

## Corrosive Properties of Polyester and Epoxy Reinforced Silane Treated Glass Fibre in Primer Coatings Application: A Comparison Study

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### Abstract

The comparison study of corrosive properties of polyester and epoxy reinforced silane treated glass fibre in primer coatings application was successfully done. The study was carried out by varying the glass fibre loadings from 0–40% to establish the optimum corrosive behaviour of the coatings. The corrosive properties was characterized using autolab potentiostat and immersion testing using 3.5% NaCl for 9 days. The result showed that the optimum corrosion rate of both coatings were found at 10% and 20% of glass fibre loadings for polyester and epoxy coatings respectively. Whilst, the corrosion rate of polyester coating as compared to epoxy coatings which is 0.77735 mmpy and 10.673 mmpy for epoxy coatings. This is due to the properties of polyester that is highest in adhesion towards the substrate and it is also due to the good interaction of GF with the resin has been achieved via silane coupling agent and dispersed well in order to protect the pores from opening and allowing the ions to enter the substrates. For immersion testing, the results followed the same trend as the corrosion rate measurement test for both of the resins.

**Keywords:** polyester; epoxy; glass fibre; primer coatings; corrosion rate

## Introduction

Generally, coating is a covering technique that is applied to the surface of the substrate. It offers many advantages like adhesion, corrosion resistance, or wear resistance. Protective coatings enhance and increase life of coated systems. It also can be used as foundation and act as primer for paint system. The primer coatings are crucial to ensure good adhesion of paint, improve durability and give protection to the substrate [1] Primer is a paint product that allows finishing paint to adhere much better than if it was used alone. It is also designed to form a binding layer that is better prepared to receive the paint. Priming layer should contain corrosion inhibitors such as epoxy or polyester that can protect underlying surface by protective actions to create a better protective coating. [2]

Epoxy resins which are any class of resins derived by polymerization from epoxides that polymerizes when mixed with catalyzing agent or hardener. Epoxy is usually used in adhesives, coatings, electrical insulation, solder mix and castings. Epoxy coatings can provide high bonding strength and high chemical resistance to substrates. Epoxy has much better physical and adhesion properties. Hence, the epoxy resin is used in various industries including the aerospace, automotive, platform oil and gas and marine industries [3]

Polyester (PE) is a class of polymers that include the functional group of ester in their main chain. PE resins commonly used as a primer, undercoat and clear coat for coating. Polyesters resins have an excellent adhesion and high strength. Besides that, due to its high in gloss, hardness and impact resistance, as well as have fast cure characteristic, make it widely used in coatings. PE resins have a very low cost, easy to process, excellent wetting properties with reinforcement, offer a good balance of properties and available in wide variety of grades [4–5]

However, both polyester and epoxy resin have disadvantages in coatings process. The polyester is easily to be attacked by acids and bases that reduce the corrosive properties [6] Polyesters resins also have low thermal resistance, poor solvent resistance, and need to be dried in dehumidifier to prevent hydrolytic degradation. The disadvantage of pure epoxy

resins is their low fracture toughness. So, the epoxy and polyester needs to be reinforced with the glass fiber for the protective corrosion application in oil and gas platform [7]

Glass fiber (GF) is a material consisting of various extremely fine fibers of glass. The advantages of GF are high temperature resistance, non flammable, corrosion-resistance, heat-insulation and high tensile strength. GF has comparable mechanical properties to other fibers such as polymers and carbon fiber. It is also a strong, lightweight material and is used for many products. GF is used for the building of boats and sports car bodies and use has broadened to the automotive and sport equipment sectors [8] However, compatibility of GF with polymeric matrix is very poor due to different polarity between PE and ER with GF that contain many (OH) which is polar [9–10]. Hence, silane coupling agent is incorporated with the coatings to improve its compatibility.

