



Corrosion Inhibition on Mild Steel in 1.0 M HCl Solution by 1-Vinylimidazolium-3-methyltriphenyl Phosphonium Dihexafluoroantimonate Salt

Ayssar Nahlé*, Ideisan Abu-Abdoun, Ibrahim Abdel-Rahman

Department of Chemistry, College of Sciences, University of Sharjah, Sharjah, P.O.Box: 27272, United Arab Emirates.

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*Author for correspondence; Email: anahle@sharjah.ac.ae; Fax: +971-6-5053820

Abstract

The effect of synthesized 1-vinylimidazolium-3-methyltriphenyl phosphonium dihexafluoroantimonate (VIMTPPF) on the corrosion inhibition of mild steel in 1.0M HCl solution was studied by weight loss measurements. The weight loss measurements were carried out on carbon steel specimens in 1.0M HCl and in 1.0M HCl containing various concentrations (5.0×10^{-4} to 5.0×10^{-7} M) of the laboratory synthesized VIMTPPF salt at temperatures ranging from 303 to 343 K. 1-vinylimidazolium-3-methyltriphenyl phosphonium dihexafluoroantimonate salt (VIMTPPF) was found to be a good efficient inhibitor for carbon steel in 1.0M HCl solution, reaching about 88 percent at the concentration of 5.0×10^{-4} M at 303K. The percentage of inhibition in the presence of this inhibitor decreased with the increase of temperature indicating that physical adsorption was the predominant inhibition mechanism since the quantity of adsorbed inhibitor decreased with increasing the temperature. This inhibitor could have industrial applications, where hydrochloric acid solutions at high temperatures are used to remove scale and salts from steel surfaces, such as acid cleaning of tankage and pipeline, and may render dismantling unnecessary.

Keywords: Corrosion, inhibitor, 1-vinylimidazolium-3-methyltriphenyl phosphonium dihexafluoroantimonate salt, Temkin adsorption isotherm.

1. Introduction

Corrosion of metals is a serious problem in many industries, installations, and civil services such as water and sewage supplies. In searching for an economic method to prevent or minimize corrosion, inhibitors are usually suggested and employed, especially in flow systems such as cooling towers. Corrosion increases running costs and reduces plant efficiency, availability and product quality.

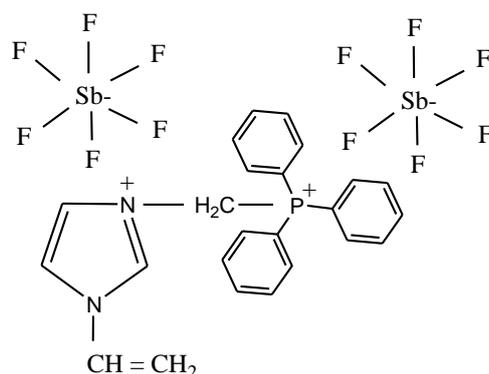
Organic compounds containing polar groups by which the molecule can become strongly or specifically adsorbed on the metal surface constitute most organic inhibitors [1,2]. These inhibitors, which include the organic N, P, S, and OH groups, are known to be similar to catalytic poisons, as they decrease the reaction rate at the metal/solution interface without, in general, being involved in the reaction considered. It is generally accepted that most organic inhibitors act via adsorption at the metal/solution interface. The mechanism by which an inhibitor decreases the corrosion current is achieved by interfering with some of the steps for the electrochemical process.

The corrosion inhibition of mild steel in aggressive acidic solutions has been widely investigated. In industries, hydrochloric acid solutions are often used in order to remove scale and salts from steel surfaces, and cleaning tanks and pipelines. This treatment may be prerequisite for coating by electroplating, galvanizing or painting techniques. The acid must be treated to prevent an extensive dissolution of the underlying metal. This treatment involves the addition of some organic inhibitors to the acid solution that adsorb at the metal/solution interface by displacing water molecules on the surface and forming a compact barrier film.

