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Corrosion Inhibition of Mild Steel by Using Carbimazole/Zn^+ System in NaCl Medium

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Abstract: The degradation of metallic materials under the effect of the environment is defined as a corrosion process. Under the aggressive environment, corrosion leads to the formation of corrosion products. The use of inhibitor substance is considered as one way to protect the metal surface against corrosion. The inhibitor is chemically adsorbed py the surface of the metal and forms a protective thin film with inhibitor effect. The protection can also be achieved by a combination of inhibitor ions and metallic surface. The current work was evaluated using corrosion inhabitation of carbon steel in NaCl solution by carbimazole\Zn system. The ability of carbomazole as a good corrosion inhibitor is enhanced by the presence of Zn^{2+} when the concentration of carbomazole increased and this may be attributed to the protective film formed on the metal surface was withstand the continuous attack of corrosive ions. Also, the formation of complex Fe-carbomizole/Zn^{+2} linkages on the anodic sites of the metal surface during the immersion time may play a role in the improvement in adsorption of inhibiter system via coverage more area of the metal surface which reduced the exposure of anode sites to the corrosive media. By using Langmuir isotherm model to identify the inhibitor mechanism performance, the values of linear correlation coefficient were close to (1) suggested that the adsorption of the studied inhibitors follows Langmuir isotherm model. Generally, values of ΔG_{ads} up to -9.7 kJ/mol are attributed to the electrostatic interaction between the inhibitor molecules and the metal surface (physical adsorption), whilst those at -10.6 kJ/mol or a little more negative are consistent with chemical bonding of the inhibitor to the sample (Chemisorption).

Keywords: Corrosion, Inhibitor, Carboimazole/Znsystem, Langmuir

1. Introduction

It well known fact that corrosion is the degradation of materials due to the chemical or electrochemical interaction between the materials and the surrounding environment. The environmental concerns have provided continuous motivation for the research community to develop new methods to reduce the impact of corrosion [1, 2]. Corrosion inhibitor is one method for corrosion protection and very useful tool used to protect metallic components in industry. Corrosion inhibitors have been used extensively in petrochemical industry due to their properties of environment-friendliness, low cost and renewability [3]. The inhibitors are substances or mixtures that in low concentration and in aggressive environment inhibit, prevent or minimize the corrosion [4, 5]. Generally, the mechanism of the inhibitor is basically based on the three processes. The inhibitor is chemically adsorbed on the surface of the metal and forms a protective thin layer with inhibitor effect or by combination between inhibitor ions and metallic surface [6, 7]. The inhibitor leads to a formation of a film to protect the base metal. Finally, the inhibitor may reacts with a potential corrosive substrates present in aqueous media and the product is a complex [8]. The use of chemical inhibitors to decrease the rate of corrosion processes is quite varied. In the oil extraction and processing industries, inhibitors have always been considered to be the first line of defense against corrosion [9]. Organic molecules can form a barrier through adsorption on the metal surface to reduce the corrosion of the metal in acidic solution [10]. Also, organic inhibitor adsorption on metal surface is influenced by organic inhibitor nature, surface charges on the metal, the type of aggressive solution, and the interaction of inhibitor with the metal surface. Metals such as mild steel and aluminum are widely used in most of the chemical industries due to its low cost and durability for fabrication of various reaction vessels, tanks, pipe components [11]. However, these kinds of materials will be subjected to a severe corrosion in aggressive environment therefore, it has to be protected. Extensive works have been made to understand the mechanism through which metals corrode in different environment [12-14]. In addition, many methods were established to reduce corrosion process includes application of protective coatings, cathodic and anodic protection, alloying and use of different class of inhibitors alone or in combination. [15]. The present work was established to study the corrosion inhabitation of carbon steel in NaCl solution by carbimazole\Zn system as an organic corrosion inhibitor. The thermodynamic parameters were calculated and discussed. Using the carbimazole substance to control the corrosion of carbon steel in the absence and the presence of zinc ion was studied. The influence of immersion period on corrosion processes also were evaluated in this study.

