Journal of Engineering and Development, Vol. 17, No.4, October 2013, ISSN 1813- 7822

Comparison Of The Corrosion Inhibition Efficiencies Of Low Carbon Steel In Different Acidic Mediums Using Some Polyethylene Glycols

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Abstract :

The inhibition of corrosion of low carbon steel (LCS) using some polyethylene glycols (PEGs) in different acidic mediums by weight loss technique was investigated. The corrosion inhibition was performed in hydrochloric acid and sulphuric acid at different acid temperatures and acid concentrations, also at various polyethylene glycol concentrations and its average molecular weights. Results obtained reveal that PEG is an active inhibitor and shows very good corrosion inhibition efficiency (IE). The IE was found to vary with acid temperature, acid concentration, polyethylene glycol's concentration and average molecular weight. Also it was found that the corrosion inhibition behavior of PEG is greater in sulphuric acid than in hydrochloric acid. The adsorption of the inhibitor on the LCS surface is in agreement with Langmuir and Frumkin adsorption isotherms. The inhibition efficiencies are increased by increase of the inhibitor's concentration and average molecular weight, decreases with an increase in the concentration and temperature of the acid. This work has demonstrated that polyethylene glycols have inhibition effect on corrosion process, and their inhibition efficiencies are between 32.11 and 95.25% in the range of experimental conditions.

مقارنة كفاءة التاكل لمعدن الفولاذ واطئ الكاربون في اوساط حامضية مختلفة بإستخدام بعض انواع البولي أثلين كليكول

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الخلاصة :

تثبيط التاكل لمعدن الفولاذ واطئ الكاربون بإستخدام بعض أنواع البولي اثلين كليكول في اوساط حامضية مختلفة تمت دراستها بإستخدام تقنية فقدان وزن المعدن. تثبيط التاكل تم انجازها بإستخدام محاليل حامض الهدروكلوريك والكبريتيك بتراكيز ودرجات حرارة مختلفة وكذلك بإستعماال تراكيز ومعدل أوزان جزيئة مختلفة للمثبط بولي اثلين كليكول. أظهرت النتائج التي تم الحصول عليها بأن المثبط فعال يعطي كفاءة تثبيط التاكل (IE) جيدة جدا. كفاءة التثبيط وجدت بأنها تتغير مع تغير تركيز وحرارة الحامض وكذلك مع تغير تركيزالمثبط ومعدل وزنه الجزيئي.كذلك وجد بأن سلوك تثبيط التاكل في حامض الكبريتيك اكبر مما هو عليه في حامض الهيدروكلوريك . ولقد وجد أن ادمصاص المثبط

على سطح معدن الفولاذ واطئ ا لكاربون تتوافق مع ادمصاص لانكموير و فرومكين. تزداد كفاءة الثبيط بزيادة تركيز المثبط ومعدل وزنه الجزيئي، وتقل بزيادة تركيز ودرجة حرارة الحامض. اثبت هذا العمل بأن المثبط له تأثيرعلى عملية التاكل وقد تراوحت كفاءة التثبيط بين 95.25-32.11 % في نطاق ظروف التجارب المختلفة.

1. Introduction :

Most of the well-known acid inhibitors used in industry are organic compounds having multiple bonds in their molecules that mainly certain nitrogen, sulphur, oxygen atoms through which they get adsorbed on the metal surface [1-4]. The effect of temperature on inhibiting process is of great importance in industry. Effective inhibitors are expected to perform under a wide range of conditions. Their inhibition action depends on factors such as the structure of the molecules, the number and type of adsorption sites, the distribution of change in the molecules and types of interaction between the molecules and the metallic surface [5-7]. Inhibitors such as aromatic compounds containing heteroatom adsorb on the metal surface and provide a barrier for the entry of aggressive ions in acid solution [8]. Concentrated mineral acids are used extensively in pickling, cleaning, descaling and oil well acidising of metallic materials cause damage of corrosion [9, 10]. Organic inhibitors are more effective with iron and are good corrosion inhibitors for acidic solutions of metals [11]. Due to the aggressiveness of hydrochloric acid and sluphuric acid in the solution against structural material, such as carbon steel, the use of corrosion inhibitor is usually required to minimize the corrosion attack [12-14].

