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An experimental approach towards anticorrosive potential of Areca fat species at aluminum/test solution (HCl/NaOH) interface

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Abstract : In the present investigation, we extracted Areca fat species from dry arecanut seed by using hexane as a solvent through soxhlet extraction apparatus. Weight loss and Tafel techniques were employed to study the inhibition role of Areca fat species on aluminum (Al) metal in both acid (0.5 M HCl) and base (0.1 M NaOH) systems. Weight loss (mass loss) studies indicated that, loss in the weight of Al decreases with increasing the amount of Areca fat species, which is due to adsorption of inhibitory (Areca fat) species to an electrode (Al) surface and obeys Langmuir adsorption mode. Mechanism of inhibition property of Areca fat species on the Al surface was discussed by kinetic and thermodynamic parameters. The nature of inhibition (type of inhibitor) was further confirmed by Tafel curves. Further, Scanning electron microscopy (SEM) and atomic force microscopy (AFM) techniques were used in order to confirm the chemical (weight loss) and electrochemical (Tafel curve) results.
Keywords : Areca fat species; Aluminum; Weight loss; Tafel curves; Scanning electron microscopy.

Introduction

Al and its alloys are the important metals for several chemical, engineering and manufacturing industries due to their unique and noble characteristics. Due to these reasons, Al is broadly utilized in many industrial processes, during this time, rust and other dust particles is created on Al surface. [1-3] To remove the unwanted rust and dust particles, many industries using acid (HCl) and base (NaOH) solutions. These solutions (HCl/NaOH) remove the rust and dust particles, but causes disintegration of Al metal, which weakness the electrode (Al) surface. Disintegration of Al metal in HCl/NaOH solution not only weakness the metal structure, but also effects on the economy. [4-7]Hence, to overcome the disintegration and economy problems in an effective way, many scientists (especially corrosion scientists) focused on synthetic corrosion inhibitors. Inhibitors are the substances which when introduced to corrosive solutions effectively block the corrosion reaction. Previous reports show that, many synthetic inhibitors possessed good inhibition action for Al metal. The corrosion inhibitory action (behavior) of synthetic inhibitors is due to adsorption of molecules from inhibitor on Al surface. Even though these inhibitors show good inhibition property, the use of application of these inhibitor molecules as corrosion inhibitors in many industries is restricted due to their expensive and toxic nature. Many inhibitors are prepared by expensive route, which is not considered by many industries because of economic issue and also majority of these inhibitors are toxic to living creatures. Due to these specific reasons, many researchers shifted interested in green (plant) products for various metal corrosion inhibition in cheap and environmentally benign way. The species present in the plant extracts contains nitrogen, sulfur, phosphorous and oxygen, which responsible for inhibition of electrode corrosion by adsorption mechanism. [8-14] Areca fat is one of the plants (natural) extracts contains capric acid, lauric acid, myristic acid, palmitic acid, which possessing electron rich elements. [15-21] No specific research report is available on Areca fat as a corrosion inhibitor for Al in both 0.5 M HCl and 0.1 M NaOH environment. Therefore, in the present study, we extracted Areca fat species from arecanut seed. The corrosion study was carried out by weight loss (gravimetric) and Tafel plot (potentiodynamic polarization) methods. Surface examination was visualized by SEM and AFM techniques.

Experimental part

Materials preparation

The chemical composition of study Al metal (A-63400 type) was shown in Table 1. The surface of Al was cleaned by sand papers, washed with acetone and dried. 0.5 M HCl and 0.1 M NaOH solution was prepared by standard procedure.

Table 1. Chemical composition of type Al specimen

Element	Cu	Mg	Fe	Si	Zn	Mn	Tl	Cr	Al
wt%	0.1	0.4-0.9	0.6	0.3-0.7	0.2	0.3	0.1	0.2	Remainder (96.9-97.8 %)

Preparation of inhibitor

130 grams of finely grind arecanut seed (dried) powder was placed in the Soxhlet extraction chamber and extraction was carried out by hexane (380 ml) for effective five hours duration. 4 different concentrations (7.5 (g/L), 15 (g/L), 30 (g/L) and 45 (g/L) for corrosion inhibition examination were prepared by resulting solution (inhibitor).