2. Experimental Procedure

2.1. Preparation of Specimens

In this work, mild steel rods with dimensions of 40 mm length and 15 mm diameter were used as substrates. The chemical composition of the investigated carbon steel is shown in Table 1. Carbon steel substrates were ground with SiC abrasive discs (180–1200 grit) and polished to mirror finish. The specimens were ultrasonically cleaned in acetone and subsequently dried to be used in weight loss method and other examination studies. Figure 1 shows the mild steel samples used in this study.

***Table 1.** Composition of mild steels.*

Comp.	C	P	S	Mn	Fe
M. Steel wt%	0.15 – 0.2	0.04	0.05 Max	0.6 – 0.9	Rest

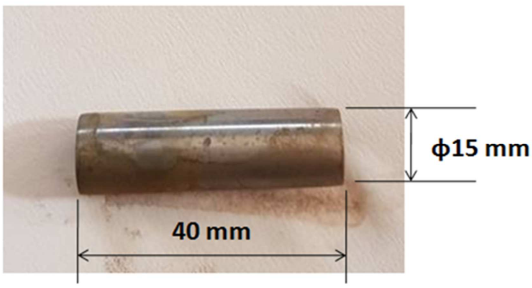

***Figure 1.** Shows the mild steel sample.*

2.2. Corrosive Media

3.5% NaCl were selected as a corroded media to conduct the corrosion behavior of carbon steel substrates. The samples were exposed to NaCl solution throughout immersion time of one and three days, at room temperature (23°C). The concentration of NaCl solutions were prepared according to the following formula (the conc. NaCl% = the amount of NaCl (g) / the volume (1L)); i.e 3.5% NaCl = 35 g of sodium chloride dissolved in 1000 ml of H_2O, pH 7.4.

2.3. Preparation of an Inhibitor

The carbomazole solutions were prepared through doped in distilled water. For the first set of experiments, at room temperature (23°C), solutions were prepared in a concentration range of 0.001M to 0.1 M. The effect of adding different ratio of Zn ions on inhibition efficiency was investigated. Every mild steel samples were weighed by an electronic balance, and then placed in the corrosion media. The duration of the immersion was 1and 3 days, at room temperature (25±2°C). After immersion, the surface of the specimen was cleaned by distilled water followed rinsing with acetone and the sample was draied and weighed again in order to calculate the corrosion rate and the inhibition efficiency (% IE). The experiments were done in triplicate and the average value of the weight loss was noted. For each experiment, a freshly prepared solution was used.

***Figure 2.** Samples in corrosion media.*

2.4. Weight Loss Method

Relevant data of the sample weight loss used in this study are determined and recorded. Carbon steel specimens, in triplicate, were immersed in 100 mL of the corrosion media and different level of of Zn^{2+} (as $ZnSO_4.7H_2O$) for a period of 24 h, and 72 h. The accumulated corrosion on the specimens were cleaned and the specimens than dried. The weight of the specimens before and after immersion was recorded. The corrosion inhibition efficiency was calculated using the following equation.

$$IE = 100\,(1 - W2 / W1)\,\% \tag{1}$$

Where; $W1$ is the corrosion rate in the absence of inhibitor and $W2$ is the corrosion rate in presence of inhibitor.

3. Results and Discussions

3.1. The Inhibitor Concentration vs Corrosion Rate (24 Hrs Immersion Time)

The effects of inhibitor concentration of the carbomazole on the corrosion of carbon steel materials (weight loss) immersed for one day in corrosive media was evaluated. This was done in the absence and presence of zinc ion, the results have been tabulated in Table 2. The weight loss values indicated that the ability of carbomazole as corrosion inhibitor is enhanced slightly, when the concentration of carbomazole increased in the presence of Zn^{2+}. This attributed to the fact that a protective film formed on the metal surface was withstand the continuous attack of corrosive ions such as Cl - ion presence in corrosive media. It well known that [16] the presence of the chloride ions creates extensive localized attack and lead to the increase of the dissolution rate of metal surface in according to the dissolution proposed mechanism. In addition, the carbomazole is organic corrosion inhibitors contain heteroatoms, such as, nitrogen, oxygen, phosphorus and sulphur as well as triple bond or aromatic ring, considered as the reaction center for the adsorption process [17]. The heteroatoms can donate their lone electron pairs to the empty d-orbitals of Fe atoms to form the coordinate covalent bonds which play a role to retard the corrosion [18]. Moreover, in some cases, the electrostatic interaction between inhibitor molecules and steel surface is also favorable for the adsorption of inhibitors [19].