Silane coupling agent is useful to improve the compatibility between the matrix and the fiber thus improving the mechanical properties of the biocomposite [11]. Besides that, silane coupling agent can increased the tensile strength and adhesion between matrix–fiber. Jia and Ling [12] found that  $\text{Al}_2\text{O}_3$  particles that being treated with silane coupling agent makes the  $\text{Al}_2\text{O}_3$  particles better distributed and have better bonding with the PA1010 matrix in  $\text{Al}_2\text{O}_3$ /PA1010 composite. In this research, 3-aminopropyltriethoxysilane (3-APE) is used as coupling agent because it can react with the hydroxyl group of GF.

## Chemicals and equipments

The chemicals used in this research were purchased from Fluka and used without further purification. The chemicals used were ethanol, silane coupling agent (3-aminopropyl triethoxysilane) , sodium chloride (NaCl), DER 354 epoxy resin, curing agent for epoxy (amine), polyester resin (SYNOLAC 9605S65 MY), curing agent for polyester which is methyl ethyl ketone peroxide (MEKP), glass fibers of 150  $\mu\text{m}$ , acetone, zinc powder and zinc sulphate. The equipments that used in this research were carbon steel plate (15 cm  $\times$  10 cm  $\times$  0.2 cm), hand brushes, high speed stirrer, Impedance Analyzer which model is Autolab PGSTAT240, siever–325 mesh model and Nichiban tape.

## Methodology

### Pre-coating of carbon steel plate

Each of the carbon steel plate (15 cm × 10 cm × 0.2 cm) surfaces was degreased by using sand papers and washed with acetone followed by distilled water. Then, allowed them to dry at room temperature.

### Preparation of ER-GF and PE-GF mixture

The 40 g of GF were immersed in 5% wt of 3-aminopropyl triethoxysilane and ethanol solution for 24 h. Then, the mixture were filtered and dried at 80°C in an oven for 6 h. The treated GF modified with 5% of 3-aminopropyl triethoxysilane were grounded using a mortar and sieved using 325 mesh screen of 150 µm. 2.8 g of zinc powder and zinc sulphate were added into distilled water and stirred roughly. The treated GF and the mixture of zinc powder and zinc sulphate were added into epoxy and polyester resin and dispersed at high speed (1500 r/min) using high speed stirrer until the uniform dispersion obtained.

### Preparation of wet paint to coat the steel plate

25 ml of the prepared coatings was applied to the surface of carbon steel plates by using hand brush to coat them for 3 layers. Lastly, the paint on the steel was left to cure for 7 days at room temperature. Then, the testing and characterization of the coatings were carried out.

## Corrosive properties testing

### Autolab PGSTAT204 (potentiostat/galvanostat)

The coated steel plate was immersed into the 3.5% NaCl solution that placed in the beaker together with platinum foil and a saturated calomel electrode which act as counter electrode and reference electrode, respectively. The coated steel plate acted as a working electrode. Then, the results obtained were analysed. The polarization analysis conducted at the open circuit potential, using an Autolab PGSTAT204 potentiostat/galvanostat. The current range was performing around 1 mA.

