

Research Article

Effect of Serine and Methionine on Electrochemical Behavior of the Corrosion of Mild Steel in Aqueous Solutions

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The pitting corrosion behaviour of mild steel in Na_2HPO_4 solutions contains chloride ion as an aggressive ion and serine and methionine as inhibitors were investigated using open-circuit potential (OCP), potentiodynamic polarization measurements, and pitting corrosion current measurements; both inhibitors shift the potential in the positive direction. The corrosion rate of the mild steel was measured in the absence and presence of the inhibitors, and the inhibition efficiency of the amino acids at a concentration of 0.02 M was calculated. The pitting corrosion current shows that increasing concentration of the inhibitor causes a decrease in pitting current density, and inhibition efficiency increases with increasing concentration of the inhibitors. The adsorption of these inhibitors on the mild steel surface obeys Langmuir isotherm, and the calculated adsorption free energy (ΔG_{ads}^0) for the inhibitors on the mild steel in 0.1 M ($\text{Na}_2\text{HPO}_4 + \text{NaCl}$) solutions was found to be $(-24.61, -29.34)$ kJ/mol for serine and methionine, respectively, which reveals strong physical adsorption of the amino acids molecules on the mild steel surface.

1. Introduction

Mild steel is one of the major construction materials, which is extensively used in chemicals and industries [1, 2]. Pitting corrosion in the presence of aggressive chloride ions Cl^- is the most frequently encountered cause of failure of mild steel. It is generally accepted that pitting proceeds by destruction of the protective oxide by adsorption of Cl^- that subsequently passes into solution [3]. Compounds that retard or stop this process when present in aggressive medium are prospective corrosion inhibitors. Its protection against pitting corrosion has attracted much attention. One of the available methods is the use of soluble inhibitors; the use of inhibitor is one of the most practical methods to protect metals from corrosion, especially in aggressive media in particular, in chemicals, and petrochemical and oil industries [4, 5]. Unfortunately, many of the inhibitors used are inorganic salts and organic compounds, with toxic properties or limited solubility [6]. Protective action of inorganic inhibitors is related to the formation of oxide film or hardly soluble salt on the metal

surface. On the other hand protective action of organic inhibitors comes from the adsorption on the oxide films. Increasing awareness of the health and ecological risks has drawn attention to finding more suitable inhibitors, which are nontoxic. Amino acids inhibitors fall into this category since they are cheap; most of them are soluble in aqueous media and are easy to produce at high purity [7]. Using amino acids as inhibitor for localized corrosion is important from the theoretical point of view, because in construction principles of prevention of localized corrosion organic weak acids or base that are accounted any substituent that can influence the electronic structure and nucleophilic properties [3]. Some studies involve the influence of amino acids on the corrosion of iron [8–10], steel [4, 11–20], aluminum [3, 7, 21] lead and its alloys [22–24], copper and its alloys [25–29], cobalt [30], vanadium [31], nickel [32] and tin [33]. The objective of the present work is to investigate the inhibition effect of serine and methionine for the pitting corrosion of mild steel in aqueous solution.

TABLE 1: The chemical composition of mild steel of the electrode (%).

C	0.175
Si	0.130
Mn	0.534
P	0.008
S	0.0094
Cr	0.0117
Mo	0.002
Ni	0.0188
AL	0.005
Co	0.0075
Cu	0.0047
Nb	0.003
Ti	0.002
V	0.005
W	0.172
Fe	99.07

2. Materials and Experimental Techniques

2.1. *Materials.* The steel samples used in this study were made from a mild steel its the chemical composition is shown in Table 1.

