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The Performance of Antifouling Paint for Prolonged Exposure in Madura Strait, East Java Province, Indonesia

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Abstract— **Antifouling paints are commonly utilized to minimize attached biofouling on the submerged marine structure. The evaluation of commercial both antifouling (AF) paints (Paint A and Paint B) for prolonged exposure has been investigated through field test in Suramadu Bridge, Madura strait, East Java Province, Indonesia. In addition, commercial anti-corrosion (AC) paints were also studied as a controlled coated specimen. The test panels containing all specimens of paint were exposure up to 3-months. Seawater quality parameters consisting of temperature, pH, salinity, conductivity, and dissolved oxygen (DO) were also measured during the test field. The coating properties, which consist of thickness, gloss, hardness, and adhesion strength, were carried out. It was found that both antifouling paints are remarkable to protect attached fouling organisms, but not anti-corrosion paints. Both antifouling paints' properties gradually decrease, such as adhesion strength and gloss, but not in their hardness. There were attached various fouling organisms such as barnacles, tubeworms, and brown algae, where barnacles mainly in the surface of both AC paints after exposure. Based on the result, there was no or less primary biocide of Cu_2O in both AF paints where that biocide can inhibit those fouling after three months of exposure. The rapid reduction of thickness for both AF paints is maybe predominantly induced by seawater current rather than pH, salinity, and temperature.**

Keywords— **Antifouling paint; seawater; corrosion; salinity; coating.**

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I. INTRODUCTION

The attachment and colonization of microorganisms such as marine biofouling have a detrimental effect on ships and static marine structures, fully immersed in seawater [1], [2]. That biological process leads to negative impacts on ships as dynamic vessels, such as increasing the frictional resistance and the time-frequency of dry-docking operation and the increase of corrosion attack [3]. In a static structure such as a bridge, jetties, docks, and others, the presence of attached biofouling can contribute to the severity of corrosion, leading to structural failures such as decreased mechanical strength and lifetime of structure [4]. Also, safety issues are faced when reducing structural defects and structural defects due to biofouling growth's aggressiveness [5]. The severity of biofouling activity depends on the following parameters: environmental condition, geographical area, and ships' trip pattern. It is most likely that surface temperature, water flow, and salinity are predominantly to take responsibility for biofouling's significant activity [6]. It is commonly shown that dense fouling occurs with high seawater temperatures [7]. Most fouling has not grown naturally in low salinity conditions but not for slime, algae, and bryozoa [6]. The tendency of growth for fouling is increasing in low flow rates and vice versa [6]. Indonesia has tropical climates as an equatorial country where the seasonal change is relatively stable for the surface temperature.

Consequently, the growth of fouling continues without the interference of session conditions but not in subtropical regions. On the other hand, the highest severity of fouling formation occurred with low activity of trip or slow-moving of vessel extensively in tropical or subtropical coastal waters. The more active vessels in a tight routine schedule are also at risk of fouling formation in those regions.

The primary action to minimize fouling is to coat the vessels or static structures with antifouling (AF) paint that the mechanism of biocides leaching from the surface of that paint [2]. AF paint usage is the most economical and proven method to mitigate structures from the attached fouling in seawater [8]. Self-polishing antifouling (SP-AF) paint commonly is applied for the most widely used A/F paint due to the consideration of the cost-benefit analysis and the optimized control of the leaching process for primary biocides such as copper or cuprous oxide (Cu_2O) [9]. Due to Tributyltin (TBT) 's harmful effects as biocides, presently, tin-free self-polishing copolymers (tin-free SPC) antifouling paint used as commercial AF paints, containing various Cu compounds. A generic formulation of AF paint mostly consists of resin, which retains the paint together and controls the release of biocide, pigment, solvent, primary biocide (copper compounds), organic or organo-metallic booster biocides, and the other additives. Those components of AF paint are built as AF paint system. However, copper has a detrimental effect on the fouling species and non-target ones [10].

In 2016, some issues of human health and the environment of copper-containing various biocides were reassessed in Europe [9]. Many commercial AF paints contain copper as primary biocides; those issues are not a significant concern in Indonesia. Furthermore, Nuraini and co-workers had only investigated AF paint's performance up to 1 month of exposure in Suramadu Bridge, Madura Straits, but not prolonged exposure. Therefore, this present work aims to investigate the performance of AF paints in prolonged exposure, which is in Suramadu Bridge, Madura Strait, Indonesia.