The aim of this study is to investigate the corrosion inhibition of low carbon steel in 3, and 4 M HCl, and 3, and 4 M H_2SO_4 solutions in absence and in presence of inhibito polyethylene glycols (PEGs) at two hours, at different acid temperatures and acid concentrations, as well as at various inhibitor's concentration and average molecular weight by weight loss technique.

2. Experimental :

2.1. Specimens

Low carbon steel of chemical composition C- 0.0526, Si- < 0.0010, Mn- 0.2080, P- 0.0202, S- 0.0264, and Fe- Balance (wt %) were used in the present work. This grade of LCS can be used for manufacturing of chain, stampings, rivets, nails, wire, seam-welded pipes, mattresses, hot and cold-rolled strip for many purposes. Specimens of area (8 x 1.5 cm^2) and thickness of 0.1 cm were first degreased with AR grade benzene and acetone and then annealed in a vacuum at 600 °C for one hour and cooled to room temperature. Samples were abraded in sequence under running tap water using emery paper of grit size 220, 320, 400, and 600, then washed with running tap water followed by distilled water, dried with clean tissue, immersed in benzene and acetone, kept in desiccators over silica gel bed until use.

2.2. Solutions

For the preparation of aggressive HCl and H_2SO_4 acid solutions AR grade HCl and H_2SO_4 were used. Approximate concentration of acids of 3, and 4 M HCl, and 3, and 4 M H_2SO_4 solutions were prepared using double distilled water. Inhibitor PEG concentrations ranging from 0.2 to 1 wt % in 3 and 4 M HCl, and in 3 and 4 M H_2SO_4 solutions were prepared. Inhibitor of average molecular weights of 1500, 4000, and 6000 g mol^{-1} were used in this study. The weight loss measurements were carried out at various experimental conditionsplan

as shown in Table (1).

Table (1): Experimental plan for weight loss measurements of low carbon steel in hydrochloric acid and in sulphuric acid solutions in absence and in presence of inhibitor PEG.

Acid temperature (°C)	Inhibitor Concentration (wt %)	Average molecular weight of inhibitor (g mol^{-1})					
		PEG 1500		PEG 4000		PEG 6000	
		Acid concentration (M)					
20	Zero	3	4	3	4	3	4
20	0.2	3	4	3	4	3	4
20	0.4	3	4	3	4	3	4
20	0.6	3	4	3	4	3	4
20	0.8	3	4	3	4	3	4
20	1.0	3	4	3	4	3	4
40	Zero	3	4	3	4	3	4
40	0.2	3	4	3	4	3	4
40	0.4	3	4	3	4	3	4
40	0.6	3	4	3	4	3	4
40	0.8	3	4	3	4	3	4
40	1.0	3	4	3	4	3	4
60	Zero	3	4	3	4	3	4
60	0.2	3	4	3	4	3	4
60	0.4	3	4	3	4	3	4
60	0.6	3	4	3	4	3	4
60	0.8	3	4	3	4	3	4
60	1.0	3	4	3	4	3	4

For these measurements the metal specimens were completely immersed in (200 cm^3) hydrochloric acid solutions or sulphuric acid solutions contained in (250 cm^3) glass beakers. They were exposed of (2 hours) at different acid temperatures of (20, 40, and 60 °C), acid concentrations of (3 and 4 M), inhibitor concentrations of (0.2, 0.4, 0.6, 0.8 and 1 wt %), and inhibitor average molecular weights of (1500, 4000, and 6000 g mol^{-1}), then the specimens were cleaned, washed with benzene and acetone, dried again. The specimens were weighted before and after immersion in acid solutions in order to estimate the weight loss of each specimen. The corrosion rate (R), percentage of corrosion efficiencies (IE %), and the surface coverage (θ), were calculated using the following equations [15, 16]:

$$R = \frac{87.6\,W}{D\,A\,T} \qquad (1)$$

$$IE\,\% = \frac{R - R_0}{R} \times 100 \qquad (2)$$

$$\theta = \frac{R - R_0}{R} \qquad (3)$$

Where:

W = weight loss of metal specimen (mg), D = density of metal specimen (7.85 g cm^{-3}),
A = total surface area of metal specimen (cm^{2}), T = exposure time (h), R = corrosion rate in absence of inhibitor (mmpy), R_0 = corrosion rate in presence of inhibitor (mmpy),
IE = corrosion inhibition efficiency (%), θ = degree of surface coverage.