Weight loss technique

Mass loss of Al metal without Areca fat species and with Areca fat species in aggressive 0.5 HCl/0.1 M NaOH) solution was monitored by gravimetric (weight loss) method. The test is performed with different immersion periods (1, 2, 3, 4, 5 and 10 hours) and temperatures (303 K, 308 K, 313 K, 318 K and 323 K) in order to examine the stability of the adsorption layer (formed from Areca fat species) and mechanism of Al corrosion inhibition. The test is repeated until constant value is obtained.

The Al corrosion rate in both 0.5 M HCl and 0.1 M NaOH environments was calculated by following relation,[22]

$$\text{Corrosion rate } (\upsilon_{corr}) \text{ (mils penetration per year) } = \frac{534W}{ATD},$$

where, T = contact time of Al metal in the test solution with and without Areca fat in hours, D = Al metal density in grams per cubic centimeter, A= Al surface area (in square inches), W= mass loss of Al in milligrams.

Corrosion inhibition efficiency of Areca fat species on Al surface was evaluated by following equation,

$$\text{Corrosion inhibition efficiency } (\eta_w) \text{ (\%)} = \frac{(W_1 - W_2)}{W_1} \times 100,$$

where, W_1 and W_2 are the loss of weight of Al in unprotected (without Areca fat species) and protected (with Areca fat species) system respectively.

Potentiodynamic polarization (Tafel plot) studies

Tafel plots are required to study the kinetics of the Al corrosion reaction. Tafel curves were obtained by submerging the 1 cm^2 of Al (working electrode) to 0.5 M HCl/0.1 M NaOH solution along with saturated standard calomel and platinum auxiliary cell in CHI660C workstation (applied potential and scan rate is ± 200 mV and 0.01 V/s.

The corrosion current density (i_{corr}) (obtained from Tafel plot) was used to calculate the corrosion inhibition efficiency of Areca fat according to the following relation,

$$\text{corrosion inhibition efficiency} \quad (\eta_p = [\,1 - \frac{i'_{corr}}{i_{corr}}\,] \times 100,$$

where, i'_{corr} = value of corrosion current density in the presence of Areca fat species extract and i_{corr} = value of corrosion current density in the absence of Areca fat species.

Surface study

Surface studies were carried out by an SEM and AFM techniques for Al surface exposed to 0.5 M HCl/0.1 M NaOH solution without and with Areca fat species for an immersion period of 2 h.

Results discussion

Gravimetric (mass loss) studies

Corrosion inhibition capacity of Areca fat species on Al in both 100 ml of 0.5 M and 0.1 M NaOH solution with different solution temperature and time was examined by weight loss technique. The parameters obtained from this method are shown in Tables 2 and 3. These tables show that, the Al corrosion rate greatly decreases with the addition of different concentrations of Areca fat species to 0.5 HCl/0.1 M NaOH environments, while corrosion inhibition capacity of Areca fat species enhances with an increase in the concentration from 7.5 (g/L) to 45 (g/L). This development is due to the presence of tenacious protective film generated by Areca fat species on Al surface. The increasing the Areca fat concentration leads to greater stability of the tenacious protective film for corrosion inhibition process. Hence, the Al corrosion rate is reduced with Areca fat species leads to increase in protection efficiency values. By increasing the solution temperature and contact time, the corrosion rate increases and protection efficiency decrease. This trend is due to the instability of protective Areca fat species film in such conditions. The stability of this film gradually decreases and undergoes degradation which left Al metal in unprotected state. Hence, Al corrosion rate increases, whereas corrosion inhibition capacity of Areca fat species decreases with a rise in both 0.5 M /0.1 M NaOH solution temperature (from 303 to 323 K) and contact time (from 1 to 10 hours).

Table 2. Corrosion parameters for Al in 0.5 M HCl/0.1 M NaOH solution obtained from weight loss (gravimetric) technique at 303 K.

