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Der Pharmacia Lettre, 2016, 8 (18):158-166
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Investigation of Quinoline Derivatives as Corrosion Inhibitors for Mild Steel in HCl 1.0 M

H. Lgaz^{1,2}, M. Saadouni³, R. Salghi^{2*}, S. Jodeh⁴, M. Elfaydy⁵, B. Lakhrissi⁵, S. Boukhris³ and H. Oudda¹

¹Laboratory of separation methods, Faculty of Science, University Ibn Tofail PO Box 242, Kenitra, Morocco

²Laboratory of Applied Chemistry and Environment, ENSA, Université Ibn Zohr, PO Box 1136, 80000 Agadir, Morocco

³Laboratory of Organic, Organometallic and Theoretical Chemistry, Faculty of Science, Ibn Tofail University, 14000 Kenitra, Morocco

⁴Department of Chemistry, An-Najah National University, P. O. Box 7, Nablus, Palestine

⁵Laboratoire d'Agroressources, Polymères et Génie des Procédés, University Ibn Tofail PO Box 242, Kenitra, Morocco

ABSTRACT

The influence of three quinoline derivatives (**8QNs**) namely, ethyl 2-(((8-hydroxyquinolin-5-yl)methyl)amino)acetate (**8QNI**), 5-((benzylamino)methyl)quinolin-8-ol (**8QN2**) and 5-(azidomethyl)quinolin-8-ol (**8QN3**) on the mild steel corrosion in 1 M HCl was studied by weight loss and electrochemical methods. Results showed that **8QN3** shows maximum inhibition efficiency of 90% at 5×10^{-3} M concentration. Polarization study revealed that the **8QNs** act as mixed type inhibitors. EIS measurements showed that the studied compounds inhibit mild steel corrosion by adsorbing on the steel surface. Results showed that inhibition efficiency increases with concentration. Adsorption of **8QNs** on the mild steel surface obeyed the Langmuir adsorption isotherm.

Keywords: Mild steel, Corrosion, HCl, Quinoline derivatives, EIS, Polarization, Thermodynamic study.

INTRODUCTION

Corrosion is defined as degradation of metals as a result of chemical reaction with the surrounding environment. Corrosion causes heavy economic losses. In the United States, the economic losses reported in 1998 were \$276 billion per year which is now exceeds to \$1 trillion dollars a year. Mild steel is widely used metal in industries because of its high strength, ease of fabrication and cost-effectiveness[1–6]. However it suffers from corrosion during acid cleaning, pickling, and descaling. Hydrochloric acid solution is used to enhance oil recovery during acidization[7–14]. The damage by corrosion generates not only high cost for renovation, replacement of various equipments, but in addition these constitute a public risk. Thus it is necessary to develop some effective corrosion inhibitors. The emphasis on the selection of corrosion inhibitor is based on the electron rich functional groups along with π -electrons inside their frameworks. Earlier various organic compounds have been reported as corrosion inhibitors[11–19]. Generally, organic inhibitors inhibit metallic corrosion by adsorbing on the surface and thereby forming a protective barrier between metal and electrolyte (1 M HCl)[20–23]. The adsorption of these inhibitors on metallic surface are influenced by several factors such as molecular size of inhibitor, nature of substituents, nature of metal and electrolyte[24–28]. Organic compounds containing heteroatoms including nitrogen, sulfur, and/or oxygen with polar functional groups and conjugated double bonds have been reported as effective corrosion inhibitor[29–32].

The compounds with quinolone functions are rarely investigated such as corrosion inhibitors[33,34]. Therefore, it is necessary to study the corrosion inhibition effect and inhibition mechanism of the quinoline derivatives which could be deemed as good potential inhibitors.

