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Inhibition of Copper Corrosion by Arecanut Seed Extracts in 0.5 M HCl and 0.5 M NaOH Environments

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ABSTRACT

In the present study, anticorrosive action of the arecanut seed extract on the surface of copper in both 0.5 M HCl and 0.5 M NaOH systems was screened by weight loss and electrochemical methods. The protection efficiency of the inhibitor is directly proportional to its concentration and inversely proportional to copper immersion time and solution temperature. The copper corrosion inhibition process is related to adsorption of arecanut seed extract constituents on the copper surface and which is associated with the Langmuir adsorption model. Morphological variations on the metal surface in unprotected and protected conditions were screened by scanning electron microscopy technique.

Introduction

Due to the aggressive nature of hydrochloric acid and sodium hydroxide solutions, the copper metal undergoes dissolution during several industrial processes. Copper corrosion is a central problem for several industries. The damage occurs due to corrosion leads to high cost for inspection, repair and replacement of various tools and raises civic and ecological risks. To control these losses, many methods are developed such as coating, upgrading materials, process control, blending production fluids and inhibitors for copper protection. Among the developed methods, more interest paid on corrosion inhibitors due to its simple, low cost and strong adsorption characteristics [1-6]. Previously, many synthesized molecules have been successfully reported as effective corrosion inhibitors for various metals. But, the majority of these compounds are toxic and expensive in nature. In addition, the majority of these compounds are also insoluble in corrosive solutions. Therefore, it is required to replace the toxic, expensive and insoluble synthetic inhibitors from low cost eco-friendly corrosion inhibitors. Hence, corrosion scientists focused on greener species for its many advantages over synthetic compounds.

Green compounds are biodegradable in nature, easily accessible, renewable and economical. There are plenty reports on natural products as environmental benign potential corrosion inhibitors for various metals. Plant extract species shows commendable inhibition activity on a metal surface by adsorption mechanism. The elements such as nitrogen, phosphorous, oxygen and sulfur present in plant extract molecules were strongly adsorbed on metal surfaces and generate protective film on the surface of the metal, which is actively participating in metal corrosion inhibition process. Hence, metal is protected from corrosion process [7-13].

Arecanut is the major marketable plantation crop Southeast Asia. It is used in leucoderma, anaemia, leprosy, dyspepsia, distension, abdominal, diarrhea, jaundice, edema and obesity diseases.

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In addition, it has de-worming, antiparasitic, antifungal, anti-bacterial and anti- oxidant properties. All these properties are due to the presence of special elements (heteroatoms) in arecanut constituents. Many scientists reported that arecanut possessing rich sources of alkaloids, polyphenols, tannins, flavanoids and fatty acids.

The main phenolic compounds present in the arecanut are epicatechin, catechin, gallic acid, leucocyanidin, procyanidin B_1, and rutin. Alkaloids are the one of the important components of arecanut and its contents about 0.3-0.7 %. Arecoline is the major alkaloid (about 0.3-0.6 %) in arecanut, other important alkaloids present in arecanuts are guavacoline, arecolidine, arecaidine and guavacine. Areca flavonoids contains isorhamnetin, luteolin, chrysoeriol, quercetin, jacareubin, 5, 7, 4' – trihydroxy - 3', 5'-dimethoxy flavanone, jacareubin, liquiritigenin and 4', 5'-dihydroxy-3', 5', 7' – trimethoxyflavonone. Tannins are another class of arecanut constituents, mainly contains procyanidin A_1, procyanidin B_2, arecatannin B_1, arecatannin B_2, arecatannin A_1, arecatannin C_1, arecatannin A_3, and arecatannin A_2. Lauric acid, palmitic acid, myristic acid, oleic acid and stearic acids are the important fatty acids present in the arecanut [14-17]. Even though many reports are available on natural extracts as corrosion inhibitors for various metals, only a few papers discussed the inhibition property of the natural constituents on copper metal. Hence, in the present study, we selected arecanut seed constituents (tender stage) for the copper corrosion inhibition process. The arecanut seed (tender) possessing high concentration of polyphenols and low concentrations of alkaloids and fatty acids. No scientific report is available on arecanut seed (tender) extract constituents as corrosion inhibitors on copper in 0.5 M HCl/0.5 M NaOH environment. The corrosion inhibition of property of the arecanut seed (tender) extract constituents was investigated through weight loss, potentiodynamic polarization and scanning electron microscopy techniques.

