



# Hindrance of the erosion of mild steel in HCl by sparfloxacin

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## Abstract

Inhibitive and adsorption properties of sparfloxacin for the corrosion of mild steel in HCl have been studied using gravimetric, gasometric and thermometric methods of monitoring corrosion. Sparfloxacin is found to inhibit the corrosion of mild steel in HCl. At 303 K, inhibition efficiency obtained for sparfloxacin from gravimetric, gasometric and thermometric methods were found to range from 79.68 to 97.47%, 86.67 to 96.67% and 76.34 to 82.30% respectively. Inhibition efficiency of sparfloxacin increased with increase in concentration of sparfloxacin but decreased with increase in temperature. Activation energies for the inhibited reactions were higher than the value obtained for the blank. Sparfloxacin is found to be an adsorption inhibitor for the corrosion of mild steel. The adsorption of sparfloxacin on the surface of mild steel was found to be spontaneous, exothermic and its adsorption characteristics were best described by Langmuir adsorption isotherm. The use of sparfloxacin as an inhibitor for the corrosion of mild steel in HCl is advocated in this study.

**Keyword:** Corrosion of mild steel, HCl, inhibition, sparfloxacin.

## INTRODUCTION

Due to its mechanical properties, mild steel is used in the construction of numerous industrial equipments and appliances (Herrag et al., 2008; Krim et al., 2008; Saddiqi and Chaubey, 2008). However, this metal is prone to corrosion attack (Eddy et al., 2008a-c). The corrosion of mild steel is severe in the presence of an aggressive medium such as acid, basic and salty solutions. Therefore industrial processes such as acid cleaning, etching, and pickling of metal in which acidic solution are made to come in contact with the metal require the use of an inhibitor. Several inhibitors have been synthesized and used for the inhibition of the corrosion of mild steel in acidic and basic medium. Most effective inhibitors are organic compounds having hetero atom in their aromatic or long carbon chain systems. For these compounds, the presence of hetero atoms enhanced the electron donating ability of the inhibitor to the metal (Maliki Alaoui et al., 2008). However, the short coming of these inhibitors is that they are not environmentally friendly.

Therefore there is need to search for green corrosion inhibitors. According to Eddy and Ebenso (2008), green

corrosion inhibitors are biodegradable and do not contain heavy metals or other toxic compound. The successful uses of naturally occurring substances to inhibit the corrosion of metals in acidic and alkaline environment have been reported by some research groups (Abdallah, 2004b; El Etre and Tantawy, 2006; Oguzie, 2007; Okafor and Ebenso, 2007; Umoren et al., 2008). Increasing efforts is also been made in researching the possibility of using some drugs as green corrosion inhibitors. For example, Abdallah (2004b, 2002) used some antibiotics to inhibit the corrosion of Al in HCl, Odoemelam and Eddy (2008a,b) used sparfloxacin and norfloxacin to inhibit the corrosion of zinc in H<sub>2</sub>SO<sub>4</sub>, Eddy (2008) used some antibiotics to inhibit the corrosion of mild steel in acidic medium, Eddy and Ekop (2008) used some microlides to inhibit the corrosion of mild steel in H<sub>2</sub>SO<sub>4</sub>.

In all these studies, the inhibition efficiencies of these drugs were not less than 90%. According to Eddy (2008), most drugs that are suitable for used as corrosion inhibitors are heterocyclic in nature and the use of antibiotics has an added advantage in corrosive environments where micro-

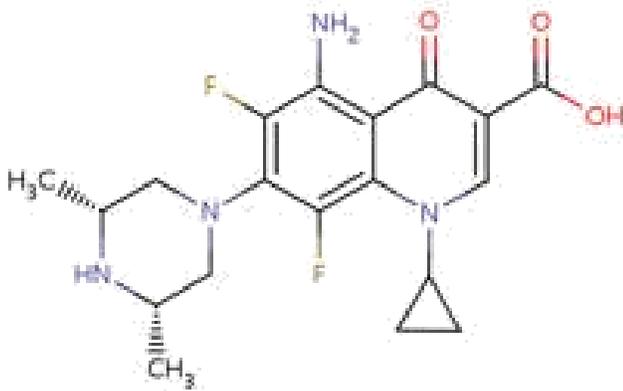


Figure 1. The chemical structure of sparfloxacin.

organism is the dominant cause. However, through a search of literature and other related database the use of sparfloxacin as an inhibitor for the corrosion of mild steel in HCl has not been reported elsewhere. The present study is aimed at investigating inhibitive and adsorption properties of spar-floxacin for the corrosion of mild steel in HCl.