In view of this, three synthesized quinolone derivatives namely ethyl 2-(((8-hydroxyquinolin-5-yl)methyl)amino)acetate (**8QN1**), 5-((benzylamino)methyl)quinolin-8-ol (**8QN2**) and 5-(azidomethyl)quinolin-8-ol (**8QN3**) were synthesized to study mild steel corrosion inhibition in 1 M HCl using weight loss, electrochemical impedance spectroscopy (EIS) and potentiodynamic polarization.

MATERIALS AND METHODS

Materials

The steel used in this study is a mild steel (Euronorm: C35E carbon steel and US specification: SAE 1035) with a chemical composition (in wt%) of 0.370 % C, 0.230 % Si, 0.680 % Mn, 0.016 % S, 0.077 % Cr, 0.011 % Ti, 0.059 % Ni, 0.009 % Co, 0.160 % Cu and the remainder iron (Fe). The mild steel samples were pre-treated prior to the experiments by grinding with emery paper SiC (120, 600 and 1200); rinsed with distilled water, degreased in acetone in an ultrasonic bath immersion for 5 min, washed again with bidistilled water and then dried at room temperature before use.

Solutions

The aggressive solutions of 1.0 M HCl was prepared by dilution of analytical grade 37% HCl with distilled water. The concentration range of quinoline derivatives used was 1×10^{-5} M to 5×10^{-3} M.

Corrosion tests

Weight loss

Gravimetric measurements were carried out at definite time interval of 6 h at room temperature using an analytical balance (precision ± 0.1 mg). The mild steel specimens used have a rectangular form (length = 2 cm, width = 2 cm, thickness = 0.08 cm). Gravimetric experiments were carried out in a double glass cell equipped with a thermostated cooling condenser containing 80 mL of test solution. After immersion period, the steel specimens were withdrawn, carefully rinsed with bidistilled water, ultrasonic cleaning in acetone, dried at room temperature and then weighed.

Electrochemical impedance spectroscopy

The electrochemical measurements were carried out using Volta lab (Tacussel- Radiometer PGZ 100) potentiostat and controlled by Tacussel corrosion analysis software model (Volta master 4) at under static condition. The corrosion cell used had three electrodes. The reference electrode was a saturated calomel electrode (SCE). A platinum electrode was used as auxiliary electrode of surface area of 1 cm^2 . The working electrode was mild steel. All potentials given in this study were referred to this reference electrode. The working electrode was immersed in test solution for 30 minutes to establish steady state open circuit potential (E_{ocp}). After measuring the E_{ocp} , the electrochemical measurements were performed. All electrochemical tests have been performed at 303 K. The EIS experiments were conducted in the frequency range with high limit of 100 kHz and different low limit 10 mHz at open circuit potential, with 10 points per decade, at the rest potential, after 30 min of acid immersion, by applying 10 mV ac voltage peak-to-peak. Nyquist plots were made from these experiments.

Potentiodynamic polarization

The electrochemical behaviour of mild steel sample in inhibited and uninhibited solution was studied by recording anodic and cathodic potentiodynamic polarization curves. Measurements were performed in the 1.0 M HCl solution containing different concentrations of the tested inhibitors by changing the electrode potential automatically from -800 to -200 mV versus corrosion potential at a scan rate of $2 \text{ mV} \cdot \text{s}^{-1}$. The linear Tafel segments of anodic and cathodic curves were extrapolated to corrosion potential to obtain corrosion current densities (i_{corr}).

Inhibitors

In continuation of our research for developing corrosion inhibitors with high effectiveness and efficiency, the present paper explores a systematic study to ascertain the inhibitive action of synthesized quinoline derivatives, on corrosion of mild steel in 1.0 M HCl solution by using weight loss measurement, potentiodynamic polarization and AC impedance. Figure 1 shows the molecular structure of the quinoline derivatives utilised in this investigation. The detail of synthesis methods referenced from the published article[6,23]