Experimental sections

Preparation of metal samples

The copper of the type C11000 (composition, oxygen-0.04 (weight %) and Cu-99.03 weight %) was used in weight loss, potentiodynamic polarization and scanning electron microscopy studies.

Preparation of inhibitor

100 grams small pieces of arecanut seed were placed in a Soxhlet extraction chamber and extraction was carried out with 350 ml distilled water for about six hours duration. After that, the resulting solution was cooled and impurities are removed through Whatman filter paper by filtration. 4 concentrations namely, 0.75, 1.5, 3.0 and 4.5 (g/L) was prepared for copper corrosion studies.

Weight loss studies

The cleaned copper metals are exposed to 0.5 M HCl/0.5 M NaOH solution without and with different concentrations of arecanut seed extract at various temperatures (303, 308, 313, 318 and 323 K) for 1, 2,3,4,5, and 10 hours. The differences in the weight of copper metal were recorded. These values are used in the determination of copper corrosion rate and protection efficiency of the inhibitor. The experiment was repeated to get concordant value.

The copper corrosion rate and protection efficiency of the arecanut seed extract was calculated using the following equations,

Corrosion rate (υ_{corr}) in mils penetration per year (mpy) $= \frac{534W}{ATD}$ [18] (1)

where, W= weight loss of copper (mg), A= area of copper metal exposed (sq inch), T= contact time of copper metal (h) and D= density of copper metal (g cm^{-3}).

Protection efficiency (η_w) in percentage $= \frac{(W_1 - W_2)}{W_1} \times 100$ (2)

where, W_2= weight loss of copper metal in inhibiting solution and W_1= weight loss of copper metal in uninhibited solution.

Potentiodynamic polarization studies

The nature of corrosion inhibition property of the arecanut seed extract on copper metal in 0.5 M HCl/0.5 M NaOH solution was understood by Tafel studies. The potentiodynamic polarization curves (Tafel plot) were obtained by the three electrode cell system (copper metal= working cell, calomel electrode= standard cell and platinum electrode= auxiliary cell) connected to potentiostat (CHI 660C). The potential of ± 200 mv with scan rate 0.01 V/s was applied to Tafel studies.

Surface studies

Copper surface submerged in 0.5 M HCl/0.5 M NaOH solution without and with inhibitor for 2 hours was visualized by scanning electron microscopy (SEM) technique in order to sustain the weight loss and Tafel plot results.

Results and discussion

Weight loss studies

The influence of different amounts of the arecanut seed extract on the protection efficiency of copper in 0.5 M HCl/0.5 M NaOH solution was examined by weight loss technique. The resulting parameters are placed in Tables 1.1, and 1.2. The resulting table clearly shows that, the protection efficiency of arecanut seed extract increases with its amount in a 0.5 M HCl/0.5 M NaOH solution. This is due to the number of molecules from arecanut seed extract adsorbed on copper metal surface in 0.5 M HCl/0.5 M NaOH solution increases with increasing inhibitor concentration.

Therefore, arecanut seed extract molecules occupy more copper active sites where a direct attack of acid/base (0.5 M HCl/0.5 M NaOH) occurs and defend the copper metal against dissolution (corrosion). In both systems, the highest copper protection achieved at 4.5 (g/L) of arecanut seed extract.

Table 1. 1 Weight loss parameters at laboratory temperature (303 K).