Sparfloxacin is a synthetic fluoroquinolone broad-spectrum antimicrobial agent. Sparfloxacin has *in vitro* activity against a wide range of gram-negative and gram-positive microorganisms. Sparfloxacin exerts its anti-bacterial activity by inhibiting DNA gyrase, a bacterial topoisomerase. DNA gyrase is an essential enzyme which controls DNA topology and assists in DNA replication, repair, deactivation, and transcription. According to Neal (2002), the physical properties of sparfloxacin include the following, Average molecular weight, 392.3998; Physical state, solid; Experimental water solubility, practically insoluble; Predicted water solubility,  $1.13 \times 10^{-1}$  mg/ml. Experimental logP/hydrophobicity, 2.5; Predicted logP, -0.06. The chemical structure of sparfloxacin is as shown in Figure 1.

## EXPERIMENTAL

Materials used for the study were mild steel sheets of dimension 5 x 4 x 0.11 cm and the chemical composition was (wt %) Mn (0.6), P(0.36), C(0.15) and Si(0.03). Each coupon was degreased in ethanol, dried in acetone and preserved in a desiccator before use. The inhibitor was supplied by LIVEMORE pharmaceutical company, Ikot Ekpene, Nigeria. All reagents used in this study were analar grade. Double distilled water was used for the preparation of all solutions. Concentrations of HCl used for gravimetric, gasometric and thermometric studies were 0.1, 2.0 and 2.0 M respectively while the concentrations of the inhibitor were  $3 \times 10^{-4}$ ,  $5 \times 10^{-4}$ ,  $8 \times 10^{-4}$ ,  $10 \times 10^{-4}$  and  $12 \times 10^{-4}$  M. Each of these concentrations was dissolved in the appropriate concentration of HCl.

### Gasometric method

Hydrogen evolution measurements were carried out at 303 and 333K as described in literature (Eddy and Ekop, 2008). From the

volume of hydrogen evolved per minutes, inhibition efficiency ( $\eta$ ), and degree of surface coverage ( $\theta$ ) were calculated using Equation 1 and 2 respectively.

$$\%I = \frac{\{1 - V'_{Ht}\}}{V^0_{Ht}} \times 100 \quad 1$$

$$\theta = \%I/100 \quad 2$$

where  $V'_{Ht}$  is the volume of hydrogen evolved at time t for inhibited solution and  $V^0_{Ht}$  is the volume of hydrogen evolved at time t for uninhibited solution.

### Thermometric method

The thermometric tests were carried out as reported elsewhere (Eddy and Ebenso, 2008). From the rise in temperature of the system per minutes, the reaction number (RN) and inhibition efficiency were calculated using equations 3 and 4 respectively:

$$RN (\text{°C minutes}^{-1}) = \frac{T_m - T_i}{t} \quad 3$$

$$\%I = \frac{RN^0 - RN'}{RN'} \times 100 \quad 4$$

where  $T_m$  is the maximum temperature attained by the system,  $T_i$  is the initial temperature, t is the time.  $RN^0$  and  $RN'$  are the reaction numbers for the blank and inhibited solutions respectively.