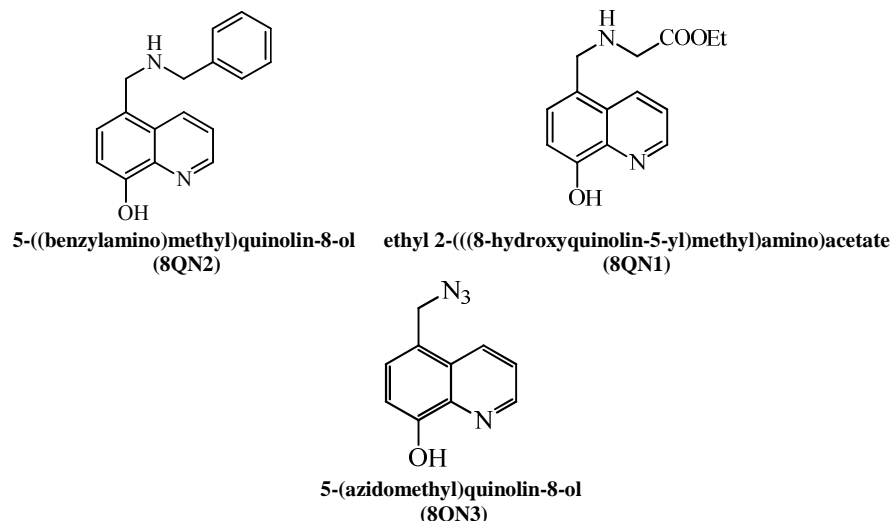


Figure 1: Chemicals structures of quinoline derivatives

RESULTS AND DISCUSSION

Effect of concentration inhibitor

Weight loss

The results obtained from weight loss study are given in Table 1. It can be seen from the results that on increasing the concentration of inhibitors, inhibition efficiency increases from 78 to 90% and shows the following order of inhibition **8QN3** > **8QN2** > **8QN1**. The corrosion rate, C_R ($\text{mg}/\text{cm}^2 \times \text{h}$), surface coverage (θ) and inhibition efficiency η_w (%) of each concentration were calculated using the following equations[1]:

$$W = \frac{\Delta m}{St} \quad (1)$$

$$\eta_w = \left(\frac{W_{\text{uninh}} - W_{\text{inh}}}{W_{\text{uninh}}} \right) \times 100 \quad (2)$$

Where Δm is the average weight loss (mg), S is the surface area of specimens (cm^2), and t is the immersion time (h), W_{uninh} and W_{inh} are corrosion rates in the absence and presence of inhibitor, respectively.

Table 1. Effect of 8QNs concentrations on corrosion data of mild steel in 1.0 M HCl

Composé	Concentration (mol/L)	W ($\text{mg}/\text{cm}^2 \times \text{h}$)	E_w (%)
HCl	1	1.1350	-
8QN3	5×10^{-3}	0.1135	90
	1×10^{-3}	0.1589	86
	1×10^{-4}	0.2270	80
	1×10^{-5}	0.2497	78
8QN2	5×10^{-3}	0.1362	88
	1×10^{-3}	0.1929	83
	1×10^{-4}	0.2837	75
	1×10^{-5}	0.3518	69
8QN1	5×10^{-3}	0.1589	86
	1×10^{-3}	0.2270	80
	1×10^{-4}	0.3178	72
	1×10^{-5}	0.4426	61

Corrosion inhibition performance of a compound depends upon its ability to get adsorbed on metal surfaces. In our investigation, we have taken quinoline derivatives containing π electrons and heteroatoms (N, O) by which they can easily get adsorbed onto the metal surface and form a protective layer thereby preventing the corrosion[24,25].

