

Electrochemical Studies of Adsorption and Inhibitive Performance of Basic Yellow 28 Dye on Mild Steel Corrosion in Acid Solutions

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Abstract

Organic corrosion inhibitors are widely used to control the corrosion of different metals in various corrosive solutions. The inhibition performance of Basic yellow 28 (BY28) dye for mild steel corrosion was investigated in 0.1 M HCl solution and in a solution of 0.1 M HCl and 1% NaCl. Two electrochemical methods including Tafel polarization and electrochemical impedance spectroscopy (EIS) measurements were used. The corrosion parameters as well as inhibition efficiencies were obtained for different concentrations of inhibitor. The inhibition efficiencies showed that the BY28 dye acts as a good corrosion inhibitor for mild steel in both solutions. The studies on adsorption isotherm of the dye on mild steel proved that the adsorption of BY28 obeys the Langmuir adsorption isotherm. The average value of $-\Delta G_{\text{ads}}$ in both solutions was more than 20 and a little less than 40 kJ mol⁻¹. Therefore, both chemisorption and physisorption phenomena were involved in the adsorption of the studied dye on mild steel surface.

Keywords: Corrosion inhibition, EIS, Polarization, Adsorption isotherm, Basic yellow 28 dye

1. Introduction

Excessive corrosion attack is known to occur on mild steel in acidic aqueous solutions. Acid solutions are widely used in different industrial processes, for example, in acid pickling of iron and steel, scale removal in metallurgy, acid cleaning of boilers and oil well acidizing; therefore, the study of steel corrosion in acid solutions and its inhibition have practical importance. Different corrosion inhibitors are used to control and reduce the corrosion of metals in corrosive solutions. Due to toxicity of most inorganic corrosion inhibitors such as chromates and nitrites and because of restrictive environmental regulations, new environmentally friendly organic compounds are replacing these inhibitors.¹⁻² Steel corrosion in acid solutions has been effectively controlled using organic inhibitors, which are usually organic substances containing nitrogen, sulfur and/or oxygen in the conjugated system. These molecules have been reported as efficient corrosion inhibitors, which act through adsorption on metal surface, form a complex with metal ions and thereby retard the metal

dissolution; heteroatoms and polar groups of the inhibitor molecule act as the reaction center for the adsorption process.³⁻¹⁹ The protection efficiency depends on adsorption ability of inhibitor molecules, where the resulting adsorption film acts as a barrier, which separates the metal from the corrosive electrolyte.¹⁶⁻¹⁷ During corrosion, the products will constitute a new distinct phase between the metal and the aqueous solution; the inhibitor thereby interacts with this interphase.¹⁸

It has been found that organic dyes, having different heteroatoms and polar groups, can also be used as corrosion inhibitors. Several authors²⁰⁻²² have reported the use of different dyes for the corrosion inhibition applications. The aim of this work is to study the adsorption characteristics and inhibition performance of Basic yellow 28 (BY28) dye on mild steel corrosion in 0.1 M HCl and in a solution of 0.1 M HCl and 1% NaCl. The molecular structure of the compound is given in Figure 1. The presence of azo and amine groups and the electron cloud on the aromatic rings may help this molecule to be adsorbed on mild steel and act as a corrosion inhibitor.

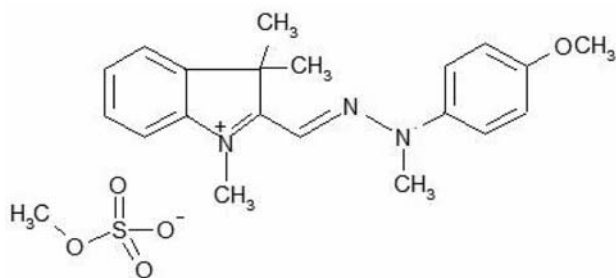


Figure 1. The molecular structure of Basic yellow 28 dye

2. Experimental

The working electrode was mild steel with the chemical composition (wt%) of: Si (1.250%), Mn (0.221%), Cr (0.018%), S (0.016%), P (0.007%), C (0.005%) and Fe (Balance). BY28 dye, was purchased from Chinachem Company and was used at concentrations of 1×10^{-5} , 1×10^{-4} , 5×10^{-4} , 1×10^{-3} and 1×10^{-2} M. The Basic Yellow 28 dye is an orange powder, easily soluble in water with molecular weight of 433.57g/mol, which its molecular structure was shown in Figure 1. Two corrosive solutions including a 0.1 M HCl (solution A) and a solution of 0.1 M HCl and 1% NaCl (solution B) were used as electrolyte. The steel specimens were abraded with SiC papers up to 1500 grade, degreased with acetone and rinsed with distilled water.