3. Results and Discussion :

The corrosion inhibition efficiencies (IE) of inhibitor polyethylene glycols (PEGs) systems obtained from weight loss data are given in Table (2) and Table (3).

Table (2): Corrosion rates and inhibition efficiencies of low carbon steel in 3 M and 4 M HCl in absence and in presence of polyethylene glycol (PEG 4000) at different acid temperatures.

Acid temperature (°C)	**Inhibitor Concentration (wt %)**	**Corrosion rate (mmpy)**		**Inhibition efficiency (%)**	
		Acid concentration (M)			
		3	**4**	**3**	**4**
20	Zero	6.84	8.68	Blank	Blank
20	0.2	0.80	2.47	88.33	71.54
20	0.4	0.75	2.34	89.10	73.11
20	0.6	0.69	2.13	89.95	75.51
20	0.8	0.66	1.97	90.30	77.31
20	1.0	0.60	1.80	91.23	79.30
40	Zero	79.10	83.63	Blank	Blank
40	0.2	13.92	30.91	82.40	63.12
40	0.4	13.37	28.30	83.10	66.23
40	0.6	12.70	26.43	83.95	68.40
40	0.8	11.86	23.96	85.00	71.35
40	1.0	11.52	22.75	86.32	72.80
60	Zero	113.97	141.35	Blank	Blank
60	0.2	39.10	78.73	65.69	44.30
60	0.4	35.78	69.94	68.61	50.52
60	0.6	32.44	65.19	71.54	53.88
60	0.8	29.69	58.45	73.95	58.65
60	1.0	25.60	50.43	77.54	64.32

Table (3): Corrosion rates and inhibition efficiencies of low carbon steel in 3 M and 4 M H_2SO_4 in absence and in presence of polyethylene glycol (PEG 4000) at different acid temperatures.

Acid temperature (°C)	Inhibitor Concentration (wt %)	Corrosion rate (mmpy)		Inhibition efficiency (%)	
		Acid concentration (M)			
		3	4	3	4
20	Zero	4.88	6.19	Blank	Blank
20	0.2	0.48	1.64	90.24	73.56
20	0.4	0.44	1.55	90.95	74.92
20	0.6	0.42	1.40	91.44	77.44
20	0.8	0.36	1.16	92.30	81.29
20	1.0	0.30	0.95	93.90	84.66
40	Zero	56.50	59.74	Blank	Blank
40	0.2	9.09	18.64	83.92	68.80
40	0.4	8.54	17.14	84.88	71.30
40	0.6	7.95	16.37	85.93	72.50
40	0.8	7.58	15.45	86.58	74.13
40	1.0	7.02	15.04	87.58	74.83
60	Zero	81.41	100.97	Blank	Blank
60	0.2	22.55	51.37	72.30	49.12
60	0.4	20.68	45.32	74.60	52.60
60	0.6	19.13	44.65	76.50	55.78
60	0.8	18.78	39.83	76.93	60.55
60	1.0	17.22	33.89	78.85	66.43

It is seen that polyethylene glycols (PEGs) inhibits corrosion of low carbon steel in 3 and 4 M HCl and in 3 and 4 M H_2SO_4 solutions at all PEGs concentrations under study. It has been observed from Table (2) and Table (3) that corrosion inhibition efficiency (IE) for PEGs increases with the increase in PEGs concentrations, and decreases with the increase in acid concentrations and acid temperatures. The influence of acid temperature on IE of PEG 1500 at various concentrations in 3 M HCl and 3 M H_2SO_4 are shown in Figure (1) and Figure (2) respectively. The IE decreases with increase in acid temperature due to desorption of inhibitor PEG from metal surface, which has led to increase in corrosion rate of the metal surface. The effect of acid temperature on IE is more effective at high temperature than at low temperature in the rage of 40-60 °C.