Electrochemical impedance spectroscopy

Fig. 2 shows the impedance responses of mild steel in the absence and presence of quinoline derivatives. The impedance parameters calculated from these plots are given in Table 2. The Nyquist plots exhibit one capacitive loop in the absence and presence of inhibitors suggesting that corrosion of mild steel was charge transfer controlled[10]. The diameter of the capacitive loops increases with increasing concentrations of inhibitors, which suggests that all the three compounds act as effective corrosion inhibitors for mild steel and show the following order of inhibition **8QN3 > 8QN2 > 8QN1**[16]. The increased diameter of the Nyquist plots in the presence of QNs suggested also that values of charged transfer resistance (R_{ct}) increase due to formation of protective film[11,14]. The electrochemical parameters, including R_{ct} , Q and n , obtained from fitting the recorded EIS data using the electrical circuit of Figure 3 are listed in Table 2. The impedance of the CPE is expressed as follows[17].

$$Z_{CPE} = \frac{1}{Q(j\omega)^n} \quad (3)$$

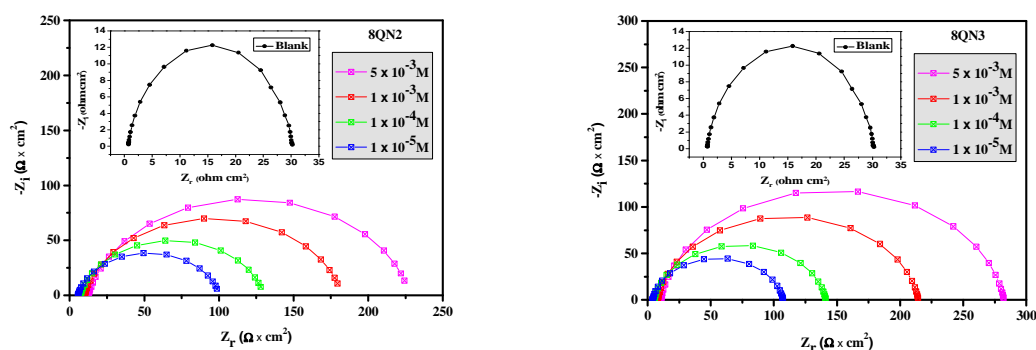
Where Q is the CPE constant, n is the phase shift which can be explained as a degree of surface inhomogeneity, j is the imaginary unit and ω is the angular frequency. Depending on the values of n , CPE can represent resistance ($n=0$), capacitance ($n=1$), inductance ($n= -1$) and Warburg impedance ($n=0.5$). The values of the interfacial capacitance C_{dl} can be calculated from CPE parameter values Q and n using the expression[30]:

$$C_{dl} = (Q \times R^{1-n})^{1/n} \quad (4)$$

The R_{ct} values were used to calculate the inhibition efficiency, $\eta_{EIS}(\%)$, (listed in Table 2), using the following equation:

$$\eta_{EIS} \% = \frac{R_{ct}^i - R_{ct}^o}{R_{ct}^i} \times 100 \quad (5)$$

Where R_{ct}^o and R_{ct}^i are the charge transfer resistance in absence and in presence of inhibitor, respectively.



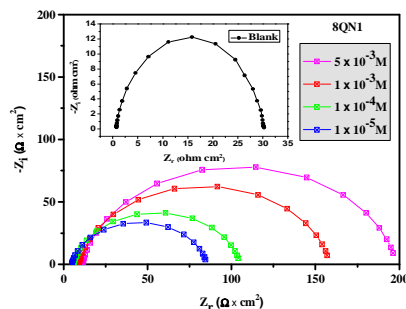


Figure 2: Nyquist diagrams of mild steel with different concentrations of 8QNs at 303K



Figure 3: Equivalent electrical circuit model

Inspection of the Table 2 reveals that values of R_{ct} increase with increasing 8QNs concentration suggesting that extent of surface coverage and $\eta\%$ increases with inhibitor concentration. The values of C_{dl} decreased (Table 2) with the addition of 8QNs, which is probably due to the decrease in the local dielectric constant and/or an increase in the thickness of the electrical double layer, suggesting that the 8QNs strongly adsorbed over the mild steel surface[5,7,11].

