Poly Acryl Amide as Acid Corrosion Inhibitor of C-steel in 15% HCl

Dr. Ghassan Burhan Yaqoob
North Oil Company
E-mail: ghassan7jul@yahoo.com

Abstract:

The effect of poly acryl amide in the 15% HCl solution of carbon steel (class L-80) was studied using weight loss method, the efficiency of inhibitor increased by increasing inhibitor concentration and decreasing temperatures. Inhibition attributed to insoluble complex adsorbed on the coupon (L-80) carbon steel surface. The iron complex was identified by FTIR. And the mechanism of inhibition was explained as Langmuir adsorption isotherm. The thermodynamic parameters of adsorption operation was calculated and explained.

Introduction:

Corrosion phenomenon occurs in the nature chemically or electrochemically decays the metallic properties of alloys making them ineffective; Corrosion of alloys is big problem in industries that need much researches and investigations, oil well acidising is widely used in oil companies for cleaning and de scaling to reduce the corrosive attack on pipe lines and well pipes inhibitors are generally used to control the metal degradation. Most of organic inhibitors contain nitrogen, sulphur and/or oxygen atoms for that the most of the organic inhibitors work by adsorption on the metal surface [1].

Physical and chemical properties of corrosion inhibitor (molecular size, and weight, steric effect, electron density at the donor atoms, aromaticity, orbital character of donating electrons [2-6], the electronic structure of molecules [7-11] is an important factors in inhibitors adsorption. Most of the corrosion inhibitors are expensive and an acceptable environmentally because of its hazardous and environmentally safe corrosion inhibitors it is very desirable [12-17].

The goal of the present work is to study the inhibitive activity of Poly acryl amide to prevent the corrosion of C-steel in 15% HCl solution by weight loss, and the effect of
temperature on carbon steel (L-80) was studied in exist of poly acryl amide and some thermodynamic parameters were founds.

**Method:**

Used (60) gr of carbon steel (L-80) has ingredients (wt%) 0.019 Cr, 1.41Mn, 0.353 Si, 0.263 C, 0.159 Ni, 0.03 P, 0.001 S and the remained Fe. with dimension of (2.5 x 4.5 x 0.7) cm. C-steel coupons surface was polished with smoothing paper, degreased with acetone, and rinsed with distilled water. the cleaned C-steel coupons were weighed before and after immersion in (50) ml of (%15) HCl solution at the times (3, 4, 5, 6) hr. The weight loss of coupon in experiments was taken in grams. The temperature (40, 60, 80, and 90)°C was adjusted using air thermostat. poly acryl amide (C₃H₅NO)n used as inhibitor was prepared by Fluka company and has the following structure:

\[-(CH₂-CH)ₙ-\]

\[\begin{array}{c}
\text{C} \\
\text{C}=\text{O} \\
\text{NH}_2
\end{array}\]

**Results and Discussion**

Figures (1, 2, 3, 4) illustrates clearly that corrosion and weight loss of carbon steel (L-80) became very low in the exists of poly acryl amide by adsorbing and making a thin film on the surface of the coupon objected the corrosion process [18], the linearity follows Langmuir equation. Table (1) has inhibition efficiency (%E) and surface coverage (Θ) surface covered by the inhibitor molecules as in equation (1, 2).

\[\%E = (1 - \frac{W_{inh.}}{W_{free}}) \times 100 \quad \text{(1)}\]

\[\Theta = (1 - \frac{W_{inh.}}{W_{free}}) \quad \text{(2)}\]

Where \(W_{free}\) and \(W_{inh.}\) Are the weight losses of C-steel in the absence and exists of inhibitors the weight loss decreases as the concentration of poly acryl amide was increased.
Effect of Temperature:

Corrosion rates increases with increasing temperature in this study corrosion rate measured by the following equation [19]:

\[ R_{corr} = \Delta W/St \] ........................ (3)

Where \( \Delta w \) is the loss of weight \( S \) is the surface area (cm\(^2\)) and \( t \) is the displayed time (hr).

