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Gravimetric Analysis of Corrosion Inhibition on Al-Mg-Si Alloy by Environmental Friendly products

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ABSTRACT

The effects of environmental friendly products i.e. natural honey, vanillin and tapioca starch on the corrosion behavior of Al-Mg-Si alloy in tropical seawater have been investigated using gravimetric measurements and the energy dispersive spectrometer. The results indicated that all the environmental friendly products were effective in reducing the rate of corrosion and abridged aluminium dissolution in seawater. The measurements have shown that the natural products are effective corrosion inhibitors for aluminium alloy immersed in tropical seawater and the inhibition efficiencies increase with increasing of concentration. The environmental friendly products established their inhibition through formation of a surface film and blocked in the porous layer due to precipitation of studied inhibitors. The energy dispersive spectrometer (EDS) studies elucidated that the breakdowns of Al₂O₃ after exposed to seawater decreased with the presence of environmental friendly products. The EDS spectrums were determined that carbonaceous, carbonyl, methoxy and hydroxyl groups as functional groups of natural products in inhibition mechanism.

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INTRODUCTION

Corrosion is a fundamental process playing an important role in economics and safety, particularly for metals. Apparently, corrosion cannot be avoided, but its severity can be prevented. Several techniques and methods have been developed to combat corrosion efficiency are continually being sought after, as a result of exorbitant amount spent on corrosion annually. Corrosion is measured by the ability of the respective metal atom to oxidize. The varying ability of a metal to lose its electrons and form a positive ion is essential in understanding the ranges of metals that are capable of corroding.

The corrosion resistance of aluminium is dependent upon a protective oxide film. The oxide film is naturally self renewing and accidental abrasion or other mechanical damage of the surface film is rapidly repaired. The conditions that promote corrosion of aluminium and its alloys, therefore, must be those that continuously abrade the film mechanically or promote conditions that locally degrade the protective oxide film and minimize the availability of oxygen to rebuild it.

Aluminium is one of the most versatile and construction materials. In pure state or alloy form, performs a unique combination of properties and characteristics, making it suitable for whole host applications ranging from cooking utensils to aircraft parts. The most popular aluminium alloys for use in corrosive environments such as seawater are the 5xxx and 6xxx series alloys because of their adequate strength and excellent corrosion resistance (Lehmhus, D. and Banhart, J., 2003). These series are highly suitable in various structural, building, marine, machinery, and process-equipment applications. Alloy 6061 is a heat treatable aluminium-magnesium-silicon alloy and well proven medium strength structural alloys that satisfied the requirements of a number of specifications in extruded shapes.

One of the means of combating corrosion in aqueous and process industry environments is the application of corrosion inhibitors. A corrosion inhibitor is a chemical substance or combination of substances that, when present in the environment, prevents, or reduces corrosion without significant reaction with the components of the environment. It is very important to add a corrosion inhibitor to decrease the rate of aluminium dissolution in such solutions. In recent years, many new corrosion inhibitors are been developed (Mishra, A.K *et al.*, 2007, Raja, P.B., and Sethuraman, M.G., 2008, Elango, A. *et al.*, 2009).

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Honey is an easily digestible foodstuff containing a range of nutritiously important complementary elements (saccharides, organic acids, amino acids, polyphenols, etc). It is considered as part of traditional medicine being effective in gastrointestinal disorders, and in healing of wounds and burns (Radojcic, I. *et al.*, 2008). Honey is good corrosion inhibitors of corrosion in aggressive media (Radojcic, I. *et al.*, 2008, Et-Etre, A.Y., 2008, Et-Etre, A.Y. and Abdallah, M., 2000). Vanillin (chemical name: 3-methoxy-4-hydroxy benzaldehyde) is an aromatic aldehyde that possesses a pleasant fragrance and the major component of natural vanilla, which is one of the most widely used and important flavoring material worldwide (Et-Etre, A.Y., 2001).