Table 2. Electrochemical impedance parameters for corrosion of mild steel in acid medium at various contents of 8QNs at 303 K

Inhibitor	Concentration (M)	R_{tc} ($\Omega \times cm^2$)	n	$Q \times 10^{-4}$ ($S^n \Omega^{-1} cm^{-2}$)	C_{dc} ($\mu F/cm^2$)	E_R (%)	θ
Blank	1.0	29.35	0.89	1.7610	91.86	-	-
8QN3	5×10^{-3}	271	0.88	0.2998	15.55	89	0.89
	1×10^{-3}	206	0.87	0.4003	19.54	85	0.85
	1×10^{-4}	135	0.86	0.7135	33.51	78	0.78
	1×10^{-5}	103	0.83	1.0701	42.51	71	0.71
8QN2	5×10^{-3}	215	0.83	0.4419	17.02	86	0.86
	1×10^{-3}	172	0.84	0.5502	22.64	82	0.82
	1×10^{-4}	122	0.84	0.8989	38.05	75	0.75
	1×10^{-5}	95	0.81	1.1743	40.91	69	0.69
8QN1	5×10^{-3}	187	0.86	0.5272	24.85	84	0.84
	1×10^{-3}	150	0.82	0.7865	29.67	80	0.80
	1×10^{-4}	99	0.81	1.1687	41.06	70	0.70
	1×10^{-5}	81	0.83	1.3321	52.68	63	0.63

Polarization curves

The polarization curves for mild steel in the absence and presence of inhibitors of different concentrations are shown in Figure 4. The values obtained from polarization curves such as corrosion current densities (i_{corr}), corrosion potential (E_{corr}), and Tafel slope (β_c) are listed in Table 2. The i_{corr} values were used to calculate the inhibition efficiency, $\eta_{PDP}(\%)$, (listed in Table 3), using the following equation[16]:

$$\eta_{PDP} \% = \frac{I_{corr} - I_{corr(i)}}{I_{corr}} \times 100 \quad (6)$$

Where, I_{corr} and $I_{corr(i)}$ are the corrosion current density in absence and presence of inhibitor, respectively.

The obtained results show that inhibition efficiency increases with decreasing values of i_{corr} and the highest inhibition efficiency 90% is obtained for 8QN3 at 5×10^{-3} M.