II. MATERIALS AND METHOD

A. *Preparation of Generic Commercial Paints*

Experimental antifouling (AF) paints were obtained from two different famous companies in Indonesia based on tin-free self-polishing copolymers. All formulation of AF paints was carried out in those companies regarding the approximate chemical composition of paints is shown in Table 1.

TABLE I
APPROXIMATE CHEMICAL COMPOSITION OF COMMERCIAL ANTIFOULING PAINT FROM BOTH COMPANIES

Substances		**Approx. w/w%**	
		A	**B**
Cu_2O		25-50	48
ZnO		1-5	8.2
CuPT		1-5	1.48
VOC	Xylene	10-25	23
	Ethylbenzene	1-5	2.5
	Methanol	0-1	-
	Naphta	-	2.5
Other substances: Plasticizer, anti-settling agent, extenders, and anti-sagging agents			

As a subtract of coating, the mild steel plate was cut into 200 mm width x 250 mm height. The thickness of the plate is 3 mm. A sandblasting machine sanded all subtracted steel according to ISO 8501-1 Sa 2.5. That standard specifies that shadows, streaks, and stains must be restricted to 5% of the surface steel area. It generally is applied to shipyards, off-shore structures, and other marine environments. All coating specimens are categorized as multiple coating due to two different layers—both commercial paints coated with a primary coat, intermediate coat and top coat (antifouling paint). AC coated plate without AF Paint as topcoat was also prepared as a control specimen. In Figure 1a and 1b, the test panels which contain anti-corrosion and antifouling paints were plugged on the pier of the bridge of Suramadu, East Java Province. Those were exposed up to 3 months.

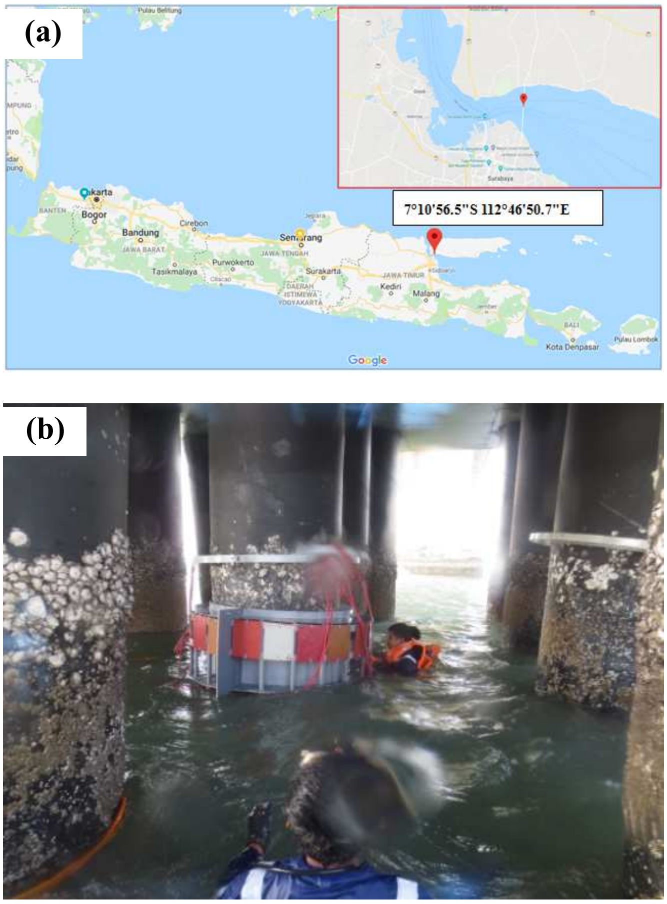

Fig. 1 (a) Location of a field test, and (b). The installation of specimens on the pier of Suramadu bridge

B. *The Test for Evaluating Coating Before and After Immersion*

The evaluation of coating property was done before and after immersion. The gloss property of coating was carried out in a specified measurement angle of 60° using Horiba Gloss Checker IG-331 according to ASTM D 523. The coating hardness used Pencil Hardness tester Elcometer 501, which refers to ASTM D3363. The coating's adhesion strength was carried out using automatic adhesion tester Elcometer 510 (ASTM D4541). The last coating property test was to measure dry thickness (DFT) coating with Thickness Gauge Elcometer 456 (ASTM B499).

