

INHIBITING EFFECTS OF L-TRYPTOPHAN ON THE CARBON STEEL CORROSION IN ALKALINE SALINE SOLUTION

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ABSTRACT

The inhibiting effect of L-tryptophan, that considered as an eco-friendly compound on the corrosion of carbon steel in saline solution (3.5% NaCl) at pH 11 was investigated potentiostatically at five temperatures in the range 293-313 K. Five concentrations of inhibitor were tested ranging from 5×10^{-4} to 1×10^{-2} mole L⁻¹. The addition of L-tryptophan to alkaline sodium chloride solution lowered the corrosion feasibility of the alloy. On the other hand, the corrosion feasibility increased with the rise of temperature. Carbon steel corrosion rate in the saline solution was decreased on increasing L-tryptophan concentration reflecting the inhibiting effect of L-tryptophan when it is present at high concentration in the saline solution. The efficiency of inhibition (%IE) of L-tryptophan increased with increasing the concentration of the inhibitor attaining a maximum value of 71.72 % when the L-tryptophan concentration in the saline solution became 1×10^{-2} mole L⁻¹, and decrease with the rise of temperature. Adsorption of L-tryptophan on the surface of carbon steel was considered to obey Langmuir isotherm. Gibbs free energy of adsorption of L-tryptophan on steel was -29.07 kJ mol⁻¹ indicates a physisorption of the inhibitor on the surface of carbon steel alloy. Quantum mechanical study using DFT method with B3LYP/6-31G basis set was applied to achieve good relation between the inhibition efficiency and the chemical structure of L-tryptophan.

Keywords: Corrosion, Carbon Steel, saline solution, Adsorption, L-tryptophan, DFT method.

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INTRODUCTION

Carbon steel is one of the most extensively used iron alloy in engineering material, tonnages in marine applications, chemical handling, manufacture of petroleum, purgation, construction and metal-processing equipment¹. Protection of metals or alloys from corrosion were studied by using many techniques. The use of organic inhibitors containing a heteroatom, like oxygen, nitrogen, sulfur and multiple bonds is usually the most efficacious method for the protection of metals against corrosion in saline solution. The chosen of inhibitors selected by its economic availability, its efficiency to inhibit the substrate material and its environmental parameters². On the other hand, most organic compounds are expensive and toxic to both human beings and environments, so the used of natural, Eco-friendly, cheap and nontoxic compounds are required³. Amino acids are low cost, non-toxic and highly soluble in aqueous media with high purity which adsorbed on the surface of a metal to reduce the rate of corrosion either by coverage effect of adsorbed inhibitor molecules on the metal surface and / or effect on the activation barriers of the anodic and cathodic reactions of the corrosion reaction⁴.

The well-known computational method that is used successfully to explain the chemical reactivity of inhibitors and their adsorption efficiency on the metal surface is density functional theory (DFT). This method is also providing suitable information to corrosion researchers in order to the investigation of corrosion inhibition mechanism theoretically. Such computations were widely used to analyze the molecular electronic structures of adsorbed inhibitors using a number of computational descriptors that give important insights into the mechanisms of corrosion inhibition. Consequently, inhibition efficiency corresponds to the molecular and structural properties of inhibitor compounds. These parameters which might be obtained through computational calculations that includes, chemical reactivity, charge distribution and the frontier molecular orbit theory, HOMO (higher occupied molecular orbital) energy, the LUMO (lower unoccupied molecular orbital) energy, the energy gap (ΔE), global hardness (η), global softness (σ), dipole moment (μ), electronegativity (χ) and electron transfer number (ΔN)^{5,6}.

This work aimed to investigate the effectiveness of L-tryptophan on the protection efficiency and kinetic of the carbon steel corrosion in saline solution at pH 11. The chemical and optimize structures of L-tryptophan are presented in Fig.-1. A correlation was made between the corrosion inhibition potential and molecular structure of L-tryptophan using DFT method.

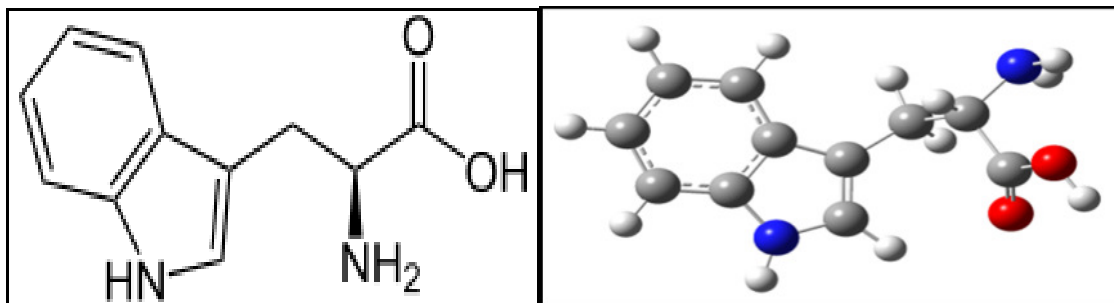


Fig.-1: The chemical and optimized structures of L-tryptophan.