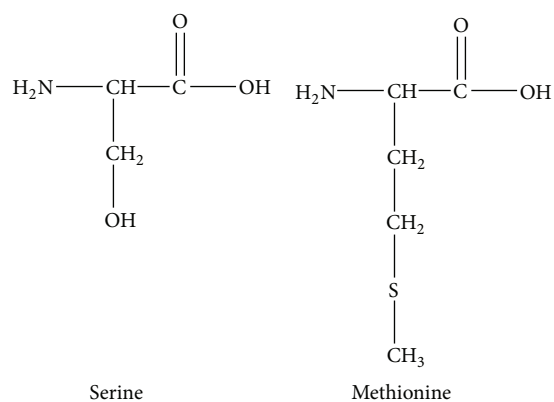
These steel samples were polished using a series of emery paper 100, 166, 320, 400, and 1200; then the specimens are washed thoroughly with double-distilled water degreased and dried with acetone. All chemicals used in this paper reagent grade, disodium hydrogen phosphate (Na_2HPO_4) as passivating agent, sodium chloride (NaCl) solution as an aggressive solution and serine (Ser.) and methionine (Met.) as corrosion inhibitors.

2.2. Techniques

2.2.1. *Open-Circuit Potential Measurements.* Open-circuit potential measurements are carried out in a conventional three-electrode test cell, the working electrode sample with an exposed area to the corrosive solution of 1 cm^2 . A saturated calomel electrode (SCE) and two graphite rods are used, respectively, as reference and auxiliary electrodes. The potential was measured using the potentiostat EG&G model 273A and recorded it as a function of time until the steady state potential was attained (Scheme 1).

2.2.2. *Potentiodynamic Polarization Measurements.* The potentiodynamic current-potential curves were recorded by changing the electrode potential automatically from -250 mV to $+250\text{ mV}$ (SCE) with a scan rate of 0.2 mVs^{-1} [34].

2.2.3. *Pitting Corrosion Current Measurements.* The simple electrolytic cell which was used, as shown diagrammatically in Figure 1. It consists of two 250 mL Pyrex glass tube endings in a fine porosity, low resistance (G4) sintered glass disc [35–37]. Two identical cylindrical mild steel electrodes



SCHEME 1

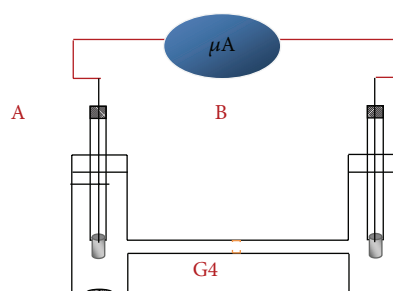


FIGURE 1: Electrical circuit used for measuring pitting corrosion currents.

with a diameter of (1.127 cm) and height of (3.059 cm) were short circuited through a low resistance micrometer (Zero Resistance Ammeter). The passivating agents used were Na_2HPO_4 solution, Cl^- ions were used as aggression ions and amino acids (Serine and Methionin) were utilized as corrosion inhibitors. The principle of this method is based on the presence of two similar electrodes in a two-compartment cell filled with a solution of the passivation of such has a concentration to stifle corrosion. When a steady state is attained, no current flows through the electrical circuit. An aggressive anion (Cl^-) is then introduced into one of the cells compartments, to initiate pitting corrosion. Local corrosion currents will flow through the circuit, and the pitting current will be measured using (Zero Resistance Ammeter). This is followed as a function of time until steady-state current values are obtained. Pitting corrosion current measurements were performed at different concentration of the inhibitor (amino acids) of (Cl^-). This was done by adding different concentrations of the inhibitor (amino acids) to the solution containing a constant amount of the (Cl^-) and recording the corrosion current.

3. Results and Discussion

3.1. *Open-Circuit Potential Measurements.* The electrochemical behaviour of mild steel in $0.1\text{ M Na}_2\text{HPO}_4$ in absence and in the presence of 0.1 M NaCl and 0.02 M of serine and methionine was studied on the basis of the change in

TABLE 2: Values of E_{im} and $E_{s.s.}$ for mild steel in 0.1 M Na_2HPO_4 + 0.1 M NaCl solution (blank) in presence of 0.02 M of different amino acids.

