. The biocide, which functions as an antifouling agent, was blended with oil, epoxy, or vinyl-based binder in traditional paints. However, some biocides are toxic and carcinogenic, yet it still has been the major choice in antifouling coating for marine applications (Vilas-Boas *et al.*, 2017; Li and Ning, 2019).

Hence, there is an increasing trend to discover the antifouling agent extracted from natural resources. Antifoulants derived from natural products have been considered one of the finest alternatives to tributyltin (TBT). Mangrove trees have long been considered a natural antifoulant due to their abundance of structurally varied bioactive chemicals like tannins (Zulkifli *et al.*, 2019). Tannins are a type of water-soluble polyphenol found in various herbaceous and woody plants. Tannin, the active ingredient in the removed mangrove bark, contributes to TZn's antifouling activity. Tannin either kills or inhibits the growth of microorganisms. The antimicrobial activity of phenolic substances alters microbial cell permeability, resulting in the loss of macromolecules from the cell core (Alexandre *et al.*, 2019).

Tannin is a polyphenolic substance that can impact Gram-positive and Gram-negative bacteria's cell membranes. As a result, it may alter the hydrophobicity and charge of the cell surface, resulting in cytoplasmic leakage (Cho *et al.*, 2001). The phenolic compound's hydroxyl group disrupts the production of the conditioning film, which is the first step in the fouling process and repels microbe attachment in seawater. Several researchers discovered the usage of tannin in antifungal coatings. Gámez-Espinosa *et al.* (2021) reported that their sol-gel coating incorporated with tannin shows a 100% inhibition of their targeted bacteria.

The application of silver nanoparticles also provides another insight into the antifungal coating. High inhibition of silver nanoparticles was discovered towards yeast, mould and bacteria in a sol-gel coating method, as reported by Żarowska *et al.*, 2019. Biologically synthesised silver nanoparticles showed excellent antifungal behaviour towards several target bacteria (Khan *et al.*, 2020). Chitosan/pectin silver nanoparticle films have been developed by Akalin *et al.*, 2022. They found that the formulated coating exhibits an antibacterial influence on the bacterial strain.

Tannic acid, a hydrolysable gallotannin, is another major gallotannin, whereas catechin is a non-hydrolysable gallotannin. Other researchers discovered that the presence of catechin, oxacillin,

ampicillin, and cefalexin boosts antibacterial activity (Lima *et al.*, 2016). As a result, developing an ecologically friendly antifouling paint is critical to reducing hazardous chemicals released by conventional biocide. To decrease fouling, researchers have recommended using natural resources such as cabbage, zinc nitrate, copper, polyaspartic acid, and plant tannin (Idora *et al.*, 2015; Suriani *et al.*, 2016; Gao *et al.*, 2019; Zulkifli *et al.*, 2019; Chasse *et al.*, 2020). Therefore, this paper explained the potential of tannin derived from *Rhizophora apiculata* as an antifouling agent in epoxy coating. Two marine port was chosen to deploy the sample as most researchers focused on the lab-based experiment. Bioassay and weight loss methods were employed to investigate the potential of the formulated coating to inhibit fouling growth on the substrate surface.

Materials and Methods

This study used mild steel grades A36 with dimensions of 25mm x 25mm x 4mm as a substrate. According to ISO8501-1:1988, the surface of mild steel was polished using a power tool cleaning procedure with a copper brush. The metal was then immersed for 2 to 3 minutes in thinner to remove any grease or grime from its surface. The metal was then placed in a desiccator to keep it from rusting.

Tannin extraction follows ASTM D6405-99:2009 Standard Practice for Extraction of Tannins from Raw and Spent Material. At room temperature, finely crushed mangrove barks were immersed in 70% aqueous acetone. The acetone was then removed under pressure, and the yield was freeze-dried. Because of their higher water solubility, tannin extracts are not immediately absorbed into the coatings. As a result, zinc nitrate was added to produce zinc tannate (TZn), a less soluble compound (Idora *et al.*, 2015).