EXPERIMENTAL

Sample Preparation

Carbon steel specimen has the following composition in weight percentage (P 0.018, Mo 0.03, Ni 0.017, C 0.19, Si 0.35, Cr 0.04, Cu 0.02, Al 0.06, and the rest iron) as revealed by emission spectroscopic analysis. Analar grade chemicals (NaCl, NaOH and L-tryptophan) were used for the preparation of the solutions. Circular working electrode samples with 2.5 cm in diameter, 1mm in thickness and with a surface area of 1 cm² were polished to mirror finish with emery paper in various grades of 320, 500, 1000, 2400 and 4000 mm, then with diamond product spray that contains ethanol with different size of diamond particles (1, 3, 6, 9) μm and then washed with ethanol, acetone and finally rinsed with distilled water.

Preparation of solutions

1. 35g of NaCl was dissolved in 1000 mL distilled water to prepare 3.5% of NaCl solution.
2. L-tryptophan was prepared in different concentrations (5×10^{-4} , 1×10^{-3} , 5×10^{-3} , and 1×10^{-2}) M by dissolving appropriate amount in 1000 mL of 3.5% NaCl solution at pH 11.
3. 1M NaOH solution was prepared to control the pH of the testing solution.

Electrochemical studies

Electrochemical measurements were performed by potentiodynamic polarization calculations that carried out in an electrochemical cell using three-electrode:

- Saturated Calomel Electrode (SCE) as a reference electrode.
- a platinum electrode as an auxiliary electrode.
- carbon steel alloy as a working electrode.

M Lab (WENKING MLab multichannel and SCI-MLab system for corrosion measurement - obtained from Bank Electronics- Intelligent controls GmbH, Germany 2007) is an instrument with high-tech for electrochemical measurements. M-Lab is worked by computer desktop Window XP.

Theoretical Studies

Gaussian 09W program was used to study the presence of a relation between computational results and the experimental inhibition efficiencies of L-tryptophan being corrosion inhibitor. Computational results were achieved by using DFT method with Beck's three parameter exchange functional along with Lee-Yang-Parr non-local correlation functional (B3LYP) and with 6-31G basis set. E_{HOMO} (highest occupied molecular orbital), E_{LUMO} (lowest unoccupied molecular orbital), ΔE (energy gap), μ (dipole

moment), E_{total} (the total energy), χ (absolute electronegativity), γ (global hardness) and ΔN (the fraction of electrons transferred) were calculated by using the above given computer code package.

RESULTS AND DISCUSSION

Polarization Measurements

Table-1 presents the data obtained from the polarization curves for corrosion of carbon steel in 3.5% NaCl solution at pH 11 for blank and inhibited solution containing various concentrations of L-tryptophan at different five temperatures 293, 298, 303, 308 and 313 K (Fig.-2). Values of corrosion current density (i_{corr}), corrosion potential (E_{corr}), cathodic (b_c), and anodic (b_a) Tafel slopes are given in Table-1.

The addition of L-tryptophan to saline solution (3.5% NaCl) at pH 11 caused an alteration in the corrosion potential (E_{corr}) of carbon steel. The range of shifting is toward more negative potentials and the effect became more intense with increasing concentration of inhibitor (Table-1). This is interpreted on the basis of the decreasing corrosion feasibility in the existence of L-tryptophan and also by its increasing concentration in the saline solution, this indicated that L-tryptophan acts more cathodic than an anodic inhibitor.

