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## Anti-Corrosive Substance as Green Inhibitor for Carbon Steel in Saline and Acidic Media

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**Abstract.** Ficus (FIC) leaf extract used as corrosion inhibitor for carbon steel alloy (C.S) in two corrosive environments (saline and acidic) with four concentrations (1, 2, 3 and 4 ppm) at varied temperature range between (298-328 K) using electrochemical polarization measurements. The importance of this work focused on the use the green chemistry that is far from the chemical materials effect. The results of polarization presented the FIC inhibitor consider a mixed type (anodic and cathodic) inhibitor. Tafel curve used to evaluate the corrosion inhibition activity. In a saline medium, the best inhibitor efficiency reaches to (87%) in 2 ppm and IE% reach to (99%) for HCl medium inhibited by 1ppm. Langmuir isotherm obeys the study by thermodynamic parameters and confirms the physical adsorption.

**Keywords:** Adsorption, Ficus, Corrosion, Inhibition, Carbon Steel

### 1. Introduction

Carbon steel constructed of iron and carbon of 0.12 – 2.00%. The more extensive definition includes alloy steels that can also retain the content of up to 10.5% alloy. Carbon steel alloys different uses depend upon the percentage of carbon in the alloy. Due to its cost and quality, mild steel is ordinarily used in nuclear, fossil fuel power plants, marine applications, shipping, chemical manufacturing, building mining and metal manufacturing equipment [1]–[3]. Corrosion is the chemical interaction between a metal with its associated elements. Because background metals subjected to damp, dry, salty, acidic, and oxygenated environments in the carbon steel, the capacity for these reactions must be recognized [4]–[6].

There are many methods for reducing or preventing the corrosion impact on carbon steel, including cathodic control, anodic control, shielding and the usage of corrosion inhibitors [7],



[8]. Inhibitor, in the other hand, is a deficient quantity substance (or a mixture of materials) to treat the metal surface that exposed to a corrosive environment that diminishes the corrosion of metal [9].

Many effective inhibitors have a problem that is detrimental to the extreme environment, including biocompatibility, biodegradability, bioaccumulation, and toxicity composition. Recently, several researchers have been studying green inhibitor employing electrochemical techniques and FTIR spectra, as well as isothermal measurements for adsorption. Inhibitors classified into cathodic type inhibitors, anodic type inhibitors, mixed type inhibitors and volatile type inhibitors of corrosion [10].

Ambrish et al. are studied an inhibition by fruits extract like Orange (*Citrus aurantium*), Pipali (*Piper longum*) and Shahjan (*Moringaoleifera*), Weight loss, electrochemical, spectroscopic potentiodynamic polarization and linear methods for polarization [11]. Cysteine amino acid studied the inhibition as a green inhibitor in medium acidic [12] and *Peganum harmala* as an inhibitor for the carbon steel in seawater [13].

This leaf revealed some natural green inhibitor that has used as corrosion protection on carbon steel by aqueous extracted of Ficus leaf extract. Ficus (FIS) is a medicinal plant commonly used and developed worldwide since ancient times. It appeared in Africa but now spread through almost all regions. The green inhibition used by a ficus leaf in this research [11].

## 2. Experimental section

### 2.1. Ficus extracts

Preparation of plant leaf extract was collected, Washed, dried, ground and stored in a laboratory until use. Weight (50) grams of plant leave powder for aqueous extracts, in a conical flask (500) mL in boiling distilled water, mixed the aqueous extract to (30 min). Then left the mixture for 24h at room temperature, after that filtrated by using Buchner filter, Four concentration of (FIS) used as inhibitor includes 1,2,3 and 4 ppm in corrosive media (synthesis seawater), at temperature range (298- 328)K [14].

### 2.2. Preparation of electrode samples

**Table 1** showed the chemical composition of C.S that used. The working electrode samples used in dimensional polarization experiments (2×2cm) with the thickness (4 mm). The C.S sample regularly polished with different SiC papers grades (between 400 – 2500) mesh, after cleaning; the polished Instances of distilled water, ethanol and acetone were dried, then desiccated. Only 1 cm<sup>2</sup> of the working electrode were exposed to a corrosive solution In the polarization analysis, using the electrode holder of the corrosion experimental [15].