## Immersion test

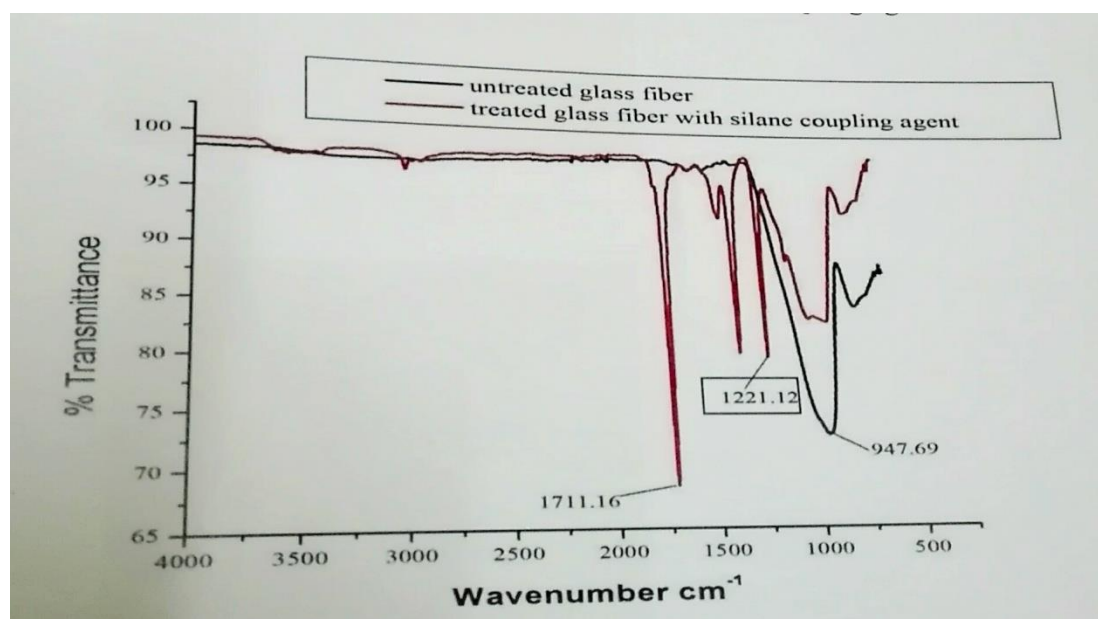
All the carbon steel plate was scratched as "X" scratch presenting to the substrates on the covering surface of the plate. Then, the coated carbon steel was immersed in sea water and salt solution (3.5% of NaCl) for 9 days at room temperature. The coated carbon steel was observed within 3 days of the time intervals and the visual observation were recorded precisely by a camera.

## Results and discussion

### Fourier Transformer–Infrared Spectroscopy (FTIR) Analysis

FTIR analysis was done in this research to determine the functional group exists as the glass fibers were treated with 3–aminopropyl triethoxysilane that acts as coupling agent. The purpose of this analysis is also to determine the cross linking between the glass fibers and the coupling agent.

Figure 1 shows the peak at  $1221.12\text{cm}^{-1}$  corresponding to silanol group from silane coupling agent confirmed the modification process has been successfully done. The FTIR spectra show that the peak of hydroxyl group of GF at  $947.69\text{cm}^{-1}$  was decreased and shifted by applying the 3–aminopropyl triethoxysilane treatment. This is due to the interaction between polar group of coupling agent and the hydroxyl group of GF. Whist the peak at  $1711.16\text{ cm}^{-1}$  corresponding to  $\text{C}=\text{O}$  functional groups [7] According to Chun & Taylor [13], silanization involves the hydrolysis of silane in order to produce silanol then condensation between silanols and covalent bond formation which form between siloxane and GF.



**Figure 1** IR spectrums for treated glass fiber by coupling agent and untreated glass fiber

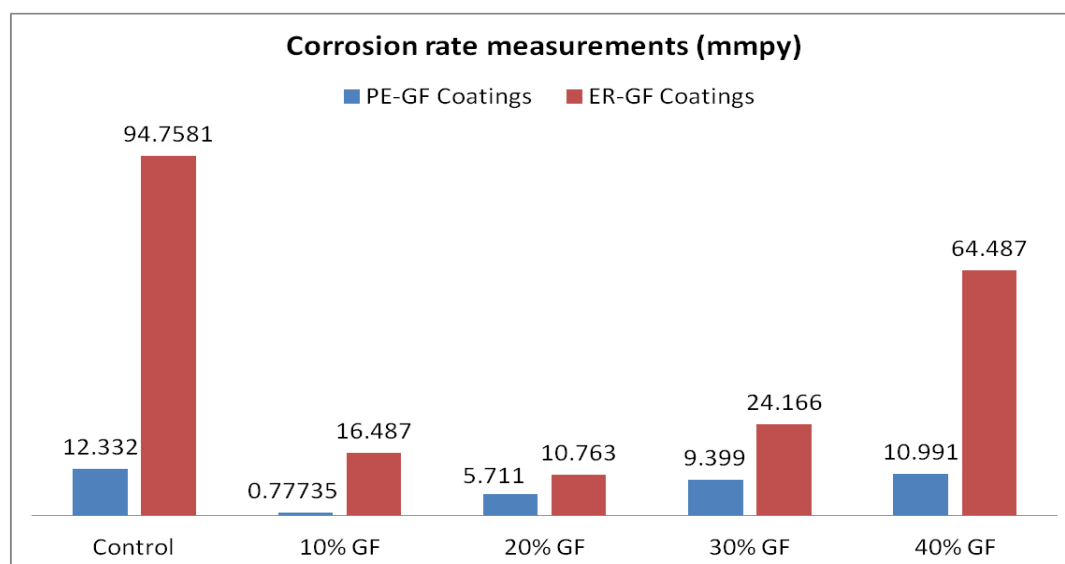
### **Autolab PGSTAT204 (potentiostat/galvanostat)**