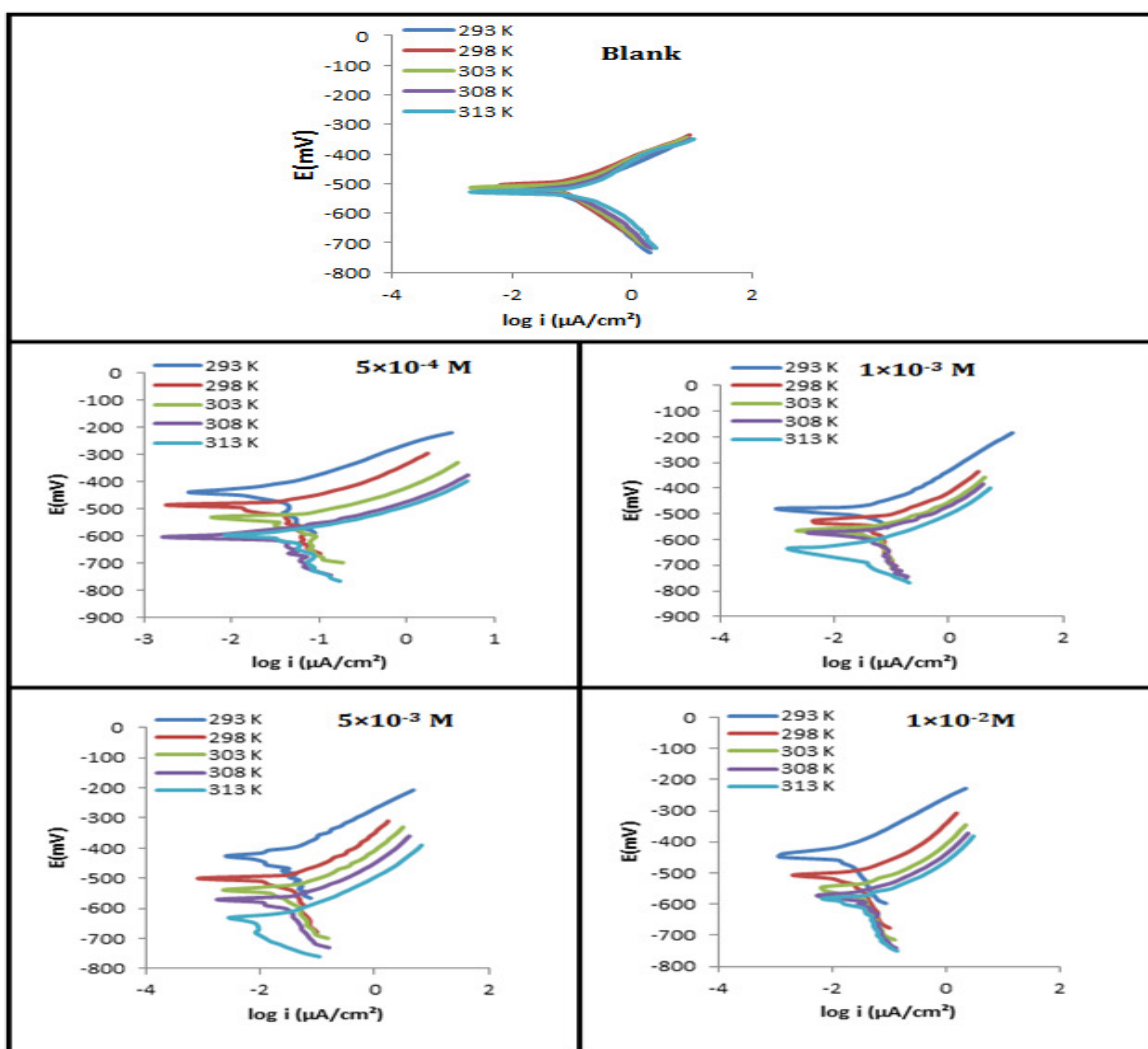


Fig.-2: Polarization curves for carbon steel corrosion in 3.5% NaCl at pH 11 for blank and inhibited solution containing various L-tryptophan concentrations at different temperatures in the range (293-313)K.

At a constant L-tryptophan concentration, E_{corr} values moved to more negative potential with increasing temperature. Thus, corrosion became more feasible with the rise of temperature (Table-1). The corrosion rate as reflected in the values of the corrosion current density (i_{corr}) decreased at 293 K with increasing L-tryptophan concentration in the saline solution as shown in Table-1. At high concentrations of L-tryptophan, i_{corr} began to decrease. At a constant L-tryptophan concentration in the saline solution, corrosion rate as reflected in the values of i_{corr} (Table-1) increased with the rise of temperature. The corrosion current density (i_{corr}) values are decreased with increasing inhibitor concentration. The i_{corr} values increases, whereas the protection efficiencies (%IE) values decrease with elevation temperature. The maximum inhibition efficiency 71.7 % was achieved at the highest L-tryptophan concentration of 1×10^{-2} M and at 293K.

