Effects of Mangrove Plant Extraction on Adhesion Strength in Coating of Epoxy

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Abstract. Steel is an alloy of ferrous metal with a maximum carbon content of 2%. This material is widely used for jetty building structures at rivers, estuary, and coastal areas. Corrosion of steel can occur due to several factors, such as the steel material itself and the surrounding environmental factors. One of the corrosion is biocorrosion that occurs due to the presence of biofouling. This study aims to determine the effect of adding extraction of mangrove (Avicennia marina) as an antifouling compound mixed in epoxy paint through adhesion strength parameter. Then the alloy was applied as a coating on the ASTM A36 steel specimen. The variation of coating composition were 100% of epoxy paint, 90%:10% (epoxy paint and extraction of Avicennia marina), 80%:20% and 70%:30%. The results showed that the adhesion strength occurred in 100% of epoxy paint (10.15 ± 0.64 Mpa). The adhesion strength was 8.86 ± 0.21 Mpa, 8.50 ± 0.03 Mpa, and 8.19 ± 0.07 Mpa at ratio of composition coating 90%: 10%, 80%: 20%, and 70%: 30%, respectively. In conclusion, the more mangrove extraction was added, the smaller the adhesion strength.

1. Introduction
The development of the industry in the maritime construction sector is getting better and better. In practice, metal is one of the primary materials used on a large scale due to its strong physical properties and abundant availability. However, there are still problems related to iron which are often the reason for the increase in production costs for maintenance of the problems that occur. One of these problems is biofouling. Biofouling is a collection of, either macro or microorganisms that attach to the surface of structures submerged in seawater, such as bacteria, fungi, algae, algae, and shellfish. This continuous adhesion of biofouling can cause further problems such as increasing the corrosion rate of the structure, weakening the carrying capacity of the rig, reducing the readability of the sensor, reducing the maximum speed of the ship, increasing the consumption of materials, and many more. So it takes more maintenance costs which can result in losses, both in cost and time.

An effort that can be conducted to minimize the adhesion of biofouling on the structure is coating. In general, coatings are used to increase the service life and reliability of structures, such as enhancing the appearance of structures, protecting against corrosion, and slowing down the maintenance life of structures. Since new problems related to biofouling were found, coatings have been started using antifouling paints, namely paints containing synthetic chemical compounds that are useful for slowing
down the adhesion of biofouling. Initially, antifouling paints were made using light chemical compounds, such as arsenic, or mercuric oxide derived from linseed oil until tributyltin (TBT) was found.

Tributyltin was considered the most efficient to inhibit the adhesion of biofouling for a long time [1]. Tributyltin contains organotin compounds that are very stable in dark places (for example the seabed) and do not quickly degrade in an environment that impacts the seabed’s pollution for a long time. Tributyltin is also very toxic to mollusks, crustaceans, and algae species such as shrimp, crab, barnacles, shellfish, algae to algae. Since 2008, TBT-based antifouling paint has been banned by the International Maritime Organization (IMO). The use of natural ingredients as antifouling agents was proven by Santi et al. (2014) [2]. Seaweed extract can inhibit film formation, thereby slowing the process of adhering to biofouling. Bazes et al. (2006) [3] and Purnama et al. (2010) [4] reported that red algae and green algae were capable of being antifouling agents due to they have active compounds as antibacterials. Amin (2017) [5] reported that Terminalia catappa or Ketapang leaf extract affects the amount of biofouling attached to the material’s surface.

Another potential antifouling substance was mangroves. Mangroves contain phenolic compounds in the form of alkaloids, tannins, flavonoids, steroids, and phenolics [6], which are antibacterial compounds and can provide toxic effects on biofouling. This study used the Avicennia marina due to this mangrove plant grown primarily on the Wonorejo Mangrove Eco-Forest area. Usage of Avicennia marina for antifouling is still limited so that the purpose of this study was to determine the effect of adding extraction of mangrove (Avicennia marina) as an antifouling compound mixed in epoxy paint.

2. Method
This research was conducted in laboratory at the Department of Ocean Engineering, Institut Teknologi Sepuluh Nopember (ITS), Surabaya. There were seven steps of the research, i.e., mangrove collection, mangrove plant extraction, preparation of material, coating process, determination of thickness coating, adhesion strength test, and statistical analysis. All steps were explained in the paragraph below this.

2.1. Collection of Mangrove
Mangrove plants with species of Avicennia marina were collected at Wonorejo Mangrove Eco Forest in East Surabaya. Around 5 kg of Avicennia marina, twigs were put into the plastic bag, brought to the laboratory. All those twigs were washed to remove soil at those plants. Figure 1 showed twigs Avicennia marina collection.

![Figure 1. Sample of Avicennia marina.](image)

2.2. Preparation of Mangrove Extraction
The process of Avicennia marina extraction was carried out using the maceration method. The maceration method was carried out for 14 days to obtain maximum extraction results. Based on Zhang et al. (2018) [7], the maceration method was a straightforward extraction method with a long extraction time disadvantage. Twigs of Avicennia marina mangrove were cleaned, then dried in an oven based on a method from Titah et al. (2013) [8]. The dried twigs were blended into powder, and then those were immersed in ethanol solvent of 96% (OneMed, Indonesia) at room temperature for seven days. The ratio of Avicennia marina and the organic solvent was 1:4, so that 1 gram of Avicennia marina powder was immersed in 4 mL of organic solvent. In this study, 3 kg of mangrove twigs were dissolved using 12 L
ethanol of 96%. Then the solution was filtered to separate the filtrate and soaking dregs. The obtained filtrate was evaporated to obtain the extract. Figure 2 showed the *Avicennia marina* mangrove twigs were in the form of liquid, and they were prepared into three concentrations to be dissolved in epoxy paint, namely 10%, 20%, and 30%.