Time (hours)	Concentration (g/L)	Copper			
		0.5 M HCl		0.5 M NaOH	
		υ_{corr} ($\times 10^{-4}$ (mpy)	ηw (%)	υ_{corr} ($\times 10^{-4}$ (mpy)	ηw (%)
1	Blank	0.615		0.384	
	0.75	0.192	68.750	0.153	60.000
	1.5	0.153	75.000	0.115	70.000
	3.0	0.115	81.250	0.076	80.000
	4.5	0.038	93.750	0.038	90.000
2	Blank	0.634		0.403	
	0.75	0.211	66.666	0.173	57.142
	1.5	0.173	72.727	0.134	66.666
	3.0	0.134	78.787	0.096	76.190
	4.5	0.096	84.848	0.057	85.714
3	Blank	0.653		0.474	
	0.75	0.230	64.705	0.205	56.756
	1.5	0.205	68.627	0.166	64.864
	3.0	0.153	76.470	0.128	72.972
	4.5	0.115	82.352	0.076	83.783
4	Blank	0.672		0.480	
	0.75	0.249	62.857	0.221	54.000
	1.5	0.221	67.142	0.173	64.000
	3.0	0.173	74.285	0.144	70.000
	4.5	0.124	81.428	0.096	80.000
5	Blank	0.692		0.499	
	0.75	0.269	61.111	0.230	53.846
	1.5	0.230	66.666	0.192	61.538
	3.0	0.192	72.222	0.161	67.692
	4.5	0.138	80.000	0.099	80.000
10	Blank	0.769		0.576	
	0.75	0.319	58.500	0.280	51.333
	1.5	0.307	60.000	0.230	60.000
	3.0	0.249	67.500	0.192	66.666
	4.5	0.153	80.000	0.126	78.000

Table 1.2. The variation of rate of copper corrosion as a function of media and substrate at 303 K.

Time (hours)	The rate of change of corrosion rate (mpy)	
	Copper in 0.5 M HCl	Copper in 0.5 M NaOH
1	16.184	10.105
2	6.604	7.070
3	5.678	6.236
4	5.419	5
5	5.014	5.040
10	5.026	4.571

Corrosion rate of copper increases with rise in contact time of electrode in both acid and alkali systems. But, the rate of reduction in the copper corrosion rate in both acid and alkali systems are small with respect to the immersion period (Table 1.2). The table also shows that, plant extract constituents are more effective for inhibition of copper corrosion in both acid and alkali systems. The order of attack of acid (0.5 M HCl) or alkali (0.5 M NaOH) on the copper surface is nearly equal to same, i.e., rate of change of copper corrosion rate (mpy) in 0.5 M HCl medium is approximately equal to the rate of change of copper corrosion rate (mpy) in 0.5 M NaOH medium.

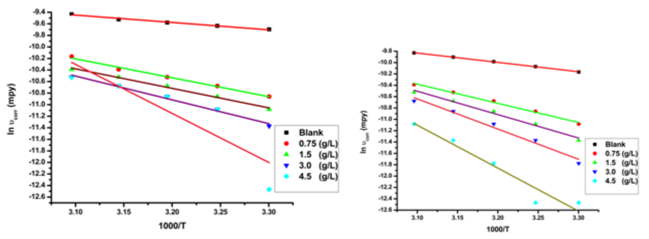
The inhibitory action of plant extract constituents on copper metal surface is almost same in both acid and alkali environments.

Table 2. Weight loss results at different solution temperatures

Temperature (K)	C oncentration (g/L)	Inhibition efficiency in percentage	
		Copper	
		0.5 M HCl	0.5 M NaOH
303	0.75	68.750	60.000
	1.5	75.000	70.000
	3.0	81.250	80.000
	4.5	93.750	90.000
308	0.75	64.705	54.545
	1.5	70.588	63.636
	3.0	76.470	72.727
	4.5	76.470	90.909
313	0.75	61.111	50.000
	1.5	66.666	58.333
	3.0	72.222	66.666
	4.5	72.222	83.333
318	0.75	57.894	46.153
	1.5	63.157	53.846
	3.0	68.421	61.538
	4.5	68.421	76.923
323	0.75	52.380	42.857
	1.5	61.904	50.000
	3.0	66.666	57.142
	4.5	66.666	71.428