**Table 1:** Composition of C.S chemicals containing.

Grade	% C	% Si	% Mn	% S	% P	% Ni	% Cr	% Mo, Cr+Ni	% Fe
C45	0.42-0.50	<0.40	0.50-0.80	<0.045	<0.045	0.40	<0.40	<0.10	97.31-97.69

### 2.3. Corrosive solution

Sodium chloride (35 gm) was dissolved in 1L distilled water for the preservation of the blank solution for the saline medium. In this study, we choose 3.5 % NaCl to avoid the problems linked to the ohmic drop[8]. Four concentrations (1, 2, 3 and 4) of the extracted solution are prepared. Initially, 100 ppm prepared by added 100 ml of the extracted solution to 1L distilled water. Every 1 ml from the 100 ppm solution represent 1 ppm. So, adding 1, 2, 3 and 4 ml to

999, 998, 997 and 996 ml of blank solution. However, the acid medium a blank solution represent HCl 0.1 M. By the dilution prepared the blank and adding 1, 2, 3 and 4 ml of extraction solution to 999, 998, 997 and 996 ml of blank solution.

#### 2.4. Corrosion tests and inhibition efficiency

The potentiostat Mlab (Germany, 2000) have been for electrochemical tests used and controlled by MLabSci software at temperature range (298-328)K. Three electrodes had been used in the corrosion cell, silver-silver chloride used as a reference electrode. In contrast, an auxiliary electrode its platinum electrode with (1 cm<sup>2</sup>) surface area of the working electrode was C.S. The final corrosive solutions (saline solution and inhibitor solution, acid solution and inhibitor solution) [16]. At each experiment onset, the working electrode must be immersed in the corrosive solution (uninhibited and inhibited) in order to get the stabilization state. The MLabSci software was used to calculate the parameters from Tafel polarization (corrosion potential  $E_{corr}$ , cathodic  $b_c$  and anodic  $b_a$  and a current density of corrosion  $i_{corr}$ ) [17], [18].

