Sodium Aliginate as Corrosion Inhibitor for Carbon Steel in 0.5 M HCl Solutions

Ameena Mohsen Al-Bonayan

Abstract—The corrosion inhibition of carbon-steel in 0.5 M HCl by sodium aliginate has been investigated using weight loss method, potentiodynamic polarization, electrochemical impedance spectroscopy (EIS) and electrochemical frequency modulation (EFM) techniques. Results obtained showed that sodium aliginate are good inhibitor and its inhibition efficiency increases with increasing sodium aliginate concentration and decrease with increase in temperature. Moreover, polarization studies clearly revealed that the presence of sodium aliginate changes the mechanism of hydrogen evolution and the metal dissolution i.e. they act as mixed type inhibitors. EIS study showed that charge transfer resistance increases with the increase of sodium aliginate concentration but the capacitance of double layer decreases. The adsorption of sodium aliginate obeys Langmuir’s adsorption isotherm. The activation parameters were determined and discussed. The mechanism of adsorption had been explained on the basis of chemical structure of sodium aliginate. It was found that there is a good agreement between the different tested techniques.

Key words — Carbon steel, HCl, Sodium aliginate, EIS, EFM.

1 INTRODUCTION

Carbon steel is an alloy of iron it is widely used in petrochemical, chemical and metallurgical industries. It is also used as a construction material owing to its excellent mechanical properties and cost effectiveness. However, it is easily undergoes corrosion in various environmental conditions. Acid solutions are widely used in industry, the most important fields of application being acid pickling, industrial acid cleaning, acid descaling and oil well acidising, because of the general aggressiveness of acid solutions, the practice of inhibition is commonly used to reduce the corrosive attack on metallic materials. Inhibitors are generally used for this purpose to control the metal dissolution. Most of the well-known inhibitors are organic compounds containing nitrogen, sulphur and/or oxygen atoms. It has been observed that the most of the organic inhibitors act by adsorption on the metal surfaces [1]. The adsorption of corrosion inhibitor depends mainly on physicochemical properties of the molecules such as functional group, steric factor, molecular size, molecular weight, molecular structure, aromaticity, electron density at the donor atoms and n-orbital character of donating electrons [2-6], and also, on the electronic structure of molecules [7-10].

The aim of the present work is to study the inhibitive action of some new sodium aliginate towards the corrosion of C-steel in 0.5 M hydrochloric acid solution using chemical and electrochemical techniques. Moreover, the effect of temperature on the dissolution of carbon steel, as well as, on the inhibition efficiency of the studied compound was also investigated and discussed.

2 EXPERIMENTAL

2.1 Materials and reagents

Carbon steel strips (BDH grade) containing (weight %): 0.2 C, 0.024 P, 0.003 Si, 0.35 Mn, and rest Fe were used in this investigation. All chemicals used were of AR grade. Specimens of C-steel strips were abraded successively by emery papers of different grades, i.e. 320, 400, 600, 800 and 1000 finally polished with a 4/0 emery paper to obtain mirror like finish, then degreased ultrasonically with ethyl alcohol and rinsed with bidistilled water several times and dried between two fitter papers. AR grade hydrochloric acid (37 %) was used for preparing the corrosive solutions.

2.2 Weight loss measurements

For weight loss measurements, rectangular C-steel specimens of size 20 x 20 x 2 mm were immersed in 100 ml inhibited and uninhibited solutions and allow to stand for several intervals at 30°C in water thermostat. Therefore, the weight losses given by:

\[ \Delta m = (m1-m2) \]

where m1 and m2 are the weights of metal before and after exposure to the corrosive solution, respectively. The percentage inhibition efficiency (\% IE) and the degree of surface coverage (θ) of the investigated compounds were calculated using equation (2):

\[ \% IE = \theta \times 100 = [1 - (\Delta \text{minh}/\Delta \text{mfree})] \times 100 \]

where \( \Delta \text{mfree} \) and \( \Delta \text{minh} \) are the weight losses per unit area in the absence and presence of additives, respectively.