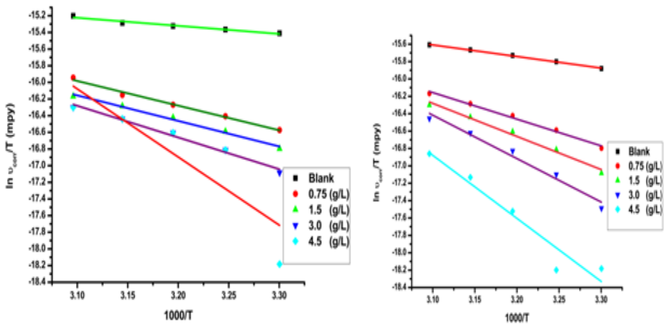
The copper corrosion rate increases and protection efficiency of arecanut seed extract decreases with the increasing corrosive solution temperature. This indicates that, arecanut seed extract molecules physically adsorbed on copper surface in a corrosive solution (0.5 M HCl/0.5 M NaOH solution). With increasing temperature, protective film formed on copper surface undergo slight desorption instead of adsorption and the strength of protective film (formed from the arecanut seed extract molecules) weakness and loses its effect. Therefore, high copper dissolution rate and low protection efficiency were noticed with improving the temperature (Table 2).

Kinetics of copper corrosion process in 0.5 M HCl/0.5 M NaOH solution without and with different amounts of the arecanut seed extract was understood by activation energy, entropy and enthalpy values.



(a) (b)

Figure 1. Arrhenius plots (without and with inhibitor), a) a) 0.5 M HCl, b) 0.5 M NaOH.
0.5 M HCl, b) 0.5 M NaOH.



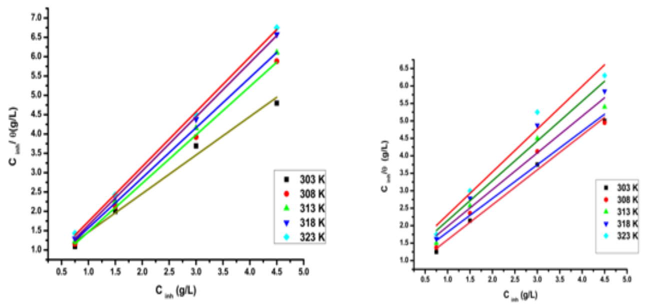
(a) (b)

Figure 2. Transition state plots, a) 0.5 M HCl, b) 0.5 M NaOH.

Activation energy was obtained from Fig.1, [19] whereas activation enthalpy and entropy were obtained from Fig.2 [20]. All these are placed in Table 3.

The activation energy values in pure 0.5 M HCl/0.5 M NaOH solution are low compared to inhibited 0.5 M HCl/0.5 M NaOH system. The higher activation energy values correlated with the enhanced double layer thickness. This suggests that, physical nature of adsorbed film on copper surface in corrosive solution. The more energy is required for the oxidation process in inhibiting system compared to uninhibited system. Hence, copper metal in 0.5 M HCl/0.5 M NaOH solution is highly protected in inhibiting system (in the presence of different amounts of arecanut seed extract).

The obtained activation enthalpy values in both unprotected and protected systems are in positive mode, which indicates the endothermic copper dissolution process in both 0.5 M HCl and 0.5 M NaOH environments. Activation entropy values in both cases move towards positive mode, which reflects the amplify in disorderness of the system. The copper corrosion inhibition mechanism was clearly understood by considering the adsorption isotherm model. On present investigation, the results of weight loss (gravimetric) are best fitted to the Langmuir adsorption model (Fig. 3) [21]. From Langmuir adsorption fit, the equilibrium constant of the inhibitor adsorption (K_{ads}) and free energy of adsorption (ΔG^o_{ads}) values were calculated and are presented in Table 4.



(a) (b)

Figure 3. Langmuir adsorption model, a) 0.5 M HCl, b) 0.5 M NaOH.

Table 3. Activation parameters.