Values of Tafel slopes (cathodic and anodic) are varied at all L-tryptophan concentrations and temperatures (Table-1). These results refer to the variation of the rate-determining step from charge transfer process to either chemical deposition or to electrochemical desorption in the cathodic reactions and also to the variation of this step in dissolution reaction of carbon steel alloy⁷.

Table-1: Corrosion parameters of carbon steel in 3.5% NaCl at pH 11 for blank and inhibited solution containing various L-tryptophan concentrations (5×10^{-4} , 1×10^{-3} , 5×10^{-3} and 1×10^{-2}) M at different temperatures in the range (293-313)K..

Inh. [M]	T [K]	- E_{corr} [mV]	i_{corr} [$\mu\text{A}/\text{cm}^2$]	Tafel slope [mV/dec]		θ	%IE
				- b_c	+ b_a		
0	293	507.1	51.09	92.5	62.6	-	-
	298	513.9	55.67	136.7	73.2	-	-
	303	516.8	66.99	120.9	81.3	-	-
	308	525.8	87.14	107.0	95.1	-	-
	313	530.3	92.28	103.5	91.9	-	-
5×10^{-4}	293	437.5	22.78	269.6	102.1	0.554	55.41
	298	488.5	25.77	262.9	77.7	0.537	53.71
	303	537.0	32.91	179.3	68.1	0.509	50.87
	308	585.3	43.13	339.5	74.2	0.505	50.50
	313	598.6	48.07	277.6	77.6	0.479	47.91
1×10^{-3}	293	486.8	20.09	94.30	68.8	0.607	60.68
	298	533.7	24.30	139.3	58.8	0.564	56.35
	303	565.1	31.15	259.8	71.2	0.535	53.50
	308	579.5	41.95	167.2	69.8	0.519	51.86
	313	629.4	45.22	285.6	86.1	0.510	51.00
5×10^{-3}	293	433.3	19.40	191.5	96.1	0.620	62.03
	298	504.6	22.88	179.3	71.2	0.589	58.90
	303	538.8	28.76	339.9	74.6	0.571	57.07
	308	576.6	38.47	285.9	75.8	0.559	55.85
	313	624.2	42.84	235.5	75.4	0.536	53.58
1×10^{-2}	293	436.6	14.45	135.9	83.7	0.717	71.72
	298	510.4	18.85	155.9	79.2	0.661	66.14
	303	544.4	24.28	343.4	68.7	0.638	63.76
	308	572.9	33.05	134.8	84.9	0.621	62.07
	313	588.4	38.16	256.1	79.5	0.587	58.65

Corrosion Protection

The protection efficiency (%IE) of L-tryptophan in inhibiting carbon steel corrosion in 3.5% NaCl solution at pH 11 was determined from the equation⁸:

$$\%IE = \frac{i_{\text{corr}}^0 - i_{\text{corr}}}{i_{\text{corr}}^0} \times 100 \quad (1)$$

Where, i_{corr}^0 and i_{corr} are respectively the corrosion current density for blank and inhibited saline solution containing various L-tryptophan concentrations at the same temperature. The calculated values of %IE, for the different tryptophan concentrations and temperatures, are given in Table-1. At 293 K, the corrosion protection (%IE) by L-tryptophan increased with increasing its concentration, attaining a maximum of 71.72 % when L-tryptophan concentration became 1×10^{-2} mole.L⁻¹ in the saline solution (Table-1).

The coverage (θ) of carbon steel's surface by the adsorbed L-tryptophan is estimated from the current density (i) at constant applied potential (E) for blank (i_{corr}) and inhibited saline solution ($i_{corr(inhi)}$) containing various concentrations of L-tryptophan by using this relation⁹:

$$\theta = \frac{i_{corr} - i_{corr(inhi)}}{i_{corr}} \quad (2)$$

Kinetics of Corrosion

The dependence of (i_{corr}) values on temperature was examined by plotting $\log(i_{corr})$ versus $1/T$ for the various L-tryptophan concentrations in saline solution in accordance with the Arrhenius equation¹⁰:

$$i_{corr} = A \exp\left(\frac{-E_a}{RT}\right) \quad (3)$$

Where i_{corr} stands for the rate of corrosion at temperature T. Activation energy (E_a) and the pre-exponential factor (A) data were obtained from the slopes and intercept respectively $\log(i_{corr})$ versus $1/T$ plots as shown in Figure-3. The resulting values of (E_a) and (A) are given in Table-2. It is noted that the activation energy (E_a) is higher in the presence of the inhibitors than in blank solution and it will increase with increasing concentration of inhibitor due to adsorption of inhibitor molecules on the metal surface, and it decreases with increasing the temperature, at high temperature occurs desorption of inhibitor molecule so the metal surface exposed to corrosion. A relatively higher value of (E_a) in the presence of L-tryptophan is combined with relatively higher pre-exponential factor A as compared with the corresponding values of (E_a) and (A) in the absence of the inhibitor. Thus, a high value of (E_a) is connected with a high value of (A) and the reverse is also true. This effect was ascribed to the presence of energetically heterogeneous reaction sites on the carbon steel surface.

