

PCAIJ, 10(4), 2015 [111-120]

Cactus as a green inhibitor for the corrosion of carbon steel in seawater

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ABSTRACT

The inhibitive effects of aqueous Cactus extracts on the corrosion of carbon steel have been investigated; in 3.5% NaClusing polarization measurements. The obtained results show an inhibition efficiency of about 94% in the presence of 1ppm Cactus at 298K. The Cactus extract acts as mixed inhibitor according to the corrosion potentials E_{corr} shifted toward either noble or active direction in the presence of inhibitor. The adsorption of used Cactus compounds on the carbon steel surface obeys Langmuir's isotherm. Theapparent activation energy, enthalpy of the dissolution process and the free energy were determined and discussed. © 2015 Trade Science Inc. - INDIA

INTRODUCTION

Carbon steel, the most widely used engineering material, despite its relatively limited corrosion resistance used in large tonnages in marine applications, nuclear powered transportation, chemical processing, petroleum production and refining, pipelines, mining, construction and metal-processing equipment^[1].

Corrosion is a surface phenomenon known as the attack of metals or alloys by their environment as air, water or soil in chemical or electrochemical reaction to form more sTABLE compounds^[2,3].

There are various methods for prevention of corrosion which basically comprises those protective measures providing separation of metal surfaces from corrosive environments or those which cater for adjustment or altering the environment. These various

KEYWORDS

Cactus; Corrosion; Carbon steel; Inhibitor; Extract.

methods of corrosion prevention include cathodic protection, anodic protection, coating and the use of corrosion inhibitor^[4].

An inhibitor is a substance (or a combination of substances) added in a very low concentration to treatthe surface of a metal that is exposed to a corrosive environment that terminates or diminishes the corrosion of a metal. These are also known as siteblocking elements, blocking species or adsorption siteblockers, due to their adsorptive properties^[5,6].

In an interesting study, investigated the extract of Cactus forcorrosion inhibition of C.S in 3.5% NaCl solution, in which the Cactus extract was obtained by aqueous extraction.Originally native toMexico, today the Cactus is commercially grown inMexico, Chile, Argentina, Morocco, Italy, and partsof California, Texas, and Florida^[7].

The present investigation presents results on the

corrosionperformance of carbon steel in 3.5% NaCl when rawCactusadditions such as corrosion inhibitors.

EXPERIMENTAL PROCEDURE

Materials

The steel used in this study is a carbon steel (C45) with a chemical composition (in wt%) of 0.42% C, 0.40 % Si, 0.50% Mn, 0.045% S, 0.40% Cr, 0.045% P, 0.40% Ni, 0.0.1% Mo and the remainder is iron (Fe). The carbon steel samples were(0.5 mm) thickness was mechanically cut into circular sample with dimensions of 2.5 cm in diameter then pre-treated prior to the experiments by grinding with emery paper SiC (120, 600 and 1200); rinse with distilled water, degreased in acetone, washed again with distilled water and then dried at room temperature before immersed in corrosivesyntheses seawater. The seawater solution was prepared by dissolved 35 g NaCl in 1L distilled water.

Preparation of cactus extract

Cleaned plant lave were grounded and kept at a laboratory temperature until use, for the preparation of aqueous extract (50) grams of Cactus laves powder were taken, placed in the conical flask containing (500) cm³ of Hot Distilled water, mixed by magnetic stirrer for (30) minutes. After that the solution stand in the laboratory temperature for 24h, then it well be filtrated using Buchner apparatus. Three volumes of Cactus extract were used as inhibitor includes 5, 10, 30, 50 mL distilled to 1L with distilled water.

Electrochemical measurements

The electrochemical measurements were carried out using Mlab (Germany, 2000) potentiostate and controlled by computer and MLabSci software which were used for data acquisition and analysis under static condition. The corrosion cell used had three electrodes, the reference electrode was a silver-silver chloride, platinum electrode was used as auxiliary electrode with 1 cm2surface area of and the working electrode was carbon steel. All potentials

Physical CHEMISTRY An Indian Journal given in this study were referred to this reference electrode. The working electrode was immersed in test solution for 15 minutes to a establish steady state open circuit potential (Eocp), then electrochemical measurements were performed in potential range (± 200) mV. All electrochemical tests have been performed in aerated solutions at (298-328) K.