Electrode	Medium	Concentration (g L^{-1})	*Ea** (kJ mol^{-1})	ΔH^* (kJ mol^{-1})	ΔS^* (J mol^{-1} K^{-1})
Copper	0.5 M HCl	Blank	10.639	8.0388	-356.412
		0.75	27.220	24.620	-311.316
		1.5	28.084	25.483	-310.081
		3.0	34.249	31.648	-292.006
		4.5	70.575	67.974	-177.715
	0.5 M NaOH	Blank	13.679	11.079	-350.177
		0.75	28.084	25.483	-310.081
		1.5	34.248	31.648	-292.006
		3.0	44.194	41.543	-262.461
		4.5	62.906	60.306	-208.138

Table 4. Thermodynamic parameters

Electrode	Medium	Temperature (K)	K_{ads} (L g^{-1})	ΔG^o_{ads} (kJ mol^{-1})
Copper	0.5 M HCl	303	2132.923	-36.718
		308	4568.922	-39.275
		313	3626.473	-39.311
		318	4087.973	-40.256
		323	3127.443	-40.169
	0.5 M NaOH	303	1677.148	-36.112
		308	1174.218	-35.795
		313	1076.356	-36.149
		318	993.561	-36.515
		323	922.594	-36.890

The higher K_{ads} values signifies the strong interaction between the arecanut seed extract molecules and copper surface. As a result, copper metal is protected with introduction of plant extract constituents (arecanut seed extract). Literature study [22-25] revealed that, ΔG^o_{ads} values closer to -40 kj/mol indicates the chemical interaction in between the plant extract species and the copper surface. Whereas, ΔG^o_{ads} values up to -20 kj/mol designates the physical interaction in between the plant extract species and the copper surface. In this study, the obtained ΔG^o_{ads} values are in between the literature values, which confirm the comprehensive adsorption mechanism (both physical and chemical adsorption).

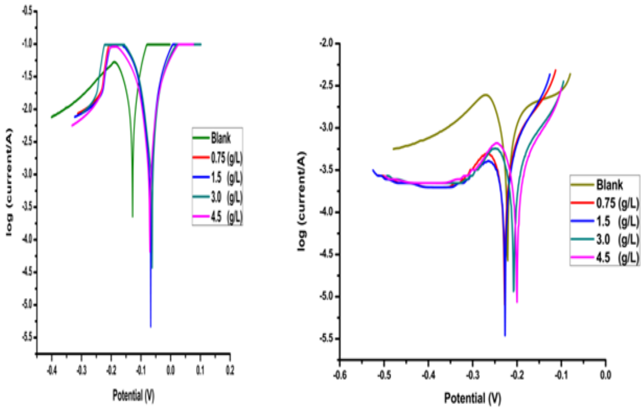
Potentiodynamic polarization technique (Tafel plot)

The potentiodynamic polarization action of copper in 0.5 M HCl/0.5 M NaOH solution was examined in relation to green inhibitor (arecanut seed extract) type and different arecanut seed extract concentrations. The corrosion current density (i_{corr}) values obtained from the Tafel curve (Fig. 4) was used in determination of protection efficiency values as per the following equation,

$$\text{Protection efficiency } [\eta p \ (\%)] = \left[1 - \frac{i'_{corr}}{i_{corr}}\right] \times 100 \qquad (3)$$

where, i'_{corr} = copper corrosion current density in protected 0.5 M HCl/0.5 M NaOH solution, and i_{corr} = copper corrosion current density in unprotected 0.5 M HCl/0.5 M NaOH solution.

The various readings from Tafel curve are summarized in Table 5. The addition of different amounts of arecanut seed extract molecules remarkably shifts the cathodic and anodic curves to lower corrosion current density values. In another way, the introduction of different amounts of arecanut seed extract to 0.5 M HCl/0.5 M NaOH solution greatly controls the both cathodic and anodic copper reactions. The molecules of the arecanut seed extract are strongly adsorbed on a copper surface and generates protective layer which blocks the movement of corrosive ions on the surface of the copper. The adsorption process increases with arecanut seed extract concentration.



(a) (b)

Figure 4. Tafel plots, a) 0.5 M HCl, b) 0.5 M NaOH.