Various nitrogen-containing compounds have been used by many authors in their corrosion inhibition investigations. These compounds included quaternary ammonium salts [3-10], polyamino-benzoquinone polymers [11], azoles [12-14], substituted aniline-N-salicylidenes [15], amides [16], heterocyclic compounds [17, 18], and cationic surfactants [19, 20]. Other authors worked on sulfur-containing inhibitors [21-28], and phosphorus-containing compounds [29]. Other studies involved the effect of addition of some ions on the

inhibition efficiency of some organic compounds. These ions included chromium [30], iodide [31]. Other authors [32] have studied the inhibitive effect of organic cations on passive film breakdown of iron in a chloride-containing borate buffer solution. Structural effect of organic compounds as corrosion inhibitors was also studied [33, 34]. Recently, research activities are geared towards developing cheap, non-toxic, and environment friendly corrosion inhibitors [35, 36]. In all these studies, the nitrogen atom(s) in the compounds showed to be able to adsorb very well on the metal surface and form protective layer, which in turns increased the corrosion inhibition with the increase in the concentration of the inhibitor, in some cases reaching 99% inhibition [5].

No studies have been reported on 1-vinylimidazolium-3-methyltriphenyl phosphonium dihexafluoroantimonate (VIMTPPF) (**Scheme 1**), in terms of studying the temperature effect on the corrosion inhibition of mild steel in 1.0M HCl solution. Mild steel was selected in our studies because high temperature aggressive acids are widely used in industries in connection to mild and low alloy steels.



Scheme 1. 1-Vinylimidazolium-3-methyltriphenyl phosphonium dihexafluoroantimonate.

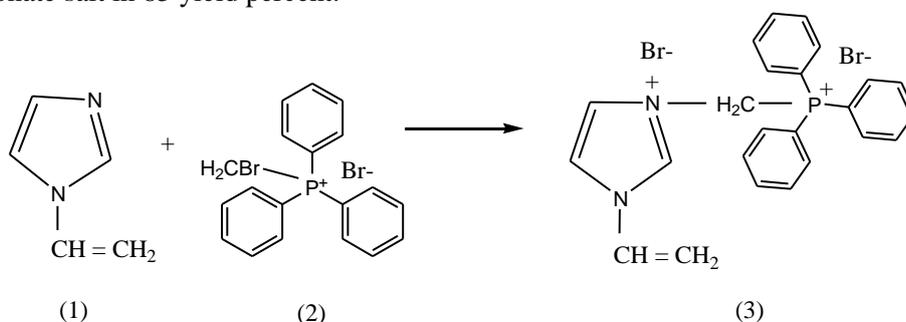
2. Experimental Details

2.1 Synthesis of 1-Vinylimidazolium-3-methyltriphenyl phosphonium dihexafluoroantimonate (VIMTPPF)

The 1-vinylimidazolium-3-methyltriphenyl phosphonium dibromide salt (3) (**Scheme 2**) was prepared by refluxing an equimolar mixture of the 1-vinylimidazole (1) and bromomethyltriphenylphosphonium bromide (2) in acetone as shown below:

2.00 grams (0.021mol) 1-vinylimidazole (1), and 10.00 grams bromomethyl-triphenylphosphonium bromide (0.023 mol) (2), in 100 mL acetone were refluxed for two hours. After cooling to room temperature, a crystalline solid of salt (3) was collected by filtration, then washed with excess diethyl ether, and dried.

The bromide salt (3) was converted to the hexafluoroantimonate by treatment with two moles potassium hexafluoroantimonate (K₂SbF₆) in methanol/water mixture 70/30 % by volume. After stirring for 20 minutes, the reaction product was filtered, washed several times with water, then with diethyl ether, and dried under vacuum at room temperature. Dissolution in chloroform and addition of excess diethyl ether gave the pure hexafluoroantimonate salt in 85 yield percent.



Scheme 2. Synthesis of 1-vinylimidazolium-3-methyltriphenyl phosphonium dibromide.

2.2 Specimen Preparation

Mild steel specimens of composition (wt %): C (0.18), Si (0.35), Mn (0.6), and Fe (balance), Rectangular specimens (1.0cm x 2.3cm x 0.3 cm) cut from large sheet of 3 mm thick mild steel (IS 226 containing 0.18% C, 0.6% Mn, and 0.35% Si) supplied by "Reliable Steel Traders", Sharjah, UAE; were used for weight loss

measurements. A 2-mm diameter hole was drilled close to the upper edge of the specimen and served to be hooked with a glass rod for immersion purposes. Prior to each experiment, the specimens were polished with 600 grade emery paper, rinsed with distilled water, degreased with acetone, dried, and finally weighed precisely on an accurate analytical balance.