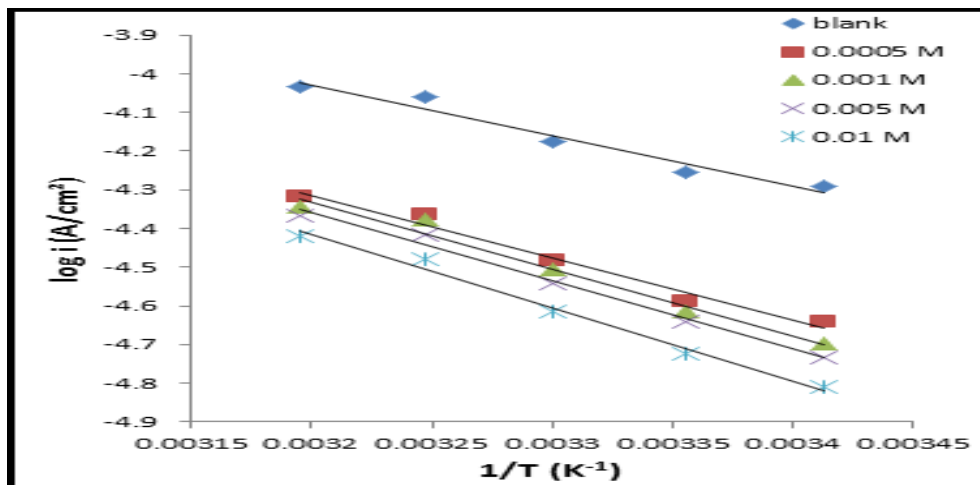


Fig.-3: $\log i_{corr}$ versus $1/T$ of carbon steel corrosion in 3.5% NaCl at pH 11 solution for blank and inhibited solution containing various L-tryptophan concentrations.

Enthalpy (ΔH) and entropy (ΔS) of activation were calculated from the transition state of a replacement formulation of Arrhenius equation that expressed as¹¹:

$$i_{corr} = \left(\frac{RT}{N_A}\right) \exp\left(\frac{\Delta S}{R}\right) \exp\left(\frac{-\Delta H}{RT}\right) \quad (4)$$

Where, i_{corr} is the corrosion current density, N is the Avogadro number, h is the Planck's constant, R is the molar gas constant and T is the absolute temperature. Equation (4) may be rearranged into linear form as :

$$\ln i_{corr} = \left(\frac{-\Delta H}{R}\right) \left(\frac{1}{T}\right) + \left[\ln \left(\frac{R}{N h}\right) + \left(\frac{\Delta S}{R}\right)\right] \quad (5)$$

ΔH and ΔS values were obtained respectively, from the slope and intercept of the plot of $\ln i_{corr}$ against $1/T$ as show in Fig.-4, and the data obtained are given in Table-3. Values of activation enthalpies (ΔH) were positive and the positivity increased with increasing L-tryptophan concentration. The positive values of ΔH in the absence and presence of the L-tryptophan indicates the endothermic nature of the carbon steel dissolution process. The entropy of activation (ΔS) data are negative and negativity decreased with increasing L-tryptophan concentration. The negative values of ΔS reveal that the activated complex in the rate determining step represents association rather than the dissociation step, this means a decrease in disorder take place^{12,13}.