2.3 Electrochemical measurements

A three electrode electrochemical cell was used. The working electrode was C-steel of surface area of 1 cm². Before each experiment, the electrode was abraded using emery papers as before. After this, the electrode was cleaned ultrasonically...
with ethyl alcohol and washed by bidistilled water. All potentials were given with reference to the saturated calomel electrode (SCE). The counter electrode was a platinum plate of surface area of 1 cm². The working electrode was immersed in the test solution during 30 min until a steady state open circuit potential (Eocp) was obtained. The polarization curves were recorded by polarization from -0.6 V to 0.2 V under potentiodynamic conditions corresponding to 1 mV/s (sweep rate) and under air atmosphere. All measurements were carried out with C-steel electrode in 1 M HCl in the absence and presence of different concentrations (10, 20, 30, 40, 50, 100, 200 and 300 ppm) of the investigated inhibitor. All experiments were carried out at 30 ± 1 °C. The inhibition efficiency and surface coverage (%) were calculated from equation (3):

\[
\% \text{ IE} = \theta \times 100 = \left[ 1 - \left( \frac{i_{corr(\text{inh.})}}{i_{corr(\text{free})}} \right) \right] \times 100
\]

Where \( i_{corr(\text{free})} \) and \( i_{corr(\text{inh.})} \) are the corrosion current densities in the absence and presence of inhibitor, respectively.

Electrochemical impedance spectroscopy measurements were performed using the same cell that used in polarization experiments. The EIS carried out over a frequency range of 1 Hz to 100 kHz, with a signal amplitude perturbation of 10 mV.

The inhibition efficiency (% IE) and surface coverage (θ) of the investigated compounds obtained was calculated from equation (4):

\[
\% \text{ IE} = \theta \times 100 = \left( \frac{R_{ct} - R^\circ_{ct}}{R_{ct}} \right) \times 100
\]

Where \( R^\circ_{ct} \) and \( R_{ct} \) are the charge transfer resistance values in the absence and presence of the inhibitors, respectively.

Electrochemical frequency modulation is a non-destructive technique as electrochemical impedance spectroscopy that can directly and rapidly give values of the corrosion current without a prior knowledge of Tafel constants. The great of the EFM is the causality factors, which serves as an internal check on the validity of the EFM measurement. With the causality factors the experimental EFM data can be verified [12-13]. Identical cell assembly was used as in impedance studies. All electrochemical measurements were carried out using Potentiostat/Galvanostat / Zra analyzer (Gamry PCI300/4). A personal computer with DC105 software for potentiodynamic, EIS300 software for EIS and EFM140 software for EMF and Echem Analyst 5.21 was used for data fitting.

3 RESULTS AND DISCUSSION

3.1 WEIGHT – LOSS MEASUREMENTS

Figure 1 shows the effect of concentration of sodium alginic acid on the weight loss vs. time of C-steel at 30°C. It is obvious that the weight loss of C-steel in presence of sodium alginate varies linearly with time, and is much lower than that obtained in blank solution. The linearity obtained indicated the absence of insoluble surface film during corrosion and that the inhibitors were first adsorbed onto the metal surface and, therefore, impede the corrosion process [14]. The calculated values of the percentage inhibition efficiency (% IE) at different concentrations of investigated compound in 0.5 M HCl at different temperatures (25-40°C) are given in Table (1). From this Table, the inhibition efficiencies increase by increasing inhibitor concentra-

![Figure 1](http://www.ijser.org)
Huggins. By far the best fit was obtained with the Langmuir isotherm is given as [15]:
\[ \theta / (1 - \theta) = K_{ads} C \]  
(5)
Where C is the inhibitor concentration and K_{ads} is the equilibrium constant of adsorption process and is related to the standard free energy of adsorption \( \Delta G^\circ_{ads} \) by Eq. (6):
\[ K_{ads} = 1 / 55.5 \exp \left( - \Delta G^\circ_{ads} / RT \right) \]  
(6)
The value of 55.5 is the concentration of water in solution expressed in mole per liter, R is the universal gas constant and T is the absolute temperature.
A plot of \( \theta / (1 - \theta) \) against C, for all concentrations of inhibitors (Figure 2) a straight line relationship was obtained in all cases with correlation coefficients (R^2) near to unity.
The deviation of the slope from unity as observed from this study could be interpreted that there are interactions between adsorbed species on the metal surface as well as changes in adsorption heat with increasing surface coverage [16, 17], factors that were ignored in the derivation of Langmuir isotherm.
The negative \( \Delta G^\circ_{ads} \) value is consistent with the spontaneity of the adsorption process and the stability of the adsorbed layer on the C-steel surface [18]. It is generally accepted that the values of \( \Delta G^\circ_{ads} \) up to -20 kJ mol\(^{-1}\) the types of adsorption were regarded as physisorption, the inhibition acts due to the electrostatic interaction between the charged molecules and the charged metal, while the values around -40 kJ mol\(^{-1}\) or smaller, were seen as chemisorptions, which is due to the charge sharing or a transfer from the inhibitor molecules to the metal surface to form covalent bond [19-20]. The \( \Delta G^\circ_{ads} \) value obtained in this study was 4.19 kJ mol\(^{-1}\) and K_{ads} value was very small (0.09 M\(^{-1}\)). These suggested that the adsorption mechanism of investigated inhibitors on C-steel in 1 M HCl solution was typical of physisorption.