An attempt at making a further contribution to research into the use of natural products as a corrosion inhibitors mean by further identifying a good products suited to our surrounding environment is therefore the objective of this study. The ultimate aim is one of economic, safety and technological benefits to industrial concerns. This paper presents the results of the study for the corrosion of unprotected aluminium alloy in tropical seawater and efficacy of natural products as corrosion resistance of this alloy against corrosion at room temperature. The results are complemented with the others finding to provide the comparison the probed inhibitors.

MATERIALS AND METHODS

The aluminium alloy used for this study was Al-Mg-Si alloy (25 x 25 x 3 mm coupons) with following chemical composition (wt); Si (0.40%), Fe (0.7%), Cu (0.15%), Mn (0.15%), Mg (0.80%), Cr (0.04%), Zn (0.25%), Ti (0.15%) and Al (reminder). The test solution used for the investigation was tropical seawater collected from Pantai Teluk Kalong, Kemaman, Terengganu (port area). The inhibitors used were natural honey (NH), vanillin (VL) and tapioca starch (TS) and the concentration range from 200-1000 ppm. Freshly seawater solution was used in all the experiments. All the experiments were conducted at room temperature (25°C).

The procedure of gravimetric measurement was performed in accordance with the Standard Practice for Laboratory Immersion Corrosion Testing of Metals by American Society of Testing and Materials (ASTM G 31). In this technique, the samples were completely submersed in chosen corrosive solution. Al-Mg-Si alloy specimens in triplicate for each measurement were immersed in a container, which contained 3000 mL of seawater with the presence and absence of various concentrations of natural products. All the test solutions were let out in the air.

Duration of exposure test was 60 days. The specimens were taken out for every 10 days, washed with distilled water, immersed in a nitric acid (HNO₃) (Merck, 99.8% purity) for 2 to 3 minutes to remove the corrosion products, cleansed in acetone (ASTM G 1), dried and weighed again to obtain the final weight. In order to obtain reliable results, the experiments were repeated three times. All the experiments were conducted at room temperature (25.0 ± 0.1 °C). The weight losses of Al-Mg-Si alloy in seawater with and without the studied corrosion inhibitors were determined as a function of the immersion time. Weight loss measured in milligrams per area (mg cm⁻²) and immersion time was calculated in days. The corrosion products in this method were removed following the ASTM G1.

The surface analytical techniques by the EDS allowed us to investigate detail the chemical composition of aluminum oxide layers. The element compositions of the specimen surface before and after immersion in seawater were studied by the energy dispersive spectrometer (EDS) model JSM-6390LA.

3. Results:

The spontaneous dissolution of Al-Mg-Si alloy in seawater containing different concentrations of NH, VL and TS was studied by weight loss measurement. The investigations were carried out in the open air at room temperature. The weight losses of Al-Mg-Si alloy in seawater with and without the studied inhibitors as corrosion inhibitor was determined as a function of the immersion time.

Fig. 1– 3 illustrate the weight losses (mg cm⁻²) of Al-Mg-Si alloy in seawater with and without NH, VL and TS as corrosion inhibitors. The plots show that the weight losses of Al-Mg-Si alloy for all samples increase with the immersion time as a result of the continuous dissolution of aluminium alloy ions. The greatest amount of corrosion rate throughout the experimental period was recorded for Al-Mg-Si alloy immersed in seawater in an absent of inhibitor. The differences can be noted between the samples exposed to seawater containing corrosion inhibitors as compared to that immersed in the absence of the corrosion inhibitor.

Initially, the dissolution of metal in seawater is slow and increases with immersion time, as indicated by increasing the weight loss by time. The presence of 200 ppm of NH, VL and TS reduce the weight loss of Al-Mg-Si alloy and further decrease in loss of weight is greatly reduced with an increase of NH, VL and TS concentrations up to 1000 ppm. This is attributed to the adsorption of NH, VL and TS molecules on the metal surface, which limits the dissolution of Al-Mg-Si alloy by blocking the corrosion site and hence decreasing the weight loss as its concentration increases. As a result, the alloy surface is efficiency separated from the medium (Li, X. *et al.*, 2008). All the three inhibitors show the similar trends in weight loss measurement.