Media	E_{im} (mV)	$E_{s.s.}$ (mV)
0.1 M Na_2HPO_4	-382	-325
0.1M NaH_2PO_4 +0.1 M NaCl (Blank)	-390	-326
(Blank) + Ser.	-351	-293
(Blank) + Met.	-392	-290

potentials with time using (OCP). The values of immersion potential (E_{im}) and steady state potential ($E_{s.s.}$) are given Table 2. The addition of NaCl to Na_2HPO_4 solution shifts the potential to more -ve direction; this gives an indication of the dissolution of the preformed protective film on the surface the steel. The presence of the amino acid in the solution shifts the $E_{s.s.}$ value most likely in the more noble direction. This behavior indicates that amino acids are adsorbed on anodic sites and, consequently, affect mainly the anodic dissolution of mild steel [24].

3.1.1. Film Thickening on the Surface of Mild Steel Electrode in Absence and in Presence of Different Amino Acids. A theory for film thickening on the surface of metal and alloys based on open-circuit potential measurements has been developed by Abd El-Kader and Shams El-Din [38]. The essence of the theory is based on the idea that the potential is determined by simultaneous anodic (film formation) and cathodic (oxygen reduction) couples, in which the anodic reaction is the rate limiting step.

The way by which the potential (E) of the mild steel approaches the stationary values is of interests, E varies linearly with the logarithm of time of immersion until steady state potential ($E_{s.s.}$) is attained

Straight lines were obtained satisfying the relation

$$E = \text{constant} + 2.303 \left(\frac{\bar{\delta}}{\beta} \right) \log t, \quad (1)$$

where t is the time from the moment of immersion in solution, $\bar{\delta}$ is the rate of film thickening per decade of time, and β is given by

$$\beta = \left(\frac{nF}{RT} \right) \alpha \delta' t, \quad (2)$$

where (α) is a transference coefficient similar to that encountered by normal electrochemical reactions, ($0 < \alpha < 1$), δ' is the width of the activation energy barrier surrounded by the ion during film formation, R is the gas constant, and T is the absolute temperature. Assuming (α) to have the value of 0.5 and δ' to the value of 1.0 nm, the constant (n) in (2) is set equal to 3.0 and β acquires the value of 58.6 nm/V [38].

Results given in Figures 2 and 3 illustrate the potential/log time curves for mild steel in 0.1 M Na_2HPO_4 in the absence and in the presence of 0.1 M NaCl and 0.02 M of for Serine and Methionine, respectively, and Table 3 illustrates the values of $\bar{\delta}$. It is observed that the rate of oxide thickening of

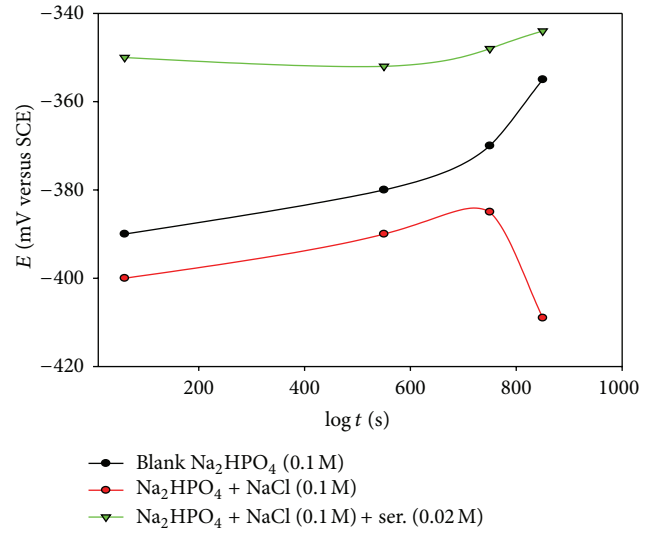


FIGURE 2: Potential-log time curves of mild steel in 0.1 M Na_2HPO_4 in absence and presence of 0.1 M NaCl and 0.02 M of serine.