### 3. Results and discussions

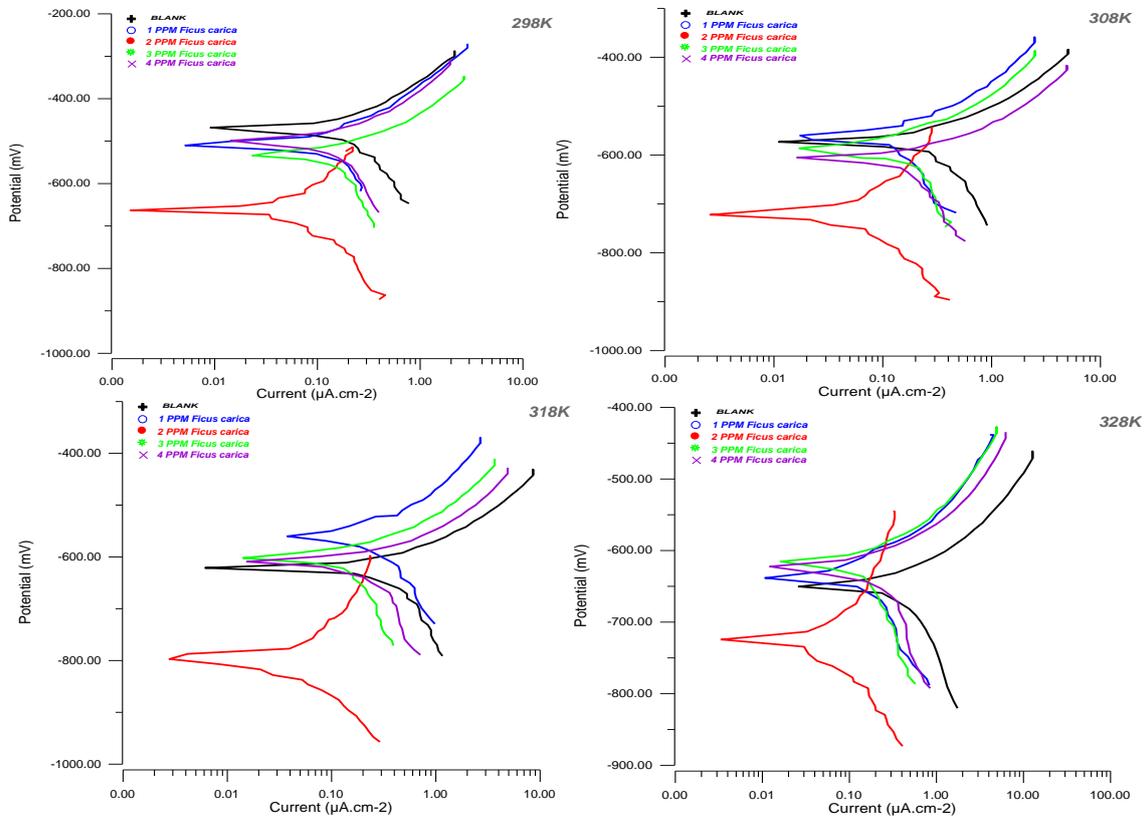
#### 3.1. Saline medium

##### 3.1.1. Corrosion behavior of C.S in Saline media

**Figure 1** shows the cathodic and anodic polarization curve for C.S in saline medium without FIC and with various concentration of FIC at a temperature range (298-328K).  $i_{corr}$  values were calculated by the cathodic tabled lines and the anodic table lines given in **Table 2** shall be extrapolated. In the case that the polarization method was used (Eq.1) to determine the inhibition efficiency (IE %). **Figure 2** reveals that the corrosion potential ( $E_{corr}$ ) for C.S in a saline solution containing FIC inhibitor. Where indicate  $E_{corr}$  shifted to more active potential compared with ( $E_{corr}$ ) for C.S in saline solution without FIC at 298 K but when the temperature increase to 308 K the corrosion potential ( $E_{corr}$ ) for C.S in saline solution contain 1ppm of FIC shifted to more noble value, but ( $E_{corr}$ ) for C.S in saline solution at 318K and 328K shifted to more noble at all concentration except (2 ppm). This different in ( $E_{corr}$ ) values were referred to FIC inhibitor act as mix inhibitor.

$$IE\% = \frac{(i_{corr})_{un} - (i_{corr})_i}{(i_{corr})_{un}} \times 100 \quad \dots\dots\dots 1$$

Where  $i_{corr} (un)$  and  $i_{corr} (i)$  are the uninhibited and inhibited corrosion current densities, respectively[15], data in **Table 2** shows ( $i_{corr}$ ) was primarily reduced when FCI was added the for all concentrations and temperatures from (280.34 to 23.05)  $\mu A.cm^{-2}$  and the best inhibition efficiency (IE %) obtained with 2 ppm that gives (IE%) reach to 87% at 318 K. ( $i_{corr}$ ) values in **Table 2** were increased with temperature increases in both without FIC and with various concentration of FIC solution. In contrast, the (IE %) inhibition efficiency of FCI decreases when temperature increase.



**Figure 1.** C.S. polarization plots in a saline medium at specific FCI concentrations at 298-328 K range.

**Table 2.** C.S in saline medium in the presence and absence different FCI concentration at temperature range 298-328 K results.

FIC /ppm	T/K	E <sub>corr</sub> /mV	I <sub>corr</sub> / μA.cm <sup>-2</sup>	-bc/ mV.dec <sup>-1</sup>	ba/ mV.dec <sup>-1</sup>	IE%	θ
3.5% NaCl	298	469.0	168.72	243.9	107.2	-	-
	308	573.5	201.57	179.1	104.2	-	-
	318	622.0	220.34	140.3	82.2	-	-
	328	651.0	280.34	137.3	79.6	-	-
1	298	510.3	23.05	39.1	54.8	86.338	0.863
	308	562.6	46.23	63.5	56.4	77.065	0.771
	318	560.2	71.46	65.0	67.7	67.568	0.676
	328	620.1	91.91	105.7	70.7	67.215	0.672
2	298	664.2	24.82	94.5	100.8	85.289	0.853
	308	722.9	30.44	100.6	125.2	84.899	0.849
	318	797.1	34.83	120.4	129.2	84.193	0.842
	328	791.7	35.03	126.1	80.9	87.504	0.875
3	298	535.7	50.00	95.5	65.9	70.365	0.704
	308	570.1	58.11	90.8	56.5	71.171	0.712

	318	602.8	62.30	85.0	55.3	71.726	0.717
	328	616.7	67.02	82.2	47.3	76.093	0.761
4	298	501.0	33.44	59.3	44.3	80.180	0.802
	308	598.7	53.68	80.8	73.1	73.369	0.734
	318	609.0	65.80	88.0	44.9	70.137	0.701
	328	622.9	89.40	99.5	53.0	68.110	0.681