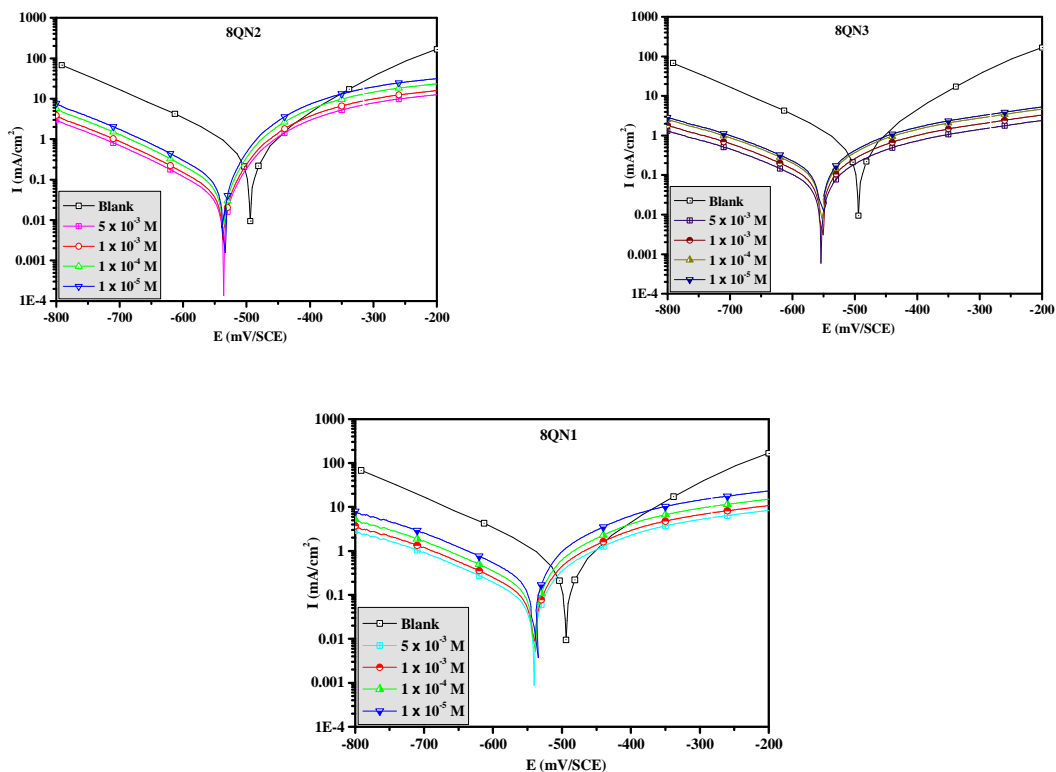


Figure 4. Potentiodynamic polarization curves of mild steel in 1.0 M HCl in the presence of different concentrations of 8QNs at 303 K

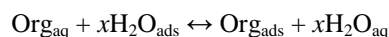
From Figure 4 it can be seen that the addition of an inhibitor modifies both cathodic and anodic polarization branches and shifts the E_{corr} towards the negative direction as compared to an inhibitor-free solution. As previously reported in the presence of inhibitor if E_{corr} shifts more than 85 mV with respect to E_{corr} in uninhibited solution, the inhibitor can be considered as cathodic or anodic type otherwise it is of mixed type[35,36]. The E_{corr} values of 8QN1, 8QN2, and 8QN3 suggested that these compounds show the mixed type of inhibition with E_{corr} shift in the negative direction which is an indication of cathodic predominance[37]. The negative shift of E_{corr} indicates more adsorption of the inhibitor on the cathodic sites and predominantly controls cathodic reactions[38]. The presence of all the three inhibitors causes a decrease in i_{corr} values in all concentrations.

Table 3. Electrochemical parameters of mild steel at various concentrations of 8QNs in 1.0 M HCl and corresponding inhibition efficiency

Inhibitor	Concentration (M)	$-E_{\text{corr}}$ (mV/SCE)	$-\beta_c$ (mV dec ⁻¹)	i_{corr} ($\mu\text{A cm}^{-2}$)	E_1 (%)	θ
HCl	1.0	496	150.19	564	-	-
8QN3	5×10^{-3}	538	170	56	90	0.9
	1×10^{-3}	539	167	79	86	0.86
	1×10^{-4}	536	171	111	80	0.80
	1×10^{-5}	537	168	145	74	0.74
8QN2	5×10^{-3}	538	152	62	89	0.89
	1×10^{-3}	539	155	92	83	0.83
	1×10^{-4}	536	152	128	77	0.77
	1×10^{-5}	537	154	155	72	0.72
8QN1	5×10^{-3}	543	159	85	0.85	0.85
	1×10^{-3}	542	157	82	0.82	0.82
	1×10^{-4}	541	153	73	0.73	0.73
	1×10^{-5}	539	158	60	0.60	0.60

Adsorption isotherm and standard adsorption free energy

The adsorption isotherm can be determined by assuming that the inhibition effect was mainly due to adsorption at the metal/solution interface. The adsorption process depends upon the electronic characteristics of the 8QNs, the nature of the metal surface, the temperature, steric effects and the varying degrees of the surface-site activity. A typical adsorption process involves the replacement of the adsorbed water molecule (H_2O_{ads}) by inhibitor molecules present in aqueous solution (Org_{aq}) at metal/electrolyte interface as represented below[39]:



where x is the number of water molecules replaced by one molecule of organic inhibitors. An attempt was made to plot the values of surface coverage (θ) derived from weight loss experiment against different 8QNs concentrations in order to obtain the best adsorption isotherm. Several adsorption isotherms such as Langmuir, Temkin and Freundlich isotherms were tested. In our present study the Langmuir isotherm gave the best fit which can be best represented by following equation[40]:

$$\frac{C_{inh}}{\theta} = \frac{1}{K_{ads}} + C_{inh} \quad (7)$$

Where C_{inh} is the concentration of inhibitor and K_{ads} the adsorptive equilibrium constant.

