



## EXPERIMENTAL AND THEORETICAL STUDIES OF POLYVINYLPIRROLIDONE-IODINE ON CARBON STEEL CORROSION IN 1M HYDROCHLORIC SOLUTION

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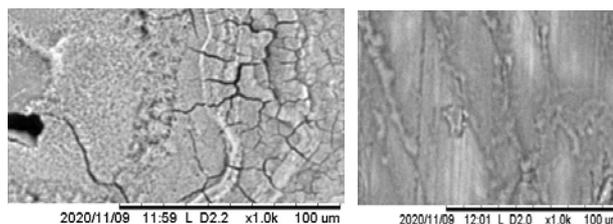
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The corrosion inhibition of polyvinylpyrrolidone-iodine (PVP-I), for carbon steel in a 1 M hydrochloric acid environment was investigated by means of weight loss, X-ray diffraction (XRD) and scanning electron microscopy (SEM) techniques. The experimental results were completed by quantum chemical calculations using density functional theory (DFT) method at the B3LYP/6-31G\* level of theory. The effects of inhibitor concentration, temperature and immersion time on the corrosion of CS were also studied. The thermodynamic parameters for both adsorption and dissolution processes were evaluated and discussed from the weight loss method. The Gibb's free energy change of adsorption was negative and the adsorption of this inhibitor on carbon steel surface in HCl medium was found to be feasible, spontaneous and it occurred by mixte adsorption with a tendency towards chemisorption. The results indicated that polyvinylpyrrolidone-iodine could form a protective layer on the metal surface, which protected the metal against corrosion.



### INTRODUCTION

Carbon steels are composed of a mixture of metallic iron and carbon which are broadly used in diverse domains like petroleum industries due to its high mechanical strength and cost-effectiveness.<sup>1</sup> To eliminate salt deposits and unwanted scales and in order to enhance oil recovery, processes such as oil well acidization, pickling and descaling that are carried out using mineral acids. But the applications of these acids are the origin of an aggressive attack and deterioration of the steel surface. The consequences of this loss damage generate great cost for replacement and renovation of several

equipment and causes public and environmental losses.<sup>2,3</sup> The most obvious way to address carbon steel destruction is by utilizing anti-corrosion inhibitors, when it is added in a little quantity to a corrosive environment regulates the rate of metal dissolution by building a preventive barrier film, which in return prevents the corrosive reaction from progressing.<sup>4-6</sup> Scientists are trying to discover and synthesize new compounds that are nontoxic, inexpensive, environment friendly and have high effectiveness in inhibiting corrosion.<sup>7,8</sup> Polymers have received major regard for replacing synthetic organic and inorganic inhibitors from they are inexpensive and dangerous for the environment. Polyvinylpyrrolidone-iodine (or

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Povidone iodine) has been studied due to its efficiency inhibition, very low toxicity, solubility, low cost, and is also used like disinfectants and antiseptics.<sup>9</sup> Preparations containing povidone iodine are employed in acute surgical care for the prevention and control of infections in various indications.<sup>10</sup> The aim of the present work is to study the inhibitive effect of polyvinylpyrrolidone-iodine on carbon steel surface in 1 M hydrochloric solution. PVP-I inhibition was confirmed through weight loss, X-ray diffraction and scanning electron microscopy analysis, this work is completed by using quantum chemical calculations. Additionally, kinetic and thermodynamic data are evaluated.

## MATERIALS AND METHODS

### Material Preparation

Carbon steel samples used as test materials contain (%wt): C: 0.37%, Mn: 0.68%, Cu: 0.16%, Cr: 0.077%, Ni: 0.059%, Si: 0.023%, S: 0.016%, Ti: 0.011%, Co: 0.009% and the remainder Fe. Before every proceeding, the specimen was abrading using emery papers with various grades. After that, the sample was washed with distilled water, acetone and finally dried. Each sample was weighed by an electronic balance ( $\pm 0.0001$  g) and then immersed in the acid solution. Polyvinylpyrrolidone-iodine, acetone and hydrochloric acid were purchased from Sigma Aldrich.