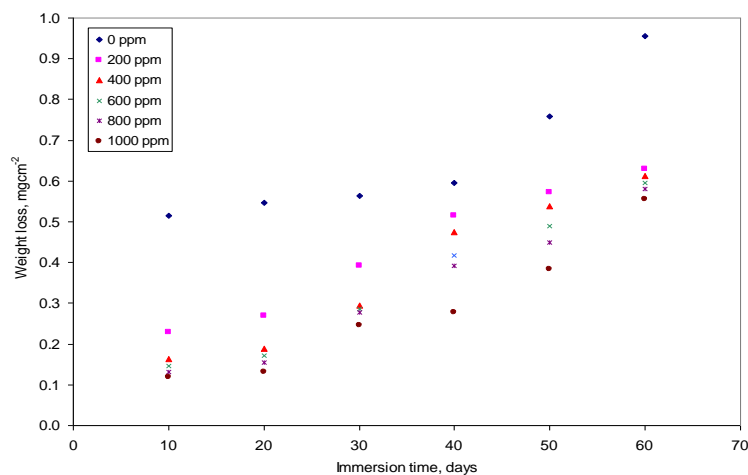


Fig. 1: Weight loss versus immersion time for Al-Mg-Si alloy in seawater containing various concentrations of NH.

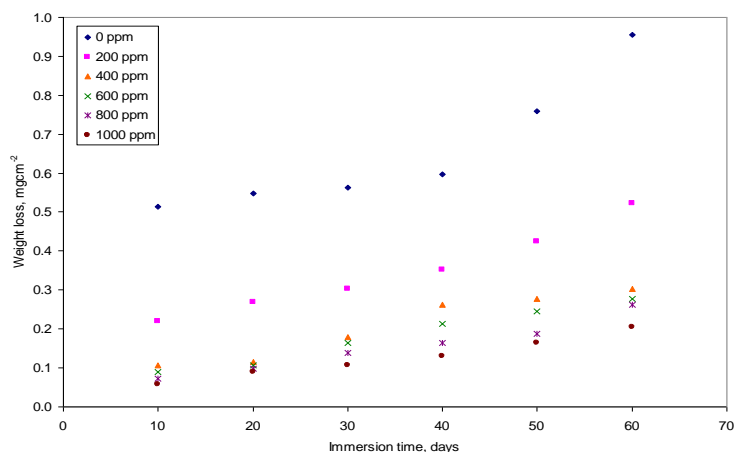


Fig. 2: Weight loss versus immersion time for Al-Mg-Si alloy in seawater containing various concentrations of VL.

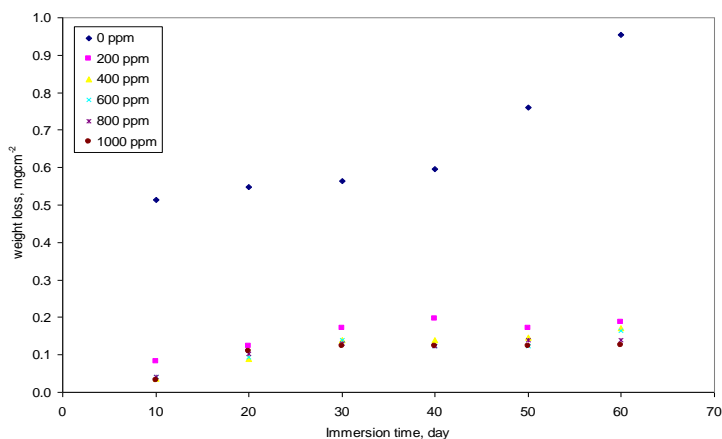


Fig. 3: Weight loss versus immersion time for Al-Mg-Si alloy in seawater containing various concentrations of TS.