As noticed previously, the adsorption process was well elucidating by using a thermodynamic model, in addition a kinetic-thermodynamic model was another tool to explain the mechanism of corrosion inhibition for an inhibitor. The apparent effective activation energies (E^*a) for the corrosion reaction of C-steel in HCl in the absence and presence of different concentrations of sodium alginolate were calculated from Arrhenius-type equation [21]:
\[ \text{Corrosion rate} = A \exp \left( -E^a \right) \]  
(7)
Where A is the Arrhenius pre-exponential factor. A plot of log (corrosion rate) vs. 1/T gave straight line as shown in (Figure 3). The entropy of activation (\( \Delta S^* \)) and the enthalpy of activation (\( \Delta H^* \)) for the intermediate complex in the transition state for the corrosion of C-steel in HCl in the absence and presence of different concentrations of sodium alginolate were obtained by applying the transition-state equation [22-24]
\[ \text{Corrosion rate} = RT / Nh \exp \left( \Delta S^*/R \right) \exp \left( - \Delta H^* / RT \right) \]  
(8)
Where h is the Planck’s constant and N is the Avogadro’s number
A plot of log k (corrosion rate) / T vs. 1 / T should give a straight lines (Figure 4), with a slope of (-\( \Delta H^* / 2.303R \)), and an intercept of [(log (RT / Nh ) + (\( \Delta S^*/2.303R \))] [25-26], from which the values of \( \Delta H^* \) and \( \Delta S^* \) were calculated, respectively. (Table 2) exhibited values of apparent activation energy, apparent enthalpies \( \Delta H^* \) and entropies \( \Delta S^* \) for C-steel dissolution in 0.5 M HCl solution in the absence and presence of sodium alginolate. The presence of inhibitors decreased the activation energies of C-steel indicating strong adsorption of the inhibitor molecules on the metal surface and the presence of these additives induces the adsorption of theses additives on the surface of C-steel. Values of the entropy of activation \( \Delta S^* \) in the absence and in presence of sodium alginolate are negative. This implies that the activated complex in the rate determining step represents an association rather than a dissociation step [27]. This means that the activated molecules were in higher order state than that at the initial stage [28-29].

Figure (2): Langmuir adsorption isotherm of sodium algininate on C-steel surface in 0.5 M HCl at 30°C

3.3 **Kinetic-thermodynamic corrosion parameters**
Table (2): Effect of concentration of sodium alginic on the activation parameters of C-steel dissolution in 0.5 M HCl duration of the experiment: 120 min. immersion.

<table>
<thead>
<tr>
<th>Concentration, ppm</th>
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<td>$E_a^*$, kJ mol$^{-1}$</td>
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<td>81.55</td>
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<td>300</td>
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</table>

Figure (3): Arrhenius plots for C-steel corrosion in 0.5 M HCl in the absence and presence of different concentrations of sodium alginic.

Figure (4): Plots of (log corrosion rate / T) vs 1/T for corrosion of C-steel in 0.5 M HCl in the absence and presence of different concentrations of sodium alginic.