Table 2. *Changes in weight loss of carbon steel immersed in 3.5% NaCl in the absence and presence of the inhibitor (the Inhibitor system is Carbomazole + Zn^{+2} and the Immersion period is one day.*

Con. Carbomozel ppm	Zn^{+2} = 0.0%			Zn^{+2} = 10.0%		
	w1	w2	w loss (mg)	w1	w2	w loss (mg)
0	35427.9	35425.2	2.7	34727.4	34724.5	2.9
20	37153.9	37152.7	1.2	36428.6	36427.2	1.4
60	38610.4	38609.4	1.0	38783.2	38781.9	1.3
100	34681.2	34680.4	0.8	34634.0	34633.2	0.8
140	36471.7	36471.2	0.5	36280.5	36280.1	0.4
180	35513.3	35513.1	0.2	35575.0	35574.6	0.4

Con. Carbomozel ppm	Zn^{+2} = 25.0%			Zn^{+2} = 50.0%		
	w1	w2	w loss (mg)	w1	w2	W loss (mg)
0	35358.3	35355.6	2.7	34606.5	34603.8	2.7
20	34474.2	34473.0	1.2	37093.0	37091.8	1.2
60	36786.4	36785.3	1.1	35448.6	35447.6	1.0
100	36202.9	36202.0	0.9	38540.1	38539.2	0.9
140	36361.9	36361.6	0.3	36429.9	36429.4	0.5
180	36643.9	36643.6	0.3	35458.2	35457.8	0.4

w1&w2 = Sample weight before and after immersed

The variation of inhibition performance of carbomazole-Zn^{2+} system for one day immersion time at various concentrations is shown in Figure 3. It was clear that the inhabitation system of mild steel rods were enhanced as the concentration increased. This may be attributed to the formation of complex Fe-carbomizole/Zn^{+2} linkages on the anodic sites of the metal surface. Also, the behavior may be attributed to the improvement in adsorption of inhibitor system via coverage more area of the metal surface which reduced the exposure of anode sites to the corrosive media. It was reported [20] that the extent of inhibition depends on the degree of the surface coverage (θ) of the iron surface with the adsorbate.

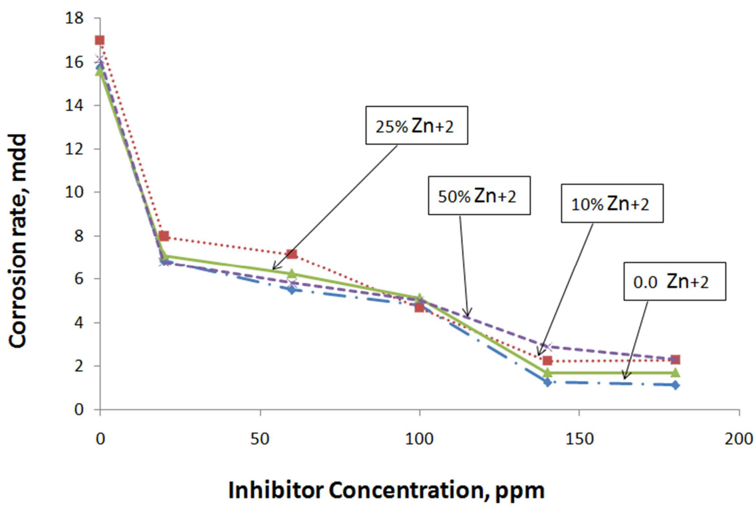

Figure 3. *Corrosion rates via inhibitor concentration for 1 day immersion time.*

3.2. The Inhibitor Concentration vs Corrosion Rate (3 Days Immersion Time)

Table 3 shows the effect of the inhibition concentrations for inhibiting mild steel surfaces in corrosive media at 25ºC for 3 days. It was observed that the corrosion rate is decreased when the immersion time increased to 3 days, for all levels of Zn^{+2} Concentrations. These decreases in corrosion rate values, suggesting strong adsorption of the inhibitor on the mild steel surface. However, the inhibition behavior of the carbomizole/Zn^{+2} system inhibitor in higher amount of Zn^{+2} (50%) was unstable as the immersion time increased. This may be due to an error percentage in the weigth loss measurement which reflected on corrosion rates.