## RESULTS AND DISCUSSIONS

The corrosion of mild steel predominantly affects the iron content of the metal. Corrosion is an electrochemical reaction indicating that there exist anodic and cathodic reactions (Atkin, 2002). During the corrosion of mild steel, the anodic reaction involves the formation of  $Fe^{3+}$  while the cathodic reaction involved the formation of hydroxyl ion or evolution of hydrogen gas. At the anode, the following reactions are likely,



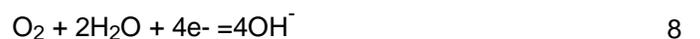
Also the main reactions that occur at cathode are as follows,

i. Hydrogen evolution (in acidic environment)



ii. If oxygen ions are present, oxygen reduction

iii. takes place in alkaline or neutral medium.



From the above, the corrosion of mild steel in HCl can be

**Table 1.** Corrosion rates of mild steel in HCl containing various concentrations of sparfloxacin.

Con. x 10 <sup>-4</sup> (M)	Weight loss	Gasometric	
	CR x 10 <sup>-5</sup> (gcm <sup>-3</sup> h <sup>-1</sup> ) at 303 K	CR (cm <sup>3</sup> /min) at 303 K	CR(cm <sup>3</sup> /min) at 333K
Blank	92.90	0.5000	2.7625
3	18.87	0.0667	1.8250
5	18.90	0.0417	1.3250
8	6.82	0.0333	1.3375
10	4.97	0.0250	1.1500
12	2.35	0.0167	1.1125

**Table 2.** Inhibition efficiencies of sparfloxacin for the corrosion of mild steel.

C(M)	Weight loss	Gasometric		Thermometric
	303K	303K	333K	303K
3	79.68	86.67	33.94	76.34
5	80.20	91.67	52.04	78.22
8	92.66	93.33	51.58	78.22
10	94.65	95.00	58.37	80.21
12	97.47	96.67	59.73	82.30

represented according to equation 5



The implication of equation 5 is that the rate of corrosion can be studied by measuring the volume of hydrogen gas evolved with time. This forms the bases of gasometric method of monitoring corrosion. Another implication of equation 9 is that as the corrosion proceeds, Fe is consumed even as Fe<sup>3+</sup> and H<sub>2</sub> are formed indicating that the rate of corrosion can also be monitored by knowing the amount of Fe that is consumed or the amount of Fe<sup>3+</sup> that is formed. This forms the bases of gravimetric analysis. Generally, stoichiometric principles requires that equation 5 can also be written as follows,

$$d[\text{Fe}]/dt = d[\text{Fe}^{3+}]/dt = d[\text{H}_2]/dt \quad 10$$

Also as the corrosion of mild steel proceeds, the temperature of the systems is expected to increase hence by monitoring the rate of change of temperature, the rate of corrosion can also be monitored.

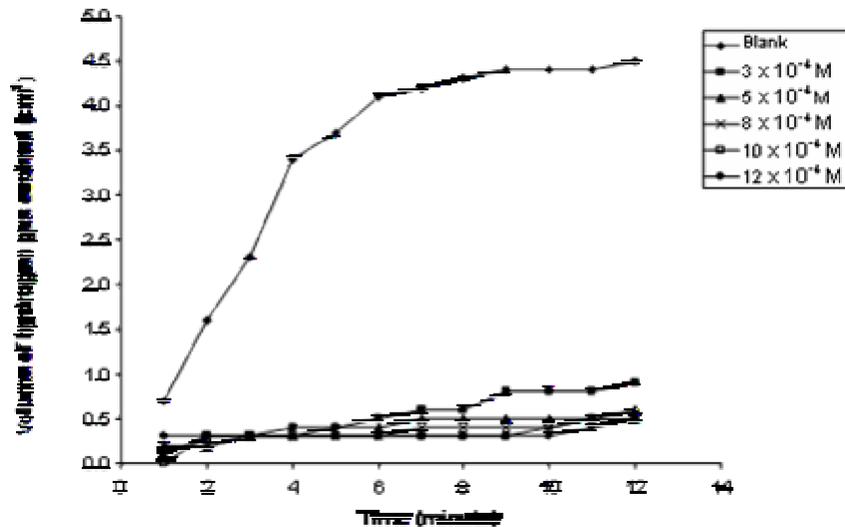
Figures 2 and 3 show the variation of volume of hydrogen gas evolved with time for the corrosion of mild steel in 2.5 M HCl containing various concentrations of sparfloxacin at 303 and 333K respectively. From the Figures, it was found that the rate of corrosion of mild steel increased with increase in the period of contact and temperature. However, the corrosion rate decreased as the concentration of sparfloxacin increases indicating that

sparfloxacin is an adsorption inhibitor for the corrosion of mild steel in HCl.