### Gravimetric method

Weight loss techniques provide more realistic results with regard to uniform corrosion than electrochemical techniques because the experimental conditions simulate real life conditions. The experiences were performed in triplicate, and the results of weight loss data were used to calculate the corrosion rate ( $CR$ ) as follows:<sup>11,12</sup>

$$CR = w / (S \times t) \quad (1)$$

where  $w$  is the weight loss (mg),  $S$  is the total area of the specimen ( $\text{cm}^2$ ) and  $t$  is the exposure time (h). The inhibition efficiency (IE %) and the surface coverage ( $\theta$ ) were calculated from the  $CR$  values:

$$IE (\%) = 100 \times (1 - CR_{inh} / CR) \quad (2)$$

$$\theta = 1 - CR_{inh} / CR \quad (3)$$

where  $CR$  and  $CR_{inh}$  denote the obtained corrosion rates in uninhibited and inhibited solution, respectively.

## RESULTS AND DISCUSSION

### Effect of Concentration and Temperature on Inhibition Efficiency

The elevation of temperature causes so many complex modifications like decomposition, desorption of inhibitor and rapid etching on the metal surface, which invited the attention of many studies towards this.<sup>13,14</sup>

The weight loss studies were carried out different concentrations of inhibitor in the temperature varying to 293 to 333 K. The increase of IE at a given temperature with the increase in concentration of the inhibitor is due to the availability of a large number of adsorption sites that are brought by many molecule concentrations of Polyvinylpyrrolidone-iodine. Figure 1 indicates that the inhibition efficiency presents a lowering trend with the elevation in temperature can be show that the molecules of inhibitor undergo desorption from the steel surface.

### Immersion time

To examine the stability of the performance of polyvinylpyrrolidone-iodine, the effect of exposure time was investigated in the presence of optimum concentration (5 mg/L) on the carbon steel samples at 303 K (Figure 2). The obtained results indicated that the immersion time had a significant effect on the weight loss and inhibition efficiency. Actually, the weight loss increased with immersion time (both the uninhibited and the inhibited samples). However, comparing the amount of weight lost, the uninhibited specimen (blank) recorded higher weight loss than the inhibited sample. Thus, the noticed increase in weight loss could be attributed to the increased time of immersion in the aggressive/unfavourable environment, making the steel to be "wet" for an extended time. There was an important increase of IE with an increase in the exposure time of the inhibited solution ( $\sim 0.5$ – $2$  h) and became almost constant (from 2 to 3 h with 88.97% to 87.29%, respectively), then it decreased after this period to 52.87% within 24 hours. The decrease of IE was affected to the desorption of corrosion inhibitors from metal surface.<sup>9,15</sup>

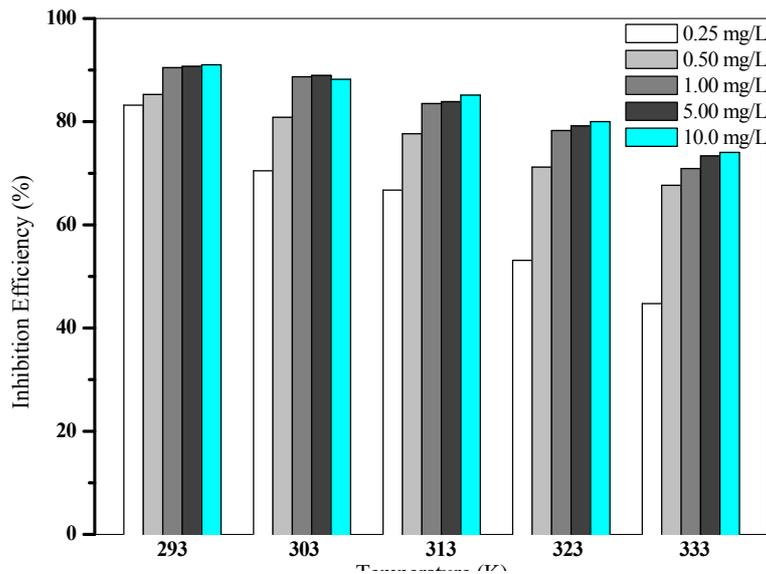


Fig. 1 – Plot of inhibition efficiency ( $IE$  %) against solution temperature (293–333 K) at different concentrations (0.25 to 10 mg/L) of polyvinylpyrrolidone-iodine.