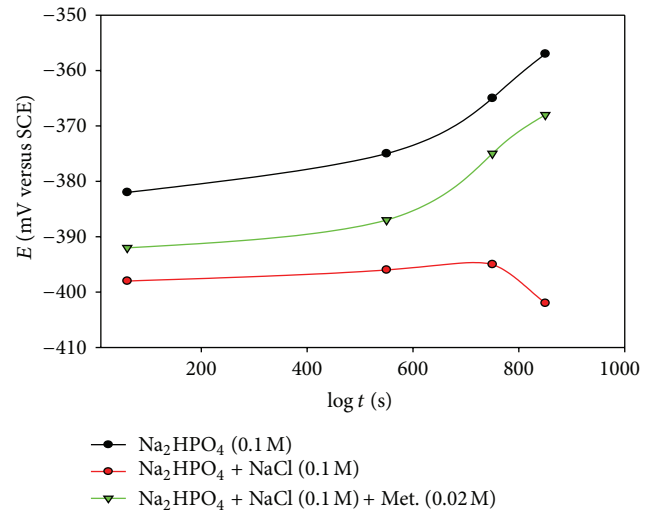


FIGURE 3: Potential-log time curves of mild steel in 0.1 M Na_2HPO_4 in absence and presence of 0.1 M NaCl and 0.02 M of Methionine. Blank Na_2HPO_4 (0.1 M).

TABLE 3: Rate of oxide thickening (nm/log t) on mild steel in 0.1 M Na_2HPO_4 solution (blank) in presence of 0.1 M NaCl and 0.02 M of different amino acids.

Test solutions	Slope	$\bar{\delta}$ (nm/log t)
0.1 M Na_2HPO_4	57.00	1.4540
0.1 M Na_2HPO_4 + 0.1 M NaCl (Blank)	64.00	1.6327
(Blank) + Ser.	58.01	1.4796
(Blank) + Met.	101.99	2.6020

mild steel immersed in aerated phosphate solution in the absence of amino acids is 1.454 (nm/log t). This indicates that the thickness of the formed oxide films increases with

TABLE 4: Electrochemical parameters of mild steel in 0.1 M Na_2HPO_4 + 0.1 M NaCl solution (blank) in presence of 0.02 M of different amino acids.

Media	R_p (Ohms)	E_{corr} (mV)	I_{corr} ($\mu\text{A}/\text{cm}^2$)	C.R. (mpy)	I.E.%
0.1 M Na_2HPO_4	4124	-350.8	2.086	1.90	—
0.1 M Na_2HPO_4 + 0.1 M NaCl (Blank)	1631	-356.5	15.030	13.90	—
Blank + Ser.	3407	-303.7	5.994	5.48	60.60
Blank + Met.	2580	-303.4	3.490	3.23	76.76

the field strength in the passive region. The presence of the amino acids the rate of oxide thickening are (1.4796 and 2.6020 ($\text{nm}/\log t$)) for Serine and Methionine, respectively. This indicates that thick oxide films are formed increasing with strength in the passive region, and also these may consist of a thin nonstructural barrier layer next to the mild steel and a thicker crystalline layer next to the barrier layer [38], showing the highest value of δ indicating a strong adsorption on the active sites of the electrode surface and the formation of thicker adsorption layer.

3.2. Potentiodynamic Polarization Measurements. Linear polarization resistance and Tafel plots are utilized to provide information about the corrosion rate of mild steel and inhibitor efficiencies in 0.1 M (Na_2HPO_4 + NaCl) solutions containing 0.02 M of Serine and Methionine, and the corrosion parameters which obtained from the linear polarization resistance and Tafel plot are given in Table 4.