2.3 Instrumentation

A 250-mL round bottom flask fitted with a reflux condenser and long glass rod which served to hook and immerse the specimen and in turn immersed in a thermally controlled water bath was used.

2.4 Measuring Procedure

The round bottom flask was filled with 100 mL of 1.0M HCl solution with and without various concentrations of 1-vinylimidazolium-3-methyltriphenyl phosphonium dihexafluoroantimonate (VIMTPPF), and placed in thermal water bath. When the required working temperature was reached, the mild steel sample was immersed in the solution, and left there for exactly six hours, after which the sample was removed, rinsed with distilled deionized water, degreased with acetone, dried, and finally weighed precisely on an accurate analytical balance. This procedure was repeated with a variety of inhibitor concentrations ranging from 5.0×10^{-7} M up to 5.0×10^{-4} M and at temperatures ranging from 303 to 343 K.

3. Results and Discussions

Weight loss corrosion tests were carried out on the mild steel in 1.0M HCl in the absence and presence of (VIMTPPF) over an immersion period of 6 hours. **Table 1** represents the corrosion rates [$\text{mg.cm}^{-2}.\text{h}^{-1}$], and the percentage efficiencies [%] for the studied inhibitor with concentrations varying from 5.0×10^{-7} M up to 5.0×10^{-4} M at 303, 313, 323, 333, and 343 K, respectively. The percentage efficiency was calculated according to the following expression:

$$\% \text{ Inhibition} = \frac{W_{Uninh.} - W_{Inh.}}{W_{Uninh.}} \times 100 \quad (1)$$

Where: $W_{Uninh.}$ = corrosion rate without inhibitor; and

$W_{Inh.}$ = Corrosion rate with inhibitor.

Table 1. Effect of concentration of 1-vinylimidazolium-3-methyltriphenyl phosphonium dihexafluoroantimonate (VIMTPPF) on the corrosion rate ($\text{mg.cm}^{-2}.\text{h}^{-1}$) and percentage efficiency of mild steel in 1.0 M HCl at various temperatures.

Concentration of Inhibitor	Temperature / K									
		303		313		323		333		343
	Corr. Rate	% Effic-ieny	Corr. Rate	% Effic-ieny	Corr. Rate	% Effic-ieny	Corr. Rate	% Effic-ieny	Corr. Rate	% Effic-ieny
1.0 M HCl	0.961	—	1.394	—	4.671	—	12.225	—	26.280	—
1.0 M HCl + 5.0×10^{-7} M	0.489	49.1	0.878	37.0	3.474	25.6	10.401	14.9	24.526	6.7
1.0 M HCl + 5.0×10^{-6} M	0.374	61.1	0.657	52.9	2.887	38.2	8.852	27.6	21.642	17.6
1.0 M HCl + 5.0×10^{-5} M	0.197	79.5	0.432	69.0	1.820	61.0	6.477	47.0	16.103	38.7
1.0 M HCl + 5.0×10^{-4} M	0.115	88.0	0.264	81.1	1.338	71.4	4.873	60.1	12.768	51.4

The plots of the corrosion rate of (VIMTPPF) as a function of concentration at 303, 313, 323, 333, and 343 K are shown in **Figures 1** and **2**. At 303 K (**Figure 1**) the corrosion rate dropped from $0.961 \text{ mg.cm}^{-2}.\text{h}^{-1}$ (1.0M