The data listed in Table (1) illustrate that the highest inhibition efficiency is happened at 40°C by using poly acryl amide as inhibitor in (%15) HCl. The activation energy \( E_a^* \) of the corrosion operation calculated using Arrhenius equation [20].

\[ \log R_{corr} = \log A - \frac{E_a^*}{2.303RT} \] ........................ (4)

Where \( R_{corr} \) is the rate of corrosion from weight loss, \( A \) is Arrhenius constant, \( R \) is the gas constant (8.31J.K\(^{-1}\).mol\(^{-1}\)) and \( T \) is absolute temperature. Figures (5, 6, 7, 8) shows Arrhenius curves between \( \log R_{corr} \) and \( 1/T \) in the exist or absence of different concentrations of poly acryl amide and from the slope of the straight lines activation energy \( E_a^* \) can be calculated from the slope of curve and in the increasing of activation energy at exists of inhibitor means decrease in the adsorption operation on coupon surface, the data listed in Tables (2, 3, 4, 5) illustrates by increasing in temperature the corrosion rate increase and surface area of coupon that is displayed to acid became greater [21]. in our study \( E_a^* \) increases with increasing the poly acryl amide concentration

Effect of adsorption:

Poly acryl amide adsorbed on coupon and covered (\( \Theta \)) part of its surface in different concentration, temperature and time and the data are listed in Table (1) and it was found the value of (\( \Theta \)) increase with increasing the concentration of poly acryl amide and decrease with rise in temperature from 40 to 90°C.

Depending on Langmuir adsorption isotherm [22], we will get equation (5)

\[ \frac{\Theta}{1-\Theta} = AC \exp (-\frac{\Delta H}{RT}) \] ........................ (5)

Where \( A \) is constant \( C \) is concentration of poly acryl amide and \( \Delta H \) is the heat of adsorption getting logarithmic scale of equation (5) convert to equation (6).
Figures (9, 10, 11, 12) shows the relations between log Θ / 1 - Θ and 1/T at different concentrations and different time of poly acryl amide. The adsorption of poly acryl amide on the (L-80) carbon steel follows Langmuir adsorption isotherm, ΔH of the adsorption can be calculated from the slope of the straight line and are given in Tables (2, 3, 4, 5). The positivity of ΔH means the endothermic adsorption behavior of poly acryl amide on (L-80) carbon steel surface.

**Thermodynamic parameter:**

The adsorption equation formulated on Langmuir as:

\[
\frac{C}{\Theta} = \frac{1}{K} + C \tag{7}
\]

Where K is the equilibrium constant for adsorption operation. Figures (13, 14, 15, 16) shows the curve of log C/ Θ agonist log C for poly acryl amide. The striate line indicates that the adsorption on surface of (L-80) carbon steel obey Langmuir absorption isotherm. The result states clearly there is no reaction between the adsorbed species. The equilibrium constant (K) for adsorption operation can be calculated from the equation (7). The thermodynamic parameters for adsorption operation, (ΔG)_{inh} and entropy (ΔS)_{inh} of the investigated poly acryl amide can be obtained from the thermodynamic relations (8,9).

\[
K = \exp \frac{-\Delta G_{inh}}{RT} \tag{8}
\]

\[
\Delta G = \Delta H - T \Delta S \tag{9}
\]

The ΔG_{inh} and (ΔS)_{inh} values at temperature range from 40 to 90°C are given in Tables (2, 3, 4, 5). The negative values of ΔG_{inh} for poly acryl amid indicates the spontaneous adsorption on the surface of coupon (L-80) carbon steel. The positive value of (ΔS)_{inh} means the molecular of inhibitor is at the continues movement to obtain high absorbability of poly acryl amide on surface of coupon (L-80) carbon steel by making the activated complex with metal.
Mechanism of inhibition:

The action of poly acryl amide as a corrosion inhibitor on carbon steel in acid media can be attributed to some factors such as structure, nature of molecule, types of adsorption sites and the ability to form complexes [23]. The inhibition mechanism of poly acryl amide is happened as a result of complex formation between Fe$^{2+}$ ion, and poly acryl amide. The iron complexes adsorbed on coupon of (L-80) carbon steel and isolated the surface metal from more corrosion happens and these complexes forming by an electron donating groups NH$_2$ and electron withdrawing group C=O that order the inhibition efficiency of the poly acryl amide.