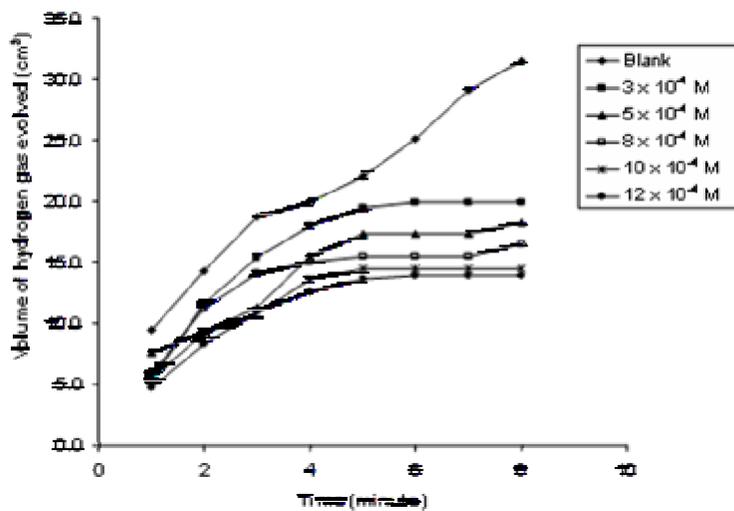
Figure 4 shows the variation of weight loss with time for the corrosion of mild steel in 0.1 M HCl containing various concentrations of sparfloxacin at 303 K. From the Figures, it was found that weight loss of mild steel increased with increase in temperature and with the period of contact but decreased as the concentration of the inhibitor increases indicating that the rate of corrosion of mild steel increases with increase in the period of contact and temperature. And that sparfloxacin inhibited the corrosion of mild steel in HCl.

Figure 5 shows the variation of temperature with time for the corrosion of mild steel in 2.5 M HCl containing various concentrations of sparfloxacin. The Figure also revealed that sparfloxacin inhibited the corrosion of mild steel in HCl and that their inhibition efficiencies increased with increase in temperature.

Table 1 shows the corrosion rates of mild steel in the absence and presence of sparfloxacin. The corrosion rates increased as the temperature increases but decreased as the concentration of the inhibitor increases indicating that these inhibitors inhibited the corrosion of mild steel in HCl by retarding its corrosion rate. Table 2 shows the inhibition efficiencies of sparfloxacin for the corrosion of mild steel in HCl. The results indicated that the inhibition efficiencies of these compounds increased with increase in concentration of the inhibitors indicating that these inhibitors are adsorption inhibitors (Odoemelam and Eddy, 2008a). Figure 6 shows the



**Figure 2.** Variation of volume of hydrogen gas evolved with time for the corrosion of mild steel in HCl containing various concentrations of sparfloxacin at 303K.



**Figure 3.** Variation of volume of hydrogen gas evolved with time for the corrosion of mild steel in HCl containing various concentrations of sparfloxacin.

tion of the inhibition efficiencies of sparfloxacin with immersion time. From the Figure, it can be seen that the stability of the inhibitor increased with increase in its concentration. After seven days of immersion, the inhibitor still retained more than 75% of its inhibition efficiency indicating that it is a good inhibitor.

The corrosion of mild steel in the presence and absence of sparfloxacin was found to first order kinetics implying that equation 11 is applicable (Eddy, 2008).