The inhibition efficiencies calculated using (3) [31, 39] are listed in Table 4:

$$\text{I.E.\%} = \frac{(\text{C.R.})_{\text{uninh.}} - (\text{C.R.})_{\text{inh.}}}{(\text{C.R.})_{\text{uninh.}}} \times 100, \quad (3)$$

where $(\text{C.R.})_{\text{inh.}}$ and $(\text{C.R.})_{\text{uninh.}}$ are the corrosion rates with and without inhibitor, respectively.

The presence of the amino acid inhibitor decreases the corrosion rate and currents as shown in Table 4 this is due to the adsorption of amino acids on the metal surface. Accordingly, the inhibitors are found to retard the dissolution of the metals and act as mixed-type inhibitors. The presence of R-S-R in molecular structure of Methionine provokes an increase of the inhibition efficiency, which can be attributed to the fact that the sulfur containing amino acids can be adsorbed as bidentate ligands in which surface coordination is taking place through both the amino group (or carboxylic group) and the -S- moiety.

3.3. Pitting Corrosion Current on Mild Steel. In 1975, A. Shams El Din and his co-workers have devised a sample model cell for measuring the pitting corrosion currents of zinc [36]. In the present part the this cell has been to measure the pitting corrosion currents of a mild steel electrode in (0.1 M Na_2HPO_4 + 0.1 M NaCl) blank solution in presence of different concentrations (2.0×10^{-2} M to 0.125×10^{-2}) of the studied amino acids as shown in Figures 4 and 5.

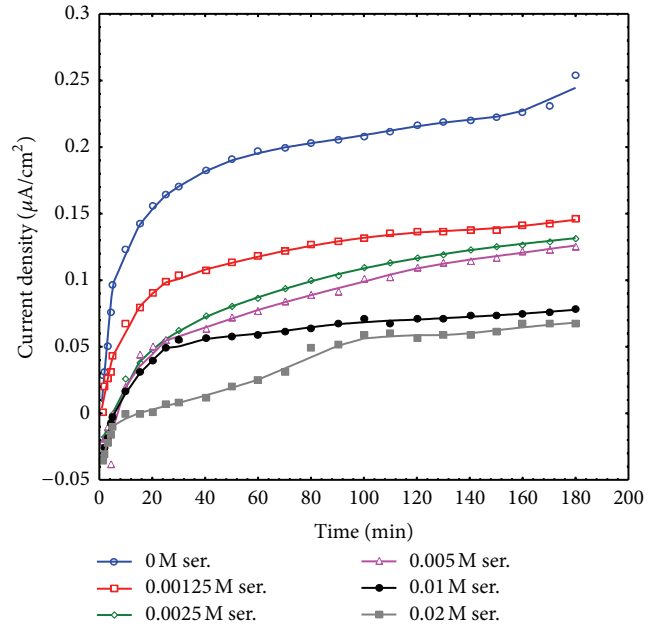


FIGURE 4: Pitting corrosion current density versus time for mild steel in 0.1 M Na_2HPO_4 + 0.1 M NaCl solution containing different concentrations of Serine at 25°C.

The inhibition activity, calculated using following equations

$$\text{I.E.\%} = \frac{I_p^o (\mu\text{A}/\text{cm}^2) - I_p (\mu\text{A}/\text{cm}^2)}{I_p^o (\mu\text{A}/\text{cm}^2)} \times 100, \quad (4)$$

where I_p^o is the pitting current density of the blank solution and I_p is the pitting current density of the inhibited solution containing Serine and Methionine, and I.E.% is the percentage of inhibition efficiency; these data are included in Table 5. These results reveal that the presence or increasing concentration of the inhibitor causes decrease in pitting current density, and the value of pitting current density (I_p) increases with increases of the time.