C. *The Parameter of Natural Seawater and Observation of Coating Specimens*

The efficacy of antifouling paints depends on several main factors, including water temperature, salinity, solar radiation, or the interaction between varieties of the organism [6], [11], [12]. In measuring the seawater environment, the apparatus used was a HACH HQ40d Advanced Portable Meter. This instrument is a handheld system for field measurements of dissolved oxygen (DO), salinity, conductivity, temperature, and pH in water. Furthermore, the observation of coating morphology and elements in paint A and paint B was conducted using Energy Dispersive X-Ray Spectroscopy (EDAX) and scanning electron microscope (SEM).

III. RESULTS AND DISCUSSION

A. The Observation of Visual Specimens After Exposures

On the preliminary work, Nuraini and co-works had investigated the same type of commercial paint A and paint B in the same research project with this recent work, but only one-month exposure [4]. Figure 2 shows that photographs of anti-corrosion and antifouling paints A after second months; third months, and three months exposures in Seawater of Suramadu Bridge. Generally, antifouling paint is proven to mitigate the attachment of fouling organisms in different exposure time. Based on the results, the growth level of fouling on anti-corrosion (AC) coating was not similar each month as shown in Figure 2 a and 2 b, even though the duration of immersion of them were the same time (1 month of exposure). In three months after exposure, the high diversity of species was found the more maturely developed organisms on AC paint compared to that paint in the first month [4].

On the other hand, there are no attached fouling organisms on all AF paints in all different exposures, as shown in Figure 2d, 2e and 2f. Figure 3 also shows that photograph photographs of anti-corrosion and antifouling paints B after second months; third months and three months exposures in seawater of Suramadu Bridge. The AF paint B has more efficacy to mitigate biofouling's growth and settlement compared to AC Paint B. In three months of exposure, the mature organisms were also found as well as at AC paint A. Furthermore, there were various biofouling organisms such as barnacles, tubeworms, brown algae, where barnacles are mostly attached in both commercial AC paints. The fouling process stages commonly consist of adhesion of the organic film, primary colonizers, secondary colonizer, and tertiary colonizer [6]. The primary colonizer and the second one act as micro fouling, while the tertiary one is categorized as macrofouling. Based on the results, barnacles and tubeworms, and brown algae are hard macrofouling and soft micro fouling, respectively [13].

B. The Evaluation of Paint Properties After Exposure

In proceeding work, Nuraini and co-worker had determined the value of gloss properties, both AF paint A and AF paint B before and after exposure of 1 month, as shown in Table 2 [4]. Figure 2 shows the gloss properties of AF paint after exposure in a prolonged time. After exposure, the fluctuation of both AF paints' gloss values takes place monthly, as shown in Figure 4. However, both AF paint's lowest gloss properties are in three months of exposure compared to those in each month. It implies that the less its gloss, the higher the surface roughness of AF paint. Both AF paint A and AF paint B are categorized as Tin-Free self-polishing copolymers (tin-free CPCs). Those paints have a self-polishing effect for reducing hull roughness, which has smooth paint surfaces during sailing [6]. The hardness of both AF paint A and paint B has classified in soft level (B) before and after exposures.