T	C (g/L)	Aluminum			
		0.5 M HCl		0.1 M NaOH	
		υ_{corr} ($\times 10^{-4}$ (mpy)	η_w (%)	υ_{corr} ($\times 10^{-4}$ (mpy)	η_w (%)
1	Blank	8.699		61.865	
	7.5	2.416	77.222	19.333	68.750
	15	2.416	72.222	16.916	72.656
	30	1.933	77.777	12.083	80.468
	45	1.449	83.333	7.249	88.468
2	Blank	11.116		62.832	
	7.5	3.866	65.217	20.541	67.307
	15	3.624	67.391	18.849	70.000
	30	2.658	76.086	12.566	80.000
	45	1.933	82.608	7.249	88.461
3	Blank	16.110		64.443	
	7.5	5.638	65.000	22.555	65.000
	15	5.638	65.000	19.333	70.000
	30	4.349	73.000	13.049	79.750
	45	3.222	80.000	8.055	87.500
4	Blank	18.124		78.540	
	7.5	6.404	64.666	27.791	64.615
	15	6.404	64.666	25.374	67.692
	30	4.954	72.666	18.124	76.923
	45	3.745	79.333	12.083	84.615
5	Blank	24.166		82.165	
	7.5	8.699	64.000	30.932	62.352
	15	8.699	64.000	28.032	65.882
	30	6.863	71.600	20.299	75.294
	45	4.929	79.600	14.499	82.352
10	Blank	42.291		86.998	
	7.5	16.916	60.000	36.249	58.333
	15	15.949	62.285	33.349	61.666
	30	12.566	70.285	31.416	63.888
	45	9.666	77.142	25.616	70.555

Table 3. Effect of test solution (0.5 M HCl/0.1 M NaOH) temperature on protection (corrosion inhibition) efficiency

T (K)	C (g/L)	Inhibition efficiency in percentage Aluminum	
		0.5 M HCl	0.1 M NaOH
303	7.5	77.222	68.750
	15	72.222	72.656
	30	77.777	80.468
	45	83.333	88.468
308	7.5	70.000	66.666
	15	70.000	66.666
	30	75.000	76.666
	45	80.000	86.666
313	7.5	68.181	63.888
	15	68.181	63.888
	30	72.727	75.000
	45	77.272	83.333
318	7.5	68.000	63.636
	15	68.000	63.636
	30	72.000	72.727
	45	76.000	81.818
323	7.5	66.666	60.606
	15	66.666	63.636
	30	70.370	69.696
	45	74.074	80.303

Activation parameters

Activation energy (Ea) (from Arrhenius plot Fig. 1 (a,b) [23], the entropy of activation (ΔS*) and enthalpy of activation (ΔH*) (from the transition state plot, Fig. 2 (a,b) [24] values were investigated in order to study the nature of the Al dissolution process in 0.5 M HCl/0.1 M NaOH. These results are placed in the Table 4**.** In all studied cases, a Ea value in protected conditions (with Areca fat species) is generally higher compared to bare (0.5 M HCl/0.1 M NaOH) solution, indicates the high energy is needed for the dissolution of the Al metal in the protected system (with Areca fat species). The obtained ΔH* and ΔS* values clearly indicates the endothermic and difficulty of the Al dissolution process in both environments.