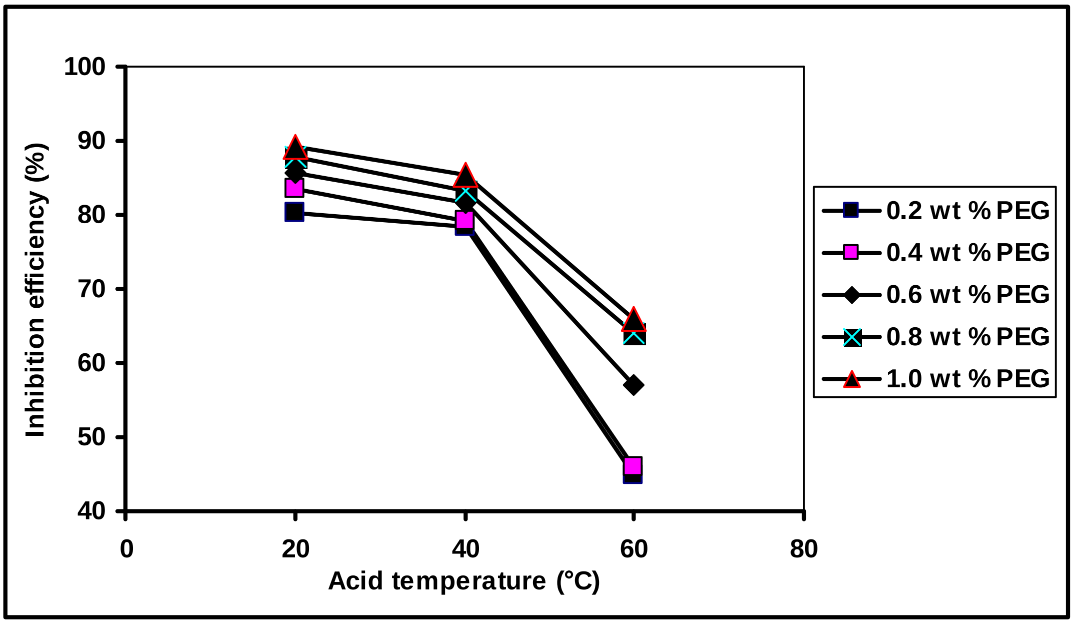


Fig. (1): ***Variation*** **of inhibition efficiency with acid temperature in 3 M HCl solution, at various PEG 1500 concentrations.**

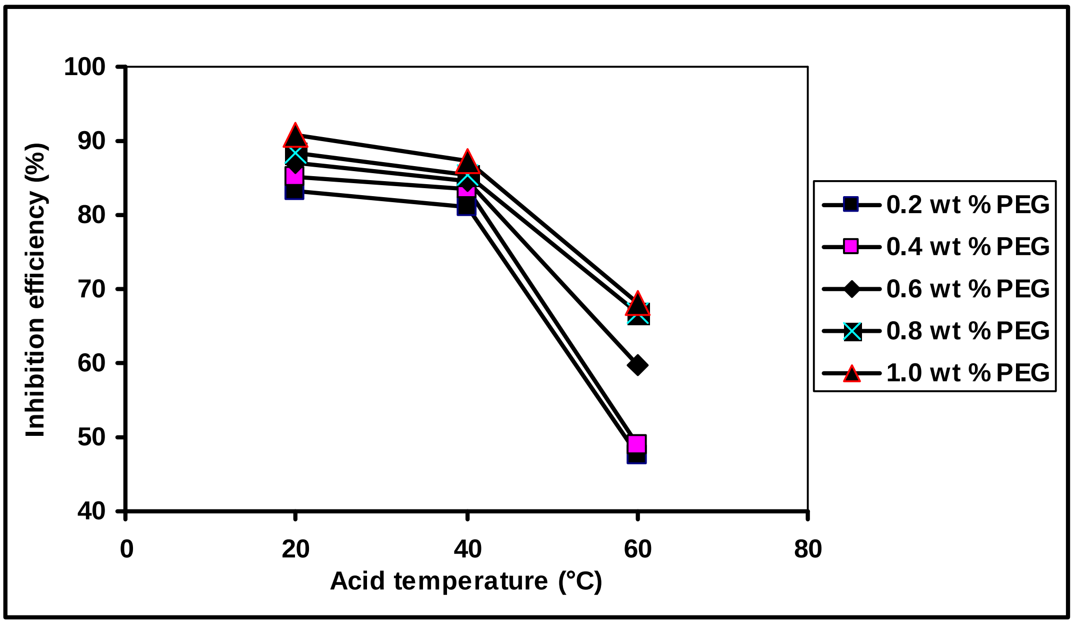


Fig. (2) ***Variation*** **of inhibition efficiency with acid temperature in 3 M H_2SO_4 solution, at various PEG 1500 concentrations**.