Three items were pipetted into 96 wells of a microtiter plate: bacteria, nutritious broth, and a sample of tannin extract. It was then placed in an incubator for 24 hours at 37°C to allow biofilms to form. The cells' optical density (OD) was measured, and the value had to be less than 0.2. The medium was then carefully removed, and the microtiter plate wells were rinsed with 200 l of distilled water per well and dried using a multichannel pipette.

The microtiter plate wells were dyed for 20 minutes at room temperature with 200 l of 0.4 percent crystal violet. The discolouration was removed, and the wells were carefully cleaned with 200 liters of distilled water. The crystal violet was solubilised in 200 l of 90% alcohol and incubated at room temperature for 10 minutes in the shaker after



Figure 1. Schematic diagram of the deployed samples holder

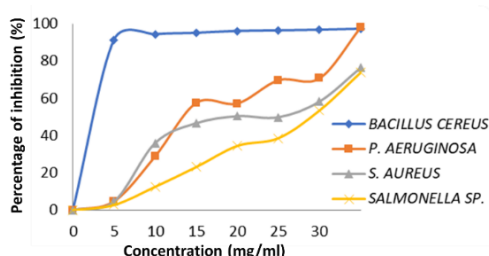


Figure 2. Percentage inhibition of zinc tannate extract towards targets bacteria

being dried for 15 minutes. The motion plate was read at 595 nm using a UV-Vis spectrophotometer. Equation. 1 was used to get the inhibition percentage (1).

$$\text{Inhibition \%} = \frac{\text{OD Negative Control} - \text{OD samples}}{\text{OD Negative Control}} \times 100\% \quad (1)$$

As an undercoat, Paralux P268HS Epoxy High Solid Zinc Phosphate Primer was used, and zinc tannate (TZn) was mixed into the paint at varied ratios. Thinner was applied as a solvent to thin the paint to flow more easily on the surface. Finally, a hardener was used to absorb moisture from the paint and speed up the drying process. The Epoxy base and hardener were packed at a 4:1 ratio. Table 1 illustrates the new paint mixture for compressing air sprayers to coat the sample. After the coating application has been completed, thickness control is performed on the dry film thickness (DFT).

To conserve freshwater resources, seawater is commonly employed as a cooling medium in ship circulation cooling systems (Gao *et al.*, 2019). As a result, natural seawater was employed in this experiment. Figure 1 shows a schematic diagram of the deployed cage containing coated and uncoated

Table 1. The formulation of antifouling paint

Type of paint	TZn (g)	Epoxy base (ml)	Hardener (ml)	Thinner (ml)	Total paint (ml)
1(control)	-	100	25	150	275
2	2	100	25	150	275
3	4	100	25	150	275
4	6	100	25	150	275

Table 2. Optimum inhibition concentration of zinc tannate at 50%

Bacteria Samples	IC ₅₀ (mg/ml)
<i>Bacillus Cereus</i>	2.6
<i>P. Aeruginosa</i>	13.75
<i>S. Aureus</i>	22.5
<i>Salmonella Sp.</i>	28.75

samples. Four cages were deployed for the immersion test, each containing five coated specimens with varying TZn concentrations.

Five pieces of specimens were hung in each cage, and the immersion time was 60 days. At both Chendering Port and Marang Jetty, the cages were submerged to the bottom. These sites were chosen because of their industrial activity, which encourages fouling organism adhesion.