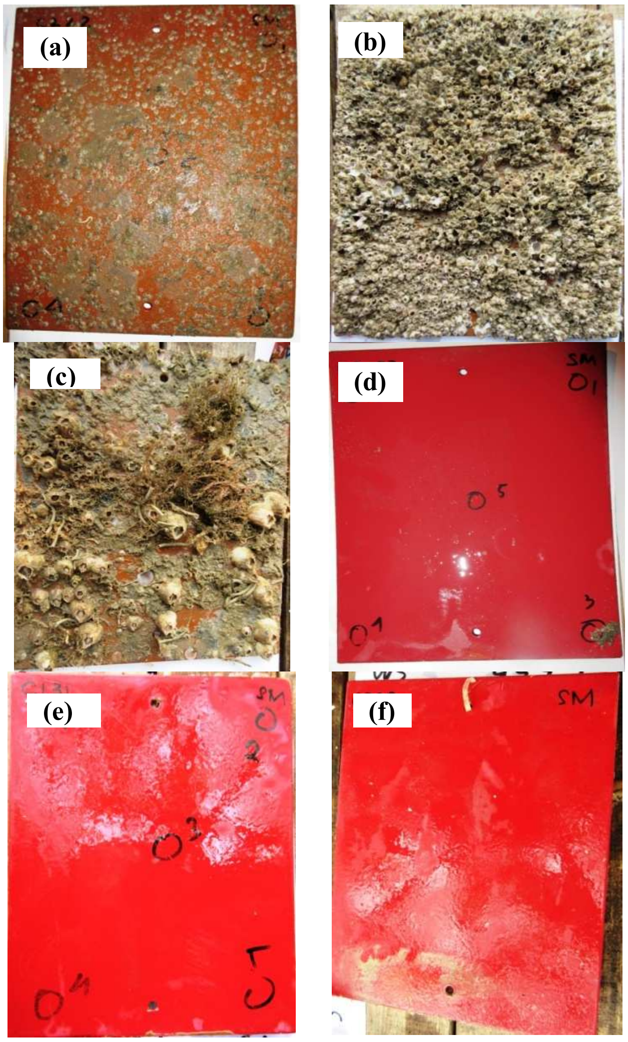

Fig. 2 Visual comparison of specimens for anti-corrosion paint A after a) second months; b) third-month exposure and c) three months of exposures, and those for antifouling paint A after d) the second month; e) third-month exposure and f) three months of exposures

TABLE II
THE PROPERTIES OF AF-A AND AF-B AFTER EXPOSURE FOR 1 MONTH [4].

	Before exposure		
AF Paint	**Gloss at 60°C (GU)**	**Hardness**	**Adhesion strength (MPa)**
A	5.8	B	3.20
B	3.0	B	1.82
	After exposure for 1 month		
A	5.6	B	2.81
B	2.8	B	2.56

AF paint A and AF paint B's adhesion strength is 3.20 MPa and 1.82 MPa, respectively. Those of AF paint A decreases in 1 month of exposure, but not that of paint B [6]. In prolonged exposure up to 3 months, both the adhesion strength of them degrades significantly, as shown in Figure 5**.** On the other hand, that of paint A slightly decreases, while that of paint B relatively increases in the second and third months, respectively. Based on the result, the type of failure is categorized as 100% cohesive pattern. Also, generally, cohesive failure takes places in the paint itself, such as abrasion, cracking due to ageing, dissolving insolvent, and so on [14]. SPC paint contains the pigments such biocide, co-biocides and so on chemically bonded in the coating system, where they leached from the coating during application in seawater [15], [16]. However, SPC paint leaves a thin leached layer without biocide and co-biocide agents. It is presumed that a thin leached layer could slightly reduce the magnitude of cohesive strength due to dissimilar both leached and un-leached layers in AF paint system. The efficacy of AF paints in controlled leaching our rate of biocides is primarily induced by water temperature, pH, and salinity [15], [17], [18].