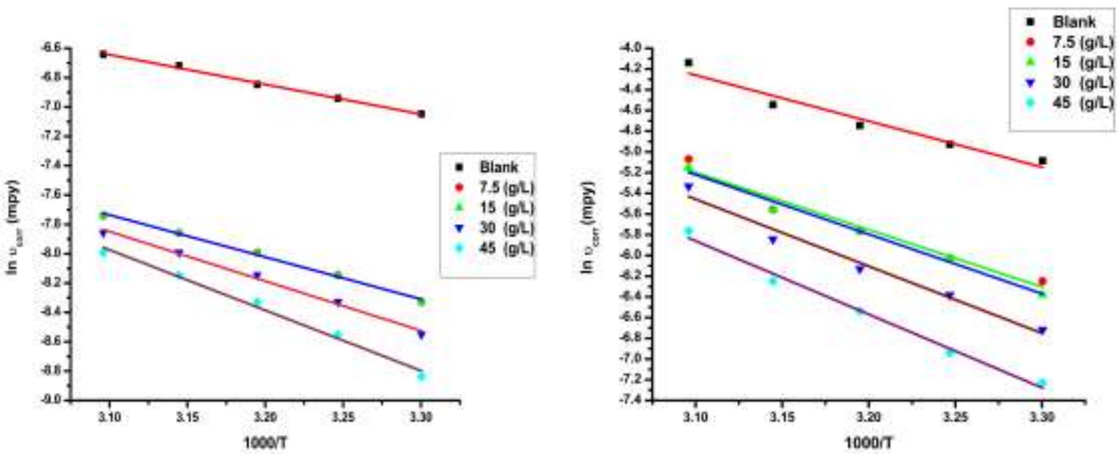


Fig. 1 (a,b). Arrhenius plots, a) Al in 0.5 M HCl (without and with inhibitor), b) Al in 0.1 M NaOH solution (without and with inhibitor)

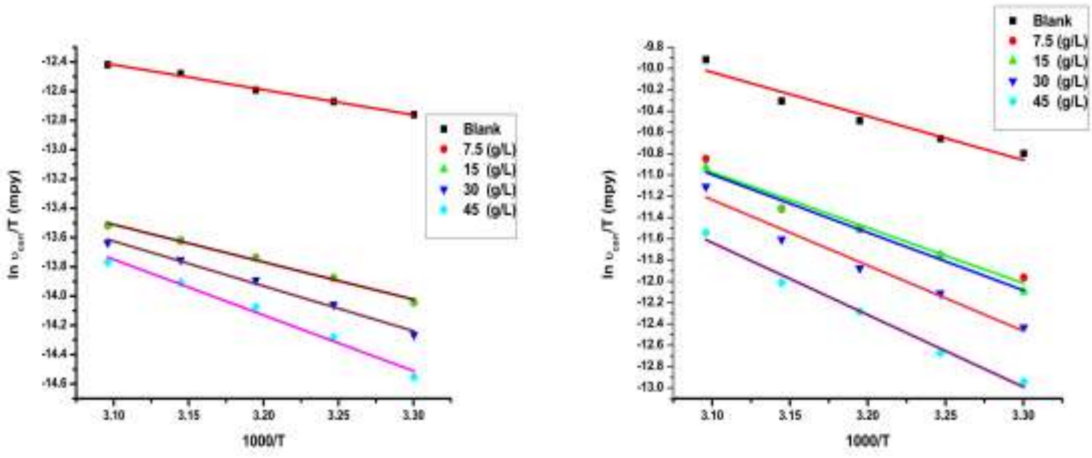


Fig. 2 (a,b). Transition state plots, , a) Al in 0.5 M HCl (without and with inhibitor), b) Al in 0.1 M NaOH solution (without and with inhibitor)

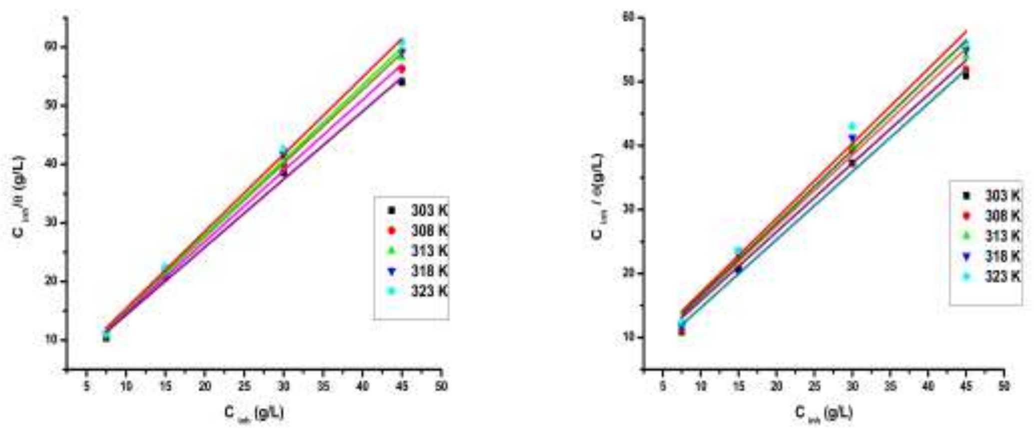


Fig. 3. Langmuir adsorption isotherm , a) Al in 0.5 M HCl (without and with inhibitor), b) Al in 0.1 M NaOH solution (without and with inhibitor)