In the presence and absence of TZn, the initial weight of the coated specimens was recorded. After 60 days of immersion, the plates were weighed again, and the data was recorded. Equation. 2 was used to get the weight %.

$$\text{Weight gain(\%)} = \frac{\text{final weight} - \text{initial weight}}{\text{total weight}} \times 100 \quad (2)$$

Results and Discussions

Inhibition of TZn towards Bacteria

In 96 well plates, four bacterium samples were pipetted. The dilution test plot in Figure 2 indicates the inhibitory efficacy of TZn against target microorganisms such as *Bacillus cereus*, *Pseudomonas aeruginosa*, *Staphylococcus aureus*, and *Salmonella Sp.* *Bacillus Cereus* was found to have the highest inhibition efficiency, followed by *P. Aeruginosa*, *S. Aureus*, and *Salmonella Sp.* in that order. Meanwhile, *Bacillus Cereus* requires the least amount of TZn (2.6 mg/ml) at 50% inhibition efficiency, while *P. Aeruginosa*, *S. Aureus*, and *Salmonella Sp.* require 13.75, 22.5, and 28.75 mg/ml, respectively, as shown in Table 2. It can be inferred that lower TZn loading is associated with a higher rate of inhibition efficiency. As the concentration of TZn, the inhibition efficiency was also found to increase.

Table 3. Average seawater parameters at the port and jetty

Location of Sample	Salinity (ppt)	Temperature (°C)	Dissolved oxygen (mg/l)
Chendering port	32.7	30.7	6.58
Marang jetty	25.36	30.46	6.02

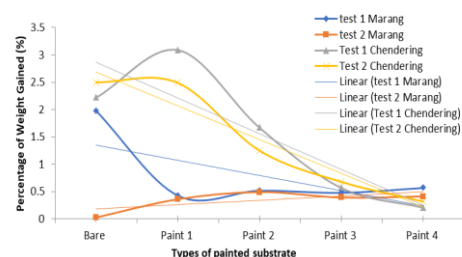
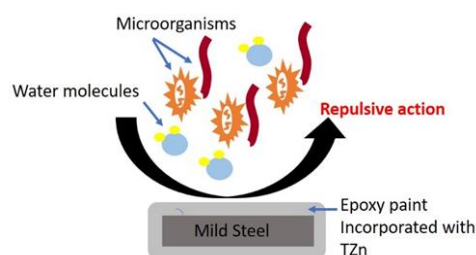
The trend was aligned with other findings reported in the literature (Ostovari *et al.*, 2009; Li *et al.*, 2016). Several authors also emphasize the ability of tannin as an antimicrobial coating. Xie *et al.* (2021) confirmed that their composite coatings consisting of Lignin/Tannin/ZnONP inhibit the growth of Gram-negative/positive bacteria growth. In addition, the application of tannic acid and tung oil to treat wood shows that both substances have inhibited the growth of brown rot fungi due to their antioxidant property (Peng *et al.*, 2021). Based on dedicated support from previous findings, it can be inferred that TZn is promising to be employed as an antifouling agent.

Weight gained percentage measurement

Figure 3 shows the weight increment percentage for all immersed samples at two separate sites. When comparing Chendering port to Marang jetty, it was discovered that Chendering port has a larger weight growth %. Paint 1 has the largest weight growth % of all the samples immersed in the Chendering port, followed by bare, paint 1, paint 2, paint 3, and paint four samples. The weight acquired percentage decreases as the concentration of TZn increases, which is linked to the antifouling inhibitory effect of TZn.

In the meantime, the trend for samples immersed in the Marang jetty shows an increase in weight gained % from bare to painted samples. When TZn is added to paint 2, paint 3, and paint 4, the percentage of weight gained increases little compared to paint 1. It could be because the samples have been covered in mud, preventing fouling organisms from attaching to their surface. It is consistent with the results of sediment transport published by previous researchers, who discovered that the Marang jetty transports more silt by river than the Chendering port (Marghany, 2003).

Table 3 shows the variance of physical properties of the surrounding water, which was gathered from earlier researchers (Gasim *et al.*, 2015; Suriani *et al.*, 2016). The salinity and dissolved oxygen levels range significantly, which aids in the growth of fouling organisms on the substrate. Furthermore, physical characteristics of seawater, such as salinity, temperature, pH, dissolved salts, and oxygen content, have been shown to have a major impact on fouling

**Figure 3. Weight gained as a percentage of painted substrate****Figure 4. Mechanism of epoxy paint incorporated with TZn towards microorganism inhibition**

on immersed solid surfaces (Tan *et al.*, 2017). Hence, the incorporation of TZn will inhibit the microorganism from accumulating on the coated mild steel surface, as shown in Figure 4.