The carbon steel plate was immersed into 3.5% NaCl solution at normal room temperature. The reference electrode that used in this study is the saturated calomel electrode (SCE), which consists of mercury covered with a paste of mercurous chloride and mercury in a chloride solution. It will gives a stable and reproducible potential in the solution. The  $E_{corr}$ ,  $I_{corr}$ , corrosion rate and polarization resistance were determined from the Tafel plot of epoxy coating that resulted from the potentiostat/galvanostat [14] The results for the five coatings can be obtained from the Tafel plot that provides a quick estimation of the corrosion rate and polarization resistance,  $R_p$ . The corrosion rate is calculated from the estimated corrosion current,  $I_{corr}$  that can be obtained from the intercept of the two linear segment of the Tafel plot.



**Table 1** Results for Tafel plot polarization of epoxy - glass fiber coatings and polyester- glass fiber coatings.

Formulations	Corrosion potential, $E_{\text{corr}}$ (mV)	Corrosion current, $i_{\text{corr}}$ ( $\mu\text{A}/\text{cm}^2$ )	Corrosion rate (mmpy)	Polarization resistance ( $\Omega$ )
Control (PE)	-382.990	1061.30	12.332	59.3000
Control (ER)	-399.990	409.48	94.758	96.0000
PE-GF 10%	-344.450	66.90	0.77735	333.000
ER-GF 10%	-489.220	554.97	16.487	309.9330
PE-GF 20%	-363.680	491.48	5.711	296.180
ER-GF 20%	-451.390	926.29	10.763	496.890
PE-GF 30%	-375.180	945.90	9.399	109.570
ER-GF 30%	-375.320	182.15	24.166	254.8550
PE-GF 40%	-389.850	808.87	10.991	95.5730
ER-GF 40%	-360.230	253.31	64.487	126.8270



**Figure 2** Corrosion rates measurements for polyester - glass fiber coatings and epoxy - glass fiber coatings with different formulations

Tafel polarization measurements showed that the higher the loadings of GF from 10–20%, the lowest the corrosion rate for PE–GF coatings and ER–GF coatings. The utilization of glass fiber in the coatings makes the corrosion protection increased and the steel plate last longer. The high values of potential of corrosion towards negative path indicates that high susceptibility of steel to corrosion as the corrosion process was related to the diffusion of water, ions and oxygen from the environment or from the polymer matrix itself [15] It was clear from these results that lowest loading of glass fiber (10–20%) was the best corrosion resistant than others. The higher the corrosion potential will shifted the graph towards positive directions, and the lower corrosion current will make the corrosion rate to be increased as well as polarization resistance. The pore or coating resistance relates to the diffusion of electrolyte through the organic coating via the pore, which can affect the barrier properties of the coating while the polarization resistance is the ionic resistance of the coating which is inversely proportional to the area of surface defects (pores). The polarization resistance role is to defend the coatings from the diffusion of ions into the substrate plate.