Table 4. Activation parameters

Electrode	Medium	C (g L^{-1})	*Ea** (kJ mol^{-1})	Δ*H** (kJ mol^{-1})	Δ*S** (J mol^{-1} K^{-1})
Aluminum	0.5 M HCl	Blank	16.830	14.230	-313.901
		7.5	23.846	21.246	-301.232
		15	23.846	21.246	-301.232
		30	28.084	25.483	-289.036
		45	34.248	31.648	-270.960
	0.1M	Blank	36.917	34.316	-231.808
	NaOH	7.5	45.880	43.280	-211.843
		15	47.732	45.131	-206.279
		30	53.782	51.181	-189.486
		45	58.921	56.321	-176.902

Table 5. Thermodynamic parameters

Electrode	Medium	Temperature (K)	K_{ads} (L g^{-1})	ΔG^{o}_{ads} (kJ mol^{-1})
Aluminum	0.5 M	303	370.111	-32.305
	HCl	308	381.965	-32.918
		313	395.153	-33.541
		318	440.200	-34.363
		323	454.167	-34.987
	0.1 M	303	250.036	-31.316
	NaOH	308	198.141	-31.237
		313	190.109	-31.637
		318	196.315	-32.227
		323	188.114	-32.619

The Areca fat species block the Al corrosion reaction by adsorption mechanism. In the present case, in both cases (Al in 0.5 M HCl environment without and with Areca fat and Al in 0.1 M NaOH solution without and with Areca fat) the adsorption of Areca fat species on the Al surface obeys Langmuir isotherm model (Fig. 3 (a,b)**.** All calculated parameters from the Langmuir isotherm plot are shown in Table 5.

The equilibrium constant for the adsorption process (K_{ads}) value is increased with 0.5 M HCl solution temperature is an indication of a formation of strong and effective adsorption layer on Al surface, whereas, in the case of Al in 0.1 M NaOH solution, the value K_{ads} decreases with increasing the 0.1 M NaOH solution temperature, which shows the thermal agitation of protective Areca fat species film on Al surface. The obtained free energy of adsorption (ΔG^{o}_{ads}) values in both systems (Al in 0.5 M HCl and Al in 0.1 M NaOH solution) is in negative mode, which greatly supports the spontaneous adsorption process. In this study, The free energy of adsorption (ΔG^{o}_{ads}) values are between the literature values, [25] which certify the applicability of both physical and chemical (comprehensive) adsorption process.

Potentiodynamic polarization technique (Tafel plot)

Fig. 4 (a-b) clearly represents the Tafel curves for Al metal immersed in 0.5 M HCl (without and with Areca fat) and 0.1 M NaOH solution (without and with Areca fat) respectively. Numerous parameters obtained from this plot are placed in the Table 6. This table shows that, the introduction of Areca fat species to 0.5 M HCl.0.1 M NaOH solution lowers the corrosion current density values, which is an indication of protection of Al metal. The protection of Al metal in 0.5 M HCl/0.1 M NaOH solution is increasing with plant extract concentration. Areca fat species covers the Al surface and blocks the attack of 0.5 M HCl/0.1 M NaOH solution on the surface the Al. The obtained corrosion potential (E_{corr}) [55 mv for Al in 0.5 M HCl system and 48 mv for Al in 0.1 M NaOH system) and cathodic Tafel slope (βc) and anodic Tafel slope (βa) values did not move toward any direction is an indication of mixed Al corrosion inhibition property of Areca fat species, which means that, Areca fat species inhibit the both anodic and cathodic reactions on Al surface. [26, 27]