3.1.2 Kinetics and Thermodynamic Studies for C.S in saline media

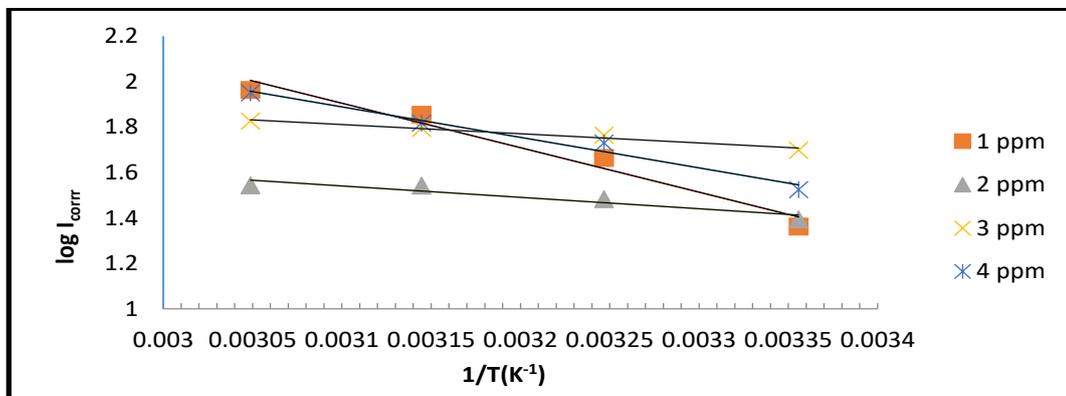
Potentiostat polarization experiments have been carried out in the temperature range 298–328K to investigate the effect of temperature on the inhibition efficiencies of FIC without FIC and with different FIC concentration. **Table 3** shows various corrosion parameters. The calculated data proposed that FIC have been adsorbed on the C.S surface at all studied temperatures and  $I_{corr}$  increased with as well as without FIC when the temperature increases in saline solutions. The cathodic reaction accompanies corrosion of metal; Temperature rise typically increases corrosion processes, resulting in higher metal dissolution levels. The Arrhenius Equation (Eq.2) Moreover, transition state Equation 3 are used to calculate the activation parameters for the corrosion process[19][20].

$$\text{Log}i_{corr} = \frac{-E_a}{2.303RT} + \text{log}A \quad \dots\dots\dots 2$$

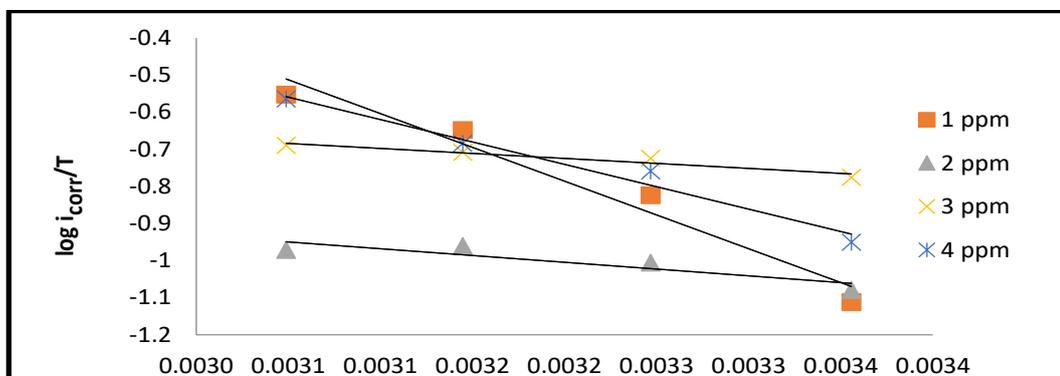
$$\text{log} \frac{i_{corr}}{T} = \text{log} \frac{R}{N_h} + \frac{\Delta S^*}{2.303R} - \frac{\Delta H^*}{2.303RT} \quad \dots\dots\dots 3$$

The graph between logarithm  $i_{corr}$  against temperature inverse 1/T (**Figure 2**) shows a linear relation with slope (-  $E_a / 2.303 R$ ). The intercept of the extrapolated line log A. The activation energy (Ea) values increased after inhibited the corrosive solution by FIC inhibitor by different concentrations, this refers to a reduction in the rate-determining step (difficult to corrosion), and the results were described in **Table 3**.