The weight loss of Al-Mg-Si alloy in uninhibited and inhibited solution of NH increased with time for 10 to 60 days of immersion time. Weight loss for Al-Mg-Si significantly decreases after addition of 200 and 400 ppm of NH for 10 to 30 days of immersion time. Further increase of 600 to 1000 of NH only contributes to lessening reduction in weight loss of Al-Mg-Si. Meanwhile, a decline of weight loss was found in constant for immersion day from 40 to 60 days. Weight loss for Al-Mg-Si extensively decreases after addition of 200 and 400 ppm of VL.

Slightly reduce in weight loss was observed after addition of 600 ppm to 1000 ppm of VL. The weight loss of uninhibited and inhibited solution increased with time for 10 to 60 days of immersion time.

Weight loss for Al-Mg-Si drastically decreases after addition of 200 ppm of TS. Further increase of TS only contributes small amount of weight loss of Al-Mg-Si increase. It means 200 ppm of TS is the adequate amounts to effectively retard the corrosion process of aluminium alloy in seawater. The weight loss of inhibited solution increased with time for 10 to 30 days of immersion time and turn out to be constant after 30 days onwards.

Fig. 4 shows the plot of weight loss versus immersion time for Al-Mg-Si alloy in seawater containing 1000 ppm of NH, VL and TS. From the figure, it can be seen that all the studied inhibitors display good inhibitive effect on the corrosion attack of Al-Mg-Si alloy in seawater. The addition of NH in the solution reduced 42% of the weight loss of uninhibited solution; meanwhile VL declined the value to 79%. TS performed the best inhibitive effect on retarding the metal from corrosion process in seawater by decreasing the weight loss up to 94%.

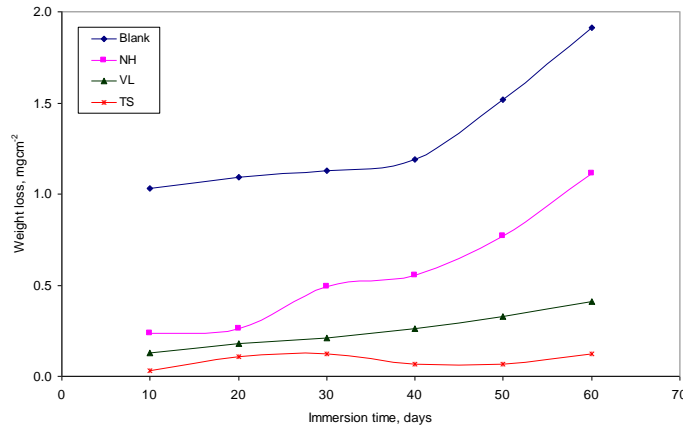


Fig. 4: Weight loss versus immersion time for Al-Mg-Si alloy in seawater containing 1000 ppm of NH, VL and TS.

Uninhibited and inhibited samples were analyzed by EDS in order to identify the composition of the corrosion products before and after immersion in seawater at 25°C. The EDS analysis of plain sample indicated that only Al and oxygen were detected, with ratio of about 2:3, which indicated that the passive film contained only Al₂O₃. Table 1 presents the percentage of passive film, Al₂O₃ obtained from the analysis of the EDS spectra and the data shows that plain surface of unexposed specimen consists of 96.06% Al₂O₃.

Table 1: The percentage of passive film, Al₂O₃ obtained from the analysis of the EDS spectra.