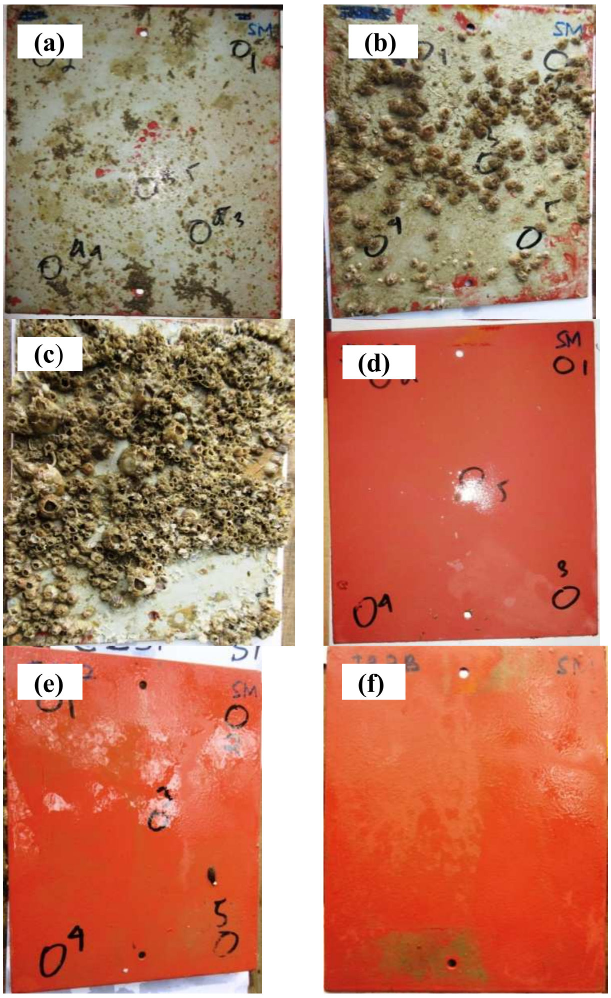

Fig. 3 Visual comparison of specimens for anti-corrosion paint B after a) the second month; b) the third month of exposure and c) three months of exposures, and those for antifouling paint B after d) the second month; e) the third-month exposure and f) three months of exposures

TABLE III
PARAMETER OF SEAWATER ENVIRONMENT IN AVERAGE VALUES AT SURAMADU BRIDGE, MADURA STRAIT.

Period	pH	Salinity	Temperature (°C)
Second month	8.45	28.7	29.05
Third month/ three month	8.60	28.5	32.25

The pH is the suitable range for attached fouling growth during exposure, where the water temperature is higher than 20°C, as shown in Table 3. However, all periods' average salinity is only 28.6 ppt, not from 32 to 35 ppt [17]. The lower salinity value is possible to have occurred because there are many estuaries near Suramadu Bridge. Also, even though it is categorized as the lower salinity, it is higher than brackish water. The release rate of biocide is related to polishing or erosion processes of surface AF paints, where vessel speed or flow seawater current can contribute to the increasing of this process. Kojima and co-workers showed that the decrease rate of paint thickness depended on seawater's flow rate [19]. The slow movements of current take places in the speed of 0.55 m/s, which is defined as ocean drift [20]. Indonesian researchers showed that the range of flow current is from 0.47 m/s to 1.3 m/s in Madura strait, which induced narrow topography of strait and semi-diurnal tide [21]. It implies that the current of Madura strait is categorized as the moderate speed of the current.

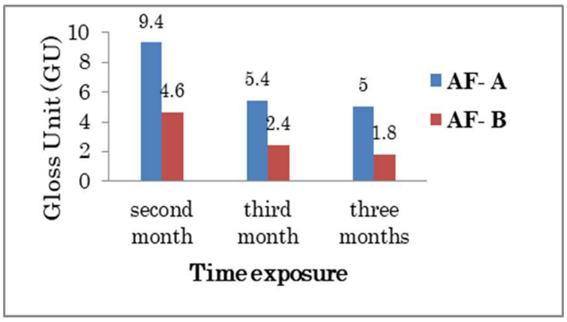

Fig. 4 Gloss properties of AF paint A and AF paint B after exposure.

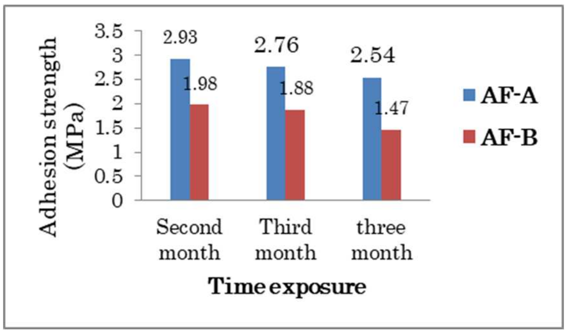

Fig. 5 Adhesion properties of AF paint A and AF paint B after exposure.