Spectra (FTIR):

The FTIR spectrum of poly acryl amide complex that adsorbed on (L-80) carbon steel coupon shows an intense peak at ~3100 cm$^{-1}$ which is a characteristic feature of N-H stretching frequency and peak at ~ 1600 cm$^{-1}$ for C=O stretching [23-25]. M-O and M-N stretching vibration in low frequency provide direct evidence for the complexation. In the vibration of M-O bond there are many dipole moment changes in comparison with M-N bond [26]. M-N vibrations occur in the region 500–600 cm$^{-1}$ and M-O occur in the region 400-500 cm$^{-1}$ and Figures (17, 18) shows FTIR peaks for polyl acryl amide and adsorbed complex [27].

Conclusions:

1- Poly acryl amid can be used as acid corrosion inhibitor.
2- The efficiency of poly acryl amide depends on concentration of poly acryl amide and temperature of the medium.
3- Poly acryl amide makes stable and insoluble complexes on carbon steel surface.
4- The chemical adsorption of poly acryl amide on coupon of (L-80) carbon steel follows Langmuir equation (absorption isotherm).
Table (1) Inhibition efficiency of poly acryl amide on coupon of (L 80) in (%15) HCl at different temperature different time and Temp.

| R_{corr} mg/cm² h | Θ | E % | Weight loss (gr) | R_{corr} mg/cm² h | Θ | E % | Weight loss (gr) | R_{corr} mg/cm² h | Θ | E % | Weight loss (gr) | R_{corr} mg/cm² h | Θ | E % | Weight loss (gr) | Tem. C° | Concentration % |
|-------------------|---|----|---------------|-------------------|---|----|---------------|-------------------|---|----|---------------|-------------------|---|----|---------------|---------|----------------|
| 5.64              | ---|    | 1.18          | 6.35              | ---|    | 1.15          | 7.83              | ---|    | 1.12          | 9.68              | ---|    | 1.09          | Blank   | 1              |
| 1.98              | .6440| 64.40 | 0.42          | 2.21 .6434| 64.34 | 0.41          | 2.77 .6428| 64.28 | 0.40          | 3.34 .6422| 64.22 | 0.39          | 40       | 4              |
| 2.43              | .7826| 78.26 | 0.40          | 2.76 .7666| 76.66 | 0.49          | 3.32 .7500| 75.00 | 0.47          | 3.92 .7567| 75.67 | 0.45          | 80       | 3              |
| 1.22              | .8086| 80.86 | 0.38          | 2.30 .7952| 79.52 | 0.43          | 2.89 .7765| 77.65 | 0.42          | 3.40 .7891| 78.91 | 0.39          | Blank    | 1              |
| 1.52              | .8608| 86.08 | 0.32          | 1.90 .8523| 85.23 | 0.31          | 2.01 .8404| 84.04 | 0.30          | 2.57 .8540| 85.40 | 0.27          | 1        | 1              |
| 0.57              | .9478| 94.78 | 0.12          | 0.52 .9523| 95.23 | 0.10          | 0.63 .9521| 95.21 | 0.09          | 0.74 .9567| 95.67 | 0.08          | 4        | 4              |
| 0.55              | .9521| 95.21 | 0.11          | 0.49 .9571| 95.71 | 0.09          | 0.57 .9574| 95.74 | 0.08          | 0.54 .9675| 96.75 | 0.06          | 5        | 5              |
| 11.18             | ---|    | 2.50          | 13.03              | ---|    | 2.3          | 13.65              | ---|    | 1.95          | 17.48              | ---|    | 1.91          | Blank    | 1              |
| 2.57              | .7640| 76.40 | 0.59          | 3.12 .7608| 76.08 | 0.55          | 3.59 .7333| 73.33 | 0.52          | 4.27 .7434| 74.34 | 0.49          | 40       | 4              |
| 2.28              | .8120| 81.20 | 0.47          | 2.59 .7956| 79.56 | 0.47          | 3.19 .7641| 76.41 | 0.46          | 3.75 .7748| 77.48 | 0.43          | 2        | 2              |
| 1.62              | .8520| 85.20 | 0.37          | 1.90 .8434| 84.34 | 0.36          | 2.48 .8205| 82.05 | 0.35          | 3.15 .8272| 82.72 | 0.33          | 3        | 3              |
| 0.64              | .9440| 94.40 | 0.14          | 0.62 .9478| 94.78 | 0.12          | 0.77 .9435| 94.35 | 0.11          | 0.90 .9528| 95.28 | 0.09          | 4        | 4              |
| 0.59              | .9480| 94.80 | 0.13          | 0.52 .9565| 95.65 | 0.10          | 0.60 .9538| 95.38 | 0.09          | 0.61 .9633| 96.33 | 0.07          | 5        | 5              |
Table (2) Thermodynamic parameters of poly acryl amide in time (3) hours.