Table 3. *Changes in weight loss of carbon steel immersed in 3.5% NaCl in the absence and presence of the inhibitor (Inhibitor system is Carbomazole + $Zn^{+2}$and the Immersion period ithree days).*

Con. Carbomozel ppm	Zn^{+2} = 0.0%			Zn^{+2} = 10.0%		
	w1	w2	w loss (mg)	w1	w2	w loss (mg)
0	36600.9	36588.47	12.4268	36760.55	36773.1	12.54302
20	37040.4	37034.94	5.450869	35429.11	35434.4	5.288897
60	36323.3	36318.99	4.301424	35343.53	35347.8	4.262965
100	34580.9	34577.76	3.13899	36407.45	36410.7	3.244243
140	36184.9	36182.83	2.076173	35448.27	35450.3	2.028642
180	34458.6	34457.21	1.384497	38516.13	38517.6	1.469402

Con. Carbomozel ppm	Zn^{+2} = 25.0%			Zn^{+2} = 50.0%		
	w1	w2	w loss (mg)	w1	w2	w loss (mg)
0	33422.8	33411.3	11.49734	35281.2	35270.08	11.11787
20	34404.4	34398.57	5.824018	34644.5	34640.23	4.263939
60	35562.7	35558.6	4.095248	34329.8	34327.54	2.235938
100	35739.2	35736.12	3.07252	33983.5	33982.55	0.94432
140	35149.6	35147.54	2.055605	34570.5	34572.28	-1.78433
180	33274.9	33273.13	1.467437	36761.7	36764.69	-2.99154

w1&w2 = Sample weight before and after immersed

In addition, the same trend was observed on the relationship between the concentration and corrosion rats into different amount of Zn^{+2} ions, see Figure 4. It can be clearly seen that as the inhibitor concentration increased the corrosion rates of mild steel decreased. Also, the inhibition molecules were stable on the metal surface, thus reduces or prevent the attached corrosive ions from reaching the metal surface, which result in less corrosion rates. Moreover, the strong inhibitive action of carbomizole/Zn^{+2} system into the corrosive media, which is attributed again to the formation of strong protective films on mild steel surface may play a key role in reduction the corrosion rates as time increased.

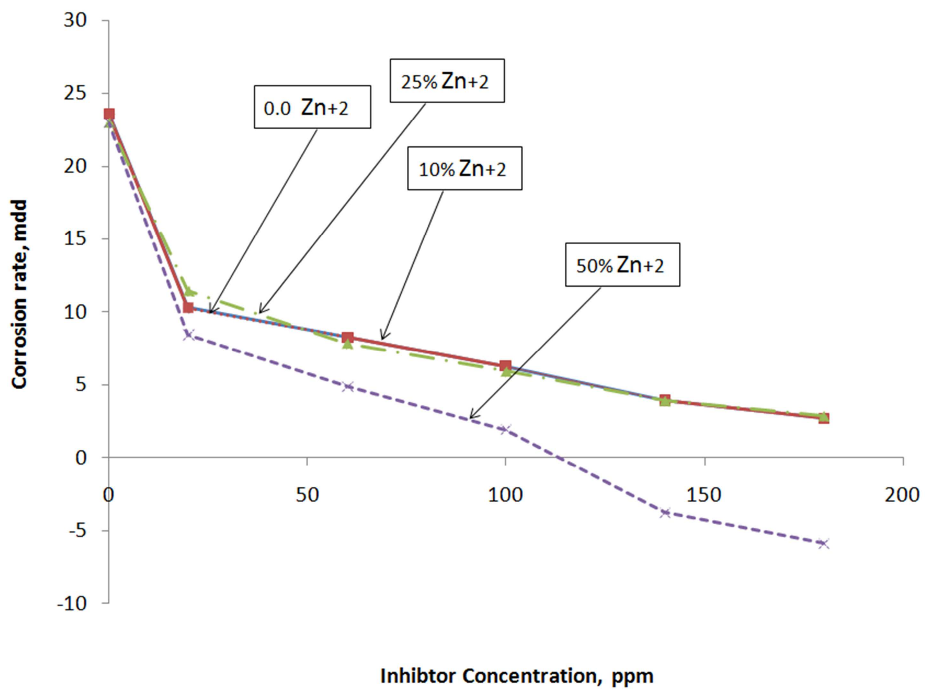

Figure 4. *Corrosion rates via inhibitor concentration for 3 days immersion time.*