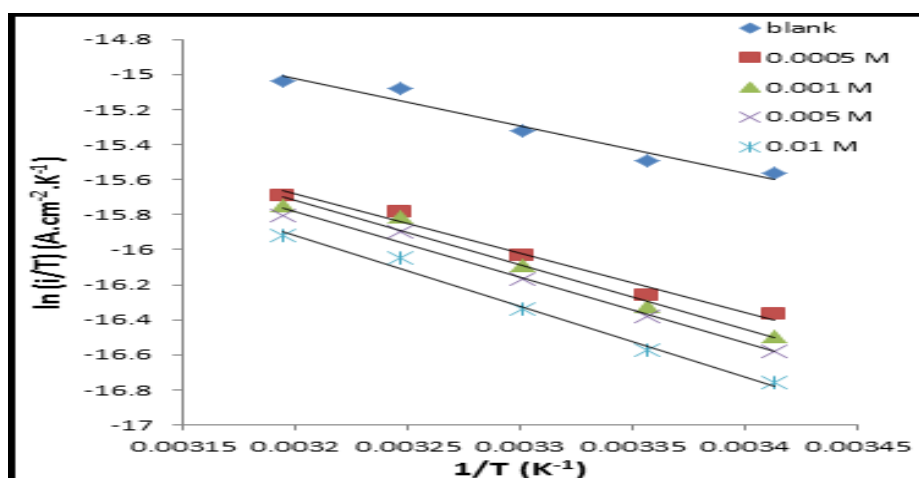


Fig.-4: Plots of $\ln (i_{corr}/T)$ versus $1/T$ for carbon steel in 3.5% NaCl at pH 11 solution for blank and inhibited solution containing various L-tryptophan concentrations.

Adsorption Isotherm

The interaction type between carbon steel surface and L-tryptophan molecules were explained by using Langmuir isotherm as expressed by the following linear equation¹⁴:

$$\left(\frac{C_{inh}}{\theta}\right) = \frac{1}{K_{ads}} + C_{inh} \quad (6)$$

C_{inh} is the L-tryptophan concentration and K_{ads} is the adsorption-desorption equilibrium constant. Values of C_{inh}/θ for L-tryptophan in the saline solution were plotted against the corresponding values of the inhibitor concentration (C_{inh}) as shown in Fig.-5 and the relationships obtained were linear. The intercept of each plot on C_{inh}/θ axis is equal to $1/K_{ads}$.

Table-3: Activation energy (E_a), enthalpy (ΔH_a), and entropy (ΔS_a) of activation for the corrosion of carbon steel in 3.5% NaCl aqueous solution at pH 11 in the absence and presence of different concentration of L-tryptophan at (293, 298, 303, 308 and 313) K.

Con. [M]	E_a [kJ.mol ⁻¹]	$A \times 10^{24}$ [molecule.cm ⁻² .s ⁻¹]	ΔH_a [kJ.mol ⁻¹]	$-\Delta S_a$ [J.K ⁻¹ .mol ⁻¹]
-	24.86	0.801	22.34	250.98
5×10^{-4}	30.63	3.82	28.10	237.98
1×10^{-3}	33.11	9.60	30.59	230.32

5×10^{-3}	33.74	11.4	31.21	228.88
1×10^{-2}	36.15	25.4	33.63	222.25

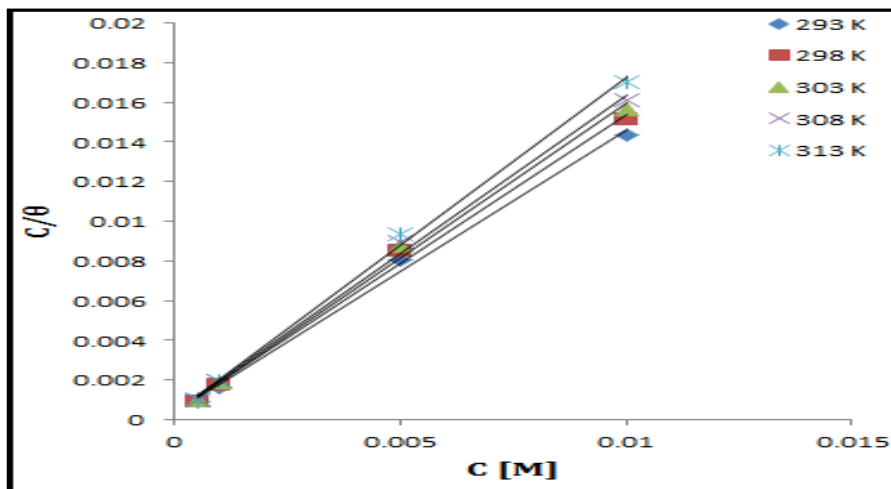


Fig.-6: Langmuir isotherm plots for the adsorption of L-tryptophan on the surface of carbon steel.