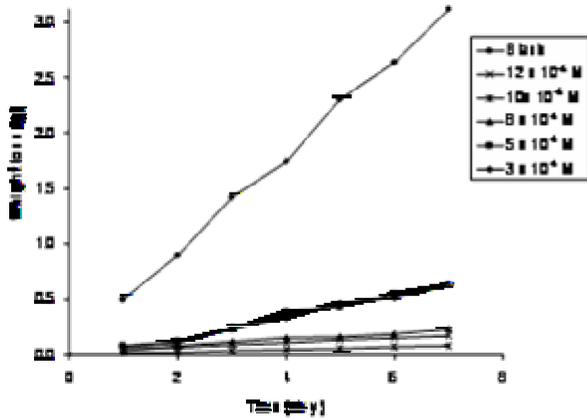
$$-\log(\text{weight loss}) = k_1 t / 2.303$$

11

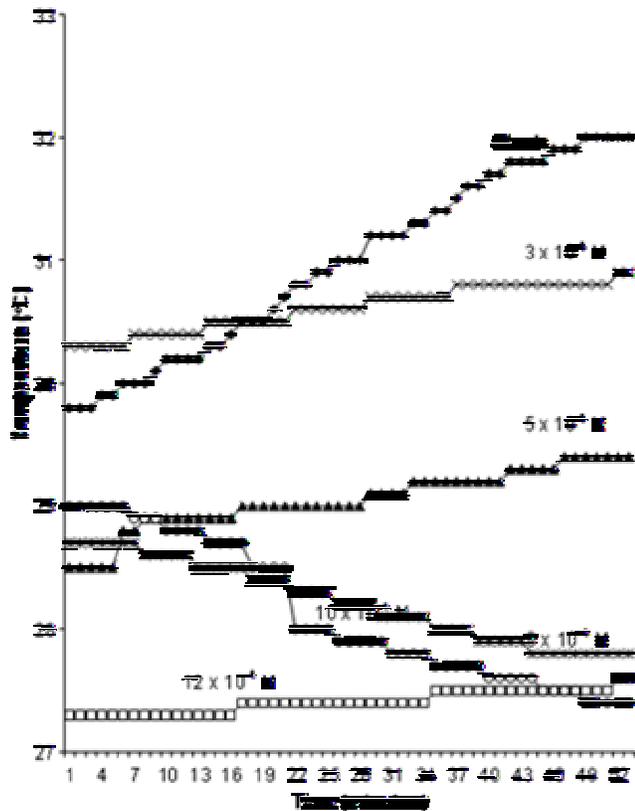
Where  $k_1$  is the first order rate constant and  $t$  is the time in days. The plots of  $-\log(\text{weight loss})$  versus time (with respect to sparfloxacin) were linear confirming that a first order kinetic is applicable to the corrosion of mild steel in the absence and presence of sparfloxacin. Figure 7 show the kinetic plots for the corrosion of mild steel in the presence of various concentrations of sparfloxacin. Also, the half life of a first order reaction is related to the rate constant according to equation 12 (Atkins, 2002; Sharma and Sharama, 2004),

$$t_{1/2} = 0.963/k_1$$

12

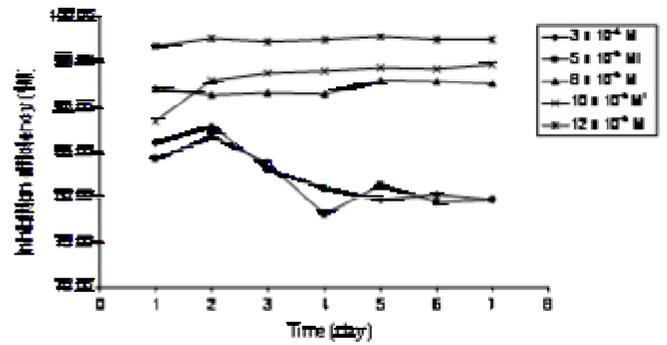


**Figure 4.** Variation of weight loss with time for the corrosion of mild steel in HCl containing various concentrations of sparfloxacin.