Straight lines were taken from the  $\text{log}i_{corr} / T$  compared with the 1/T plot showed (**Figure 3**) in the pitch, of- ( $-\Delta H^*/2.303 R$ ), and the  $[(\text{log} (R/N_h) + (\Delta S^*/2.303 R))]$  intercept from which values of  $\Delta S^*$  and  $\Delta H^*$  were determined by EQ.3, respectively. Correct lines were produced.



**Figure 2.** Log icorr compared to 1/T plot for 3.5% NaCl corrosion containing specific Ficus concentrations



**Figure 3.** plots of  $\log i_{\text{corr}}/T$  vs  $1/T$  for the corrosion of C.S in 3.5% NaCl in the absence and the presence of FCI.

$\Delta H^*$  value for the transition state of corrosion for C.S in saline medium increase after inhibited by FIC from (10.467)  $\text{kJ}\cdot\text{mol}^{-1}$  to (5.147 – 34.877)  $\text{kJ}\cdot\text{mol}^{-1}$ . The highest  $\Delta H^*$  value occurred with 1ppm of FCI. However, the entropy of activation value  $\Delta S^*$  for the corrosion of C.S in 3.5 % NaCl solutions were slightly affected when FCI was added to the corrosive solution, as shown in **Table 3**.

**Table 3:** Kinetic parameters of a saline medium in presence FCI and absence.

$C_{\text{FIC}}$ in 3.5% NaCl	$\Delta H^*/\text{kJ}\cdot\text{mol}^{-1}$	$-\Delta S^*/\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$	$E_a/\text{kJ}\cdot\text{mol}^{-1}$	$A \text{ Molecules}\cdot\text{cm}^{-2}\cdot\text{S}^{-1}$
Without	10.467	178.8	13.0652	$2.0\cdot 10^{28}$
1ppm	34.877	101.0	37.4748	$5.6\cdot 10^{31}$
2ppm	6.995	194.4	39.5935	$7.5\cdot 10^{26}$
3ppm	5.147	195.0	27.7454	$7.0\cdot 10^{26}$
4ppm	23.107	137.8	25.7050	$6.8\cdot 10^{29}$

### 3.2. Acid medium

#### 3.2.1. Corrosion behavior of C.S. in acidic medium

The curves in the polarization of corrosion C.S. immersed in acidic media with and without presence FIC inhibitor at variable temperatures were recorded. The curves of polarization of C.S in HCl without FIC shows ( $E_{\text{corr}}$ ) ranged between (-469.1 to 441.1 mv), and this potential shifted to (-945 to -1063 mV) after inhibited the corrosive media (HCl) by different concentration of FIC. This value more active potential; this indicates the FIC extract in acidic media act as a cathodic inhibitor [17].

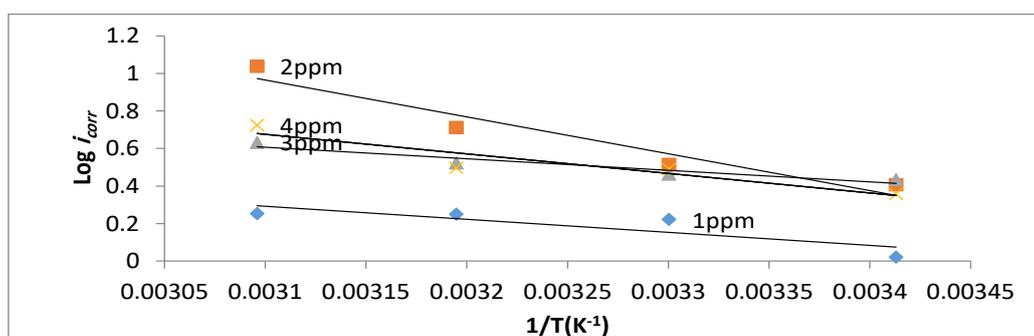
**Table 4:** Corrosion data for C.S in an acid medium in present FCI and absence at temperature range 298-328 K.