Using K_{ads} values we can calculate the values of ΔG°_{ads} through the following equation:

$$\Delta G^{\circ}_{ads} = -RT \ln(55.5K_{ads}) \quad (7)$$

ΔG°_{ads} are adsorption Gibbs free energy of L-tryptophan at Temperature. 55.5 is the concentration of water in solution in mol/L. Values of K_{ads} and are tabulated in Table-4. The negative sign of ΔG°_{ads} values indicated that the adsorption of inhibitors to form a stable adsorbed protective layer on carbon steel was spontaneous. ΔG°_{ads} values are ranging from $-31.02 \text{ kJ mol}^{-1}$ to $-29.07 \text{ kJ.mol}^{-1}$ for L-tryptophan at the temperature range 293 K to 313 K. The values of ΔG°_{ads} ranging between -40^1 to -20 kJ.mol^{-1} meaning that the adsorption of these molecules involves chemisorptions as well as physisorption^{15,16}. The molecular structure of the amino acid investigates indicates that they have strong potential to become an effective corrosion inhibitor due to the presence of nitrogen atom that is a derivative of indole. The results show also, that L-tryptophan adsorbed on the surface of carbon steel and fill the empty orbital of it through the indole ring with π -electrons, nitrogen and oxygen atoms and with the lone pair electrons in its molecule⁷. Adsorption Gibbs free energy change (ΔG°_{ads}) data were plotted versus temperature (T) in order to evaluate enthalpy change (ΔH°_{ads}) and entropy change (ΔS°_{ads}) of adsorption process according to the following equation:

$$\Delta G^{\circ}_{ads} = \Delta H^{\circ}_{ads} - T \Delta S^{\circ}_{ads} \quad (8)$$

Where, ΔH°_{ads} and ΔS°_{ads} are respectively, the slope and intercept of linear relation as indicated in Fig.-7, and the data obtained are given in Table-4. The negative value of (ΔH°_{ads}) shows that the adsorption process of L-tryptophan on the carbon steel surface is exothermic in nature. The value of the entropy change of activation (ΔS°_{ads}) is positive, this indicates an increase in randomness during the adsorption process¹⁷.

Computational Study

Quantum chemical calculations of L-tryptophan molecule were achieved by using DFT that based on the following relations¹⁸:

$$X = \frac{(I+A)}{2} \quad (9)$$

$$Y = \frac{(I-A)}{2} \quad (10)$$

Where χ is absolute electronegativity and γ is global hardness. According to Koopman's theorem, the E_{HOMO} and E_{LUMO} of the inhibitor molecule are represent the ionization potential (I), and the electron affinity (A), respectively, according to the following relations¹⁸:

$$I = -E_{\text{HOMO}}, \quad A = -E_{\text{LUMO}}$$

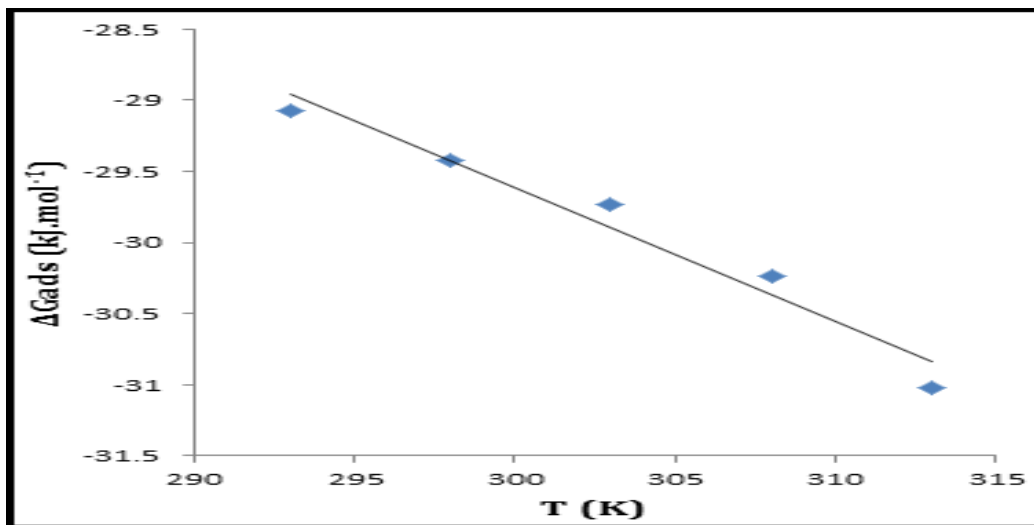


Fig.-7: The changing of Gibbs free energies (ΔG_{ads}) with temperature.