Compound Al ₂ O ₃ (mass%)								
Seawater containing natural products (ppm)								
NH			VL			TS		
200	600	1000	200	600	1000	200	600	1000
52.67	60.86	75.22	55.23	65.96	80.21	57.09	67.57	85.96

The EDS spectrum of Al-Mg-Si alloy in seawater with the presence of 1000 ppm of NH shows in Figure 5. The spectrum elucidates the existence of carbon (C), oxygen (O) and sulphur (S); due to the carbon, oxygen and sulphur atoms of the NH. These data show that carbonaceous material containing O and S atoms has covered the specimen surface. This layer is absolutely due to the inhibitor, because the carbon signal and the high contribution of the oxygen signal are absent on the specimen surface exposed to uninhibited seawater.

The EDS spectrum of Al-Mg-Si alloy immersed in seawater contained 1000 ppm of VL and TS portray in Figs. 6 and 7. The results show an additional line characteristic for the existence of C (due to the carbon atoms of the vanillin and tapioca starch). These data show that carbonaceous material has covered the specimen surface. This layer is absolutely due to the inhibitor, because the carbon signal is absent on the specimen surface exposed to uninhibited seawater.

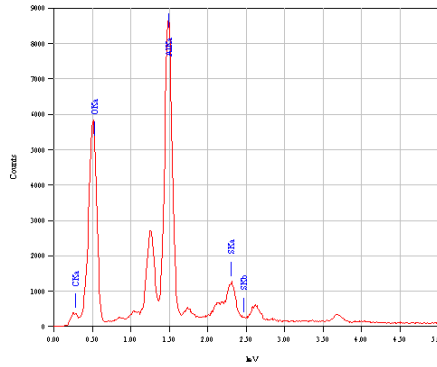


Fig. 5: EDS analysis of Al-Mg-Si alloy immersed in seawater contained 1000 ppm of NH.

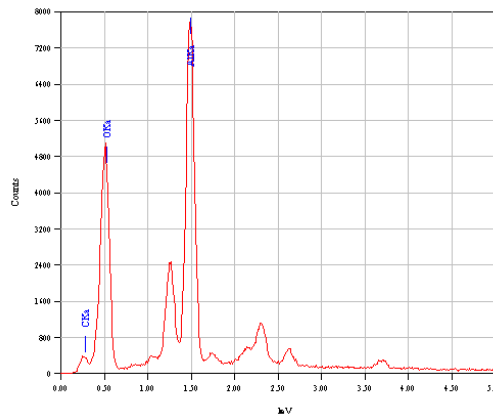


Fig. 6: EDS analysis of Al-Mg-Si alloy immersed in seawater contained 1000 ppm of VL.

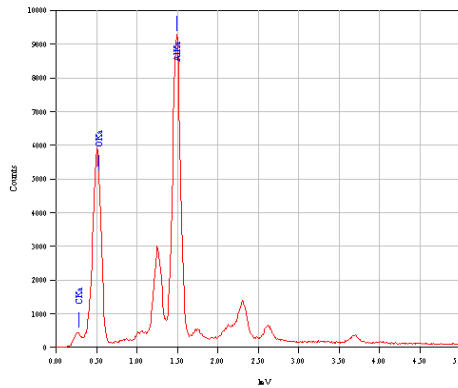


Fig. 7: EDS analysis of Al-Mg-Si alloy immersed in seawater contained 1000 ppm of TS.

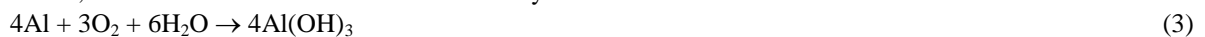
4. Discussion:

The corrosion process of Al-Mg-Si alloy in seawater could be explained on the basis that the surface of aluminium alloy is covered by passive film. When immersed in seawater, the outer surface of the film is dissolved on the other hand, aluminium atoms in substrate will diffuse toward surface, or oxygen toward substrate, and combine to form passive film (Amin, M.A. *et al.*, 2009).