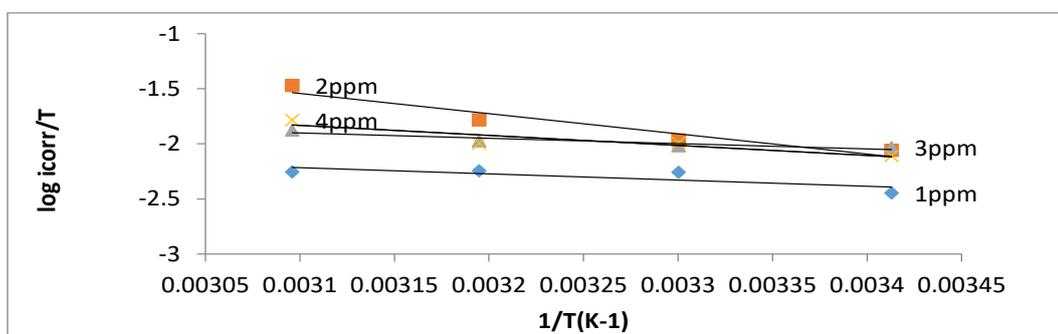
FiC /ppm	T/K	$-E_{\text{corr}}$ /mV	$i_{\text{corr}}$ / $\mu\text{A.cm}^{-2}$	IE%	$\theta$
0.1M HCl	298	460.1	131.24	--	--
	308	457.6	182.19	--	--
	318	468.5	288.89	--	--
	328	441.1	528.72	--	--
1	298	989	1.05	99.1	0.991
	308	945	1.67	99	0.99
	318	966	1.78	99.3	0.993
	328	965	1.79	99.6	0.996
2	298	1016	2.55	98	0.98
	308	1012	3.28	98.1	0.981
	318	1035	5.16	98.2	0.982
	328	1035	10.93	97.9	0.979
3	298	1008	2.72	97.9	0.979
	308	1030	2.91	98.4	0.984
	318	1031	3.34	98.8	0.988
	328	1038	4.3	99	0.99
4	298	1043	2.28	98.2	0.982
	308	1034	3.1	98	0.98
	318	1047	3.15	98.9	0.989
	328	1063	5.31	98.9	0.989

Data in **Table 4** shows that the ( $i_{\text{corr}}$ ) were reduced after adding FIC inhibitor to acidic corrosive media, where ( $i_{\text{corr}}$ ) value of C.S in HCl with FIC decrease from (131.24 – 528.72)  $\mu\text{A.cm}^{-2}$  to (1.05 – 10.93)  $\mu\text{A.cm}^{-2}$  in different concentration at temperature ranged (298 – 328)K and this decrease in ( $i_{\text{corr}}$ ) which give IE% reach to 99.6% for 1ppm FIC at 328K [21], [22]. The lowest IE% were obtained with 2 and 3 ppm at 328K and 298K respectively.

### 3.2.2. Kinetics and thermodynamic studies for C.S in Acidic media

Temperature effect on C.S corrosion kinetic mechanism in 0.1M HCl to get extra knowledge on the electrochemical demeanour of metallic materials in acidic medium and for protection, C.S. Adding FIC inhibitor in corrosive media (0.1M HCl) lead to increase ( $E_a$ ) from (16.36  $\text{kJ.mol}^{-1}$ ) to (19.966–33.32  $\text{kJ.mol}^{-1}$ ) for different concentration which means that add FIC to corrosive media lead to decrease the corrosion rate [23]. The ( $E_a$ ) values decreased with increasing the concentration of FIC from 1ppm to 4ppm.

**Figure 4.** Plot of  $\log i_{\text{corr}}$  versus  $1/T$  for the corrosion of C.S in 0.1m HCl solution containing various PLE with (1, 2, 3 and 4) ppm.



**Figure 5.** Arrhenius plots of  $\log I_{\text{corr}}/T$  vs  $1/T$  for the corrosion of C.S in 0.1M HCl solution containing various FCI with (1, 2, 3 and 4) ppm.

**Table 5:** Kinetic parameters in presence FCI and absence.

$C_{\text{FCI}}$ in 0.1M HCl	$\Delta H/\text{kJ.mol}^{-1}$	$-\Delta S/\text{J.mol}^{-1} .\text{K}^{-1}$	$E_a/\text{kJ.mol}^{-1}$	$A \text{ Molecules.cm}^{-2} .\text{S}^{-1}$
Without	33.795	-0.127	16.360	$2.562 \times 10^{13}$
1ppm	-10.767	-0.20662	33.32335	1.69276E+26
2ppm	-15.0929	-0.21832	32.64903	1.92463E+30
3ppm	-9.23888	-0.20533	21.79521	1.97832E+26
4ppm	-17.4105	-0.17868	19.96664	4.87525E+27

The result of **Table 5** expresses the thermodynamic feasibility of the corrosion reaction of C.S inhibited by FIC inhibitor. Negative  $\Delta S^*$  values increased after adding FIC inhibitor in corrosive media (0.1M HCl) that mean decrease in disorder occurs in the move of reactants to activated complex. The nature of corrosion transition state change from endothermic to exothermic transition state process after inhibited corrosion reaction of C.S by FIC.