RESULTS AND DISCUSSION

Corrosion behavior

Figure (1) shows the polarization curves of carbon steelin 3.5% NaCl medium in absence and presence three concentrations of Cactus extract. The anodic and cathodic current–potential curves were extrapolated up to their interaction point where corrosion current density (i_{corr}) and corrosion potential (E_{corr}) are obtained. The electrochemical parameters such as corrosion currentdensity (i_{corr}), corrosion potential (E_{corr}), cathodic and anodicTafel slopes (bc&ba) and inhibition efficiency obtained from polarizationmeasurements are listed in TABLE 1.

The data listed in TABLE (1) indicates that the corrosion potentials E_{corr} shifttoward either noble or active direction in the presence of inhibitor, i.e., Cactus extract acts as mixed inhibitor. It is clear from the results of electrochemical polarization measurements that the addition of inhibitor causes a decrease in the current density. The values i_{corr} of C.S in the inhibited solution are smaller than those for theinhibitor free solution. The change in cathodicTafel slope (bc) values (TABLE 1), suggest that the reaction mechanism of the hydrogen reductionis change due to the effect of Cactus adsorption.

The inhibition officiency IE (%) can be calculated using the equation given below^[8]:

$$IE\% = \frac{(i_{corr})a - (i_{corr})p}{(i_{corr})a} \times 100$$
(1)

Where $(i_{corr})_a$ and $(i_{corr})_p$ are the corrosion current density (μ A.cm⁻²) in the absence and the presence of the inhibitor respectively.

The best efficiencies were obtained in presence of 10 mL/L (1ppm) of Cactusextract whichgive IE reach to 94.22% at 298K as shown in Figure (2).



Figure 1 : Polarization curves for C.S corrosion in 3.5% NaCl in absence and presence different concentration of cactus at temperature range (298-328)K

Adsorption isotherm

The involves studies the assumption that the corrosion reactions are prevented from occurring over the area (or active sites) of the metal surface covered by adsorbed inhibitor species, whereas these corrosion reactions occurnormally on the inhibitor-free area^[9]. Adsorption isotherms are usually used to describe the adsorption process.

The most frequently used isotherms include Langmuir, Temkin, Frumkin, Hill deBoer, Parsons, Flory-Huggins, Dhar- Flory-Huggins, Bockris-Swinkels an thermodynamic/kinetic model. Adsorption of the inhibitor compounds depends upon the charge and the nature of the metal surface, electronic characteristics of the metal surface onadsorption of solvent and other ionic species, temperature of the corrosion reaction and the electrochemical potentialat the metal solution interface^[10-14]. The establishment of adsorption isotherms that describe the adsorption of a corrosion inhibitor can provide important clues to the nature of the metal- inhibitor interaction. Adsorption of the organic molecules occurs as the interaction energy between molecule and metal surface is higher than that between the H₂O molecule and the metalsurface^[15].

In order to obtain the adsorption isotherm, the

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Tem	p./K	-OCP/mV	-E _{corr} /mV	$I_{corr}/\mu A.cm^{-2}$	-bc/ mV.Dec ⁻¹	ba/ mV.Dec ⁻¹	CR/ g.m ⁻² .d ⁻¹	P.L/mm.a ⁻¹	IE%	θ	Rp
Free 3.5% NaCl	298	541	520.0	168.72	243.9	107.2	42.1	1.92	-	-	191.7
	308	568	573.5	201.57	179.1	104.2	46.1	2.11	-	-	141.9
	318	592	622.0	220.34	140.3	82.2	55.3	2.53	-	-	102.1
	328	609	651.0	280.34	137.3	79.6	64.8	2.99	-	-	78.1
0.5ppm inhibitor	298	535	592.0	22.8	48.7	81.4	5.677	0.265	86.5	0.865	580.3
	308	562	610.0	32.09	121.2	54.6	7.99	0.372	84.1	0.841	509.3
	318	649	676.9	39.19	124.9	53.5	9.758	0.455	82.2	0.822	415
	328	685	713.2	59.94	121.8	73.2	14.93	0.696	78.6	0.786	331.2
1 ppm inhibito	298	379	416.5	9.68	42.0	34.8	2.4	0.112	94.3	0.943	853.7
	308	475	482.0	17.88	32.0	34.8	4.5	0.208	91.1	0.911	404.8
	318	612	626.9	29.18	54.0	48.2	7.3	0.339	86.8	0.868	378.9
	328	628	677.0	37.79	70.5	68.2	9.5	4.39	86.5	0.865	398.3
3ppm inhibitor	298	470	476.6	25.22	49.8	39.0	6.3	0.215	85.1	0.851	376.6
	308	526	553.5	36.66	43.2	54.8	9.1	0.395	81.8	0.818	286.1
	318	587	614.7	52.37	79.0	58.3	13.1	0.721	76.2	0.762	278.1
	328	614	643.7	66.90	43.1	67.4	16.7	0.685	76.1	0.761	170.6
om Ditor	298	500	527.3	15.45	22.8	31.9	3.8	0.179	90.8	0.908	373.7
	308	579	618.5	28.20	33.0	47.5	7.1	0.327	86.0	0.860	299.8
5pJ nhil	318	601	626.4	69.65	86.0	56.2	17.3	0.808	68.4	0.684	211.9
	328	622	639.0	89.25	106.3	51.1	22.2	0.885	68.2	0.682	167.9