HCl in the absence of the inhibitor) to $0.489 \text{ mg.cm}^{-2}.\text{h}^{-1}$ when $5.0 \times 10^{-7} \text{ M}$ of (VIMTPPF) was present in the 1.0M HCl. This corrosion rate continued to decrease slightly to reach $0.374 \text{ mg.cm}^{-2}.\text{h}^{-1}$ (61.1 % inhibition) at a concentration of $5.0 \times 10^{-6} \text{ M}$, and continued to decrease to reach $0.197 \text{ mg.cm}^{-2}.\text{h}^{-1}$ when the inhibitor's concentration was $5.0 \times 10^{-5} \text{ M}$; and finally, at higher concentration ($5.0 \times 10^{-4} \text{ M}$) the corrosion rate decreased further to reach $0.115 \text{ mg.cm}^{-2}.\text{h}^{-1}$ (88.0 % inhibition). At 313 K (Figure 1), the corrosion rate decreased as the inhibitor concentration increased from $5.0 \times 10^{-7} \text{ M}$ to reach $0.264 \text{ mg.cm}^{-2}.\text{h}^{-1}$ (81.1 %) at $5.0 \times 10^{-4} \text{ M}$.

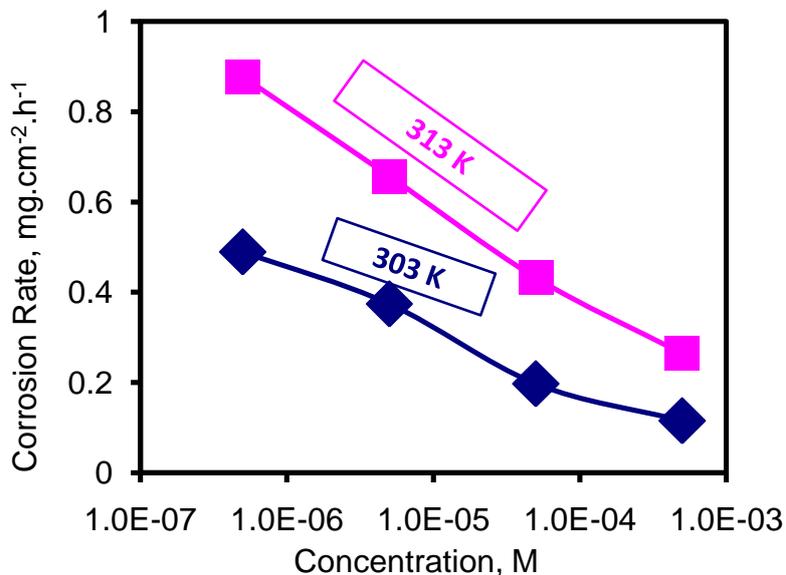


Figure 1. Effect of concentration of VIMTPPF on the corrosion rate ($\text{mg.cm}^{-2}.\text{h}^{-1}$) of mild steel in 1.0 M HCl at various temperatures. ◆303 K ■ 313 K

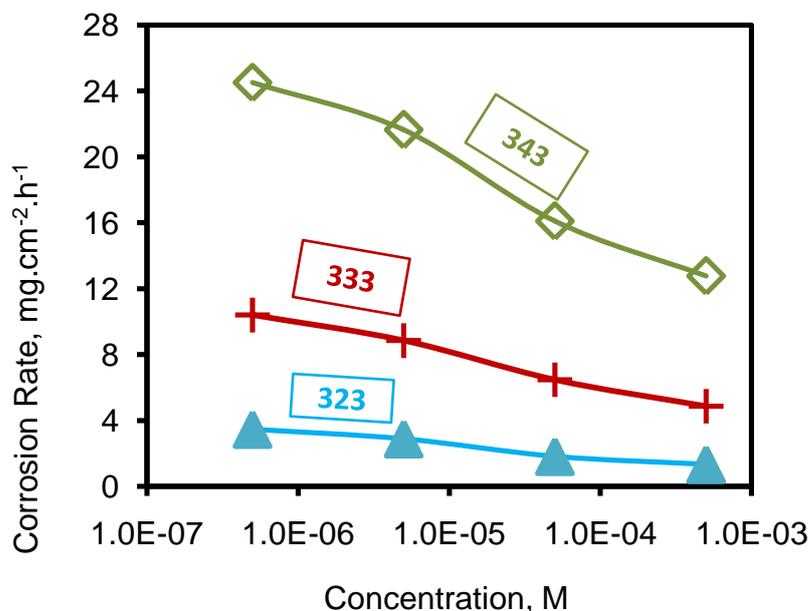


Figure 2. Effect of concentration of VIMTPPF on the corrosion rate ($\text{mg.cm}^{-2}.\text{h}^{-1}$) of mild steel in 1.0 M HCl at various temperatures. ▲323 K+333 K◇343 K

In Figure 2, the corrosion rate at 323, 333, and 343 K are shown respectively as a function of the concentration of (VIMTPPF). It can be clearly seen that the presence of the (VIMTPPF) inhibitor at these high temperatures has acted as a corrosion inhibitor reaching a percent inhibition of 71.4%, 60.1%, and 51.4% when $5.0 \times 10^{-4} \text{ M}$ inhibitor was employed at 323K, 333K, and 343 K respectively.