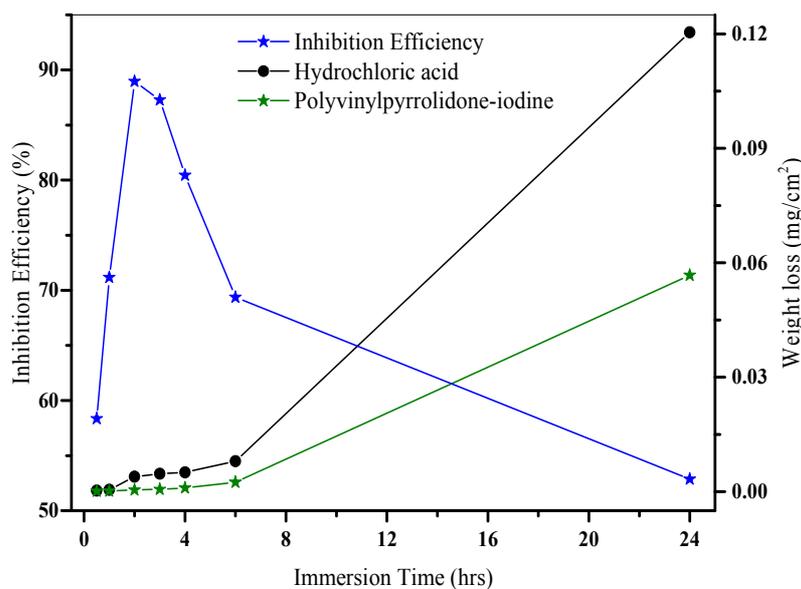


Fig. 2 – Variation of inhibition efficiency and weight loss vs immersion time for carbon steel at 5 mg/L of polyvinylpyrrolidone-iodine.

### Activation energy calculations

The Arrhenius equation has been applied strongly to prove the effect of temperature change on the inhibition action of polyvinylpyrrolidone-iodine. It is clarified by using the eq 4:<sup>16</sup>

$$CR = k \times \exp(-E_a/RT) \quad (4)$$

where  $CR$  is the corrosion rate of carbon steel,  $k$  is Arrhenius pre-exponential factor,  $E_a$  (kJ/mol) is the

activation energy,  $R$  is the gas constant (8.314 J/mol K) and  $T$  is the temperature (K).

Values of  $E_{act}$  for the corrosion reaction with and without of different concentrations of inhibitor were obtained from the slope of the plot of  $\ln(CR)$  against  $1/T$  (Figure 3) and given in Table 1. The values of activation entropy  $\Delta S_{act}$  and activation enthalpy  $\Delta H_{act}$  can be calculated by the intercept and slope of the above plot (Figure 4), using the following transition state equation (5).<sup>17</sup>

$$\ln(CR/T) = \ln(R/K_4h) + \Delta S_{act}/R - \Delta H_{act}/RT \quad (5)$$

where  $h$  is Plank's constant and  $N_A$  is Avogadro number. A plot of  $\ln(CR/T)$  versus  $1/T$  gave a straight line with an intercept of  $(\ln(R/N_A h) + (\Delta S_{act}/R))$  and a slope of  $(-\Delta H_{act}/R)$  from which the values of  $\Delta S_{act}$  and  $\Delta H_{act}$  can be calculated and presented (Table 1).

The  $\Delta G_{act}$  of the corrosion process can be determined at any temperature by using the equation:

$$\Delta G_{act} = \Delta H_{act} - T\Delta S_{act} \quad (6)$$

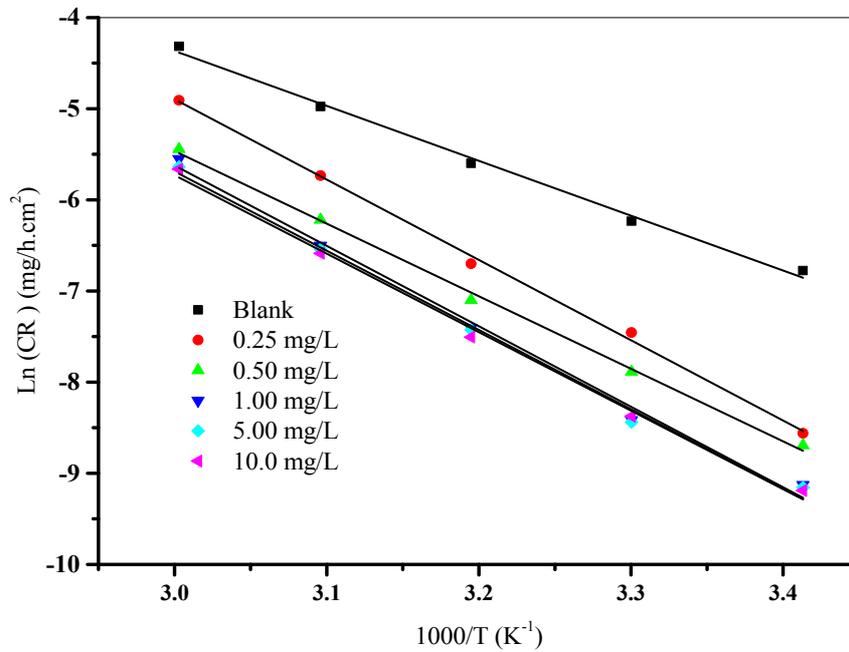


Fig. 3 – Ln (CR) versus  $1/T$  curves for carbon steel dissolution without and with various concentrations of polyvinylpyrrolidone-iodine in 1 M HCl solution.