Table 1 shows the optimum formulation of ER–GF coatings for the corrosion rate at 20% of glass fibers which give the lowest corrosion rate at 10.673 millimeter per year (mmpy). This is due to the utilization of GF make steel plate do not corrode and last longer. It shows that at 20% loading of glass fibers, the coating exhibits better barrier property among the other four coatings. ER–GF 20% also give the highest polarization resistance,  $R_p$  at 496.890  $\Omega$  which indicates that the higher the value of  $R_p$ , the higher the resistance of the epoxy coating towards the corrosion [16] For PE–GF coatings, the optimum formulation was PE–GF 10% as the corrosion rate was the lowest which was 0.77735 mmpy. The utilization of 10% glass fiber corrosion protection is increased and make the steel plate last longer. According to Ramesh. D., & Vasudevan. T., [15] as compound of the treated glass fiber which has loadings of 10% gives the optimum results due to the particles with lower loadings of the pigments completely surrounded by the binder and protected from the diffusion of electrolytes as the pores or voids were closely packed. At 10% of GF loadings also gives the highest  $R_p$  value at 333.000  $\Omega$  which is higher the resistance towards electrolyte.



Then, the other formulations in ER–GF coatings which have the loadings of GF at 10%, 30% and 40% gives the corrosion rates at 16.487 mmpy, 24.166 mmpy and 64.487 mmpy, respectively. Whilst, PE–GF coatings, the other formulations at 20%, 30%, and 40% of GF loadings give the highest corrosion rate at 5.711 mmpy, 9.399 mmpy, and 10.991 mmpy, respectively. It is believed, the higher the content of the glass fibers on the coating, the resins cannot block the pores on the coating. So, the electrolyte solution gradually permeated into the coating that gives the higher corrosion rate. The corrosion activity will happen faster on the substrate if the corrosion rate is higher. This is in line with the finding from Yue *et al.*, [14] With the presence of treated glass fiber in the coating formulations will improved the corrosion rate as well as protected the steel of plates by performed a good interaction with the resin. The good interaction of glass fiber with the resin has been achieved via silane coupling agent and dispersed well in order to protect the pores from opening and allowing the ions to enter the substrates and hence corroded the surface of coatings. However, the increment in the value of glass fiber loadings also increased the micro-defects in polymer coating which will lead to the weak coating barrier property.

The control sample has the highest corrosion rate due to the penetration of ions into the substrate as the substrates was not covered with the glass fiber that have higher corrosion resistance. The different barrier properties of coatings are due to the contents of glass fibers. When the solution diffuses into the coating, the glass fibers can avoid the water from permeating and extend the water diffusion path. Water can pass through coating of control sample and reach the substrate surface easily because coating has none pigment. So, the corrosion rates increase compared to the other coatings [7]

## Immersion test

The results of immersion testing in seawater and salt solution follows the same trend with tafel polarization measurements which is the higher the loadings of GF from 10–20%, the lowest the corrosion occur on steel plate for PE–GF coatings and ER–GF coatings. The utilization of glass fiber in the coatings makes the corrosion protection increased and the steel plate last longer.

The optimum result that give the good barrier property was at 20% volume fraction of glass fibers on epoxy coating. It gives no contact between metal substrate and electrolyte as they cannot pass through the coating surfaces. It also shows the optimum result for corrosion as it only shows small flacking without the corrosion at "X" scratched after 9 days of immersion. The highest corrosive resistance of coating for PE-GF coatings was at 10% of GF loadings as the plate was not corroded at all as well as the blistering and flaking was not occurred. This is due to the glass fiber content that is suitable enough to inhibit the corrosion to occur. The content of glass fiber in the coating gives no contact between the electrolyte and metal substrate and the coating exhibits better barrier property than other formulations of coatings [7]

For the control sample in the epoxy coatings, the rust is observed on the "X" scratched. It is because at 0% glass fiber, epoxy resin alone and does not have the inhibition properties to combat corrosion on the carbon steel plate. ER have poor resistance towards electrolyte, so coating resistance is low and corrosion occurred. As for 10% of glass fibers loadings, the epoxy coating shows that the coating started to peel off after 9 days of immersion showing improved corrosion properties as compared to control sample. A little bit flacking and corrosion was occurred at "X" scratched in formulation 4 (30% GF) and 5 (40% GF).