Figure 3 shows the plots of the % inhibition versus the concentration of the inhibitor at temperatures of 303, 313, 323, 333, and 343 K, respectively. This figure showed that the % inhibition was sufficiently affected by the increase of temperature (303 to 343K), when all concentrations (5.0×10^{-7} to 5.0×10^{-4} M) of inhibitors were employed; and the presence of the inhibitor has greatly increased the % inhibition at all temperatures. The data obtained from the weight loss measurements (**Table 2**) were plotted in accordance to Arrhenius equation:

$$\ln \text{rate} = -\frac{E_a}{RT} + \text{const.} \quad (2)$$

Where: E_a = activation energy [kcal.mol^{-1}], R = gas constant [kcal.mol^{-1}],
 T = absolute temperature [K], and const. = constant

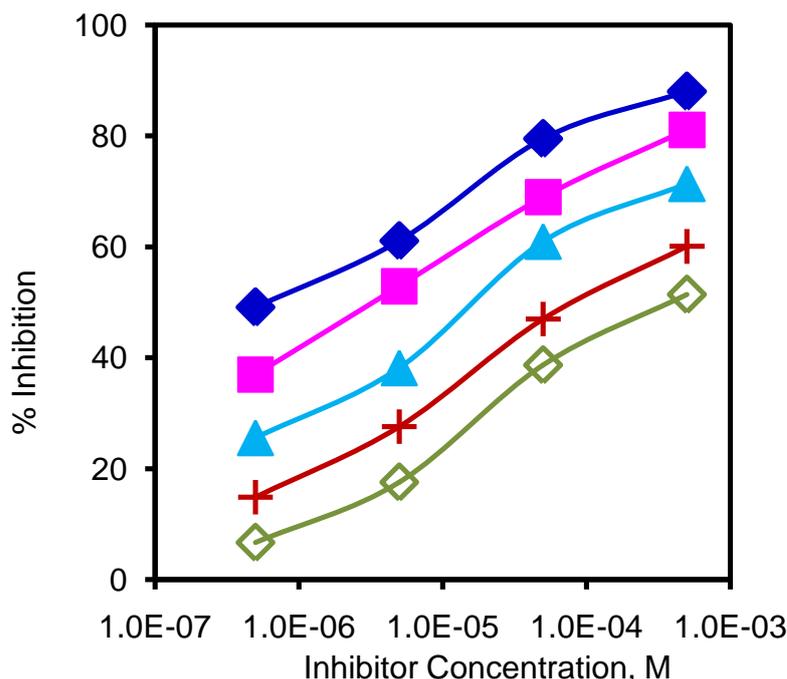


Figure 3. Effect of concentration of VIMTPPF on the percent inhibition of mild steel in 1.0 M HCl at various temperatures. ◆303 K ■ 313 K ▲323 K +333 K ◇343 K

Table 2. The Data obtained from the weight loss measurements for Arrhenius Equation: $(1/T)$ against \ln Corrosion Rate.

$(1/T) \times 10^3 \text{ K}^{-1}$	Ln Corrosion Rate ($\text{mg.cm}^{-2}.\text{h}^{-1}$)				
	1.0 M HCl	1.0 M HCl + 5.0×10^{-7} M	1.0 M HCl + 5.0×10^{-6} M	1.0 M HCl + 5.0×10^{-5} M	1.0 M HCl + 5.0×10^{-4} M
3.30	-0.03978	-0.71539	-0.9835	-1.62455	-2.16282
3.19	0.332177	-0.1301	-0.42007	-0.83933	-1.33181
3.10	1.541373	1.24531	1.06022	0.59884	0.29118
3.00	2.503483	2.3419	2.18064	1.86826	1.58371
2.92	3.268808	3.19973	3.07464	2.77901	2.54694