Electrochemical experiments, including Tafel polarization and EIS measurements, were carried out using an Autolab PGSTAT30 Potentiostat-Galvanostat. A conventional three-electrode configuration was used for electrochemical studies. The working electrode was prepared from a 1 cm \times 1 cm mild steel sheet, mounted in a polyester resin with the uncovered area of 1 cm². A saturated calomel electrode (SCE) and a platinum electrode were used as the reference and counter electrodes, respectively. EIS tests were performed immediately after 30 minutes immersion of working electrode at 298 ± 1 K in open circuit potential over a frequency range between 10 kHz and 10 mHz; a sine wave with 10 mV amplitude was applied. EIS data were analyzed using Zview2 software and the double layer capacitance, C_{dl} , and charge transfer resistance, R_{ct} , were obtained from Nyquist plots as described elsewhere.^{19,23} The Tafel polarization measurements were made just after EIS tests. The potential was swept from $E_{ocp} - 250$ mV to $E_{ocp} + 250$ mV with a scan rate of 2 mVs⁻¹.

3. Results and Discussions

3. 1. Tafel Polarization Measurements

In order to study the effect of BY28 dye on the corrosion behavior of mild steel, Tafel polarization curves for different concentrations of inhibitor in the solutions A and

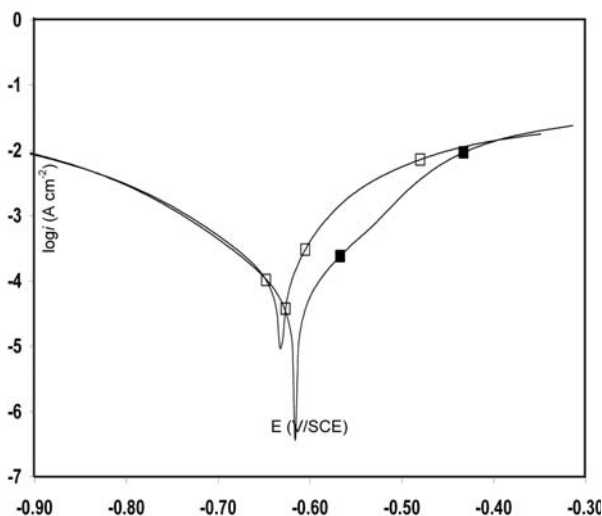
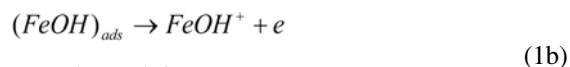
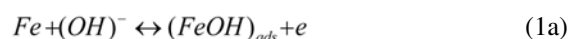


Figure 2. Tafel polarization plots for mild steel corrosion in 0.1 M HCl (□) and 0.1 M HCl solution containing 1% NaCl (■)

B were plotted. Figure 2 indicates two Tafel plots obtained for mild steel corrosion in A and B uninhibited solutions.

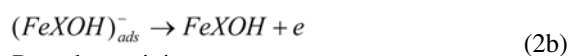
It can be seen from Figure 2 that the anodic current density in solution B is lower than solution A. However, the cathodic branches in both solutions are coincident with each other. Therefore, the corrosion current density in solution B is relatively less than the corrosion current density obtained for mild steel in the solution A. The decrease in corrosion current density in the presence of NaCl, may be attributed to the participating of halide ions in forming intermediates on a corroding steel's surface in acid solutions. It is well known that the anodic dissolution of steel in aqueous solutions is significantly facilitated by hydroxyl ions, which form intermediate catalytic complexes on the metal surface:



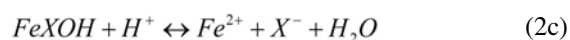
Rate determining step



Halide ions may replace hydroxyl ions adsorbed on the surface of the metal, thus leading to a reduction in the catalytic effect of the hydroxyl ions:



Rate determining step



where X represents the halide ion. This ability is initiated by the specific adsorption of the anion onto the metal surface.²⁴

The Tafel plots for different concentrations of BY28 have also been represented in Figures 3 and 4 for solutions A and B respectively. Electrochemical corrosion parameters including corrosion potential (E_{corr}), cathodic and anodic Tafel slopes (b_c , b_a) and corrosion current density (i_{corr}), obtained by extrapolation of Tafel lines, are given in Table 1. The inhibition efficiencies, $\eta_p\%$, in both solutions were calculated from the following equation and are also given in Table 1.²⁵

$$\eta_p \% = \left(\frac{i_{corr}^0 - i_{corr}}{i_{corr}^0} \right) \times 100 \quad (3)$$

where i_{corr}^0 and i_{corr} are the corrosion current densities in the absence and presence of inhibitor, respectively.