3.4 Polarization curves

Figure 5 illustrates the polarization curves of carbon steel in 0.5 M HCl solution without and with various concentrations of sodium alginic at 30°C. The presence of sodium alginic shifts both anodic and cathodic branches to the lower values of corrosion current densities and thus causes a remarkable decrease in the corrosion rate. The parameters derived from the polarization curves in (Figure 5) are given in Table 3. In 0.5 M HCl solution, the presence of the sodium alginic cause a remarkable decrease in the corrosion rate i.e., shifts both anodic and cathodic curves to lower current densities. In other words, both cathodic and anodic reactions of carbon steel electrode are retarded by the sodium alginic in HCl solution. The Tafel slopes of $\beta_a$ and $\beta_c$ at 30°C do not change remarkably upon addition of the sodium alginic, which indicates that the presence of sodium alginic does not change the mechanism of hydrogen evolution and the metal dissolution process. Generally, an inhibitor can be classified as cathodic or anodic type if the shift of corrosion potential in the presence of the inhibitor is more than 85 mV with respect to that in the absence of the inhibitor [30, 31]. In the presence of investigated compound, $E_{corr}$ shifts to more negative but this shift is very small (about 12 mV), which indicates that sodium alginic can be arranged as a mixed-type inhibitor, with predominant cathodic effectiveness. The % inhibition increased with increasing the concentration of the compounds, the inhibition efficiency of sodium alginic measured by polarization method is in accordance with that obtained from weight-loss measurements.
Concentrations of sodium alginate at 30°C

Table (3): Data from Potentiodynamic polarization of carbon steel in 0.5 M HCl containing various concentrations of sodium alginate at 30°C

<table>
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<tr>
<th>Conc. ppm</th>
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<th>Icorr, µA cm⁻²</th>
<th>-Ecorr, mV vs SCE</th>
<th>β+, V dec⁻¹</th>
<th>β-, V dec⁻¹</th>
<th>% IE</th>
<th>θ</th>
<th>Corrosion rate, mm y⁻¹</th>
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<td>10</td>
<td>511.6</td>
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<td>229.0</td>
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<td>106.9</td>
<td>171.5</td>
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<td>487.0</td>
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<td>109.0</td>
<td>179.9</td>
<td>61.29</td>
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<td>88.6</td>
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<td>300</td>
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<td>486.0</td>
<td>95.6</td>
<td>141.9</td>
<td>76.54</td>
<td>0.7654</td>
<td>45.58</td>
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</tr>
</tbody>
</table>

Figure (5): Potentiodynamic polarization curves for the corrosion of C-steel in 0.5 M HCl in the absence and presence of different concentrations of sodium alginate at 30°C

3.5 ELECTROCHEMICAL IMPEDANCE SPECTROSCOPY

Figures 6 and 7 show the Nyquist and Bode diagrams of carbon steel in 0.5 M HCl solutions containing different concentrations of sodium alginate at 30°C. All the impedance spectra exhibit one single depressed semicircle. The diameter of semicircle increases with the increase of sodium alginate concentration. The impedance spectra exhibit one single capacitive loop, which indicates that the corrosion of steel is mainly controlled by a charge transfer process [32] and the presence of the sodium alginate does not change the mechanism of carbon steel dissolution [33]. In addition, these Nyquist diagrams are not perfect semicircles in 0.5 M HCl that can be attributed to the frequency dispersion effect as a result of the roughness and inhomogeneous of electrode surface [34]. Furthermore, the diameter of the capacitive loop in the presence of inhibitor is larger than that in the absence of inhibitor (blank solution), and increased with the inhibitor concentration. This indicates that the impedance of inhibited substrate increased with the inhibitor concentration. [33,34]. This behavior is usually attributed to the inhomogeneity of the metal surface arising from surface roughness or interfacial phenomena [35], which is typical for solid metal electrodes [36]. Generally, when a non-ideal frequency response is presented, it is commonly accepted to employ the distributed circuit elements in the equivalent circuits. What is most widely used is the constant phase element (CPE), which has a non-integer power dependence on the frequency [37]. Thus, the equivalent circuit depicted in Figure 8 is employed to analyze the impedance spectra, where Rs represents the solution resistance, Rct denotes the charge-transfer resistance, and a CPE instead of a pure capacitor represents the interfacial capacitance. The impedance of a CPE is described by the equation 9:

\[
Z_{CPE} = Y_0^{-1} \left( j\omega \right)^{-n}
\]

Where Y0 is the magnitude of the CPE, j is an imaginary number, ω is the angular frequency at which the imaginary component of the impedance reaches its maximum values, and n is the deviation parameter of the CPE: -1 ≤ n ≤ 1. The values of the interfacial capacitance Cdl can be calculated from CPE parameter values Y0 and n using equation 10 [38]:

\[
C_{dl} = Y_0 (\omega_{max})^n
\]

Where the values of the parameters such as Rs, Rct, through EIS fitting as well as the derived parameters Cdl and η% are listed in Table 4. The order of inhibition efficiency obtained from EIS measurements is in accordance with that obtained from weight-loss and polarization measurements.