Anodic and cathodic processes of aluminium corrosion in seawater are dissolution of aluminium and reduction of dissolved oxygen, respectively, as



Hence, Al^{3+} reacts with OH^- to form aluminum hydroxide near the aluminium surface as below



and the hydroxide precipitates on the surface due to its low solubility product. Aluminium hydroxide changes gradually to aluminium oxide, resulting in the formation of passive film (Rosliza, R. *et al.*, 2008):



However, this nature oxide film does not offer sufficient protection against aggressive anions and dissolution of aluminium substrate occurs when exposed to corrosive solution.

Seawater predominantly consists of about 3.5% of sodium chloride (NaCl) and many other ions. Chloride ions are very strong and could easily penetrate the passive film. Thus, dissolution of the aluminium substrate occurs and results in corrosion. The adsorption of the corrosion inhibitor competes with anions such as chloride. By assuming that the corrosion inhibitor molecules preferentially react with Al^{3+} to form a precipitate of salt or complex on the surface of the aluminum substrate, the anodic and cathodic processes subsequently suppressed by inhibitor molecules. Thus, this result suggests that the protective film that was formed comprise aluminium hydroxide, oxide and salts or complexes of the corrosion inhibitor anions.

The weight loss measurement results indicated that introduction of NH, VL and TS obviously minimize the weight loss and abridged aluminium dissolution in seawater. The weight loss in inhibited systems decreased with increasing concentration of inhibitors, suggesting that the inhibiting effect of Al-Mg-Si alloy in seawater is concentration dependent. Similar observations are found for 200, 400, 600 and 800 ppm of inhibitors concentration. Therefore these inhibitors can be considered as efficient inhibitors of Al-Mg-Si alloy in seawater.

The EDS analysis of exposed specimen indicated that 50% of passive film, Al_2O_3 was breakdown after exposed to seawater. Data in Table 3 indicated that the breakdowns of Al_2O_3 percentage for three studied inhibitors were decreased with the increasing of inhibitor concentrations of NH, VL and TS; hence decreased the dissolution of Al-Mg-Si substrate. It means the NH, VL and TS acts as protective film for Al-Mg-Si alloy from the aggressive solution. The protective film coverage is increased with the increasing of inhibitor concentration.

Conclusions:

The corrosion inhibition studies of the aluminium alloy were carried out at room temperature using seawater and the results indicate that NH, VL and TS are an effective corrosion inhibitor of aluminium alloy in that particular solution. The gravimetric results show that the introduction of NH, VL and TS obviously minimizes the weight loss and abridged aluminum dissolution in seawater. The inhibition efficiency increases with the increasing of corrosion inhibitor concentration. The corrosion inhibition process of Al-Mg-Si alloy in seawater using three selected natural products as corrosion inhibitors show that the corrosion rate of the alloy significantly reduced upon the addition of studied inhibitors.

The EDS spectrums reveal that the presence of C, O, and S for NH as elements which take place in the inhibition mechanism. The carbonyl, methoxy and hydroxyl groups arranged around the aromatic ring are determined as functional groups of VL in inhibition process. The C atoms in TS are recognized by the EDS analysis, where these atoms involve in the adsorption process in alloy surface. The formation of precipitates of oxides/hydroxides of these inhibitors results in improved corrosion resistance.

Based on the results from EDS studies, it can be concluded that the TS gave the best protection of Al-Mg-Si alloy from the corrosion attack in seawater, following by VL and NH. The protection of passive film is increased with the increasing in inhibitor concentrations.

It is explored and proven in this research that NH, VL and TS carry tremendous potential for industrial usage. Unlike the pure synthetic product that requires enormous investment scale; NH, VL and TS can be produced at any type of industrial scale, which is potentially capable of eradicating the disparity among the communities, especially in the third world. Furthermore, the potential usages of these natural products discussed in this research are in line with the recent trend of the environment-friendly concept. However, resolution of the problem of whether the origin of these effects is associated with an application of aluminium alloys must await the results of further experimental studies.

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