(2)

TABLE 1 : Corrosion kinetic parameters for carbon steel in absence and presence different concentration of cactusin 3.5% NaCl at different temperature range(298-328)K

degree of surface coverage (θ) for various concentrations of theinhibitor has been calculated according to equation(2):

$$\theta = \left[1 - \frac{i_{(corr)p}}{i_{(corr)a}}\right]$$

The plot of C_{inh}/θ versus C_{inh} yields a straight linewith regression coefficient, R^2 , almost equal to 1. Proving that the adsorption o the inhibitors from 3.5% NaCl solution on the carbon steel surfaceobeys the Langmuir adsorption isotherm (Figure 3)

$$\frac{C_{inh}}{\theta} = \frac{1}{K_{ads}} + C_{inh}$$
(3)

 C_{inh} is the inhibitor concentration; **è** is the fraction of the surface covered. K_{ads} is the adsorption coefficient (Equilibrium constant).

 K_{ads} represents the strength between adsorbate and adsorbent. Larger values of the K_{ads} imply more efficient adsorption and hence better inhibition efficiency^[16-17]. K_{ads} value decreased in this study with increase in the temperature indicating that adsorption of Cactus on the carbon steel surface was unfa-

Physical CHEMISTRY An Indian Journal vorable at higher temperature.

The equilibrium constant for the adsorption process is related to the standard free energy of adsorption by the expression^[18]:

$$K_{ads} = \frac{1}{55.5} \exp\left[\frac{-\Delta G_{ads}}{RT}\right]$$
(4)

This equation can also be expressed as:

 $\Delta G_{ads} = -2.303 RT log(55.5 K_{ads})$ (5) where, ΔG_{ads} is Gibbs free energy of adsorption, T is the temperature in Kelvin and K_{ads} is the equilibrium constant for the adsorption process and 55.5 is the molar concentration of water in solution. K_{ads} value was calculated from the intercept of the Figure (3) and listed in TABLE (2). The negative values of ΔG_{ads} indicates the spontaneity of the adsorption process and the stability of the adsorbed species on the mild steel surface^[19,20]. Usually the values around -20 kJ mol⁻¹ or lower are consistent with physisorption, while those higher than -40kJ mol⁻¹ involve chemisorption^[21]. When charged species are adsorbed on the metal surface, there is possibility





Figure 2 : The relation between inhibitor efficiency (IE%) and concentration of inhibitor in 3.5% NaCl solution



Figure 3 : Langmuir isotherm plot for the adsorption of cactus on the surface of carbon steel

of coulombic interaction between adsorbed cation and anion thereby causing increase in the Gibb's free energy even if no chemical bonds are formed^[22].

The values of ΔG_{ads} for the Cactusare listed in TABLE (2). These values indicate physical adsorption of Cactus on carbon steel surface.

Assuming the thermodynamic model, corrosion inhibition of carbon steel in presence of Cactus can be better explained using the enthalpy of adsorption ΔH_{ads} and entropy of adsorption ΔS_{ads} which can be calculated from the integrated vantHoff equation^[23,24]:

$$logK_{ads} = \frac{-\Delta H_{ads}}{2.303RT} + \frac{\Delta S_{ads}}{2.303R} + \frac{\log 1}{55.5}$$
(6)

To calculate the enthalpy of adsorption and entropy of adsorption, $\log K_{ads}$ was plotted against 1/T (Figure 4) and straight line was obtained with slope equal to $(-\Delta H_{ads}/2.303R)$ and intercept equal to $(\Delta S_{ads}/2.303R+\log 1/55.5)$. The calculated values of the heat of adsorption and entropy of adsorption are listed in TABLE (2) also.