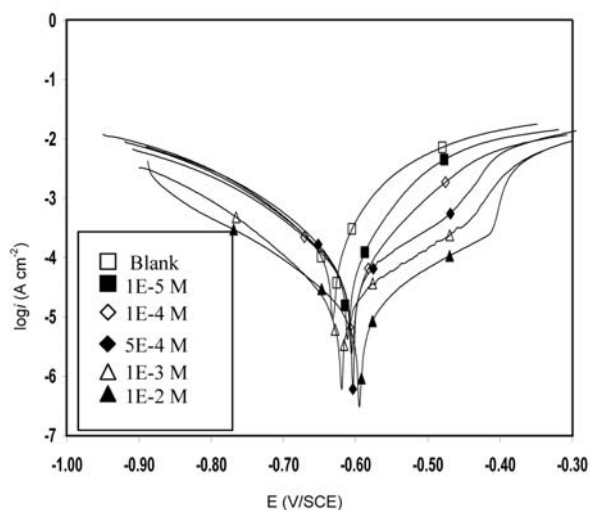


Figure 3. The Tafel polarization curves for mild steel in 0.1 M HCl solution (solution A) in the absence and presence of different concentrations of inhibitor

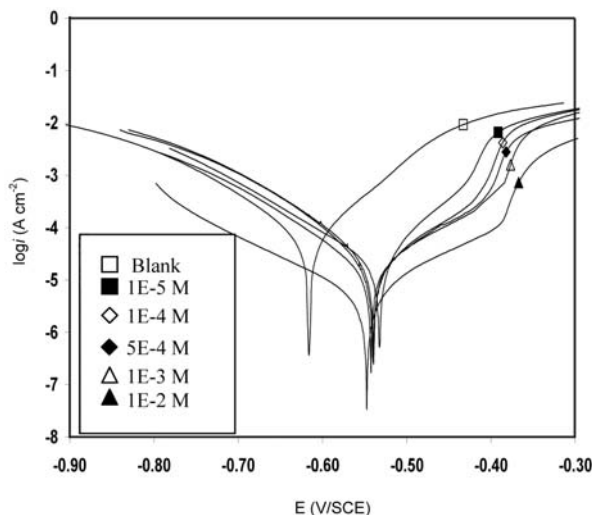


Figure 4. The Tafel polarization curves for mild steel in 0.1 M HCl solution containing 1% NaCl (B) in the absence and presence of different concentrations of inhibitor

Figures 3 and 4 show that the anodic current densities decrease in the presence of inhibitor. The cathodic reduction reaction was inhibited just when the high concentrations of inhibitor were used. It means that the anodic dissolution of metal is more inhibited by the BY28 dye. Therefore, in the presence of inhibitor, the corrosion potential shifts toward the positive potentials. These results indicate that the BY28 dye, in both solutions, acts as an inhibitor with predominantly control of anodic reaction. It is due to the adsorption of dye molecules and formation of a protective layer on metal surface. This protective film retards the anodic dissolution of Fe to Fe^{2+} . However, the BY28 dye cannot significantly inhibit the cathodic reaction, i.e. hydrogen evolution reaction.²⁶

It is also seen from data presented in Table 1 that the inhibition efficiencies (in both solutions A and B) enhance significantly as the inhibitor concentration increases. Ma-

Table 1: Electrochemical parameters for mild steel corrosion in 0.1 M HCl (solution A) and 0.1 M HCl solution containing 1% NaCl (solution B) in the absence and presence of different concentrations of Basic yellow 28

Solution	C_{inh} (M)	R_p ($\Omega \text{ cm}^2$)	E_{corr} (mV/SCE)	b_a (mV/dec)	b_c (mV/dec)	i_{corr} (A cm^{-2})	$\eta_p\%$
Solution A	Blank	132	-631	92	60	1.20×10^{-4}	–
	1.0×10^{-5}	254	-611	90	51	5.57×10^{-5}	45
	1.0×10^{-4}	409	-605	76	70	3.87×10^{-5}	68
	5.0×10^{-4}	857	-604	76	78	1.95×10^{-5}	84
	1.0×10^{-3}	1356	-620	84	100	1.46×10^{-5}	88
	1.0×10^{-2}	2208	-594	90	110	1.00×10^{-5}	92
Solution B	Blank	321	-630	62	76	4.63×10^{-5}	–
	1.0×10^{-5}	814	-532	70	90	2.10×10^{-5}	54
	1.0×10^{-4}	940	-539	80	76	1.80×10^{-5}	67
	5.0×10^{-4}	1046	-542	93	88	1.38×10^{-5}	70
	1.0×10^{-3}	1099	-540	109	72	1.06×10^{-5}	77
	1.0×10^{-2}	2532	-548	107	112	6.30×10^{-6}	86

ximum inhibition efficiencies were obtained in the concentration of 1 ± 10^{-2} M.