C. *Surface Morphology the AF Paints After Exposure*

The cross-sections of AF Paint A were observed before and after the field tests, as shown in Figure 6. Figure 6a shows three different layers of coating, where the top coating is AF coating in 171.9 µm of the average thickness before exposure. However, there is no appearance of the AF layer in all different exposures. The copper element concentration as the main biocide is little or no in increasing the exposure time, as shown in Table 4. For comparison, in Figure 7a, Paint B consists of three layers of coating. An outer coating layer is classified as AF paint. The average thickness of AF paint B is thicker than that of AF paint A before field exposure. The decrease of copper concentration in AF paint B took place gradually in increasing the exposure time. However, there is less, or no concentration of copper in AF paint B compared to that in AF paint A after exposure, as shown in Table 4.

Furthermore, Tin-free self-polishing copolymers (tin -free SPC) paint had been reported that the range of life is between 3 and 5 years in many papers [6], [15], [22], [23]. Based on present fieldwork, both commercial AF Paint A and AF Paint B's lifetime tend to be shorter than their designed lifetime in Suramadu Bridge, Madura strait. The lifetime of AF paint is related to AF coating, which prevents the attachment of fouling with a slow release of binder phase, thus controlling the thickness of the leached layer (biocide depleted layer).

However, this layer is not able to be observed due to the rapid depletion of AF paint.

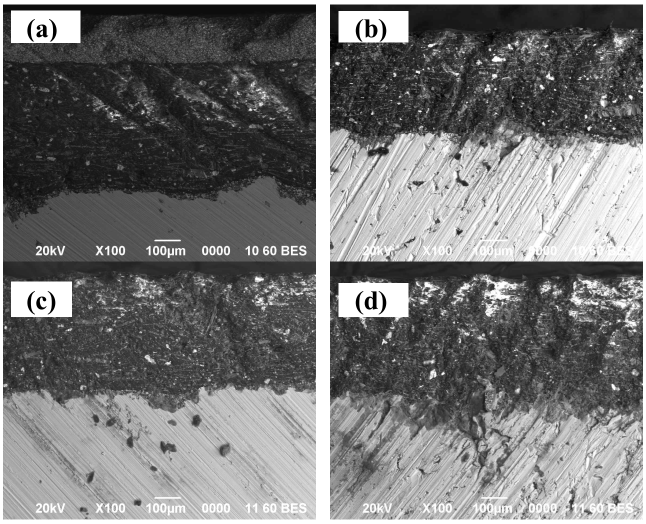

Fig. 6 Cross-section side of AF paint A surface (a) before and after (b) second months; (c) third month, and (d) three months of exposure in Suramadu Bridge, Madura strait.

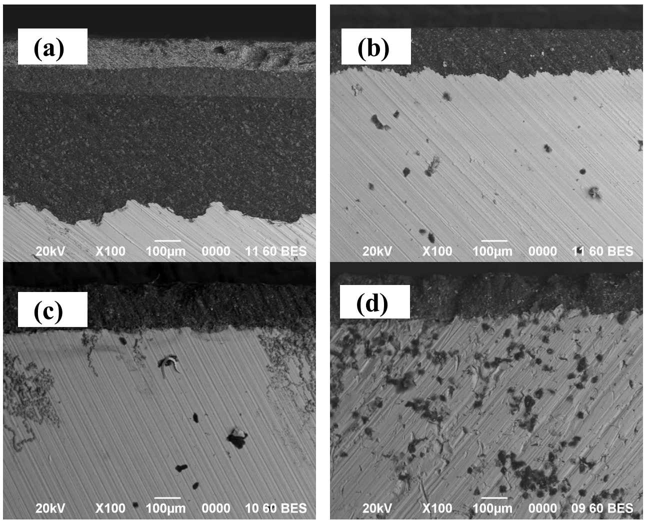

Fig. 7 Cross-section side of AF paint a surface (a) before and after (b) second months; (c) third month, and (d) three months of exposure in Suramadu Bridge, Madura strait.