| Conc. % | SA (J.mol⁻¹.k⁻¹) | GA (k.J.mol⁻¹) | HA (K.J.mol⁻¹) | Ea* (K.J.mol⁻¹) | K (K.J.mol⁻¹) | Temp. K | Conc. % |
|---------|------------------|----------------|----------------|-----------------|--------------|---------|---------|
| 1       | 41.91            | -13.00         | 120            | 44              | 1.81x10²     | 313     | 333     |
|         | 43.60            | -14.40         |                |                 | 1.88x10²     | 353     | 363     |
|         | 47.90            | -16.80         |                |                 | 3.12x10²     | 313     | 333     |
|         | 47.40            | -17.10         |                |                 | 2.94x10²     | 353     | 363     |
| 2       | 45.10            | -14.00         | 111            | 55              | 2.22x10²     | 313     | 333     |
|         | 39.97            | -13.20         |                |                 | 1.19x10²     | 353     | 363     |
|         | 43.65            | -15.30         |                |                 | 1.88x10²     | 353     | 363     |
|         | 43.00            | -15.50         |                |                 | 1.72x10²     | 353     | 363     |
| 3       | 42.72            | -13.30         | 72             | 109             | 1.69x10²     | 313     | 333     |
|         | 41.10            | -13.60         |                |                 | 1.38x10²     | 353     | 363     |
|         | 43.83            | -15.40         |                |                 | 1.96x10²     | 353     | 363     |
|         | 42.34            | -15.30         |                |                 | 1.61x10²     | 353     | 363     |
| 4       | 52.25            | -16.30         | 57             | 135             | 5.26x10²     | 313     | 333     |
|         | 51.22            | -17.00         |                |                 | 4.76x10²     | 353     | 363     |
|         | 52.56            | -18.50         |                |                 | 5.50x10²     | 353     | 363     |
|         | 51.67            | -18.70         |                |                 | 5.00x10²     | 353     | 363     |
| 5       | 54.36            | -17.00         | 17             | 158             | 7.14x10²     | 313     | 333     |
|         | 52.90            | -17.60         |                |                 | 5.88x10²     | 353     | 363     |
|         | 53.02            | -18.70         |                |                 | 5.88x10²     | 353     | 363     |
|         | 52.11            | -18.90         |                |                 | 5.26x10²     | 353     | 363     |
Table 3 Thermodynamic parameters of poly acryl amide in time (4) hours.

| Conc. |  |  |  |  |  |  |  |  |
|-------|-------|-------|-------|-------|-------|-------|-------|-------|
|       | SΔ (J.mol⁻¹.K⁻¹) | GΔ (J.mol⁻¹) | HΔ (K.J.mol⁻¹) | Ea* (K.J.mol⁻¹) | K (K.J.mol⁻¹) | Temp. K | Conc. % |
| 1     | 41.87 | -13.00 | 107 | 51 | 1.81x10² | 313 | 1 |
|       | 43.56 | -14.40 |   |   | 1.88x10² | 333 |   |
|       | 47.61 | -16.70 |   |   | 3.03x10² | 353 |   |
|       | 46.85 | -16.90 |   |   | 2.77x10² | 363 |   |
| 2     | 40.17 | -12.50 | 76 | 70 | 1.25x10² | 313 | 2 |
|       | 39.86 | -13.20 |   |   | 1.19x10² | 333 |   |
|       | 42.99 | -15.10 |   |   | 1.75x10² | 353 |   |
|       | 42.35 | -15.30 |   |   | 1.63x10² | 363 |   |
| 3     | 41.71 | -13.00 | 57 | 109 | 1.53x10² | 313 | 3 |
|       | 40.71 | -13.50 |   |   | 1.35x10² | 333 |   |
|       | 42.93 | -15.10 |   |   | 1.75x10² | 353 |   |
|       | 35.96 | -13.00 |   |   | 1.53x10² | 363 |   |
| 4     | 49.34 | -15.40 | 44 | 126 | 3.84x10² | 313 | 4 |
|       | 48.78 | -16.20 |   |   | 3.57x10² | 333 |   |
|       | 51.96 | -18.30 |   |   | 5.00x10² | 353 |   |
|       | 50.25 | -18.20 |   |   | 4.16x10² | 363 |   |
| 5     | 52.47 | -16.40 | 24 | 172 | 5.55x10² | 313 | 5 |
|       | 51.12 | -17.00 |   |   | 4.76x10² | 333 |   |
|       | 50.77 | -17.90 |   |   | 4.54x10² | 353 |   |
|       | 50.20 | -18.20 |   |   | 4.16x10² | 363 |   |
Table (4) Thermodynamic parameters of poly acryl amide in time (5) hours.