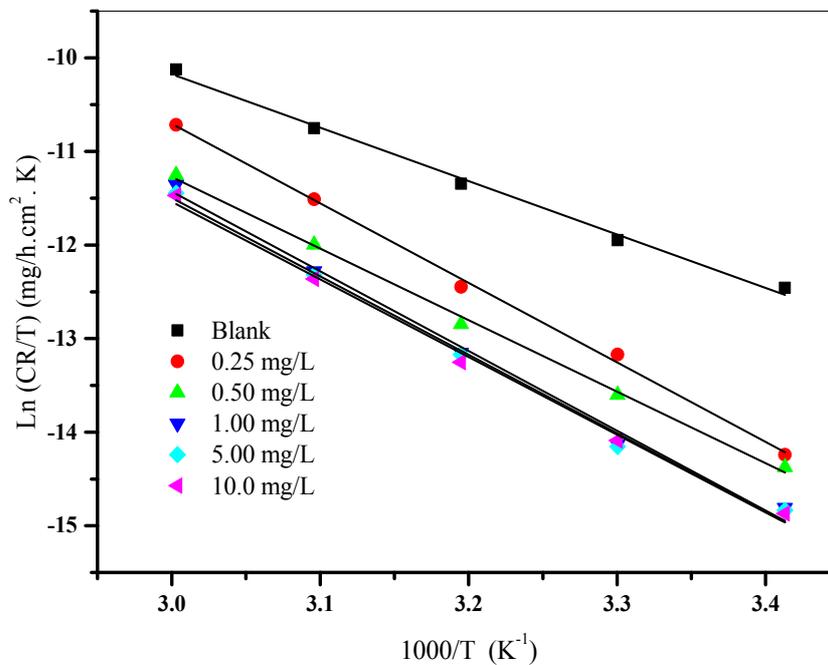


Fig. 4 – Ln (CR/T) versus  $1/T$  curves for carbon steel dissolution without and with various concentrations of polyvinylpyrrolidone-iodine in 1 M HCl solution.

Table 1

The thermodynamic activation parameters in the absence and presence of different concentrations of polyvinylpyrrolidone-iodine in 1 M hydrochloric acid

C (mg/L)	R <sup>2</sup>	E <sub>act</sub> (kJ/mol)	ΔH <sub>act</sub> (kJ/mol)	ΔS <sub>act</sub> (J/mol K)	ΔG <sub>act</sub> (kJ/mol)				
					293K	303K	313K	323K	333K
Blank	0.997	50.07	47.47	-139.71	88.40	89.80	91.20	92.59	93.99
0.25	0.999	73.27	70.67	-74.52	92.50	93.25	93.99	94.73	95.48
0.50	0.999	66.19	63.59	-100.48	93.03	94.03	95.04	96.05	97.05
1.00	0.996	73.42	70.82	-80.07	94.28	95.08	95.88	96.68	97.48
5.00	0.997	72.32	69.72	-83.95	94.31	95.15	95.99	96.83	97.67
10.0	0.997	71.59	68.99	-86.45	94.32	95.18	96.04	96.91	97.77

The values of activation energy obtained (Table 1) in presence of inhibitor are higher than those obtained in free acid solution (blank) showing that the adsorption of polyvinylpyrrolidone-iodine in 1M hydrochloric acid is a physical adsorption. The increase of E<sub>act</sub> is assigned to the appreciable decrease in the adsorption of PVP-I on carbon steel surface by increase in the temperature. The positive signs of enthalpy activation without and with different concentrations of PVP-I reflect the endothermic nature of the steel dissolution.<sup>18</sup> ΔS<sub>act</sub><0 in the presence of various concentrations of polyvinylpyrrolidone-iodine implying that a decrease in disorder occurred when proceeding from the reactant to the product. The positive values of free Gibbs energy mean a non-spontaneous corrosion reaction and they increase with increasing of both the concentration of inhibitor and temperature. The higher values of

ΔG<sub>act</sub> of the process with presence of inhibitor when compared to that in its absence are attributed to its physisorption, while the opposite one is the case with chemisorptions.