TABLE IV
THE COMPOSITION OF COPPER (CU) IN AF PAINTS BEFORE AND AFTER EXPOSURE IN MADURA STRAIT

Time exposures	**Wt%**	
	Paint A	**Paint B**
Before	38.34	42.75
Second month	0.18	0.25
Third month	0.00	0.09
Three months	0.00	0.28

In the protective mechanism of SPC-AF coating, seawater enters the paint matrix, dissolve such biocides, and diffuse out into the bulk phase again in slow reaction [6]. Generally, the thin leached layer of AF coating consists of depleted primary biocide and co-biocide particles such Cu_2O and ZnO, respectively which is appeared clearly after exposure, but not this recent work as shown Figure 6 and Figure 7. This condition can be elucidated through the main factor of flow current rate in Madura Strait during field test compared to pH, salinity, and temperature. The high release rates of biocide from the paint bulk are induced by a high flow rate of seawater [19], where the leached layer is easy to polish or erode gradually. However, the primary role of pH, salinity, and water temperature is not predominantly to induce erosion AF surface speed. Also, there are not both commercial AF layer coatings, which appeared clearly after exposure, as shown in Figure 6 and Figure 7. In Figure 2 and Figure 3, the function of fouling protection still worked on both AF paints up to 3 months of exposure. It presumes that the remaining AF layer on both paints still exists. Therefore, it may be predicted that both commercial AF paints' efficacy will be decreased rapidly from 6 months of exposure in Madura strait.

D. Proposed Mechanism of Antifouling Paint Against Biofouling

In recent work, both as-received commercial antifouling paints A and B are categorized as tin-free-polishing copolymer (tin-free SPC). Those paints are based on silyl acrylate (SA) polymers as primary binder according to the technical data sheet of them, as shown in Table 1. The service life of tin-free SPC AF paint is mostly related to its paint's performance, which inhibits the growth and settlement of biofouling with slow rate release of binder [24]. Furthermore, in the mechanism of antifouling paint to protect the attachment of biofouling, seawater enters the paint matrix, dissolves such biocides, co-biocides, and other additives diffuse out into the bulk paint again in slow reaction [24]. The thin leached layer of antifouling paint consists of depleted primary biocide such as Cu_2O [24]. Also, in self-polishing copolymer (SPC) AF paint, the leaching release of paint consist of into initial leaching and steady-state leaching releases [24]-[26].

Table 4 shows element composition of copper as the main biocide in AF paint A and AF Paint B. based on the results, the high concentration of Cu has indicated the presence of primary biocide compounds (Cu_2O) in both the AF paints. Furthermore, the mechanism of dissolution the primary biocide of Cu_2O in seawater in the following chemical equation:

$$Cu_2O_{(s)} + 2H^+_{(aq)} + 4Cl_{(aq)} \rightarrow 2CuCl^-_{(aq)} + H_2O_{(l)} \quad (1)$$

The high level of salinity is influenced by the presence of a high concentration of chloride ions which increase the dissolution rate of Cu_2O [2]. When cuprous oxide meets seawater, it produces soluble hydrated Cu(I) chloride complexes. The hydrolysis process could shift from Cu^+ to Cu^{2+} as the main biocidal species. The mechanisms of controlling the release rate of biocide consist of chemical reactions and diffusion where sea water-soluble pigment dissolution, binder reaction and paint polishing process take place simultaneously [2]. That mechanism could affect the consistency for the thin thickness of leached layer SPC-AF paint [24], [27]. It implies that the absence of biocides in the silyl acrylate matrix leaves behind small pores in that matrix and enhances the paint's total wetted area. The hydrolysis mechanism occurs throughout the leached layer, where there is an alteration of binder's wettability from hydrophobic to hydrophilic [24]. Partially reacted binders are susceptible to be eroded by the flowing seawater and exposed at a less reacted paint surface (self-polishing effect). The less reacted paint surface comprises copper biocide enriched matrix which protects further the attachment of marine biofouling.

IV. CONCLUSION

Both commercial antifouling paints' performance showed valuable results compared to anti-corrosion paints in Suramadu Bridge after exposure. Both AF paints are remarkable to protect attached various biofouling organisms, but not anti-corrosion paint. Both AF paint's coating properties gradually decrease such adhesion strength and gloss, but not their hardness after the field test. It was found that various biofouling macro-organisms had grown densely such as barnacles, tubeworm, and brown algae without antifouling protection in anti-corrosion paint after seawater exposure. After three months of exposure, there was no or less main biocide based on copper in both AF paints. The rapid reduction of thickness for both commercial AF paints may be predominantly affected by seawater current compared to the factors of pH, salinity, and temperature.

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