For corrosion of steel in acidic solution, the main anodic reaction is oxidation of Fe to Fe^{2+} and the main cathodic one is the reduction of H^+ ions. In the presence of inhibitor, the inhibitor is first adsorbed on steel surface and therefore, impedes the dissolution of metal by merely blocking the active sites of the metal surface. On the other hand, the surface area available for H^+ ions is decreased, while the actual reaction mechanism remains unaffected. The presence of a free part of metal surface ($1-\theta$), where θ is the degree of surface coverage by inhibitor molecules, permits a free access of H^+ ions to the mild steel surface.²⁷ When high concentrations of inhibitor are used, the uncovered area of the metal surface ($1-\theta$) decreases remarkably; therefore, the cathodic current density also shows significant reduction, while it remains almost unaffected in low concentrations of inhibitor.

In the anodic part of polarization curves, a significant inhibition was observed at low over potentials, which may suggest formation of a protective layer of adsorbed species at the metal surface. However, inhibitor has little effect on the anodic current densities at overpotentials more than ≈ -200 mV, which is usually defined as »desorption potential«. It means that the inhibition mode of inhibitor depends on the electrode potential. So, desorption of inhibitor protective films occurs at potentials more positive than »desorption potential«, and the anodic current density remains almost constant. It may be the result of significant dissolution of mild steel, leading to desorption of inhibitor film from the metal surface.²⁷ On the other hand, a current plateau is seen in the anodic branch of polarization curves for high concentrations of the inhibitor. This plateau is also another evidence for the adsorption of inhibitor molecules on steel sur-

face, which leads to the formation of a protective film, inhibits the dissolution of metal and creates the anodic current plateau.

3. 2. Electrochemical Impedance Spectroscopy (EIS) Measurements

The corrosion behavior of mild steel in solutions A and B in the absence and presence of BY28 dye was also investigated using electrochemical impedance spectroscopy. Nyquist plots for mild steel in uninhibited and inhibited solutions A and B containing various concentrations of inhibitor are shown in Figures 5 and 6 respectively. As it can be seen, the impedance of mild steel has significantly changed after the addition of inhibitor.

The EIS results can be interpreted using an equivalent circuit of the electrical double layer shown in Figure 7, which has been used previously to model the iron/acid interface.²⁸ The double layer capacitance and charge transfer resistance were obtained from Nyquist plots. The inhibition efficiencies from EIS results were calculated using the following equation.²⁵

$$\eta_z(\%) = \left(\frac{R_{ct} - R_{ct}^0}{R_{ct}} \right) \times 100 \quad (4)$$

where R_{ct}^0 and R_{ct} are the charge transfer resistances in the absence and presence of inhibitor, respectively. The electrochemical parameters derived from the Nyquist plots as well as the inhibition efficiencies, η_z , are given in Table 2.

It is clear that, R_{ct} increases and C_{dl} decreases as the inhibitor concentration increases. The decrease in C_{dl} could be attributed to the adsorption of the inhibitor forming protective adsorption layer.²⁹ The inhibition effi-