3.3. Inhibitor Concentration vs the Efficiency (One Day Immersion Time)

The inhibitor efficiency (IE) of the carbomizole/Zn^{+2} system has been evaluated at different concentration values by using the equation 1. The IE values have been tabulated in table 4. It can be seen that the influences of adding Zn^{+2} in a different concentrations levels have a small effects on the efficiency of corrosion inhibition system. These results suggested that the changes in amount of Zn^{+2} to transport carbomizole towards the metal surface do not play a critical role in improving the inhibitor efficiency. This may be due to the fact that zinc ions in the bulk solution are precipitated as zinc hydroxide.

Table 4. *Corrosion rate and the inhibitor efficiencyfor different concentrations (Inhibitor system is Carbomazole + Zn+2 and the Immersion period is one days)*

C. Mozl ppm	The inhibitor efficiency (IE%); Immersion time: One Day			
	Zn^{+2} = 0.0%	Zn^{+2} = 10.0%	Zn^{+2} = 25.0%	Zn^{+2} = 50.0%
0	0	0	0	0
20	56.33	56.42	50.10	63.16
60	64.84	64.84	65.67	78.53
100	73.41	73.37	74.06	91.05
140	83.30	83.28	82.81	116.34
180	88.37	88.50	84.79	125.4

3.4. Influence of Inhibitor Concentration on the Efficiency

The values of inhibition efficiencies obtained from weight loss measurements for various concentrations of carbomizole/Zn^{+2} systemfor 3 days are presented Table 5. It can be seen, from Figure 4 the inhibition efficiencies increased with increasing concentration of all inhibitors. This implies that more inhibitor molecules adsorbed onto the mild steel surfaces with increasing inhibitor concentration. It does not change their inhibitive performance as the addition of Zn^{+2} ions into the inhibition media. This could be due to aggregation of the inhibitor molecules at metal/solution interface, hence forming micelles that blocked the corrosion sites. This was supported in the literature [21], where the adsorption of inhibitors will decrease the contact between the metal surface and the aggressive medium. Therefore, this decreases the corrosive effect of aggressive medium on the metal surface.

Table 5. *Corrosion rate and the inhibitor efficiency for different concentrations (Inhibitor system is Carbomazole + Zn^{+2} and the Immersion period is three days).*

C. Mozl ppm	The inhibitor efficiency (IE%); Immersion time: Three Days			
	Zn^{+2} = 0.0%	Zn^{+2} = 10.0%	Zn^{+2} = 25.0%	Zn^{+2} = 50.0%
0	0	0	0	0
20	56.33	56.42	50.10	63.16
60	64.84	64.84	65.67	78.53
100	73.41	73.37	74.06	91.52
140	83.30	83.28	82.81	106.03
180	88.37	88.50	84.79	125.4

3.5. Adsorption Isotherm and Adsorption Parameters

In order to gain more information about the mode of adsorptionof the inhibitor system on the mild steel surface, the experimentaldata have been tested langmuir adsorption isotherm for one and three days immersion times. The weight loss results and the addition of Zn^{+2} into corrosion media via the inhibitor concentrations were used to calculatethe adsorption isotherm parameters. Inhibitor molecules were adsorbed on the metal surface if theinteraction between molecule and metal surface was higherthan that of the corrosive media molecule and the metal surface. The linearity of the Langmuir was plottedin Figure 4 and 5. The adsorption mechanism can be investigated by fitting the surface coverage (θ) values obtained from the calculated inhibition efficiencies into Langmuir isotherm. Langmuir isotherm gave the best fit with values of correlation coefficients (R2) and slopes near unity. The Langmuir isotherm model is given by the following equation:

$$\text{Langmuir Isotherm } C / \theta = 1 / K_{ads} + C \qquad (2)$$

where C is the inhibitor concentration in the bulk solution, K_{ads} represents the adsorption equilibrium constant and θ (η/100) represents the fraction of the metal surface covered by the inhibitor. The adsorption equilibrium constant (K_{ads}) is related to the free energy of adsorption (ΔG_{ads}) by the equation:

$$\Delta G°_{ads} = - RT \ln (C_w K_{ads}) \qquad (3)$$

Where *C*w is the concentration of water (1000 g /L), R is the universal gas constant, and T is the absolute temperature. The unit of Cw is expressed in 1000 g/L instead of 55.5 mol L-1 for consistency with K_{ads} units (L g-1). The adsorption equilibrium constant is obtained from the reciprocal of the intercepts from the straight lines on the C/θ axis. Figure 5 and 6 showed the Langmuir isotherm plots of C/θ against C for the adsorption of different concentrations of carbomizole/Zn^{+2} molecules on MS surfaces at 25°C for 1 & 3 days. The values of linear correlation coefficient close to unity suggest that the adsorption of the studied inhibitors follows Langmuir isotherm model. Generally, values of ΔG_{ads} up to -9.7 kJ/mol are attributed to the electrostatic interaction between the inhibitor molecules and the metal surface (physical adsorption), whilst those at -10.6 kJ/mol or a little more negative are consistent with chemical bonding of the inhibitor to the sample (Chemisorption).

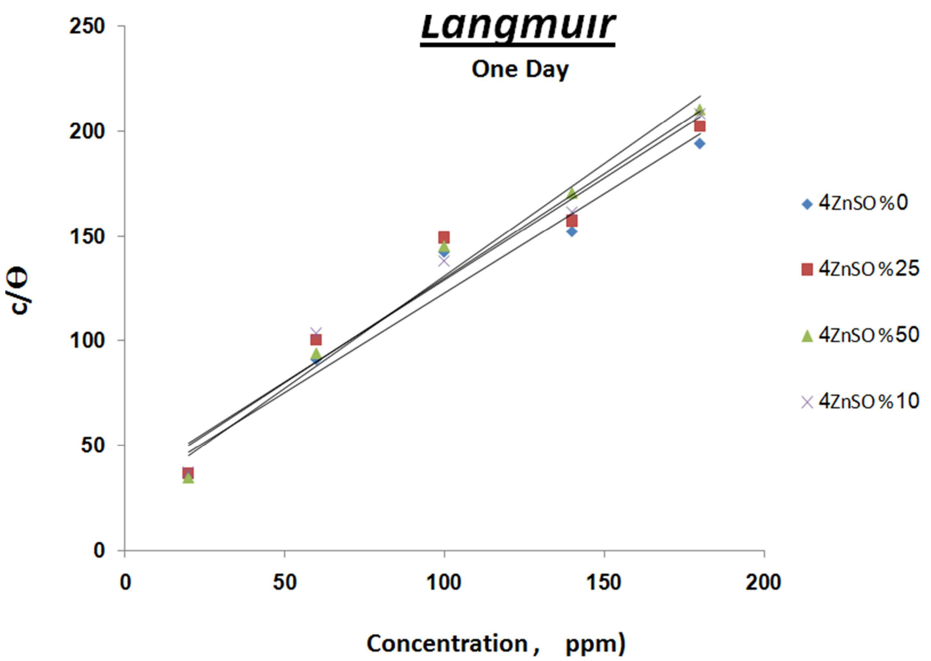

Figure 5. *Langmuir isotherm plots for adsorption inhibitor of carbomizole/Zn^{+2} On mild steel for one day immersion time.*