The effect of inhibitor PEG concentration on corrosion inhibition efficiency (IE) at different acid temperature in the range of 20-60 °C are given in Figure (3) and Figure (4). It can be seen that IE increases with increasing inhibitor concentration as a result of more inhibitor molecules being adsorbed on the LCS surface when the inhibitor concentration was increased.

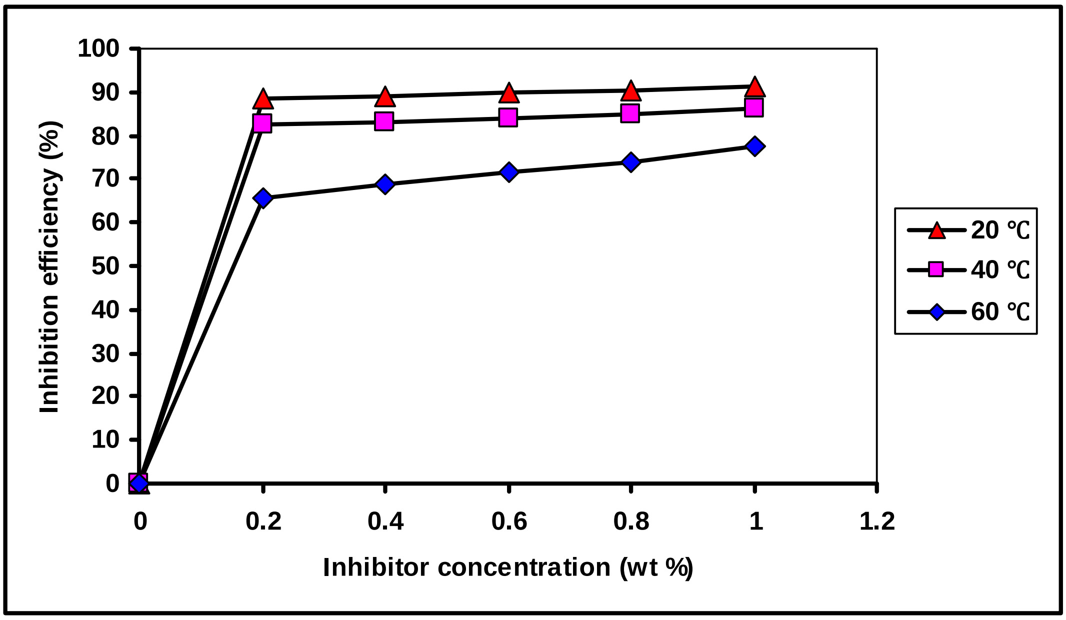


Fig. (3) ***Variation*** **of inhibition efficiency with polyethylene glycol (PEG 4000) concentrations in 3 M HCl solution, at different acid temperatures.**