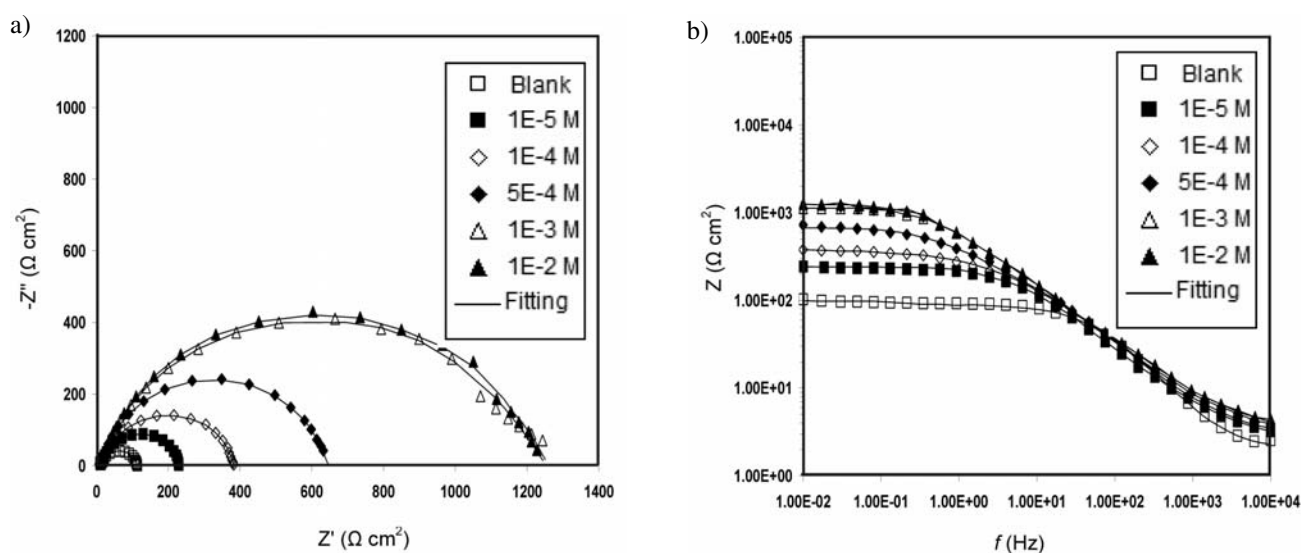


Figure 5. The (a) Nyquist and (b) Bode-Magnitude plots for mild steel in 0.1 M HCl solution (A) in the absence and presence of different concentrations of inhibitor (dots: experimental points; lines: fitting results)

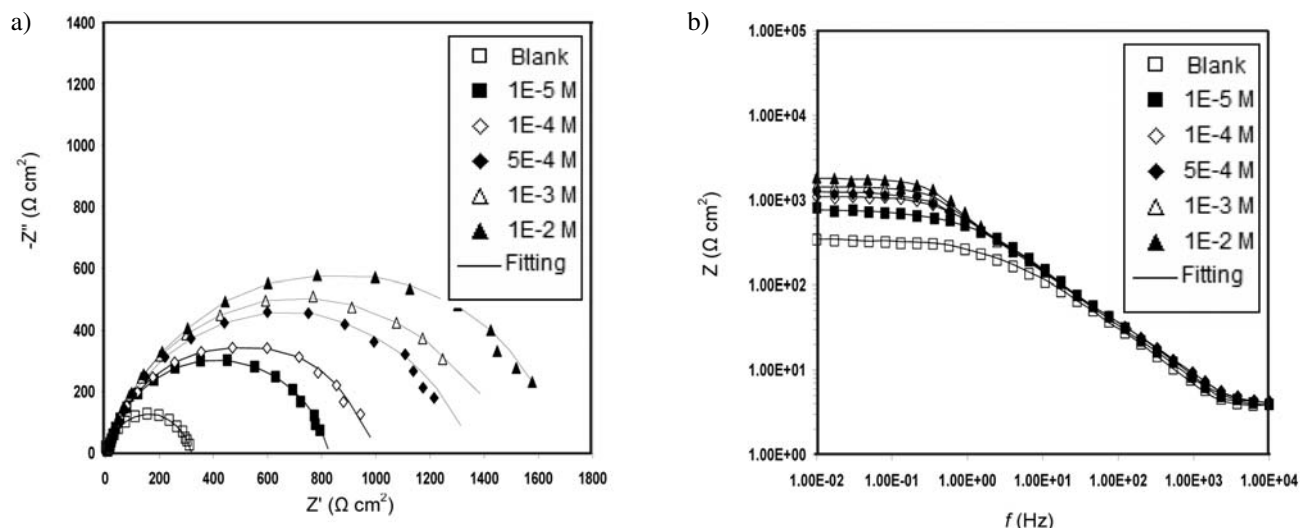


Figure 6. The (a) Nyquist and (b) Bode-Magnitude plots for mild steel in 0.1 M HCl solution containing 1% NaCl (B) in the absence and presence of different concentrations of inhibitor (dots: experimental points; lines: fitting results)

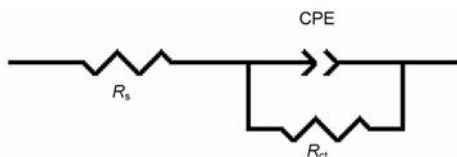


Figure 7. The electrochemical equivalent circuit used for fitting the experimental EIS data for mild steel corrosion in acidic solutions

ciencies enhance as the concentration of the inhibitor increases. They are also in good agreement with the polarization results. The increase in R_{ct} and inhibition efficiencies along with the decrease in C_{dl} with the enhancement of BY28 concentration are reasons for the formation of a protective layer of inhibitor molecules on mild steel surface in both electrolytes. The adsorption isotherms of BY28

on mild steel in solutions A and B have been discussed in the following section.