Scanning electron microscopy (SEM)

Figure 5(a) shows the digital image of the surface for a coated sample without TZn loading. The surface exhibits a smooth finishing with uneven roughness can be spotted. Figure 5b shows a digital image for a coated sample with 6 g of TZn loading. The sample's surface exhibits a dark small sport and a rough surface that indicates the presence of TZn on the sample's surface.

Tubeworms and molluscs are attached to the substrate surface in a shot made with a digital camera of samples. In comparison to Figure 6 (b), where the metal coating does not contain TZn extract, the size of fouling organisms was greater in Figure 6a. Figure 5(b) shows the TZn-coated substrate, which has less fouling organism adhesion than the control substrate. The result could synergistically affect the chloride ion in seawater (Guerraf *et al.*, 2018).

The attachment of fouling organisms was more obvious in samples without TZn than in samples with TZn, as illustrated in Figures 7 (a) and (b) by comparing scanning electron microscope (SEM) micrographs. Researchers experimenting with copper and brass found comparable results (Ren *et al.*, 2020). Hence it implies that TZn minimises the likelihood of fouling organisms attaching to the substrate surface.

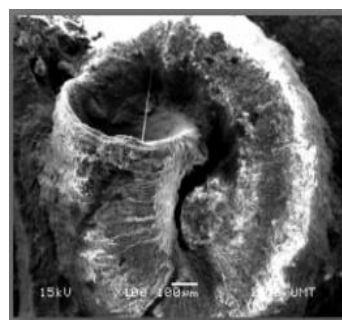


(a)

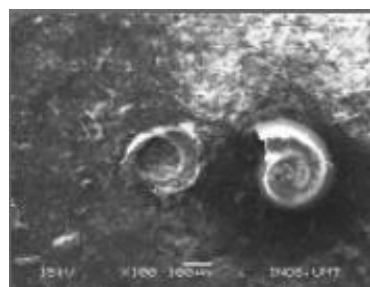


(b)

Figure 5. Photograph of the substrate before immersion a) a control sample without TZn loading and b) a sample with 6g of TZn loading



(a)

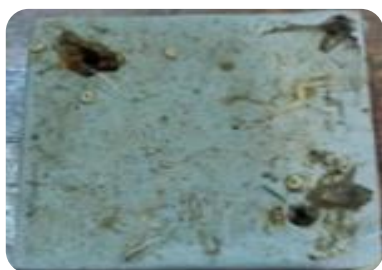


(b)

Figure 7. SEM micrographs of the surface of A36 after 60 days of immersion at x100 magnification a) a control sample without TZn loading and b) a sample with 6g of TZn loading



(a)



(b)

Figure 6. Photograph of the mild steel after immersion a) a control sample without TZn loading and b) a sample with 6g of TZn loading

Conclusions

Antifouling effects of zinc tannate (TZn) were identified after it was included in epoxy paint. TZn inhibition efficiency was better than 60% for all target bacteria, with *Salmonella Sp.* significantly inhibited at 50% TZn loading. Because of the higher reported physical factors such as salinity and dissolved oxygen, Chendering port has a bigger impact on fouling attachment. Because the weight obtained for substrate with TZn loading is lower than paint without TZn, adding TZn to the epoxy paint has reduced fouling adhesion. Digital photography and scanning electron microscopy reveal that tubeworms and molluscs dominate fouling attachment on a coated substrate without TZn, whereas fouling attachment on a coated substrate loaded with TZn is smaller. The presence of TZn inhibits fouling growth, according to this research. It can be concluded that TZn has a high potential for usage as an antifouling agent and for promoting natural products to replace environmentally dangerous substances to reduce fouling organisms.

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