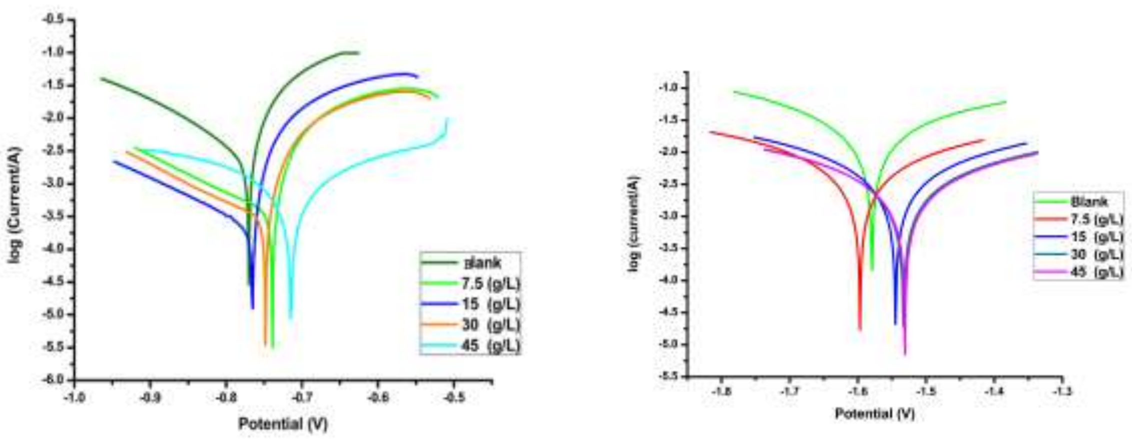


Fig. 4. Tafel plots, a) Al in 0.5 M HCl (without and with inhibitor), b) Al in 0.1 M NaOH solution (without and with inhibitor)

Table 6. Potentiodynamic polarization (Tafel plot parameters) parameters

Electrode	Medium	Concentration ($g\ L^{-1}$)	E_{corr} (mV)	$i_{corr}\times10^{-3}$ (A)	βc ($V\ dec^{-1}$)	βa ($V\ dec^{-1}$)	η_p
Aluminum	0.5 M HCl	Blank	-770	8.916	6.679	6.031	
		7.5	-739	1.800	5.624	5.843	79.81
		15	-765	1.520	5.496	6.353	82.95
		30	-748	1.471	5.693	6.017	83.50
		45	-715	1.116	3.971	4.813	87.48
	0.1M	Blank	-1579	17.000	5.307	4.364	
	NaOH	7.5	-1597	4.022	5.031	4.760	76.341
		15	-1545	3.324	5.123	4.760	80.447
		30	-1533	2.304	5.092	4.804	86.447
		45	-1531	2.246	5.035	4.841	86.788

Scanning electron microscopy technique

Fig. 5 (a, b) shows the SEM topography for Al in 0.5 M HCl and Al in 0.1 M NaOH solution (without and with Areca fat) respectively. Al surface exposed to corrosive solutions (0.5 M HCl/0.1 M NaOH solution) is greatly damaged, as a result large number of scratches appeared on Al surface. But, in another case (Al metal with Areca fat species), improvement in the Al surface morphology was observed, which is due to corrosion inhibition action of Areca fat on the Al surface in both 0.5 M HCl/0.1 M NaOH mediums. Therefore, majority scratches on Al surfcae are filled up and become smooth.