The negative value of ΔH_{ads} indicates that the adsorption of Cactus molecules is an exothermic process.

Kinetic parameters for the corrosion inhibition process

Temperature plays an important role in understanding the inhibitive mechanism of the corrosion process. To assess the temperature effect, experiments were performed at 298 - 328K inuninhibited and inhibited solutions containing different concentrations of Cactus and the corrosion rate was evaluated as presented in TABLE 3. The relationship between the corrosion rate (CR)of carbon steel in 3.5% NaCl solution and temperature (T) is expressed by the Arrhenius equation^[18,25-27]:

$$logCR = \frac{-E_a}{2.303RT} + \log \Box$$
(7)

where,Ea is the apparent effective activation energy, R molar gas constant and A the Arrhenius pre expo-

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Temp.	K _{ads}	$\Delta G_{ads} / kJ.mol^{-1}$	R ²	- ΔS_{ads} / J.K ⁻¹ .mol ⁻¹	-AH _{ads} / kJ.mol ⁻¹
298	38.168	-18.98	0.989		
308	666.667	-26.94	0.998	256	86
318	3.438	-13.88	0.996	5.36	.81
328	3.598	-14.45	0.996		

 TABLE 2 : Thermodynamic parameters for adsorption of the inhibitors on the surface of carbon steel in 3.5% NaCl solution



1/T Figure 4 : The relationship between logK_{ads} and 1/T.

TABLE 3 : Activation energy (Ea), pre exponential factor (A) and the thermodynamic parameters for the corrosion activation complex of C.S in 3.5% NaCl solution in the absence and presence of Cactus concentration over the temperature range 293 -323 K

C	$\Delta G_a/kJ.mol^{-1}$				AII /lrI mol ⁻¹	$-\Delta S_a/$	Ea/kI mal ⁻¹	A		
Cinh	298	308	318	328	ΔП _а /КЈ.ШОІ	kJ.mol ⁻¹ .K ⁻¹	La/KJ.11101	Molecules.cm ⁻² ·S ⁻¹		
without	63.75	65.54	67.32	69.11	10.47	0.179	13.1	1.96E+28		
0.5ppm	68.71	70.26	71.81	73.36	22.54	0.155	25.1	8.59E+28		
1ppm	70.65	71.85	73.05	74.26	34.75	0.120	37.4	5.45E+30		
3ppm	68.40	69.88	71.37	72.85	24.14	0.149	26.7	1.86E+29		
5ppm	69.61	93.41	94.89	96.38	47.67	0.074	50.3	1.52E+33		

nential factor.

Figure (5) show the relation between logarithm of corrosion rate versus 1/T whichgave straight line with regression coefficient close to unity. The values of apparent activationenergy (Ea) obtained from the slope (-Ea/2.303R) of the lines and the pre exponential factor (A)obtained from the intercept (log A) are listed in TABLE (3).

The activation energy data listed in TABLE (3) show an increasing with adding Cactus in comparison to the uninhibited solution. The larger value of E_a was obtained with 5 ppminhibitor which reach to 50.3 kJ.mol⁻¹.

Furthermore, eq. (8),could be also used to determine the thermodynamic parameters of activated

Physical CHEMISTRY Au Indian Journal complex for corrosion process in absence and present of inhibition:

$$\frac{\log CR}{\log R} = \frac{\log R}{1 + \Delta S} - \Delta H$$

T Nh [•] 2.303R ⁻ 2.303RT (8) Where CR (i_{corr}) is the corrosion rate, E_a is the apparent activation energy, R is the universal gas constant (8.314 J mol⁻¹ K⁻¹), T is temperature, A is the Arrhenius pre-exponential factor, h is the Plank's constant (6.626176 x 10⁻³⁴Js), N is the Avogadro's number (6.022 x 10²³ mol⁻¹), Δ S_a is the entropy and Δ H_a is the enthalpy of activation. The plot of log CR/T vs. 1/T obtained straight lines were obtained with the slope of (- Δ H_a / 2.303 R) and an intercept of [(log (R/Nh) + (Δ S_a, respectively were calculated,