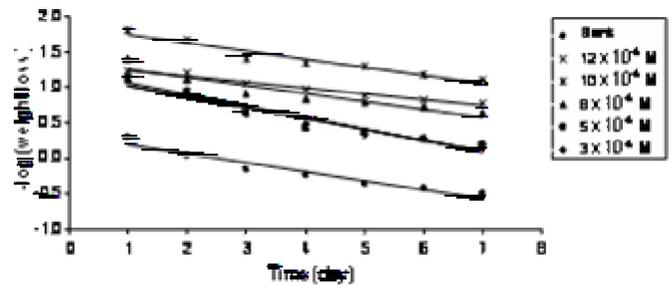


**Figure 5.** Variation of temperature with time for the corrosion of mild steel in H<sub>2</sub>SO<sub>4</sub> containing various concentrations of sparfloxacin.

Values of the rate constants and half lives obtained from the slopes of the kinetic plots are recorded in Table 3. The results revealed that the half lives of mild steel in the presence of the inhibitors were higher than the half



**Figure 6.** Variation of inhibition efficiency of sparfloxacin with time.



**Figure 7.** Kinetic plot for the corrosion of mild steel in HCl containing various concentrations of sparfloxacin.

life obtained for the blank solution indicating that the inhibitors increased the half life of mild steel in HCl.

The effect of temperature on the corrosion of mild steel in the absence and presence of sparfloxacin was studied using the Arrhenius equation,

$$CR = A \exp(-E_a/RT) \quad 13$$

Where CR is the corrosion rate of mild steel, A is Arrhenius or pre-exponential factor,  $E_a$  is the activation energy, R is the gas constant and T is the temperature. Assuming at the temperatures  $T_1$  (303K) and  $T_2$  (333K), the corrosion rates of mild steel are  $CR_1$  and  $CR_2$  and that A is constant, then equation 9 simplifies to equation 14

$$\log(CR_2/CR_1) = E_a/RT (1/T_1 - 1/T_2) \quad 14$$

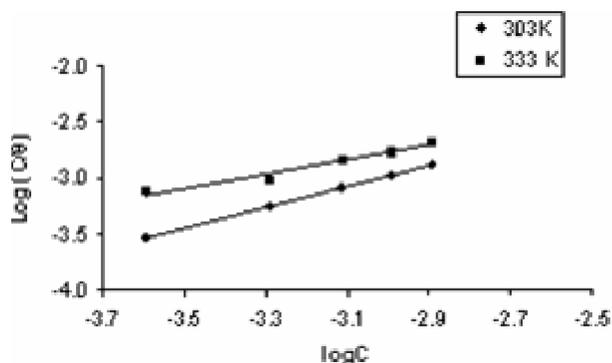
Values of  $E_a$  calculated from equation 14 are recorded in Table 4.  $E_a$  values were found to range from 21.10 to 27.09 KJ/mol indicating that the mechanism of adsorption of sparfloxacin on the surface of mild steel is physical adsorption (Eddy et al., 2008a-c). These values were also found to be higher than the value of 5.67 KJ obtained for the blank indicating that sparfloxacin retarded the corrosion of mild steel. Also,  $E_a$  values were found to increase with increase in the concentration of

**Table 3.** Kinetic parameters for the corrosion of mild steel in the presence of various sparfloxacin.

C x 10 <sup>-4</sup> (M)	Parameters		
	k <sub>1</sub>	t <sub>1/2</sub>	R <sup>2</sup>
Blank	0.3719	1.86	0.9341
3	0.3537	1.96	0.9298
5	0.2902	2.39	0.9327
8	0.2605	2.66	0.8859
10	0.1905	3.64	0.9780
12	0.2607	2.66	0.9477

**Table 4.** Activation energies and heat of adsorption of sparfloxacin on the surface of mild steel.

C (M)	E <sub>a</sub> (KJ/mol)	Q <sub>ads</sub> (KJ/mol)
Blank	5.76	-
3	11.14	-53.24
5	11.65	-48.59
8	12.44	-54.02
10	12.89	-54.67
12	14.14	-62.38



**Figure 8.** Langmuir isotherm for the adsorption of sparfloxacin on the surface of mild steel.

the inhibitor indicating that there is an increase in the surface area of adsorption due to the ease of adsorption as the concentration of the inhibitor increases.