Without inhibitor With inhibitor

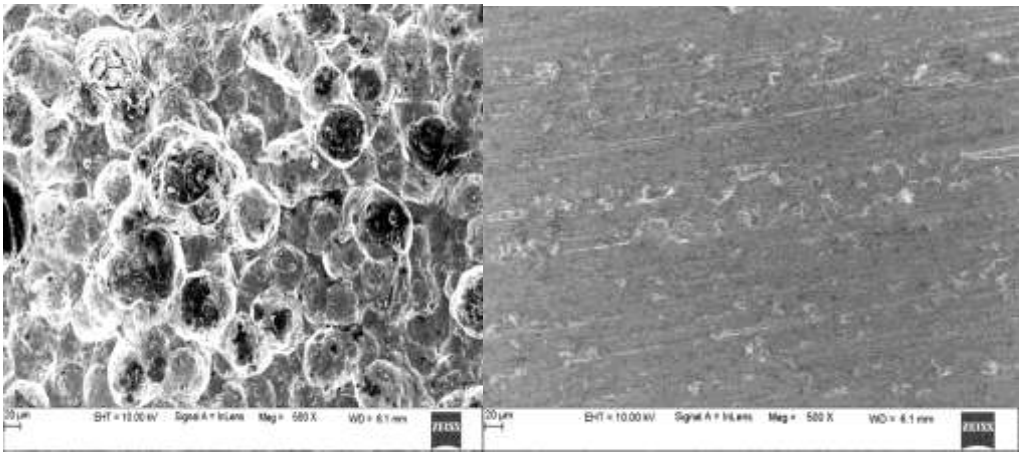


Fig. 5 a. (Al in 0.5 M HCl solution)

Without inhibitor With inhibitor

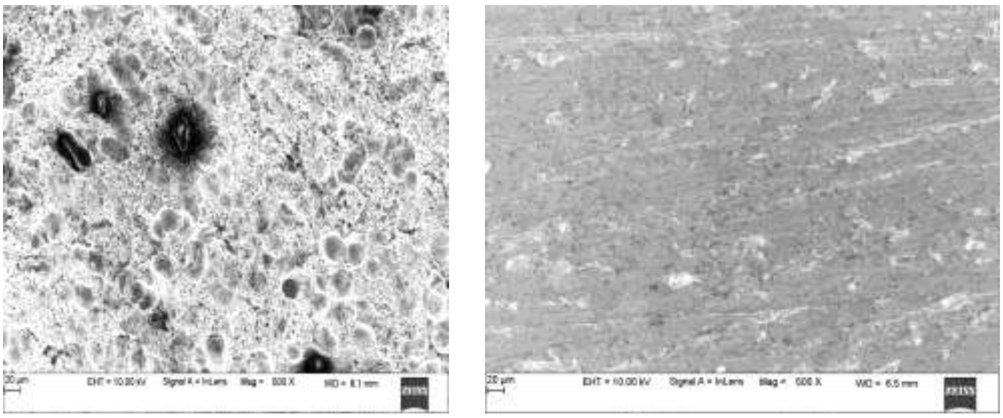
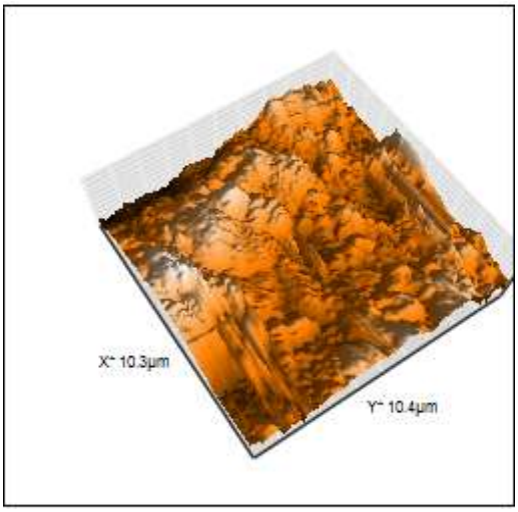


Fig. 5 b. (Al in 0.1 M NaOH solution)
Fig. 5 (a, b). SEM images of Al specimen

Atomic force microscopy technique

Closer look at Fig. 6 a, b reveals that, Al surface in 0.5 M HCl /0.1 M NaOH solution was highly damaged with huge number of deep voids owing to the aggressive nature of the HCl/NaOH solution on the surface of the Al and average roughness (Sa) value was equal to 137.94 nm (for Al in 0.5 M HCl system) and134. 9 nm (for Al in 0.1 M NaOH solution) [Table 7], with the introduction of Areca fat, notable upgraded surface morphology of Al was observed and average roughness were equal to 96.061 nm (Al in 0.5 M HCl plus Areca fat) and 56.586 nm (Al in 0.1 M NaOH plus Areca fat) [Table 7], indicating of a lessening in the Al corrosion rate. The reduction in the roughness value is an indication of adsorbed protective film over the surface of the electrode and which fully supports the results of SEM studies.