2.3. Preparation of material  
This research was conducted using ASTM A36 steel as a specimen that is often used in structures with variations in the concentration of the extract to be mixed in the paint. ASTM A36 steel specimens were prepared with the dimension of 90 x 40 x 10 mm. There were eight plates for adhesion strength testing. After the specimens had been cut, then those surface specimens were cleaned using sandpaper with grid 60. The grinding process is used as a grinding tool to obtain good results. Figure 3 showed the specimens were ready to use for the following experiments, coating process, and adhesion strength test.

2.4. Process of Coating  
The NIPPON 0848 of epoxy paint (Nipsea Paint and Chemical, Gresik) was used in this study. The coating process was carried out using a brush with two applications. The process was conducted for two days to get a dry specimen. These steps must be conducted due to the specimens were dry before they re-apply the previous coating. According to the technical data sheet NIPPON 8048 [9], repainting can only be conducted seven hours after the initial painting. The combination of epoxy paint and mangrove extract was carried out on this coating in Table 1.
2.5. Determination of Dry Thickness Coating
After the coating on all specimens was completely dry, the testing of dry film thickness coating as conducted to obtain the thickness coating was similar at all specimens’ sides. The dry film thickness coating was measured using the Dry Film Thickness Gauge. In this study, Dry Film Thickness measurement was carried out at three points for one specimen.

2.6. Adhesion Strength Test
The adhesion strength testing was carried out using the pull-off method based on ASTM 4541-02 (2017) [10]. This testing aimed to determine the adhesion of the paint to the test material. The adhesive pull-off tester lever was pressed until the dolly was released from the specimen. After that, the portable adhesive tester displayed the value of the adhesive strength of the paint on the specimen. The adhesion strength testing was carried out using the Self Aligning Adhesion Tester type V based on ASTM 4541-02 standard method [10]. This adhesion strength testing results were shown on the adhesion between the glue particles and the epoxy coating, as shown in Figure 4.

![Figure 4. Results of Adhesion Strength Testing.](image)

2.7. Statistical Analysis
The experimental data of coating composition, dry film thickness coating (DFT), and adhesion strength were subjected to an analysis of variance (ANOVA) using SPSS Statistics for Windows version 21.0 (SPSS, Inc., Chicago, IL). Statistical significance was defined as p < 0.05.

3. Results and discussion
Figure 5 shows the results of dry film thickness coating (DFT) measurement. The dry film thickness coating results were 258.17 ± 1.65 µm, 256.5 ± 0.24 µm, 254.67 ± 0.47 µm, and 253.17 ± 1.65 µm at code of specimens of E100M, E90M10, E80M20, and E70M30, respectively. ANOVA one-way analysis of coating composition and dry film thickness coating (DFT) was not significantly different. It indicated that the dry thickness coating on all specimens was similar. So that all specimens can be used for the next experiment, i.e., adhesion strength testing.

![Figure 5. Results of dry film thickness coating testing.](image)
The results of the adhesion strength testing on all specimens are shown in Figure 6. The adhesion strength results were 10.15 ± 0.64 Mpa, 8.86 ± 0.21 Mpa, 8.50 ± 0.03 Mpa, and 8.19 ± 0.07 Mpa at code of specimens of E100M, E90M10, E80M20, and E70M30, respectively. Based on ANOVA, the one-way analysis of coating composition and adhesion strength was significantly different. It indicated that the adhesion strength on all specimens was not similar. According to ISO 4624 (2016) [11], the results of the adhesion strength test on all of these specimens were upper than the acceptance criteria (>5 Mpa). Figure 8 showed a decrease in adhesion strength that occurred along with the addition of mangrove extract. The more mangrove extract was added, the adhesion strength of the paint and specimen was decreased. The highest adhesion strength greatest occurred in the specimen of E100M0, composition without the addition of mangrove extract. It reached 10.15 ± 0.64 Mpa. The lower results of the adhesion test with the lowest strength occurred in the E70M30 specimen with a composition of 70% epoxy and 30% mangrove extract. The remaining 5% influenced this by the volume of 96% ethanol in the mangrove extract. This condition can cause a dilution of a mixture of resin and epoxy hardener, which affects the hardness of the coating layer (Atmaji, 2016) [12]. This addition can cause the coating layer to be more ductile. So that, the more mangrove extract was added, the more ductile and thinner the coating layer was less. This condition leads to a decrease in the adhesion strength and an increase in the cohesive strength of the layer.

![Figure 6. Results of Adhesion Strength Test](image_url)

Pratama [13] reported the effect of adding thinner on painting adhesion. The thinner addition of 60% showed a minimal adhesion strength. It reached 0.67 Mpa. It was lower than the thinner addition thinner of 33% (0.91 Mpa). It proved that the more mixtures combined in the resin coating, the lower the adhesion strength of the coating layer and the greater the cohesion strength of the coating. Another study reported that one species of a mangrove plant, *Rhizophoraceae*, contains large amounts of tannins. These plants were used as adhesive for plywood and particleboard. In general, the liability of tannin was acceptable, except for low water resistance [14]. Another study reported that combining rice husk ash/alkali-activated (RHA/AA) ratio of 0.25 and curing temperature at 75 °C based geopolymer composite coating (GCC) could reach an optimum adhesion strength of 4.7 MPa [15]. The microstructure analysis revealed that coating with high adhesion strength had good interfacial bonding with the substrate.

4. Conclusion
More mangrove extract was added to the epoxy coating so the smaller adhesion strength. The less mangrove extract was added to the epoxy coating, so the more significant the adhesion strength. In conclusion, the addition of mangrove extract to the epoxy coating paint affected the adhesion strength of the specimen. These results will be used for the next steps of research.
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