### Inhibition Mechanism and Adsorption Isotherm

Basic details on the interaction among the carbon steel surface and the inhibitor can be furnished by the adsorption isotherm.<sup>19</sup> The process of adsorption can be considered as replacement action between the PVP-I in the solution phase [PVP-I]<sub>sol</sub> and water molecules at the metal surface [H<sub>2</sub>O]<sub>ads</sub> to give the polymer compounds adsorbed on the steel surface [PVP-I]<sub>ads</sub> and thus increased inhibition efficiency due to subsequent equation:



where,  $x$  is the size ratio and simply equals the number of adsorbed water molecules substituted by a single inhibitor molecule.

In order to get the isotherm, the linear relation among  $C_{inh}/\theta$  values and  $C_{inh}$  must be establish. The best fit (Figure 5) indicates that the Polyvinylpyrrolidone-iodine is adsorbed on metallic surface according to the Langmuir isotherm model. The adsorption equilibrium constant ( $K_{ads}$ ) can be determined by applying the equation 7.<sup>20</sup>

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

The equilibrium constant is a thermodynamic variable that is linked to the free Gibbs energy, and can be determined by using van't Hoff relation:<sup>21</sup>

$$\Delta G_{ads} = -RT \ln (A' \times K_{ads}) \quad (8)$$

where R is the gas constant, T is the absolute temperature (K),  $A'$  is the density of water (1000 g/L) and  $K_{ads}$  is the adsorption equilibrium constant obtained from the isotherm and.

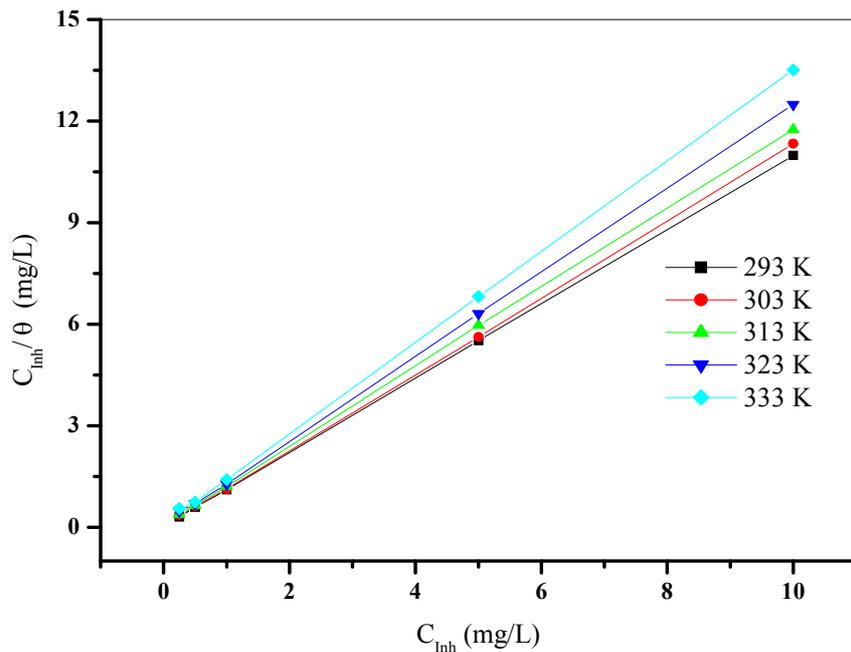


Fig. 5 – Langmuir adsorption isotherms for Polyvinylpyrrolidone-iodine.

Enthalpy of adsorption can be calculated by the Gibbs-Helmholtz equation, which is defined as follows:<sup>22</sup>

$$\Delta G_{ads}/T = \Delta H_{ads}/T - \Delta S_{ads} \quad (9)$$

The change of  $\Delta G_{ads}/T$  with  $1/T$  draw a straight line with a slope that equals  $\Delta H_{ads} = -33.89$  kJ/mol

and intercept equals  $\Delta S_{ads} = 29.92$  J/mol K (Figure 6). It can be shown that the value of  $\Delta G_{ads}/T$  decreases versus  $1/T$  in a linear manner (Figure 6). The equilibrium constants  $K_{ads}$  and the free energy for the adsorption process are summarized in Table 2.