Figure 5 shows the curves of the variation of C_{inh} / θ according to the concentration C_{inh} for the quinoline compounds. The linearity of these curves indicates that the adsorption of our inhibitors on the surface of mild steel in 1 M HCl, is according to the Langmuir isotherm model. The validity of this approach is confirmed by the strong correlation ($R^2 = 0.999$).

The values of K_{ads} obtained from the reciprocal of intercept of Langmuir isotherm line are listed in Table 6, together with the values of the Gibbs free energy of adsorption ΔG_{ads}° calculated from the equation:

$$K_{ads} = \left(\frac{1}{55.5}\right) \exp\left(-\frac{\Delta G_{ads}^\circ}{RT}\right) \quad (8)$$

Where R is gas constant and T is absolute temperature of experiment and the constant value of 55.5 is the concentration of water in solution.

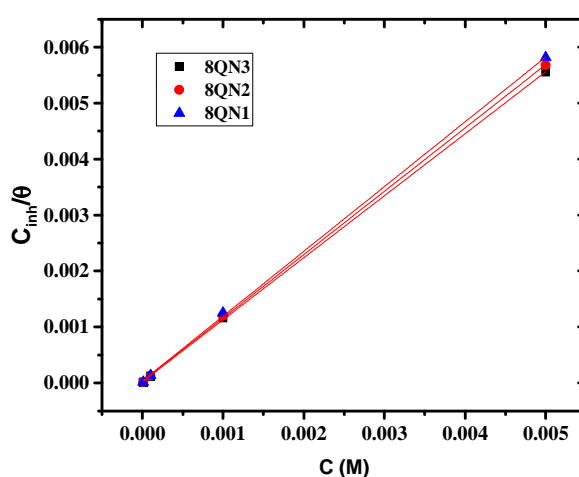


Figure 5. Adsorption isotherm according to Langmuir's model derived from weight loss measurement

Table 4. Thermodynamic parameters for the adsorption of 8QNs in 1 M HCl on the carbon steel at 303 K

Inhibitors	R^2	K_{ads} (L/mol)	ΔG_{ads} (kJ/mol)
8QN3	0.99994	46794	-37
8QN2	0.99990	34319	-36
8QN1	0.99992	27413	-35

Generally, a higher value of K_{ads} associated with higher tendency to adsorb on mild steel surface. In our present study the K_{ads} value of different 8QNs follows the order: 8QN3 > 8QN2 > 8QN1 which is in accordance with the order of the $\eta\%$. Literature study reveals that the value of ΔG_{ads} up to -20 kJ/mol or less negative is related to the electrostatic interactions between inhibitor and metallic surfaces (physisorption), while the value of ΔG_{ads} is around -40 kJ/mol or more negative related to the charge sharing between inhibitor and metallic surfaces (chemisorption)[41]. In our present investigation values of ΔG_{ads} vary from -37 to -35 kJ/mol (Table 4) that signifying the both physical and chemical *i.e.* physiochemisorption of 8QNs[16,25,41]. The negative sign of ΔG_{ads} ensures the spontaneity of the adsorption process[41].

CONCLUSION

Results show that 8QNs act as good corrosion inhibitors for mild steel in 1 M HCl and their inhibition efficiency increases with increasing concentration. The negative value of ΔG_{ads} suggests that 8QNs adsorb spontaneously and their adsorption obeys the Langmuir adsorption isotherm. The polarization study revealed that the 8QNs act as mixed type inhibitors with cathodic predominance. The presence of the 8s increases the charge transfer values and therefore inhibits mild steel corrosion.

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