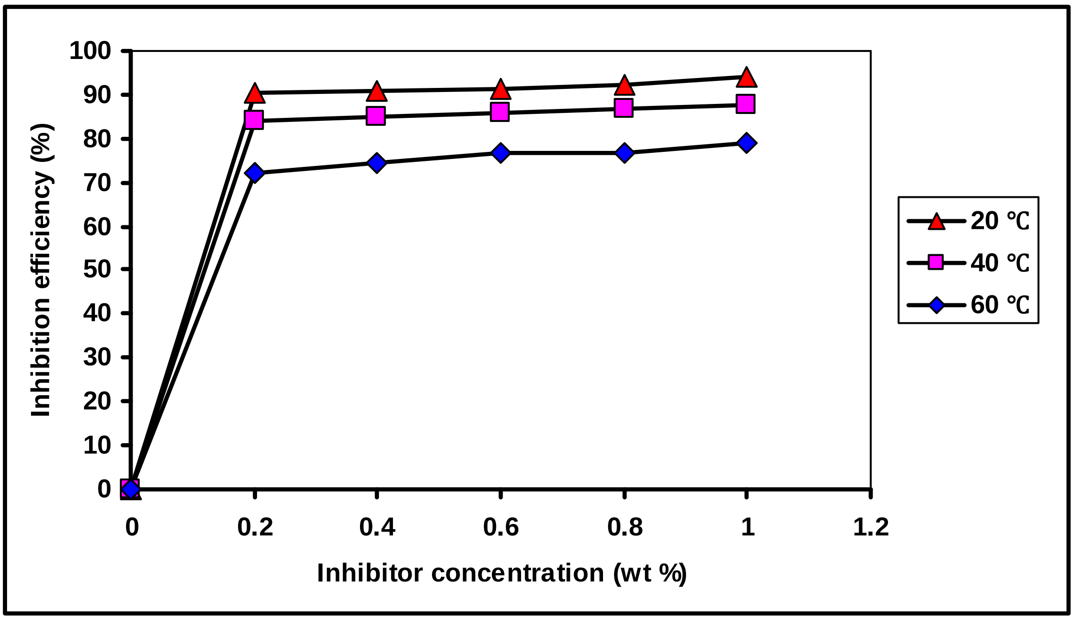


Fig. (4) Variation of inhibition efficiency with polyethylene glycol (PEG 4000) concentrations in 3 M H_2SO_4 solution, at different acid temperatures.

The variation of inhibition efficiency with PEG concentration in 3 M HCl and 3 M H_2SO_4 solutions, at 40 °C and at various PEG average molecular weights can be shown in **Figure (5)** and **Figure (6)** respectively. The IE is increased with increase of inhibitor concentration and its average molecular weight, due to increasing of adsorption of PEG onto LCS surface, and increasing the number of carbon atoms in the molecule of PEG respectively.

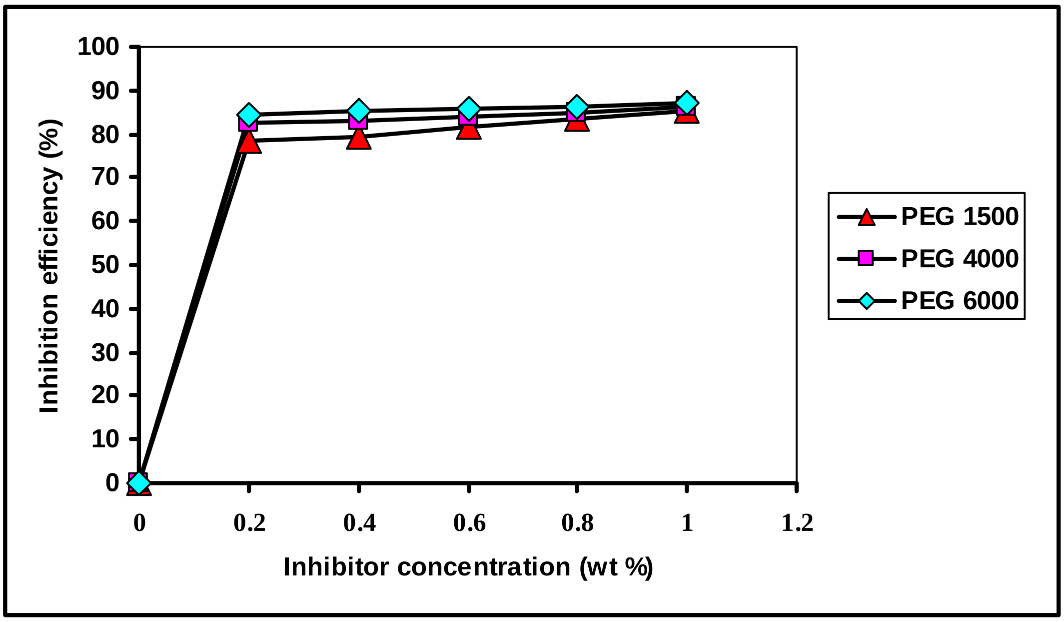


Fig. (5) Variation of inhibition efficiency with polyethylene glycol (PEG) concentrations in 3 M HCl solution, at different average molecular weights of polyethylene glycol and at 40 °C.