| Conc. | SΔ (J.mol⁻¹.k⁻¹) | GA (K.J.mol⁻¹) | HΔ (K.J.mol⁻¹) | Ea* (K.J.mol⁻¹) | K (K.J.mol⁻¹) | Temp. K | Conc. % |
|-------|------------------|----------------|----------------|-----------------|--------------|---------|---------|
| 1     | 41.99            | -13.00         | 143            | 1.81x10²        | 313          | 313     |
|       | 39.46            | -13.00         | 333            | 3.33x10²        | 353          | 363     |
|       | 43.18            | -15.10         | 363            | 3.22x10²        |              |         |
|       | 44.19            | -15.90         | 353            | 1.81x10²        |              |         |
| 2     | 40.30            | -12.50         | 116            | 1.25X10²        | 313          | 333     |
|       | 39.38            | -13.00         | 353            | 1.13X10²        | 333          | 363     |
|       | 43.95            | -15.40         | 353            | 1.96X10²        | 353          |         |
|       | 44.12            | -15.90         | 353            | 1.96X10²        | 363          |         |
| 3     | 41.86            | -13.00         | 105            | 1.49X10²        | 313          | 333     |
|       | 40.25            | -13.30         |                | 1.26X10²        | 333          |         |
|       | 43.92            | -15.40         |                | 1.92X10²        | 353          |         |
|       | 43.26            | -15.60         |                | 1.81X10²        | 363          |         |
| 4     | 48.51            | -15.10         | 86             | 3.33X10²        | 313          |         |
|       | 48.00            | -15.90         |                | 3.22X10²        | 333          |         |
|       | 51.80            | -18.20         |                | 5.00X10²        | 353          |         |
|       | 50.90            | -18.40         |                | 4.54X10²        | 363          |         |
| 5     | 47.82            | -14.90         | 68             | 3.12X10²        | 313          |         |
|       | 49.75            | -16.50         |                | 4.00X10²        | 333          |         |
|       | 50.90            | -17.90         |                | 4.54X10²        | 353          |         |
|       | 50.60            | -18.30         |                | 4.34X10²        | 363          |         |
Table (5) Thermodynamic parameters of poly acryl amide in time (6) hours.