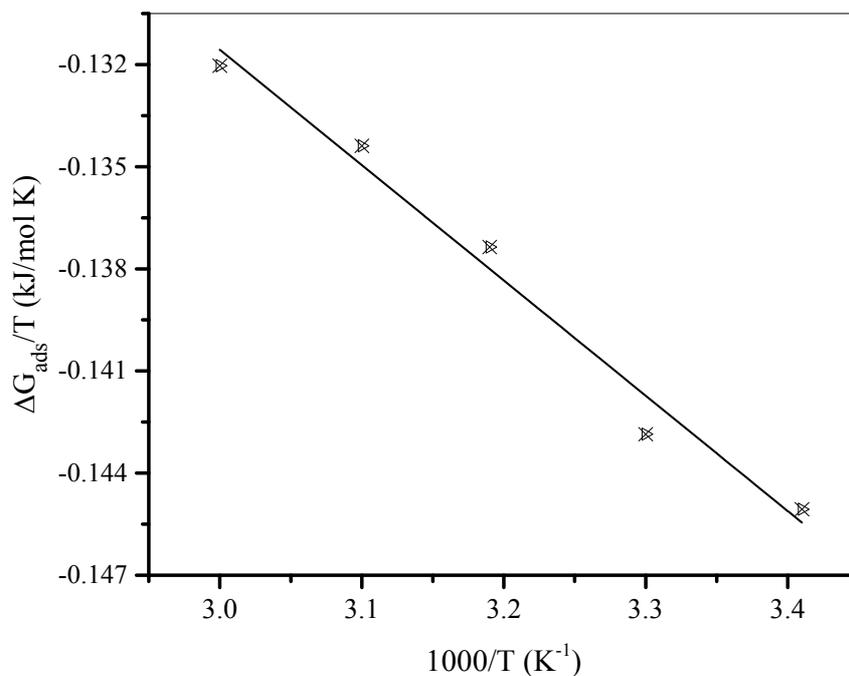


Fig. 6 – The plot of  $(\Delta G_{ads}/T)$  versus reciprocal of absolute temperature,  $1/T$ .

Table 2

Adsorption values calculated from the Langmuir isotherm

T (K)	R <sup>2</sup>	K <sub>ads</sub> (L/mg)	ΔG <sub>ads</sub> (kJ/mol)
293	1	38.47	-42.54
303	0.999	29.07	-43.29
313	0.999	15.39	-43.06
323	0.999	10.25	-43.35
333	0.999	8.03	-44.01

A high value of  $K_{ads}$  indicates that the inhibitor is readily and highly adsorbed on the steel surface, mean it exhibit a better inhibition efficiency of the inhibitor.<sup>1,9</sup> Enthalpy adsorption values can be both the exothermic or endothermic reaction following on the sign of  $\Delta H_{ads}$ . In the literature, exothermic processes are linked to either chemical or physical adsorption whereas endothermic processes are linked to chemical adsorption. The negative values of  $\Delta H_{ads}$  (-33.89 kJ/mol) for the inhibitor suggest an exothermic nature for adsorption. The positive value of  $\Delta S_{ads}$  indicates that the disordering has increased and the negative value of  $\Delta S_{ads}$  shows that the destruction on the steel surface has been lowered. From Table 2, the calculated values of free energy are between -42.54 and -44.01 kJ/mol. In general, when the values of  $|\Delta G_{ads}|$  are 20 kJ/mol or less, the adsorption of inhibitor on steel surface is referred as physisorption. On the other hand, the values of  $|\Delta G_{ads}|$  are 40 kJ/mol or more, the adsorption is considered to be chemisorptions. The increase of Gibb's free energy involves a rise in the spontaneity and stability of the adsorption at a

higher temperature. According to the obtained values of  $\Delta G_{ads}$ , it can be proposed that the polyvinylpyrrolidone-iodine implies both chemical and physical adsorption with a tendency towards chemisorption.

### Scanning Electron Microscope (SEM)

Surface analysis of carbon steel sample before and after of the immersion time in the absence and presence of polyvinylpyrrolidone-iodine in 1 M hydrochloric were examined in order to prove if the inhibition is due to the formation of a protective layer against acid attack on the metal surface (Figure 7).<sup>9, 23</sup> The morphology in Figure 7 (left) shows a pitting and cracking corrosion surface of carbon steel in 1M HCl solution. There are less cracks and pits noticed in the SEM image (Figure 7 right) in the presence of Polyvinylpyrrolidone-iodine which implies a formation of a protective layer on carbon steel surface.<sup>24</sup>