Adsorption Isotherm. Action of inhibitors on metal surface is often expressed by an adsorption isotherm; many adsorption isotherms were proposed to calculate the thermodynamic parameters pertaining to inhibitor adsorption. The models considered were [24, 40]

$$\text{Langmuir isotherm } \frac{\theta}{1 - \theta} = k_{\text{ads}} \cdot C, \quad (5)$$

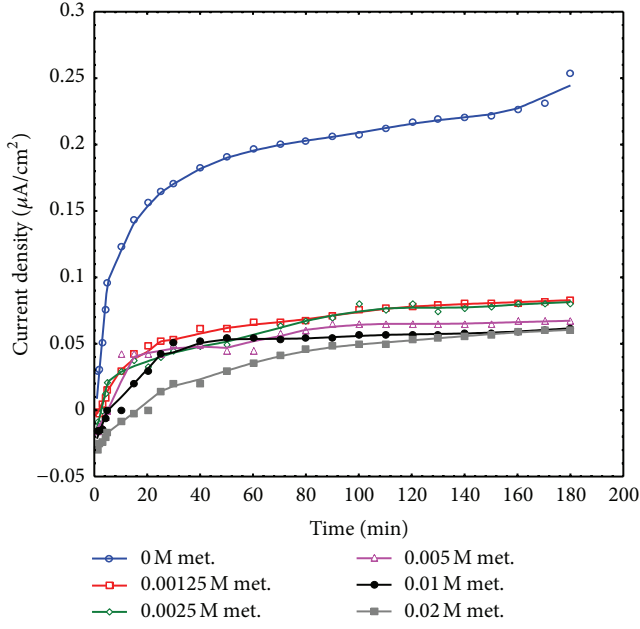


FIGURE 5: Pitting corrosion current density versus time for mild steel in 0.1 M Na_2HPO_4 + 0.1 M NaCl solution containing different concentrations of Methionine at 25°C.

TABLE 5: Concentrations effect of serine and methionine on the pitting current ($\mu\text{A}/\text{cm}^2$) and their corrosion inhibition efficiency (I.E.%) on mild steel in 0.1 M Na_2HPO_4 + 0.1 M NaCl solution.

Conc. of amino acids	Properties	Blank	Serine	Methionine
0.02 M	I_p ($\mu\text{A}/\text{cm}^2$)	0.228	0.063	0.057
	I.E.%	—	72.20	74.70
0.01 M	I_p ($\mu\text{A}/\text{cm}^2$)	0.228	0.074	0.059
	I.E.%	—	67.39	74.12
0.005 M	I_p ($\mu\text{A}/\text{cm}^2$)	0.228	0.119	0.066
	I.E.%	—	47.58	71.05
0.0025 M	I_p ($\mu\text{A}/\text{cm}^2$)	0.228	0.126	0.078
	I.E.%	—	44.73	65.49
0.00125 M	I_p ($\mu\text{A}/\text{cm}^2$)	0.228	0.140	0.081
	I.E.%	—	38.52	64.47

where θ is the surface coverage, k_{ads} is the equilibrium constant of the adsorption process, and C the inhibitor concentration.

$$\frac{C}{\theta} = \frac{1}{K} + C. \quad (6)$$

The degree of surface coverage (θ) for different concentrations of these compounds can be evaluated from pitting current measurement:

$$\theta = \frac{(I_p^0) - (I_p)}{(I_p^0)} \quad \text{or} \quad \theta = \frac{\text{I.E.}(\%)}{100}. \quad (7)$$