Figure 4 represents the Arrhenius plot of the corrosion of mild steel in 1.0 M HCl solution (\ln corrosion rate as a function of $1/T$) with and without the presence of VIMTPPF at concentrations ranging from 5.0×10^{-7} to 5.0×10^{-4} M. From this Figure, the slope ($-\frac{E_a}{R}$) of each individual line was determined and used to calculate the activation energy according to **equation 2**, and taking $R = 1.987 \times 10^{-3} \text{ kcal.mol}^{-1}$ (**Table 3**). The increase of

concentration of (VIMTPPF) (from $5.0 \times 10^{-7} \text{ M}$ to $5.0 \times 10^{-4} \text{ M}$), increased the activation energies for the corrosion of mild steel in 1.0 M HCl (initially $18.27 \text{ kcal.mol}^{-1}$) (Table 2). The increase in the activation energies for the corrosion is attributed to a decrease in the adsorption of the inhibitor on the metal surface as the temperature increased; and subsequently, an increase in the corrosion rate will result due to the greater exposed area of the metal surface to the acid.

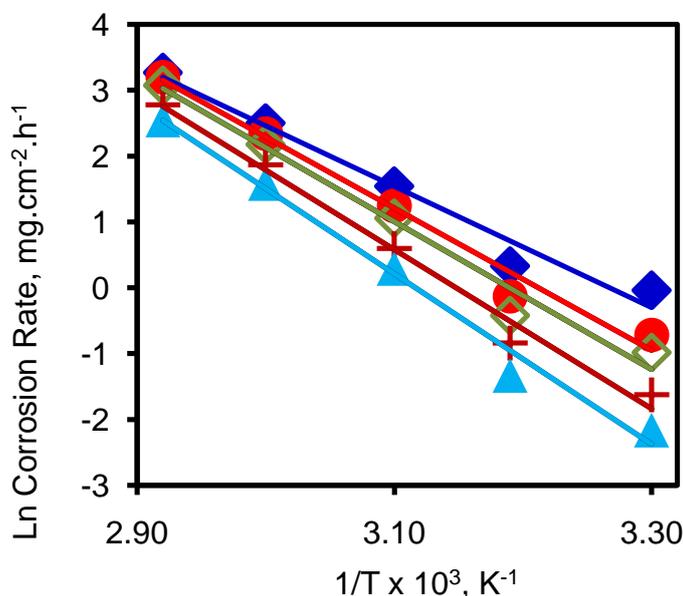


Figure 4. Effect of temperature on the corrosion rate of mild steel in 1.0 M HCl solution with and without 1-vinylimidazolium-3-methyltriphenyl phosphonium dihexafluoroantimonate (VIMTPPF) at various concentrations. \blacklozenge 1.0 M HCl \blacktriangle $5.0 \times 10^{-4} \text{ M}$ $+$ $5.0 \times 10^{-5} \text{ M}$ \blacklozenge $5.0 \times 10^{-6} \text{ M}$ \bullet $5.0 \times 10^{-7} \text{ M}$

The results in Table 3, show that the activation energy (E_a) for the corrosion of mild steel in the presence of the inhibitor are higher compared to the activation energy in the absence of inhibitor at all concentrations ranging from $5.0 \times 10^{-4} \text{ M}$ to $5.0 \times 10^{-7} \text{ M}$ (from about 26 to 21 kcal.mol^{-1}). This can be attributed to the fact that higher values of E_a in the presence of inhibitor compared to its absence are generally consistent with a physisorption, while unchanged or lower values of E_a in inhibited solution suggest charge sharing or transfer from the organic inhibitor to the metal surface to form coordinate covalent bonds [37].

Table 3. The activation energy (E_a) for the corrosion of mild steel in 1.0 M HCl with and without 1-vinylimidazolium-3-methyltriphenyl phosphonium dihexafluoroantimonate (VIMTPPF) inhibitor at various concentrations.