The fraction of electrons transferred (ΔN) was then calculated from this relation¹⁹:

$$\Delta N = \frac{(\chi_{\text{Fe}} - \chi_{\text{inh}})}{2(\eta_{\text{Fe}} + \eta_{\text{inh}})} \quad (11)$$

Where $\chi_{\text{Fe}} \approx 7.0$ eV is a theoretical value of iron and $\eta_{\text{Fe}} = 0$ is taken assuming that $I = A$ for bulk metals. The higher value of ΔN indicates higher inhibition efficiency of the inhibitor (L-tryptophan) and the higher value of dipole moment indicates that L-Tryptophan is aromatic compound can donate electrons to form strong bond¹⁹. literature survey shows that the adsorption of the inhibitor on the metal surface basis of the donor-acceptor interactions between the π -electrons of the heterocyclic compound and the empty d-orbital of the metal surface atoms. The high value of (E_{HOMO}) of the molecule indications its tendency to donate electrons to the acceptor molecules at low energy empty molecular orbitals and enhance the inhibition efficiency. The lower E_{LUMO} values refer to its ability to accept electrons. When value ΔE_{gap} decreases, the reactivity of L-Tryptophan inhibitor increases, as a result increase the inhibition efficiency of it²⁰. Values of molecular parameters in gas and aqueous phases were listed in Table-5.

Table-4: Thermodynamic functions for L-tryptophan adsorption on the surface of carbon steel alloy in 3.5% NaCl solution at pH 11.

T [K]	K_{ads} [M^{-1}]	$-\Delta G^{\circ}_{\text{ads}}$ [kJ.mol^{-1}]	$-\Delta H^{\circ}_{\text{ads}}$ [kJ.mol^{-1}]	$\Delta S^{\circ}_{\text{ads}}$ [$\text{J.K}^{-1}.\text{mol}^{-1}$]
293	2739.73	29.07	1.293	94.4
298	2583.98	29.42		
303	2409.64	29.73		
308	2427.18	30.24		
313	2710.03	31.02		

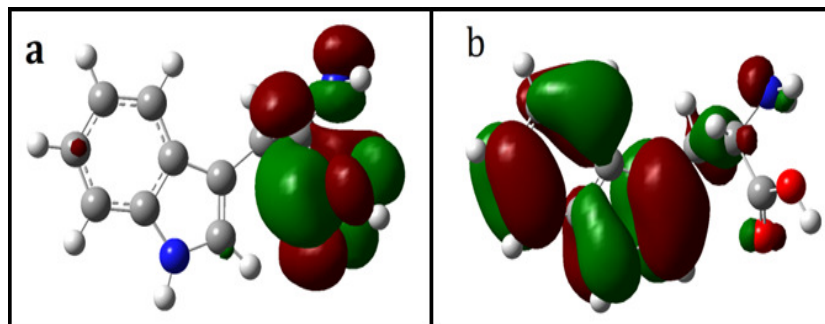


Fig.-8: (a) the energy of lowest unoccupied molecular orbital (E_{LUMO}) and (b) the energy of highest occupied molecular orbital (E_{HOMO}) of L-tryptophan.

Table-5: Quantum parameters of L-tryptophan in gas and aqueous phases

Terms	gas phase	aqueous phase
$-E_{LUMO}$ (eV)	0.2969	0.0174
$-E_{HOMO}$ (eV)	5.5523	5.2642
ΔE (eV)	5.2555	5.2467
μ (Debye)	3.6237	3.0281
$-E_{total} \times 10^5$ (kcal.mol ⁻¹)	4.2833	4.2832
χ (eV)	2.9246	2.6408
γ (eV)	2.6277	2.6234
ΔN (eV)	0.7755	0.8308

CONCLUSION

L-tryptophan was considered to be good eco-friendly inhibitor for carbon steel corrosion in alkaline saline solution with maximum inhibition efficiency of 71.72 % at 10^{-2} mol. L⁻¹ inhibitor concentration and temperature of 293 K. Potentiostatic studies revealed that L-tryptophan acted as anodic type inhibitor; also the corrosion process of carbon steel in studied environment was kinetically controlled. Furthermore, the kinetic data indicated that the corrosion of steel was both easier and faster in the absence of L-tryptophan as compared with corrosion in presence of it. On the other hand, L-tryptophan inhibited the corrosion of carbon steel through physical adsorption. The inhibiting function was permitted on the thermodynamic grounds of the adsorption process as indicated by the negative ΔG°_{ads} values. Finally, a correlation has been obtained between experimental inhibition efficiency and some quantum chemical parameters studied namely: E_{HOMO} , E_{LUMO} , μ , E_{total} , χ , γ and ΔN for both gas and aqueous phases.

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