| SΔ (J.mol\(^{-1}\).k\(^{-1}\)) | GΔ (K.J.mol\(^{-1}\)) | HΔ (K.J.mol\(^{-1}\)) | Ea* (K.J.mol\(^{-1}\)) | K (K.J.mol\(^{-1}\)) | Temp. K | Conc. % |
|---------------------------------|----------------------|----------------------|----------------------|----------------------|--------|--------|
| 42.02                           | -13.00               | -17.50               | 155                  | 63                   | 1.81x10\(^2\) | 313     |
| 43.70                           | -14.40               | -17.30               | 3.70x10\(^2\)       | 3.33x10\(^2\)       | 333     |
| 49.44                           | -17.60               | 134                  | 68                   | 1.29x10\(^2\)       | 353     |
| 44.85                           | -15.70               | 2.12x10\(^2\)       | 2.17x10\(^2\)       | 363     |
| 44.99                           | -16.20               | 2.08x10\(^2\)       | 1.92x10\(^2\)       | 363     |
| 41.51                           | -12.90               | 93                   | 78                   | 1.44x10\(^2\)       | 313     |
| 40.31                           | -13.40               | 2.80x10\(^2\)       | 1.28x10\(^2\)       | 333     |
| 44.45                           | -15.60               | 1.92x10\(^2\)       | 2.08x10\(^2\)       | 353     |
| 43.78                           | -15.80               | 4.00x10\(^2\)       | 3.70x10\(^2\)       | 363     |
| 47.56                           | -14.80               | 88                   | 88                   | 3.03x10\(^2\)       | 313     |
| 47.41                           | -15.70               | 2.94x10\(^2\)       | 4.54x10\(^2\)       | 333     |
| 50.95                           | -17.90               | 4.34x10\(^2\)       | 3.80x10\(^2\)       | 353     |
| 50.65                           | -18.30               | 4.00x10\(^2\)       | 3.70x10\(^2\)       | 363     |
| 46.91                           | -14.60               | 2.77x10\(^2\)       | 2.63x10\(^2\)       | 313     |
| 46.49                           | -15.40               | 2.63x10\(^2\)       | 4.00x10\(^2\)       | 333     |
| 49.81                           | -17.50               | 3.70x10\(^2\)       | 3.70x10\(^2\)       | 353     |
| 49.26                           | -17.80               |                      |                      | 363     |

Fig. (1) The weight loss–time plot for (L-80) C-steel in the presence and absence different concentrations of poly acryl amide at (40) °C and %15HCl.

Fig. (2) The weight loss-time plot for (L-80) C-steel in the presence and absence different concentrations of poly acryl amide at (60) °C and %15HCl.
Fig. (3) The weight loss–time plot for (L-80) C-steel in the presence and absence different concentrations of poly acryl amide at (80) oC and %15HCl.

Fig. (4) The weight loss–time plot for (L-80) C-steel in the presence and absence different concentrations of poly acryl amide at (90) oC and %15HCl.

Fig. (5) The (log R_{corr} - 1/T) plot of (L-80) C steel in the presence and absence different concentrations of poly acryl amide at time of (3) hr and %15HCl.

Fig. (6) The (log R_{corr} - 1/T) plot for (L-80) C steel in the presence and absence different concentration of poly acryl amide at time of (4) hr and %15HCl.
Fig. (7) The \( \log(R_{corr} - 1/T) \) plot for (L-80) C steel in the presence and absence of different concentrations of poly acrylamide at time of (5) hr and 15% HCl.

Fig. (8) The \( \log(R_{corr} - 1/T) \) plot for (L-80) C steel in the presence and absence of different concentration of poly acrylamide at time of (6) hr and 15% HCl.

Fig. (9) The \( \log(\Theta/(1-\Theta) - 1/T) \) plot for (L-80) C steel in the presence and absence of different concentrations of poly acrylamide at time of (3) hr and 15% HCl.

Fig. (10) The \( \log(\Theta/(1-\Theta) - 1/T) \) plot for (L-80) C steel in the presence and absence of different concentration of poly acrylamide at time of (4) hr and 15% HCl.
Fig. (11) The \((\log \theta/(1-\theta)-1/T)\) plot for (L-80) C steel in the presence and absence of different concentrations of poly acrylamide at time of (5) hr and %15HCl.

Fig. (12) The \((\log \theta/(1-\theta)-1/T)\) plot for (L-80) C steel in the presence and absence of different concentration of poly acrylamide at time of (6) hr and %15HCl.

Fig. (13) The \((\log C/\theta - \log C)\) plot for (L-80) in the absence and presence of different concentration of poly acrylamide at time of (3) hr and %15HCl.

Fig. (14) The \((\log C/\theta - \log C)\) plot for (L-80) in the absence and presence of different concentration of poly acrylamide at time of (4) hr and %15HCl.
Fig. (15) The (log C/Ω - log C) plot for (L-80) in the presence and absence of different concentration of poly acryl amide at time of (5) hr and %15HCl.

Fig. (16) The (log C/Ω - log C) plot for (L-80) in the presence and absence of different concentration of poly acryl amide at time of (6) hr and %15HCl.

Fig. (17) The FTIR spectrum for poly acryl amide
Fig. (18) The FTIR spectrum of poly acryl amide complex on C-steel
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