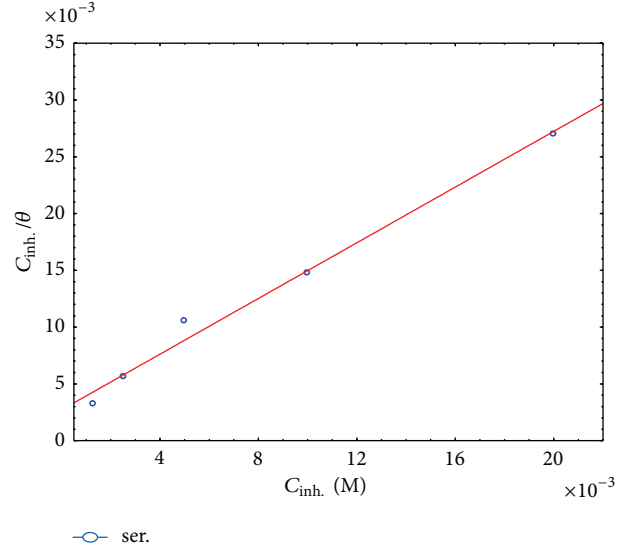


FIGURE 6: Adsorption isotherm plot for $(C_{\text{inh.}}/\theta)$ versus C for mild steel in 0.1 M Na_2HPO_4 + 0.1 M NaCl solution containing different concentrations of serine at 25°C.

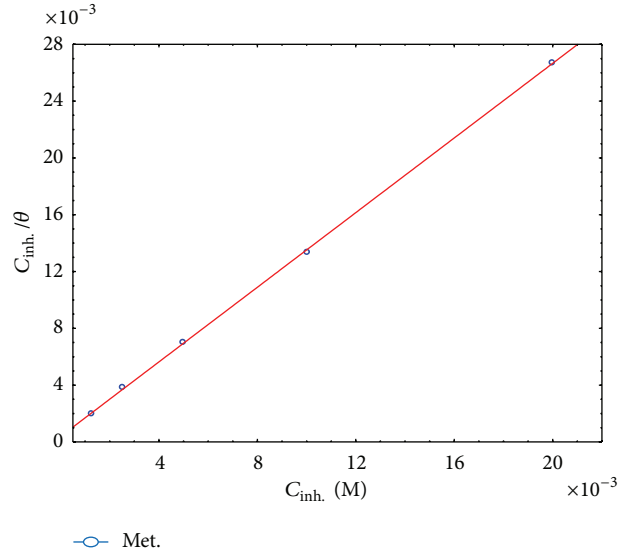


FIGURE 7: Adsorption isotherm plot for $(C_{\text{inh.}}/\theta)$ versus C for mild steel in 0.1 M Na_2HPO_4 + 0.1 M NaCl solution containing different concentrations of methionine at 25°C.

The obtained values of K lead to the adsorption free energy (ΔG_{ads}^0) obtained according to the equation [14, 18].

$$K = \frac{1}{55.5} \exp \frac{-\Delta G^0}{RT}. \quad (8)$$

The value of 55.5 is the concentration of water in the solution in moles. The results of the adsorption of Serine and Methionine on mild steel electrode in 0.1 M Na_2HPO_4 + 0.1 M NaCl solution are presented in Figures 6 and 7; plotting $C_{\text{inh.}}/\theta$ against $C_{\text{inh.}}$ of amino acids gives straight lines with unit slope value. Figures 6 and 7 indicate that adsorption of amino acids

on steel surface follows Langmuir's adsorption isotherm, free energy of adsorption for Methionine and Serine solutions is -24.61 , -29.34 kJ/mol, respectively. The values of ΔG_{ads}^0 for Serine and Methionine are <40 kJ/mol indicating that amino acids are physically adsorbed on the metal surface. The negative value of ΔG_{ads}^0 indicated the spontaneous adsorption of inhibitor on the surface of mild steel [1, 4, 16, 31, 41, 42].

4. Conclusion

- (i) Serine and Methionine revealed good inhibitory effects against mild steel pitting corrosion in aqueous solution.
- (ii) The open-circuit potential indicates that amino acids are adsorbed on anodic sites. This together with the decrease in the pitting current may indicate that these inhibitors can block the pits which are the anodic site, and inhibition increases with increasing the inhibitor concentration.
- (iii) The adsorption of Serine and Methionine on mild steel follows the Langmuir adsorption isotherm and the free energy of adsorption (-24.61 , -29.34) kJ/mol for Serine and Methionine, respectively, indicating the physical adsorption of the amino acid molecules on the metal surface.

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