### 3.3. Adsorption study

The isotherm adsorption can be determined by assuming that the adsorption on the interface between metal and solution is due to the inhibition effect. Adsorption isotherm can provide information adsorption inhibitors on surface metal. The surface coverage values ( $\theta$ ) must be calculated as a function of the concentration of an inhibitor in order to achieve the isotherm.  $\theta$  values can be estimated easily by the ratio of ( $\%IE/100$ ), where  $\%IE$  is an efficiency of inhibition determined by polarization method. Therefore, the isotherm that better serves adsorption of inhibitors in the C.S surface must be calculated empirically. Several isothermal adsorption mechanisms were tested (i.e. Frumkin, Langmuir, Temkin, Freundlich). A better explanation of the adsorption activity of this inhibitor was found with Langmuir adsorption isotherm.

#### 3.3.1 Saline medium

The isotherm Langmuir is given by the following equation (4) as shown in **Table 6**:

$$\frac{\theta}{C_{\text{inh}}} = \frac{1}{K_{\text{ads}}} + C_{\text{inh}} \quad \dots\dots\dots (4)$$

Where  $C_{inh}$  is the concentration of inhibitor,  $K_{ads}$  (constant equilibrium of the adsorption process), and  $\theta$  (surface coverage). Plot  $C_{inh}/\theta$  agent C to product a straight line, as shown in figures (6) with regression coefficient,  $R^2$ , almost equal to 1.

**Table 6:** Langmuir adsorption parameters for the adsorption of FIC on the surface of C.S in saline medium

T/K	$K_{ads}$	$\Delta G/kJ.mol^{-1}$	$\frac{\Delta H}{kJ.K^{-1}.mol^{-1}}$	$\Delta S/kJ.mol^{-1}$	$R^2$
298	0.746102	- 9.22688	- 2.55863	22.37665	0.9742
308	0.698568	- 9.36790		22.10802	0.9895
318	0.690179	- 9.64011		22.26879	0.9841
328	0.675083	- 9.88293		22.33019	0.9701

In the saline medium, the FIC inhibitor has observed as Langmuir isotherm and the relation there is the insignificance of the adsorbed molecules. The free energy of adsorption ( $\Delta G_{ads}$ ), is calculated from Eq. (6). The negative value of  $\Delta G_{ads}$  and the high values of the adsorption constant indicate spontaneous adsorption of these inhibitors on C.S and increase as the inhibition efficiency increases. These results mean that this substance's inhibition activity occurs from the chemical adsorption of specific molecules on the C.S surface.

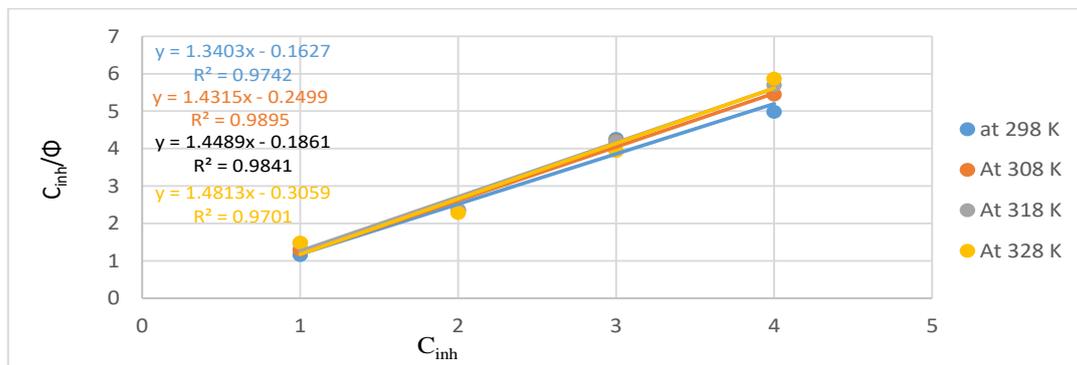
$$K_{ads} = \frac{1}{55.5} \exp \left[ \frac{-\Delta G_{ads}}{RT} \right] \dots\dots\dots(5)$$

$$\Delta G_{ads} = -2.303RT \log(55.5K_{ads}) \dots\dots\dots(6)$$

The thermodynamic model considering, it is possible to illustrate further the corrosion inhibition of C.S in the presence of (FIC) using the adsorption enthalpy  $\Delta H_{ads}$  and adsorption entropy [ $\Delta S_{ads}$ ] that can be determined from the integrated Hoff equation:

$$\log K_{ads} = \frac{-\Delta H_{ads}}{2.303RT} + \frac{\Delta S_{ads}}{2.303R} + \log \frac{1}{55.5} \dots\dots\dots (7)$$