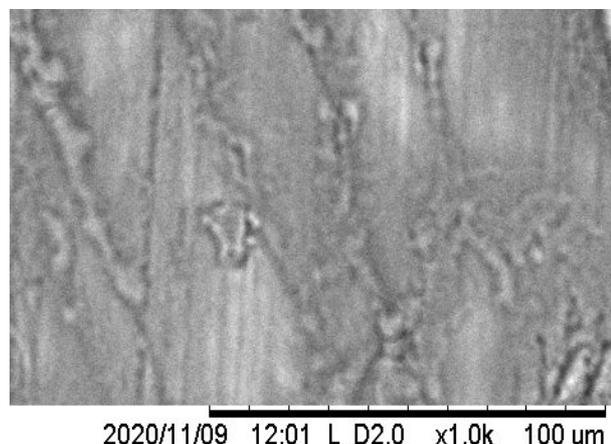
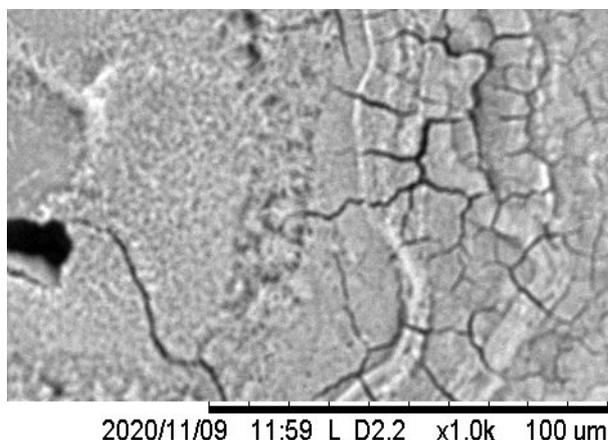


Fig. 7 – Scanning electron micrograph of carbon steel sample after immersion in 1M HCl (left) and in presence of 10 mg/L of PVP-I (right).

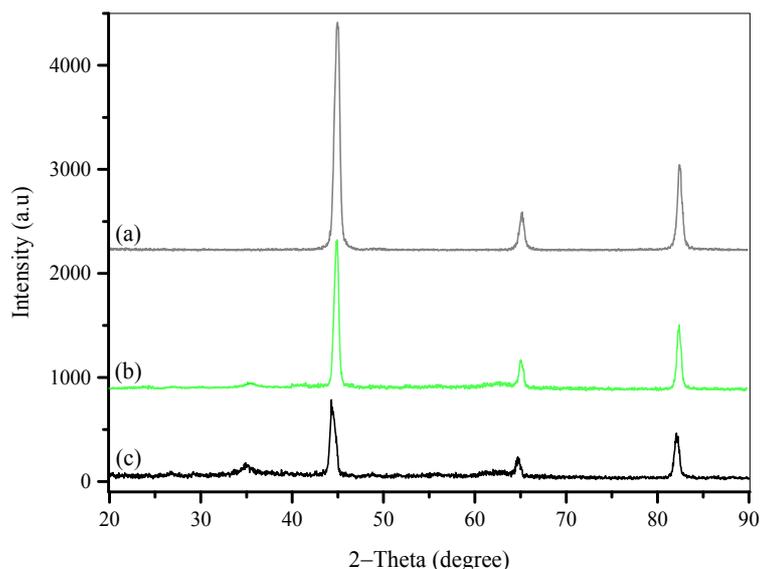


Fig. 8 – XRD pattern of (a) carbon steel (polished), carbon steel in 1 M HCl in presence (b) and absence (c) of polyvinylpyrrolidone-iodine.

### X-Ray Diffraction (XRD)

Analysis of X-Ray diffraction patterns of the surface of the carbon steel samples polished and immersed in 1 M HCl are seen in Figure 8. A series of peaks at 35.1°, 44.9°, 65.3°, and 82.5° are observed, and they are in compliance with the standard iron and chlorides of iron patterns. Peak at 2-Theta = 35.1° can be assigned to FeCl<sub>2</sub>. In Figure 8a is presented the polished sample surface before exposure time in acidic environment, the peaks due to iron appears at 2-Theta = 44.9°, 65.3° and 82.5°. The XRD pattern of the carbon steel immersed in 1 M hydrochloric solution containing 10 mg/L of polyvinylpyrrolidone-iodine is shown in Fig. 8b. It can be observed in Fig. 8c that the surface of carbon steel immersed in HCl alone contains also a peak of FeCl<sub>2</sub> (intensity of peak is 135 a.u). But in the presence of PVP-I (Fig. 8b), the peaks intensity of Fe alone is observed at 2-Theta = 44.9°, 65.3° and 82.5° which are very high, and on the other hand a very small amount 27 a.u of FeCl<sub>2</sub> resulting of the corrosion product compared with Fig. 8c. The formation of a protective layer on the surface of carbon steel in the presence of polyvinylpyrrolidone-iodine is clearly reflected from these observations.