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Figure 5 : Arrhenius Plot of log CR Versus 1/T for the corrosion of C.S in 3.5% NaCl solution in absence and presence containing various cactus concentrations



0.0030.003050.00310.003150.003201009250.00330.003350.0034

Figure 6 : Plot of log CR/T Versus 1/T for the corrosion of C.S in 3.5% NaCl solution in absence and presence various cactus concentrations

while the free energy ΔG_a was calculated using the following equation^[2];

$$\Delta G \mathbf{a} = \Delta H \mathbf{a} - T \Delta S \mathbf{a} \tag{9}$$

The positive value of enthalpy ΔH in the absence and presence of various concentration of inhibitor reflects the endothermic nature of carbon steel activation complex forming meaning that dissolution of carbon steel is difficult^[24]. It is evident from the table that the value (Ea) of increased in the presence of the inhibitor than the uninhibited solution indicating higher protection efficiency and the highest value of ΔH was in the presence 5 ppm of the inhibitor. On comparing the values of entropy (ΔS) listed in TABLE (3), it isclear that entropy increased in the presence of the studied inhibitor compared to free 3.5% NaClsolution, which means a more dis-ordering in activation complex was happen in present of inhibition.

The values of ΔG were positive and showed slightly increase with increasing temperature, which means that inhibitor cause to reduced the thermodynamic feasibility of corrosion.

Adsorption mechanism

To explanation the mechanism of inhibitor adsorption, it is necessary to establish the adsorption

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modes of the inhibiting species (whether molecular or ionic)^[28]. The dominantadsorption mode will be dependent on factors such as the extract composition, type of corrosive medium ions as well as chemicalchanges to the extract. The Cactus including polyphenolic compounds (Alue-emodin, Myricetin, Kaempferol and Taxifolin)^[29] and the

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chemical structures is shown in Figure (8). From Figure 8 shows that these compounds contain many O atoms in functional groups, π -electrons and O– heterocyclic rings in cause inhibitors are the reasons of its inhibition effect. The data obtained from the temperature dependence of the inhibition process suggest a physical adsorption. Where they will

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Figure 10 : FTIR spectra of cactusfilm formed on the carbon steel surface after immersion in 3.5% NaCl solution

adsorbed on the metal/solution interface by more the following ways: 1) donor-acceptor interactions between the π -electrons of aromatic rings and vacant d-orbital of surface iron atoms, and 2) interaction between unshared electron pairs of hetro atoms and vacant d-orbital of iron surface atoms. These process drastically reduce the transport of aggressive ions to the metal surface thereby controlling the corrosion of carbon steel^[31].

Results of FTIR spectroscopy

The main constituent of Cactus is water (80-95%), followed by small amounts of carbohydrates (3-7%), fiber (1-2%),and protein (0.5-1%); other compounds are only partly known andhave not been quantitatively determined^[31]. Figure (8) shows structures of most important phytochemicals found in Cactus^[29]. Figure (9) shows a reference transmission spectrum of extract Cactus. The spectrum shows a multi peak band at 3340-3406 cm⁻¹which indicates the presence of the OH group, also it shows a peak at 1731 cm⁻¹ due to the presence of the carbonyl group for ketone. In this spectrum, there are bands at 1251 cm⁻¹ and 2937 cm⁻¹ indicating the presence of COC and CH groups and the CO group for secondary alcohol shows at 1074 cm⁻¹.

In comparison between Figure (9) and Figure (10) it is clear that ketonic carbonyl bond and C-H bonds were disappeared due to the adsorption process occur on metal surface. The absorption frequency of -OH and C-O-C bands have been shifted to more frequency values and the C-O band in secondary alcohol was shifted to 1030 cm⁻¹ after inhibitor adsorption as shown in Figure (10).

CONCLUSION

Cactus was found to be a good inhibitor forcarbon steel corrosion in 3.5% NaCl.

The Cactus extract acts as mixed inhibitor because the corrosion potentials E_{corr} shift toward either noble or active direction in the presence of inhibitor.

The adsorption model obeys Langmuir adsorption isotherm and was a physisorption type.

The activation energy value increased with increasing Cactus concentration.

The positive value of Gibbs free energy of corrosion process activation complex indicates the nonspontaneous of the corrosion process. Moreover,



the negative values of entropy of activation decreasing with increase inhibitor concentration that indicate increasing in the disordering of corrosionprocessactivation complex.

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