The value of enthalpy was (-2.55863 kJ.mol<sup>-1</sup>); its negative value indicates to the exothermic process.



**Figure 6.** Langmuir isotherms plots for the adsorption of FIC on carbon steel in a saline medium.

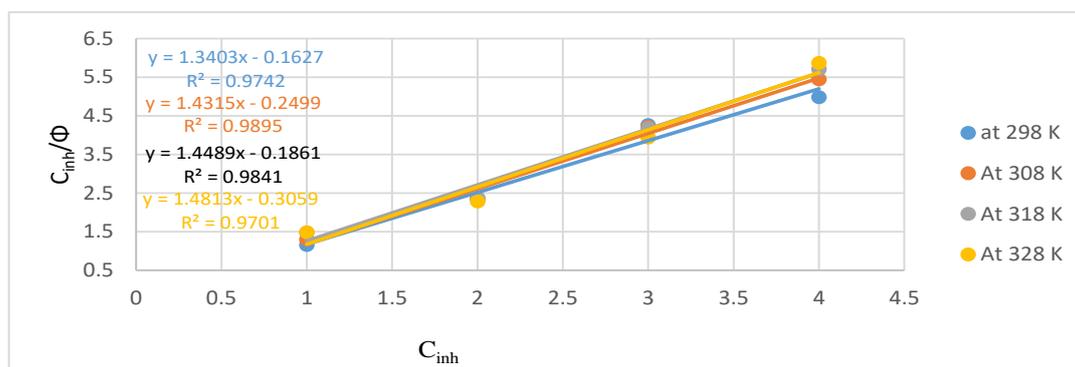
### 3.3.2. Acidic medium

**Table 7** shows the adsorption isotherm of FIC on C.S. surface at acidic medium, and it shows the  $K_{ads}$ ,  $\Delta G$ ,  $\Delta H$ ,  $\Delta S$  and  $R^2$  values. The value of  $\Delta G$  refer to the adsorption of these inhibitors on C.S surface are spontaneous. While  $\Delta H$  value indicates an endothermic process of adsorption an acidic medium, as in saline medium the entropy change value show little effect by temperature.

**Table 7:** Langmuir parameters of FIC adsorption on the surface of C.S in HCl environment.

T/K	$K_{ads}$	$\Delta G/\text{kJ.mol}^{-1}$	$\Delta H/\text{J.K}^{-1}.\text{mol}^{-1}$	$\Delta S/\text{kJ.mol}^{-1}$	$R^2$
298	0.978857	-9.89972	339.441	34.35959	1
308	0.978857	-10.2319		34.3226	1
318	0.988924	-10.5912		34.37303	1
328	0.989218	-10.925		34.34296	0.9999

FIC inhibitor obeyed the Langmuir isotherm in an acidic medium according to  $R^2$  value, which equal to one by graphically  $C/\theta$  and the concentration as shown in **Figure 7**.



**Figure 7.** The graphically Langmuir thermal of FIC inhibition on the surface of carbon steel.

## 4. Conclusions

An extract Ficus leaf used as an inhibitor of corrosion CS in various temperatures (298, 303, 318 and 328 K). Saline and the acidic medium used in this study. The examination of Tafel measurements for the inhibited solutions shows that the application of FCI inhibits the dissociation of Fe and acts as an anti-corrosive with an effective inhibition abundant to 84 % at 333 K in 2 ppm and 99.6 % in 1 ppm at 328 K for the acid medium (HCl). According to the results, we can classify as a mixed-inhibition type in two media. Corrosion rates and  $E_a$  for the C.S with solutions of FCI were compared to C.S in solution without FCI. The adsorption of FCI on the CS surface applied to Langmuir rule and concluded as physisorption type. Spontaneous interaction between FCI and CS according to  $\Delta G_{ads}^\circ$  values in negative for all medium studied.

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