Without inhibitor



With inhibitor

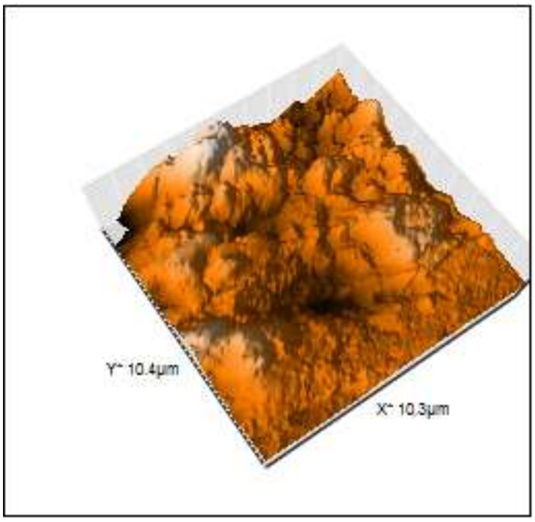
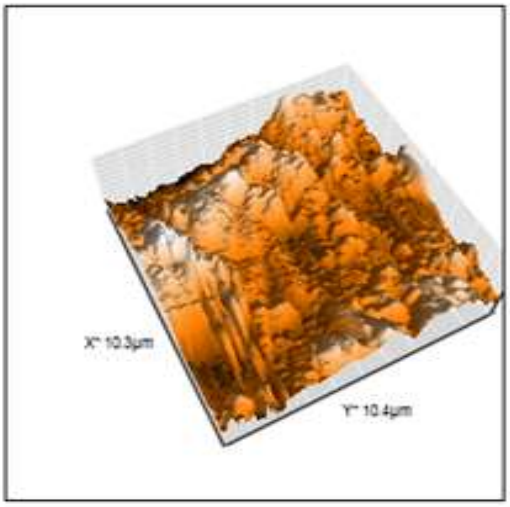


Fig. 6 a. (Al in 0.5 M HCl solution)

Without inhibitor



With inhibitor

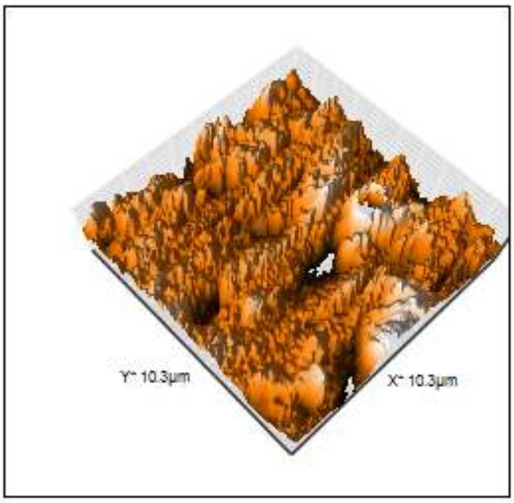


Fig. 6 b. (Al in 0.1 M NaOH solution)

Fig. 6 (a,b) AFM images of Al specimen

Table 7. AFM parameters

System	Average roughness (Sa) value
Al+0.5 M HCl	137.94 nm
Al in 0.5 M HCl plus Areca fat	96.061 nm
Al+0. 1 M NaOH	134. 9 nm
Al+0.1 M NaOH plus Areca fat	56.586 nm

Conclusion

Present investigation clearly shows the corrosion inhibition capacity of Areca fat species on the Al surface in both 0.5 M HCl and 0.1 M NaOH environments. Weight loss studies showed the inhibition action of Areca fat species through adsorption mode, which follows the Langmuir isotherm model. Mixed nature (type) of Areca fat for Al corrosion inhibition was confirmed by Tafel plots. Surface analysis by SEM and AFM techniques also validates the weight loss and Tafel study results.

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Conflicts of Interest

The authors declare no conflict of interest.

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