The control sample for PE-GF coatings displays the most corroded steel plate followed by others formulation. According to Golgoon. A. *et al.*, [17] the corrosion occurred in control sample because polyester have poor resistance to electrolyte and thus make the coating resistance reduced and corrosion occurred. For PE-GF at 20–40% of GF loadings, the corrosion occurs as the content of glass fiber was too high which do not able to protect the coating as the more micro-defects occurred as stated in the research studied by Yongsheng, H. *et al.*, [7]

The immersion test results by using 3.5% of NaCl solution as a medium solution shows the higher rate of corrosion when compared to the immersion in seawater as the salt solution was more aggressive towards carbon steel plate than sea water [18]. For this testing, the trends were similar as the sea water immersion testing whereas the best formulation for

epoxy coating in NaCl solution is at 20% of glass fibers loading is suitable enough to inhibit corrosion properties. It shows that no corrosion and blister occurred at the “X” scratched. The different contents of the glass fibers in five coatings give different barrier properties of these coatings [8] The best corrosive resistance properties of coating in PE–GF was at 10% GF loadings, while the most corroded properties was obtained for other formulations. According to Yongsheng, H. *et al.*, [7] in his research, glass fiber functions to inhibit the corrosive medium to pass through the coating. It means that the contact between electrolyte and metal substrate was not occurred which resulting in no corrosion occurred. The loading of glass fiber in formulation 2 (PE–GF 10%) is the optimum as no micro–defects appeared on the coatings surface.






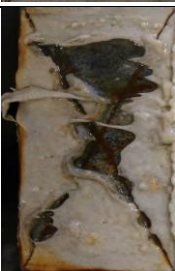



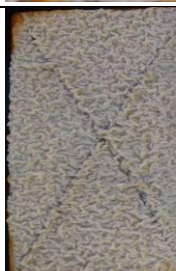
The coatings for control sample in ER–GF coatings and PE–GF coatings were most corroded after 9 days of immersion. The control sample have no glass fiber added to the coating indicates that the water can pass through the coating and reach the surface of the carbon steel plate easily. It is apparent that the glass fiber can prevent the water from permeating into the substrate. As for other formulations, a small corrosion occurred at “X” scratched. It is due to too high content of glass fibers added into epoxy coating which increase the micro–defects that will result to the weak coating barrier property of the coating [7]

Table 3 Immersion test for PE-GF coatings in seawater and salt solution after 9 days.

	PE-GF 0%	PE-GF 10%	PE-GF 20%	PE-GF 30%	PE-GF 40%
Seawater					

Salt solution					
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Table 4 Immersion test for ER-GF coatings in seawater and salt solution after 9 days.

	ER-GF 0%	ER-GF 10%	ER-GF 20%	ER-GF 30%	ER-GF 40%
Seawater					
Salt solution					

## Conclusion

Corrosive properties of polyester and epoxy reinforced silane treated glass fibre in primer coatings application were studied. FTIR spectrums showed that the silane coupling agent grafted onto the glass fiber. The alkyl group of the coupling agent has compatibility with the resin and so the glass fiber can combine with epoxy and polyester resin tightly processed by silane coupling agent. The results confirmed the modification of glass fiber with 3-aminopropyl triethoxysilane has been successfully done. The corrosion rate for epoxy coatings tested by using galvanostat shows that the optimum formulation is at 20% of GF loadings as the corrosion rate was 10.673 mmpy while for polyester coatings, the optimum result was at 10% of GF loadings with 0.77735 mmpy. Here it can be conclude that polyester is better than epoxy due to the lowest corrosion rate. This is due to the

properties of polyester that is highest in adhesion towards the substrate and it is also due to the good interaction of GF with the resin has been achieved via silane coupling agent and dispersed well in order to protect the pores from opening and allowing the ions to enter the substrates. For immersion testing, the results followed the same trend as the corrosion rate measurement test for both of the resins. However, the higher loadings of GF lead to the corrosion of the coatings of epoxy and polyester. When the loadings were increased, the protection ability of the coatings is reduced as more of micro-defects occurred in the coating system and would affect the corrosive properties.

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