The heat of adsorption of sparfloxacin on the surface of mild steel was calculated using equation 15 (Eddy et al., 2008a-d).

$$Q_{ads} = 2.303R[\log(\theta_2/1-\theta_2) - \log(\theta_1/1-\theta_1) \times (T_2T_1)/(T_2 - T_1)] \quad 15$$

Where R is the gas constant,  $\theta_1$  and  $\theta_2$  are the degree of surface coverage at temperatures,  $T_1$  and  $T_2$  respectively. Values of  $Q_{ads}$  calculated from equation 11 are also recorded in Table 4. These values ranged from -48.59 to

-62.38 KJ/mol indicating that the adsorption of these inhibitors on the surface of mild steel is exothermic.

Adsorption isotherm is necessary in studying the mechanism of adsorption and also the adsorption characteristics of the inhibitor. The general form of adsorption isotherm can be written according to equation 16.

$$f(0,x)\exp(-2a\theta) = KC \quad 16$$

Where  $f(0,x)$  is configuration factor,  $\theta$  is the surface coverage, C is inhibitors concentration; K is adsorption constant and a is the molecular interaction parameter. Data obtained for the degree of surface coverage were used to fit curves for different adsorption isotherm and it was found that Langmuir adsorption isotherm best described the adsorption characteristics of sparfloxacin on the surface of mild steel.

Langmuir adsorption isotherm relates the degree of surface coverage ( $\theta$ ) to the concentration of the inhibitor in the bulk electrolyte according to equation 17 which simplifies to equation 18.

$$C/\theta = 1/K + C \quad 17$$

$$\log(C/\theta) = \log C - \log K \quad 18$$

From equation 14, a plot of  $\log(C/\theta)$  versus  $\log C$  should give a straight line if the assumptions of Langmuir are valid. Figure 8 shows Langmuir adsorption isotherms for the adsorption of sparfloxacin on the surface of mild steel. Values of adsorption parameters deduced from Langmuir isotherms are recorded in Table 5. From the plots, values of the slopes and  $R^2$  were very close to unity indicating adherence to Langmuir adsorption isotherm (Acharya and Upadhyah, 2004; Eddy, 2008). The application of Langmuir isotherms to the adsorption of sparfloxacin and amoxil on the surface of mild steel indicated that there is no interaction between the adsorbate and the adsorbent. According to Odoemelam and Eddy (2008a), Langmuir adsorption isotherm is an ideal isotherm for physical adsorption where there is no interaction between the adsorbate and adsorbent. The equilibrium constant of adsorption is related to the free energy of adsorption according to equation 19 (Ashassi-Sorkhabi et al., 2006; Ebenso, 2003).

$$\Delta G_{ads} = -2.303RT\log(55.5K) \quad 19$$

Values of free energy calculated from equation 15 are recorded in Table 5. These values are negative and are less than the threshold value of -40 KJ/mol indicating that the adsorption of sparfloxacin on the surface of mild steel is spontaneous and favoured the mechanism of physical adsorption.

## Conclusion

From the results and findings of the study, we conclude

**Table 5.** Values of adsorption parameters for the adsorption of sparfloxacin on the surface of mild steel.

Langmuir adsorption parameters				
Temperature	logK	slope	$\Delta G_{ads}$ (KJ/mol)	$R_2$
303	0.005	0.9343	10.12	0.9999
333	0.1018	0.660	11.74	0.9686

that sparfloxacin is good inhibitors for the corrosion of mild steel in HCl. Sparfloxacin inhibited the corrosion of mild steel by been adsorbed on the surface of mild steel. The adsorption of sparfloxacin on the surface of mild steel is exothermic, spontaneous and is best described by Langmuir adsorption isotherm. Based on the variation of inhibition efficiencies with temperature, values of activation energies and free energy changes been lower than the threshold values of 80KJ/mol and -40 KJ/mol respectively, the mechanism of adsorption of sparfloxacin is physical adsorption. Therefore we recommend the use of this inhibitor for the corrosion of mild steel in HCl.

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