3. 4. Adsorption Isotherm for Basic Yellow 28 Dye

The adsorption isotherms can provide basic information on the interaction of inhibitor molecules and metal surface. The surface coverage, ($\theta = \eta/100$), values for each concentration of inhibitor were calculated from both polarization and EIS measurements. The data were then tested graphically for fitting a suitable adsorption isotherm. The most common isotherms such as Langmuir, Temkin and Freundlich isotherms were examined. It was found that the plots of C_{inh}/θ versus C_{inh} , from both polarization and EIS results, yields straight lines with the corre-

Table 2: Electrochemical impedance parameters for mild steel in 0.1 M HCl solution (A) and 0.1 M HCl solution containing 1% NaCl (B) in the absence and presence of Basic yellow 28

Solution	C_{inh} (M)	R_{ct} (Ωcm^2)	C_{dl} ($\mu F cm^{-2}$)	n	$\eta_c\%$
Solution A	Blank	102	6.65×10^{-5}	0.86	–
	1.0×10^{-5}	221	3.55×10^{-5}	0.85	53
	1.0×10^{-4}	377	1.30×10^{-5}	0.86	73
	5.0×10^{-4}	635	0.76×10^{-5}	0.84	84
	1.0×10^{-3}	1220	0.02×10^{-5}	0.83	91
	1.0×10^{-2}	1275	0.01×10^{-5}	0.85	92
Solution B	Blank	308.6	1.92×10^{-5}	0.86	–
	1.0×10^{-5}	838.4	3.84×10^{-6}	0.87	63
	1.0×10^{-4}	1030.0	2.07×10^{-6}	0.74	70
	5.0×10^{-4}	1279.0	2.74×10^{-6}	0.79	76
	1.0×10^{-3}	1498.0	1.75×10^{-6}	0.75	79
	1.0×10^{-2}	1798.0	1.25×10^{-6}	0.73	83