System	Activation Energy, E_a (kcal.mol^{-1})			
	$5.0 \times 10^{-4} \text{ M}$	$5.0 \times 10^{-5} \text{ M}$	$5.0 \times 10^{-6} \text{ M}$	$5.0 \times 10^{-7} \text{ M}$
1.0 M HCl	18.27	18.27	18.27	18.27
1.0 M HCl + Inhibitor	25.71	24.00	22.29	21.44

Table 4 shows the surface coverage of various concentrations of (VIMTPPF) (from $5.0 \times 10^{-7} \text{ M}$ to $5.0 \times 10^{-4} \text{ M}$) on mild steel surface as a function of temperature. These values were extracted from the corresponding % efficiency values reported earlier in Table 1. The plot of surface coverage, θ , against the natural logarithm of the concentration, $\ln C$, for mild steel in the presence of the various inhibitor concentrations is shown in Figure 5. After examining the data and adjusting them to different theoretical adsorption isotherms, it was concluded that all inhibitors were adsorbed on the mild steel surface according to Temkin isotherm:

$$-2a \theta = \ln K C \quad (3)$$

Where: a = molecular interaction constant,
 θ = degree of coverage,
 K = equilibrium constant for the adsorption reaction, and
 C = concentration of the inhibitor.

Table 4. Effect of concentration of VIMTPPF on surface coverage for mild steel in 1.0 M HCl at various temperatures.

	Temperature / K				
	303	313	323	333	343
Concentration of Inhibitor	Surface Coverage θ				
1.0 M HCl + 5.0x10 ⁻⁷ M	0.491	0.370	0.256	0.149	0.067
1.0 M HCl + 5.0x10 ⁻⁶ M	0.611	0.529	0.382	0.276	0.176
1.0 M HCl + 5.0x10 ⁻⁵ M	0.795	0.690	0.610	0.470	0.387
1.0 M HCl + 5.0x10 ⁻⁴ M	0.880	0.811	0.714	0.601	0.514

The equilibrium constant for the adsorption reaction, K , is related to the standard free energy of adsorption via the following equation given by Damaskin et al [1]:

$$K = \frac{1}{55.5} \exp\left(-\frac{\Delta G}{RT}\right) \quad (4)$$

Where: K = equilibrium constant for the adsorption reaction,
 55.5 = concentration of water [mol.L⁻¹],
 ΔG = standard free energy [kcal.mol⁻¹],
 R = gas constant [kcal.mol⁻¹], and
 T = absolute temperature [K].

According to equation 3, the straight lines shown in **Figure 5** will have the following slopes and intercepts:

$$\text{Slope} = -\frac{1}{2a} \quad (5)$$

$$\text{Intercept} = -\frac{1}{2a} \ln K \quad (6)$$

Combination of equations (5) and (6) leads to the following relationship:

$$\text{Intercept} = \text{Slope} \cdot (\ln K) \quad (7)$$

from which the equilibrium constant for the adsorption reaction, K , can be calculated:

$$K = e^{\left(\frac{\text{Intercept}}{\text{Slope}}\right)} \quad (8)$$

The standard free energy of adsorption of the inhibitor, ΔG^0 , can be calculated from the results in **Figure 5** used to calculate the equilibrium constant, K , and equation 4 at various temperatures (303 to 343 K).

The enthalpy of adsorption, ΔH^0 , for the inhibitor can be calculated from the following equation:

$$\Delta H^0 = E_a - RT \quad (9)$$

The entropy, ΔS^0 , can be calculated at various temperatures for the inhibitor using the following equation:

$$\Delta G^0 = \Delta H^0 - T\Delta S^0 \quad (10)$$

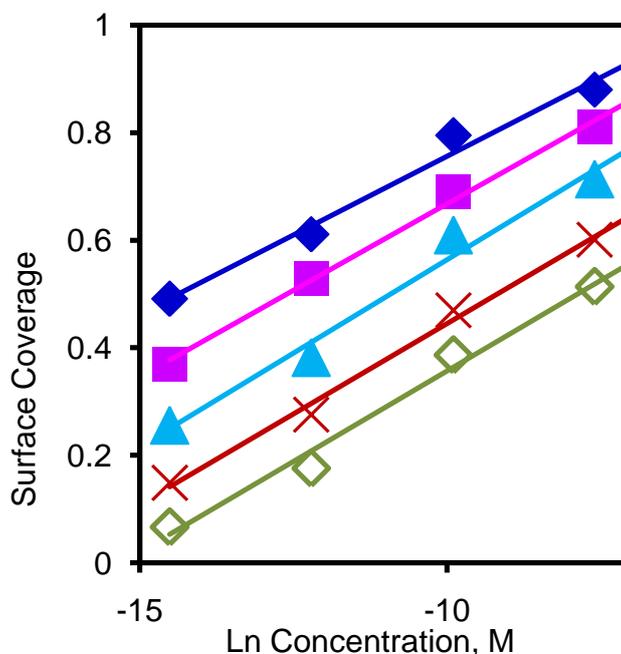


Figure 5. Effect of concentration of VIMTPPF on the surface coverage of mild steel in 1.0 M HCl at various temperatures. ◆303 K ■313 K ▲323 K ✕333 K ◇343 K

Tables 5 to 7 show the thermodynamic data obtained in the presence of the inhibitor at 5.0×10^{-4} M. These thermodynamic quantities represent the algebraic sum of the values for adsorption and desorption. The negative value of ΔG^0 indicates the spontaneous adsorption of inhibitor on the surface of the mild steel. The standard free energy, ΔG^0 , varies from $-16.60 \text{ kcal.mol}^{-1}.\text{K}^{-1}$ at 303 K to $-13.89 \text{ kcal.mol}^{-1}.\text{K}^{-1}$ at 343 K. The adsorption process is believed to be exothermic and associated with a decrease in entropy (ΔS) of solute, while the opposite is true for the solvent. The gain in entropy which accompanies the substitutional adsorption process is attributed to the increase in the solvent entropy. This agrees with the general suggestion that the values of ΔG^0 increase with the increase of inhibition efficiency as the adsorption of organic compound is accompanied by desorption of water molecules off the surface [25, 38].

The high inhibition efficiency may be attributed to the preferred flat orientation of this compound on the metal surface. The interaction occurs between the delocalized π -electrons of the four aromatic rings (three benzene rings and one imidazole ring), the double bond ($-\text{C}=\text{C}-$), and the lone pair of electrons on N atom with the positively charged metal surface [39].

Table 5. The enthalpy of adsorption (ΔH) for mild steel in 1.0 M HCl in the presence of 5.0×10^{-4} M VIMTPPF at various temperatures (303 K – 343 K).

Enthalpy, ΔH (kcal.mol ⁻¹)				
303 K	313 K	323 K	333 K	343 K
25.11	25.09	25.07	25.05	25.03

Table 6. The Free energy of adsorption (ΔG_{ads}) for mild steel in 1.0 M HCl in the presence of VIMTPPF at various temperatures (303 K – 343 K).

Free Energy, ΔG (kcal.mol ⁻¹)				
303 K	313 K	323 K	333 K	343 K
-16.60	-15.25	-14.29	-13.69	-13.89

Table 7. The change in entropy (ΔS) for mild steel in 1.0 M HCl in the presence of VIMTPPF at various temperatures (303 K- 343 K).

Entropy, ΔS (kcal.K ⁻¹ .mol ⁻¹)				
303 K	313 K	323 K	333 K	343 K
0.137	0.129	0.122	0.116	0.113

Conclusion

1-vinylimidazolium-3-methyltriphenyl phosphonium dihexafluoroantimonate (VIMTPPF) was found to be a very efficient inhibitor for carbon steel in 1.0M HCl solution, reaching about 88 % at 5.0×10^{-4} M at 303 K, a concentration which is considered to be very moderate.

1-vinylimidazolium-3-methyltriphenyl phosphonium dihexafluoroantimonate (VIMTPPF) is a potential corrosion inhibitor since it contains not only a nitrogen atom, but also three phenyl rings. It was apparent from the molecular structure that this compound would be adsorbed onto the metal surface through the lone pair of electrons of nitrogen and p-electrons of the four aromatic rings as well as the double bond that is attached to imidazole ring. The percentage inhibition in the presence of this inhibitor decreased with the increase of temperature, which indicated that physical adsorption was the predominant inhibition mechanism.

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