### Quantum chemical calculation

Molecular geometry optimization and quantum chemical calculations were performed using Gaussian 9.0 software. The fundamental basis of Density Functional Theory (DFT) is the use of

electron density in place of wave function for the energy calculations. The DFT study was done at 6-31G\* basis set combined to B3LYP method. The calculated parameters are presented in Table 3 and include the lowest unoccupied molecular orbital energy ( $E_{LUMO}$ ), highest occupied molecular orbital energy ( $E_{HOMO}$ ) and energy gap ( $\Delta E_{gap}$ ). Further calculations using the obtained parameters included the determination of the global hardness ( $\eta$ ), softness ( $S$ ), back-donation of charges ( $\Delta E_{back-donation}$ ) and the number of electrons transferred ( $\Delta N$ ) were calculated. The following Eqs (10–15) were used:<sup>25-31</sup>

$$\Delta E_{gap} = E_{LUMO} - E_{HOMO} \quad (10)$$

$$\mu = 0.5 (E_{LUMO} + E_{HOMO}) \quad (11)$$

$$\eta = 0.5 (E_{LUMO} - E_{HOMO}) \quad (12)$$

$$S = 1/\eta \quad (13)$$

$$\Delta E_{back-donation} = -\eta/4 \quad (14)$$

$$\Delta N = (\chi_{Fe} - \chi_{Inh}) / 2(\eta_{Fe} + \eta_{Inh}) \quad (15)$$

where  $\chi_{Inh}$  and ( $\chi_{Fe} = 7.0$  eV/mol) are the absolute electronegativities of inhibitor and iron,  $\eta_{Inh}$  and ( $\eta_{Fe} = 0$  eV/mol) are the absolute hardness of the inhibitor and iron, respectively.

The high value of HOMO energy expresses a tendency to lose electrons to appropriate gain molecule of low empty molecular orbital energy. The gap energy level of molecule is also important parameter that needs to be considered. The smaller value of  $\Delta E_{gap}$  of an inhibitor involved higher inhibition efficiency.

Table 3

Quantum chemical parameters derived from the B3LYP/6-31G\* level

Substrate /(ev)	E <sub>HOMO</sub>	E <sub>LUMO</sub>	$\eta$	S	$\Delta E_{\text{gap}}$	$\Delta N$	$\Delta E_{\text{back-donation}}$
PVP-I	3.02	8.49	2.73	0.36	5.47	2.33	-0.68

In general, good corrosion inhibitor is the compound which not only donates electrons to unoccupied orbital of the metal, but also accepts to liberate electrons from the metal surface. Softness (S) and chemical hardness ( $\eta$ ) are important chemical properties to measure molecular reactivity and stability.<sup>32</sup> Soft molecule is more reactive (small  $\Delta E_{\text{gap}}$ ) than a hard molecule because a hard molecule has a large  $\Delta E_{\text{gap}}$ .<sup>33</sup> The energy of the back-donation involves that when ( $\eta > 0$ ) and  $\Delta E_{\text{Back-donation}} < 0$ , the charge transfer to a molecule, followed by a back-donation from the molecule, is energetically favored.<sup>34</sup> The  $\Delta N$  is about 2.33 eV and shows that the Polyvinylpyrrolidone-iodine is a strong electron donor. In the literature, if the value of  $\Delta N < 3.6$  eV, the IE increases by increasing  $\Delta N$  capacity of inhibitor to donate electrons to the metal surface.

## CONCLUSION

The effectiveness of polyvinylpyrrolidone-iodine as an inhibitor for the corrosion of carbon steel in 1 M hydrochloric acid medium was studied by using weight loss, X-ray diffraction, scanning electron microscopy and quantum chemical methods. The following conclusions can be drawn on the basis of the present investigation.

The PVP-I showed good corrosion inhibition performances for carbon steel in 1 M HCl solution and their action increased with increasing inhibitor concentration, while efficacy decreased with increasing temperature.

The adsorption mechanism of PVP-I on steel surface obeys the Langmuir isotherm model.

Gibbs free energy of adsorption shows that the process is spontaneous and physicochemical adsorbed with a tendency towards chemisorption.

SEM and XRD images reveal that polyvinylpyrrolidone-iodine form a protective layer that defense the surface from direct impact of corrosion.

Quantum chemical parameters confirm the ability of PVP-I molecule to adsorb on a carbon steel surface.

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