Hence, higher copper protection rate (lower copper corrosion rate) was observed with improvement in the inhibitor concentrations. Furthermore, the arecanut seed extract molecules do not impact much on the corrosion potential (E corr) [26] [variation is less than 85 mv compared to bare solution] and cathodic (βc), anodic (βa) Tafel constant [only marginal variation] values, which signifies the mixed copper corrosion inhibition role of arecanut seed extract. This shows that, arecanut seed extract molecules strongly blocks the copper anodic and cathodic reaction in 0.5 M HCl/0.5 M NaOH solution by adsorption of arecanut extract molecules on a copper surface**.**

Surface studies

Scanning electron microscopy (SEM) technique

The Fig.5 (a, b) shows the micrographs of copper surface without and with the arecanut seed extract. Without arecanut seed extract, the copper surface is highly corroded (more number of scratches) because of aggressive corrosive ions attack on copper surface. But, in the presence of the arecanut seed extract, the smooth copper surface was observed, which is an indication of adsorption of arecanut seed extract molecules on a copper surface in 0.5 M HCl/0.5 M NaOH solution. Hence, metal is protected from corrosive ions.

Table 5. Tafel plot results.

Electrode	Medium	Concentration (g L-1)	Ecorr (mV)	$i_{corr}\times10^{-3}$ (A)	βc (V dec^{-1})	βa (V dec^{-1})	η_p (%)
Copper	0.5 M HCl	Blank	-128	89.610	4.572	0.004	
		0.75	-67	24.690	3.520	1.412	72.447
		1.5	-67	22.180	4.221	1.354	75.248
		3.0	-62	20.070	4.574	2.199	77.602
		4.5	-70	9.116	5.988	5.153	89.827
	0.5 M NaOH	Blank	-221	3.417	5.154	2.089	
		0.75	-228	0.745	4.771	8.810	78.194
		1.5	-227	0.649	3.556	10.551	80.983
		3.0	-208	0.574	5.303	9.560	83.184
		4.5	-200	0.499	5.733	10.900	85.373

without inhibitor with inhibitor

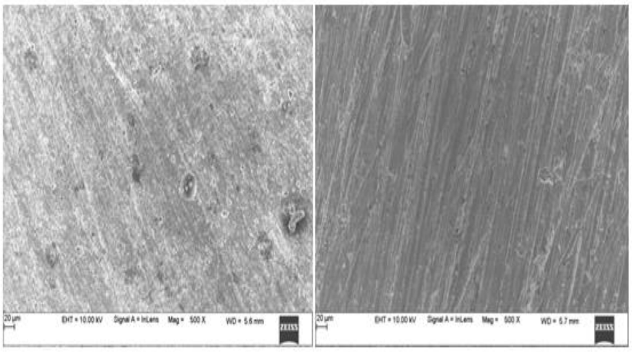


Figure 5 a. Copper metal in 0.5 M HCl system.

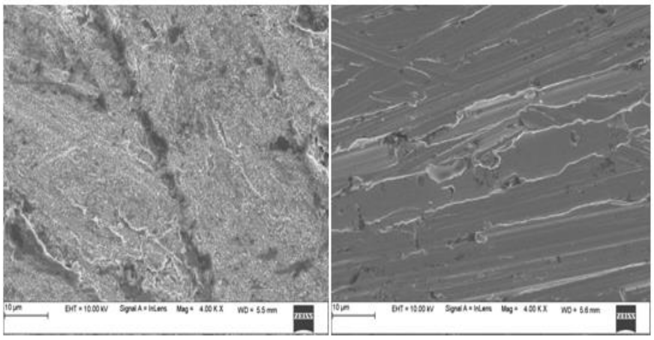


Figure 5 b. Copper metal in 0.5 M NaOH system.

Figure 5 a, b SEM images of copper specimen in acid and alkali environments.

Conclusion

The studied arecanut seed extract act as an effective green corrosion inhibitor for copper in both 0.5 M HCl and 0.5 M NaOH environments. The adsorption of arecanut extract molecules on the surface of the copper is spontaneous and follows the Langmuir adsorption theory. The potentiodynamic polarization study indicates that, arecanut seed extract molecules inhibit the both anodic and cathodic reactions of copper. SEM figures confirm the protective role of the arecanut seed extract on copper surface in 0.5 M HCl/0.5 M NaOH systems.

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