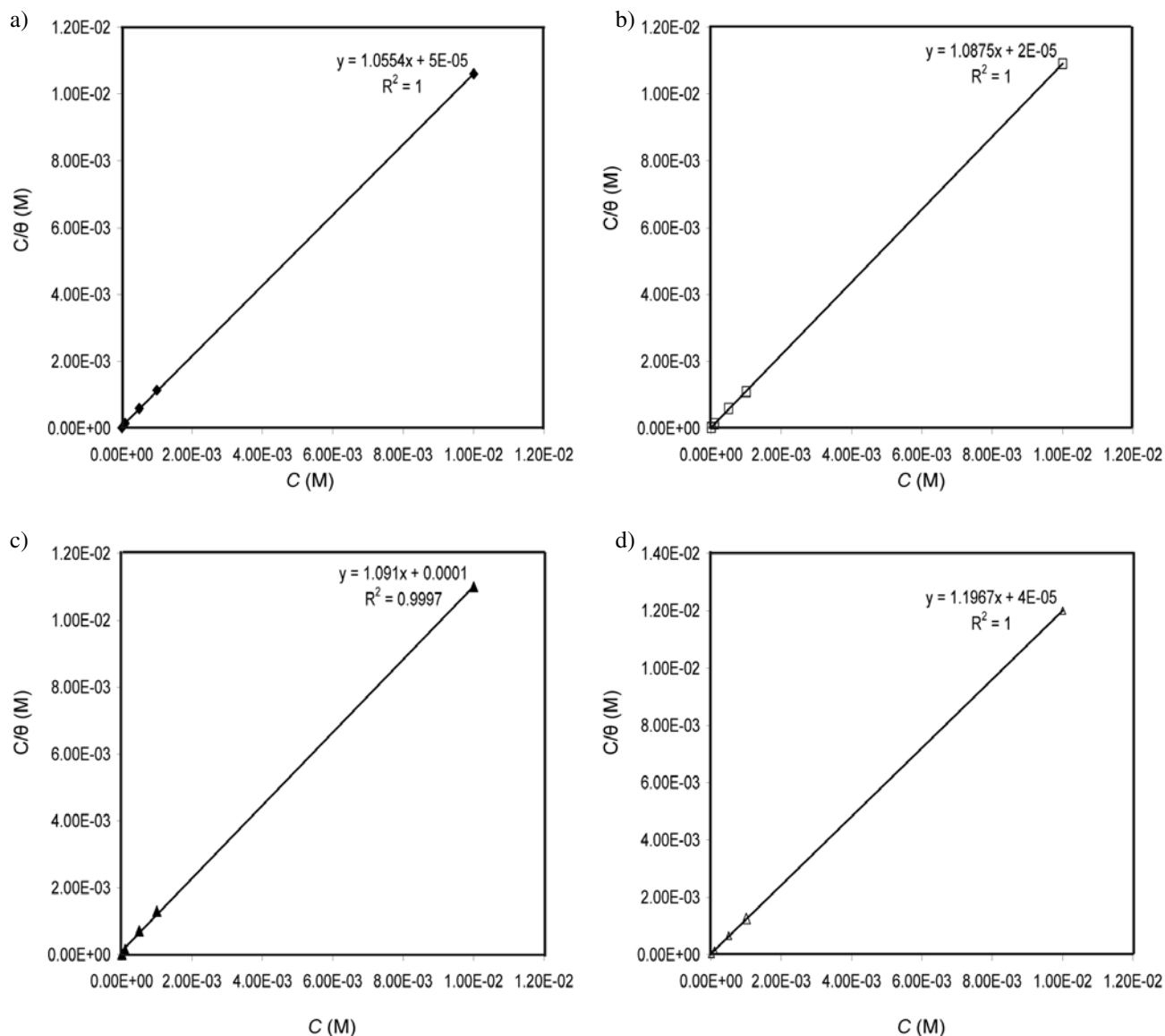


Figure 8. The Langmuir isotherm plots for adsorption of Basic yellow 28 dye on mild steel surface in acidic solutions (a) solution A, polarization data; (b) solution A, EIS data; (c) solution B, polarization data; (d) solution B, EIS data

lation coefficients of about ≈ 1.0000 . These plots as well as the corresponding equation and regression coefficient are shown in Figure 8.

It can be deduced that the adsorption of BY28 dye on the surface of mild steel in the solutions of 0.1 M HCl (A) and 0.1 M HCl containing 1% NaCl (B), obeys the Langmuir adsorption isotherm, which is presented by the equation below.²⁷

$$\frac{C_{inh}}{\theta} = C_{inh} + \frac{1}{K_{ads}} \quad (5)$$

where C_{inh} is the inhibitor concentration, θ is the degree of coverage of metal surface, and K_{ads} is the equilibrium constant for adsorption-desorption process. The values of θ

are proportional to the inhibition efficiencies ($\theta = \eta/100$). The equilibrium constants, K_{ads} , were calculated from the reciprocal of the intercept of the isotherm line. The free energy of inhibitor adsorption on mild-steel surface can be evaluated using the following equation.²⁷

$$\Delta G_{ads} = -RT \ln(55.5 K_{ads}) \quad (6)$$

The values of K_{ads} and ΔG_{ads} , obtained from polarization and EIS results, as well as their average values in both solutions A and B are listed in Table 3.

The negative values of ΔG_{ads} indicate spontaneous adsorption of BY28 molecules on the mild steel surface and strong interaction between inhibitor molecules and metal surface.²⁷

Table 3: The values of K_{ads} and ΔG_{ads} for adsorption of Basic yellow 28 on mild steel from polarization and EIS data and the average values

Solu- tion	Polarization		EIS		Average	
	K_{ads} (M^{-1})	ΔG_{ads} (kJ mol^{-1})	K_{ads} (M^{-1})	ΔG_{ads} (kJ mol^{-1})	K_{ads} (M^{-1})	ΔG_{ads} (kJ mol^{-1})
A	2.0 ± 10^4	-34.5	5.0 ± 10^4	-36.8	3.5 ± 10^4	-35.6
B	1.0 ± 10^4	-32.8	2.5 ± 10^4	-35.0	1.8 ± 10^4	-33.9