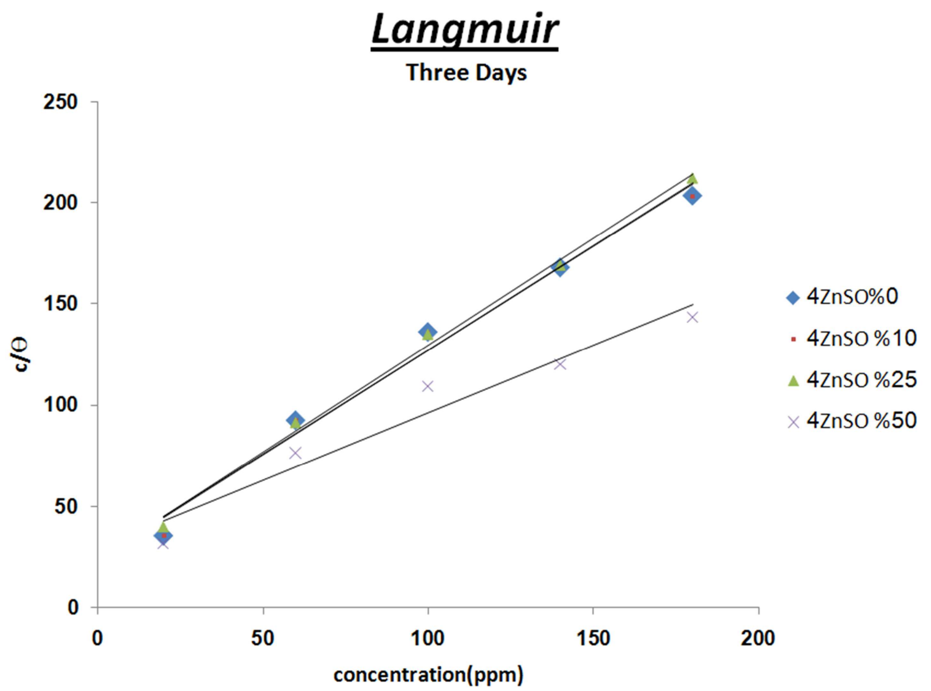

Figure 6. *Langmuir isotherm plots for adsorption inhibitor of carbomizole/Zn^{+2} On mild steel for three days immersion times.*

3.6. The Parameters of K_{ads} and ΔG_{ads} Values

In the current work the values of 1/y obtained were more than unitywhich indicates that each molecule of Carbimazol involved in the adsorption process was attached to more than one activesite on the metal surface. K_{ads} represents the strength betweenadsorbate and adsorbent. Larger values of the K_{ads} implied more efficient adsorption and hence better IE. K_{ads} value slightly decreased in the presence of Carbimazol with higher value of Zn^{+2} indicating that the adsorption of the inhibitor onthe mild steel surface was unfavorable at higher immersed time. The equilibrium constant for the adsorption process was relatedto the standard free energy of adsorption. The negative values of ΔG_{ads}ensure the spontaneity of adsorption process and stability of the adsorbed layer on mild steel surface.

Table 6. *K_{ads} and ΔG_{ads} parameters value for one and three days immersion time.*

Parameters	**One Day**			**Three Days**		
Concentration of carbomazole with Zn+	**With out**	**25%**	**50%**	**With out**	**25%**	**50%**
K_{ads}	1.22	1.27	1.27	1.09	1.09	0.72
ΔG_{ads}	10.730	10.628	10.730	10.335	10.335	9.672

4. Conclusions

The ability of carbomazole as a corrosion inhibitor is enhanced slightly when the concentration of carbomazole increased in presence of Zn^{2+}. This attributed to the protective film formed on the metal surface to withstand the continuous attack of corrosive ions. The formation of complex Fe-carbomizole/Zn^{+2} linkages on the anodic sites of the metal surface during the immersion time plays a role in the improvement in adsorption of inhibiter system via coverage more area of the metal surface which reduces the exposure of anode sites to the corrosive media. The influences of adding Zn^{+2} in different concentration level have a little effect on the efficiency of corrosion inhibition system. This is due to the fact that zinc ions in the bulk solution are precipitated as zinc hydroxide. The values of linear correlation coefficient close to unity suggest that the adsorption of the studied inhibitors follows Langmuir isotherm model. Also, values of ΔG_{ads} up to -9.7 kJ/mol are attributed to the electrostatic interaction between the inhibitor molecules and the metal surface (physical adsorption), whilst those at -10.6 kJ/mol or less are consistent with chemical bonding of the inhibitor to the sample (Chemisorption).

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