Table (4): Electrochemical kinetic parameters obtained from EIS technique for the corrosion of C-steel in 0.5 M HCl at different concentrations of sodium alginate at 30°C

<table>
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<th>Conc. ppm</th>
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<th>Rct, Ohm cm⁻²</th>
<th>Rs, Ohm cm²</th>
<th>CPE x10⁻⁶</th>
<th>n</th>
<th>fₘ₉₉ Hz</th>
<th>Clf, µF cm⁻²</th>
<th>θ</th>
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<td>10</td>
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<td>137.9</td>
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<td>80.804</td>
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small AC signal. Intermodulation spectra obtained from EFM measurements of carbon steel in 0.5 M HCl solution, in the absence and presence of different concentrations of sodium alginate at 30°C are presented in Figure 9. Each spectrum is a current response as a function of frequency. The calculated corrosion kinetic parameters at different concentrations of the sodium alginate in 0.5 M HCl at 30°C (Icorr, βa, βc, CF-2, CF-3 and %IE) are given in Table 5. From this Table, the corrosion current densities decreased by increasing the concentration of sodium alginate, the inhibition efficiencies increase by increasing sodium alginate concentrations. The causality factors CF-2 and CF-3 in Table 5 are very close to theoretical values that according to EFM theory [39] should guarantee the validity of Tafel slopes and corrosion current densities. Values of causality factors (Table 5) indicate that the measured data are of good quality. The standard values for CF-2 and CF-3 are 2 and 3, respectively. The causality factor is calculated from the frequency spectrum of the current response. If the causality factors are approximately equal to the predicted values of 2 and 3, there is a causal relationship between the perturbation signal and the response signal. Then the data are assumed to be reliable [12]. When CF-2 and CF-3 are in the range 0-2 and 0-3, respectively, then the EFM data is valid. The deviation of causality factors from their ideal values might due to that the perturbation amplitude was too small or that the resolution of the frequency spectrum is not high enough also another possible explanation that the inhibitor is not performing very well [13].

Table (5): Electrochemical kinetic parameters obtained from EFM technique for the corrosion of C-steel in 0.5 M HCl at different concentrations of sodium alginate at 30°C

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<tr>
<th>Conc. ppm</th>
<th>Icorr. µA cm⁻²</th>
<th>βa, V dec⁻¹</th>
<th>βc, V dec⁻¹</th>
<th>% IE</th>
<th>CF-2</th>
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3.6 ELECTROCHEMICAL FREQUENCY MODULATION TECHNIQUE (EFM)
The EFM is a nondestructive corrosion measurement technique that can directly give values of the corrosion current without prior knowledge of Tafel constants. Like EIS, it is a
The free energy of adsorption value is not very greater than -40 kJ mol⁻¹ should indicate contribution of physical adsorption. If adsorption of organic molecules on the solid surfaces cannot be considered only as purely physical or as purely chemical adsorption phenomenon. In addition to the chemical adsorption, inhibitor molecules can also be adsorbed on the steel surface via electrostatic interaction between the charged metal surface and charged inhibitor molecule if it is possible. The adsorption of organic molecules on the solid surfaces may lie (adsorbed) flat on metal surface so, it covers larger area.

5 CONCLUSION

Sodium alginate is good inhibitor and act as mixed type but mainly act as cathodic inhibitor for carbon steel corrosion in 0.5 M HCl solution. The results obtained from all electrochemical measurements showed that the inhibiting action increases with the inhibitor concentration and decreases with the increasing in temperature. Double layer capacitances decrease with respect to blank solution when sodium alginate is added. This fact confirms the adsorption of the investigated compound molecules on the carbon steel surface. The adsorption of sodium alginate on the carbon steel surface at different temperature was found to obey the Langmuir adsorption isotherm and this adsorption is physisorption. The values of inhibition efficiencies obtained from the different independent quantitative techniques used show the validity of the results.

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