It is well known that the values of $-\Delta G_{\text{ads}}$ around 20 kJ mol^{-1} or lower are indicative of the electrostatic interaction between charged organic molecules and the charged metal surface (i.e. physisorption); those around 40 kJ mol^{-1} or higher involve charge sharing or transfer from the organic molecules to the metal surface to form a coordinated bond (i.e. chemisorption). The value of $-\Delta G_{\text{ads}}$ for adsorption of BY28 molecules on mild steel surface is less than 40 kJ mol^{-1} , which indicates the physical adsorption. However, since the value of $-\Delta G_{\text{ads}}$ is very close to 40 kJ mol^{-1} , it could also be concluded that in addition to the electrostatic interactions, there may be some other interactions.³⁰ Therefore, the adsorption mechanism of the BY28 dye molecules on steel in both solutions A and B involves two types of interaction, chemisorption and physisorption.³¹ The values of $-\Delta G_{\text{ads}}$ in solution B are a little less than solution A. The presence of more chloride ions, in solution B, may cause the metal surface charge to be more negative; so that, the physical electrostatic interactions between the positively charged dye molecules and metal surface are a little stronger and the $-\Delta G_{\text{ads}}$ values are less compared to the solution A.

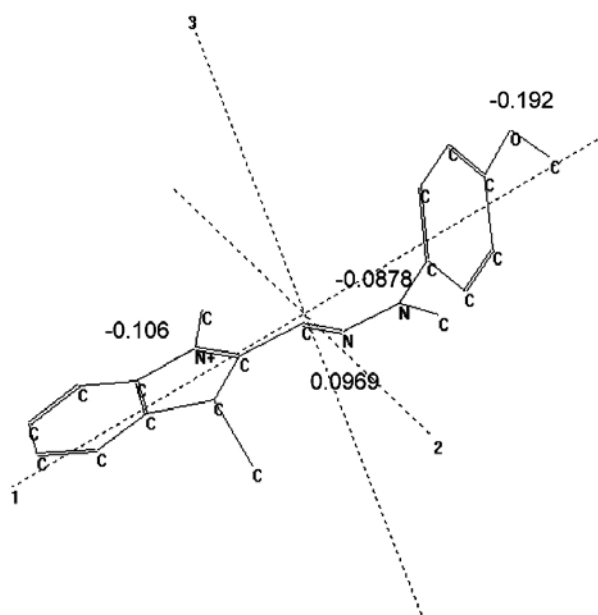


Figure 9: The optimized structure of the BY28 dye and the partial atomic charges of heteroatoms

In order to get more information on the probable centre of adsorption, the structure of the BY 28 dye was optimized by applying AM1 semi-empirical method. The optimized structure of the molecule and the partial atomic charges of heteroatoms are given in Figure 9.

It is seen that the oxygen atom has relatively more partial charge and is in a suitable spatial situation for adsorption; therefore, it is a suitable site for adsorption of the molecule on steel surface. The azo nitrogen atom can also be a center for better adsorption of the molecule.

4. Conclusion

The corrosion inhibition of mild steel in two acidic solutions (including 0.1 M HCl solution and a solution of 0.1 M HCl and 1% NaCl) were studied by Basic yellow 28 dye, BY28. It was shown that BY28 acts as a good corrosion inhibitor in both solutions. The electrochemical studies including Tafel polarization and EIS measurements were used for investigation of the corrosion behavior of mild steel in the presence of BY28. The inhibition efficiencies were calculated from these studies and it was shown that the inhibition efficiency of BY28 increases with the increase of its concentration in corrosive solution. The adsorption isotherm of dye molecules was investigated and it was shown that the dye molecules are adsorbed on steel surface, create a protective layer, and decrease the corrosion rate. The adsorption of BY28 dye molecules were spontaneous and obey the Langmuir adsorption isotherm. The values of $-\Delta G_{\text{ads}}$, in both solutions, were a little less than 40 kJ mol^{-1} that indicates the chemisorption of BY28 molecules on mild steel. However, the physical interactions also play a little role in the adsorption process. The optimization of the geometry of dye molecule introduced the most reactive atoms for adsorption on mild steel surface.

5. References

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Povzetek

S Taflovo polarizacijo in elektrokemijsko impedančno spektroskopijo (EIS) smo raziskovali barvilo osnovno rumeno (Basic yellow 28, BY28) kot inhibitor korozije jekla v raztopinah 0.1 M HCl ter v 0.1 M HCl z dodatkom 1 % NaCl. Rezultati kažejo, da je BY28 učinkovit inhibitor korozije v obeh raztopinah. Proučevali smo tudi adsorpcijo BY28 na jeklo ter ugotovili, da jo lahko opišemo z Langmuirjevo adsorpcijsko izotermo. Iz povprečnih vrednosti Gibbsove proste energije adsorpcije, ΔG_{ads} , pa lahko sklepamo, da pri adsorpciji BY28 na površino jekla poteka tako kemisorpcija kot fizikalna adsorpcija.