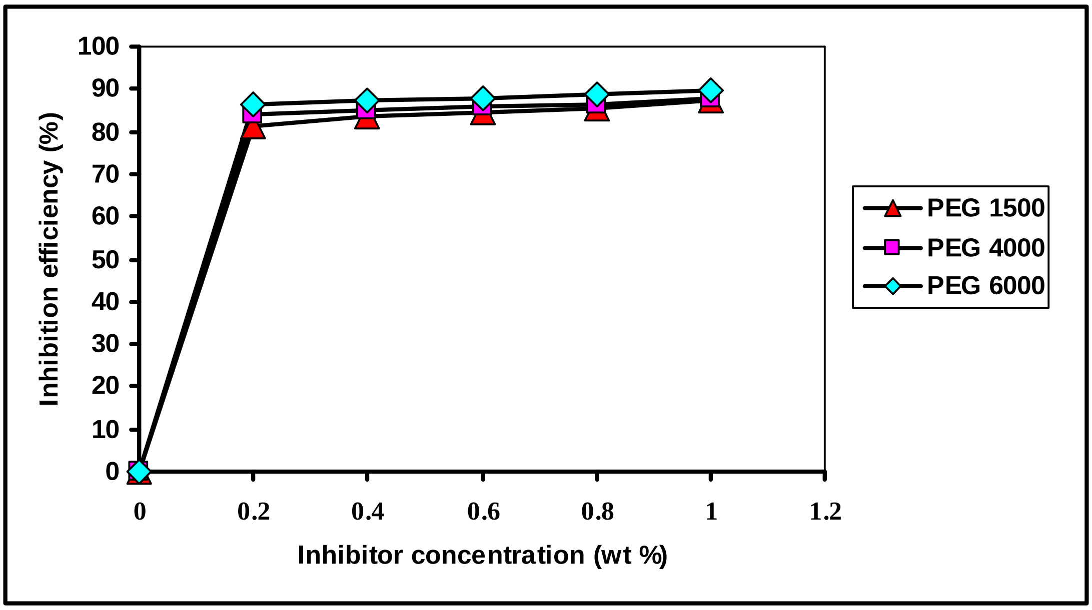


Fig. (6) Variation of inhibition efficiency with polyethylene glycol (PEG) concentrations in 3 M H_2SO_4 solution, at different average molecular weights of polyethylene glycol and at 40 °C.

The influence of inhibitor concentration on IE of PEG for various acid concentrations at 40 °C is given in **Figure (7)** and **Figure (8)** respectively. The IE increases with rising in inhibitor concentration due to increasing the adsorption of inhibitor on the metal surface, and it is decreased significantly by increasing in the acid concentration, because of an increasing in corrosion rate. This is primarily due to the fact that the amounts of hydrogen ions, which are the active species, are increased, as acid concentrations increased.

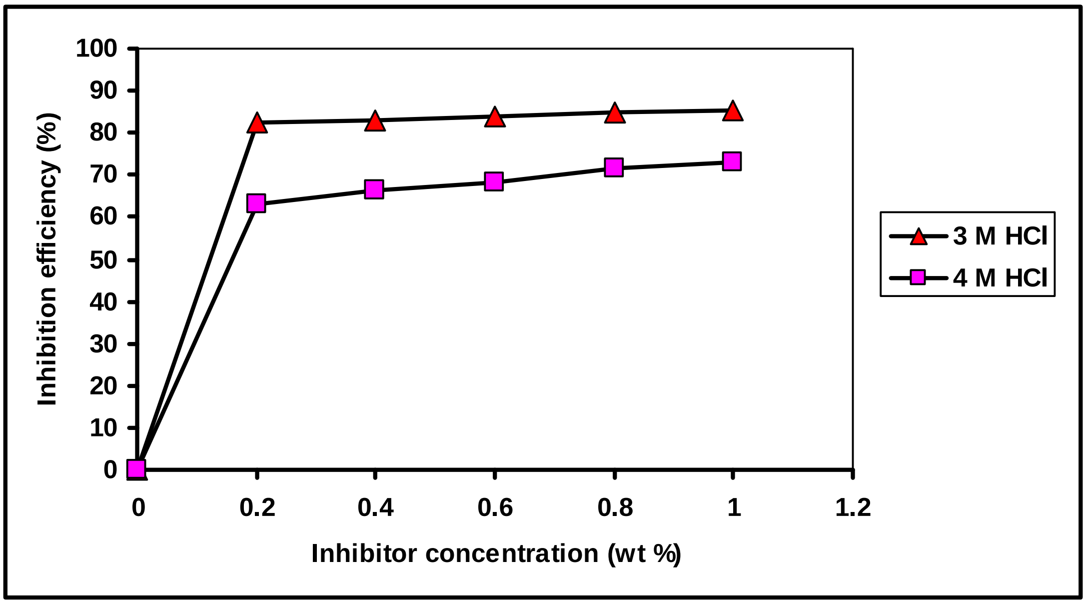


Fig. (7) Variation of inhibition efficiency with polyethylene glycol (PEG 4000) concentrations, at differet HCl concentrations and at 40 °C.

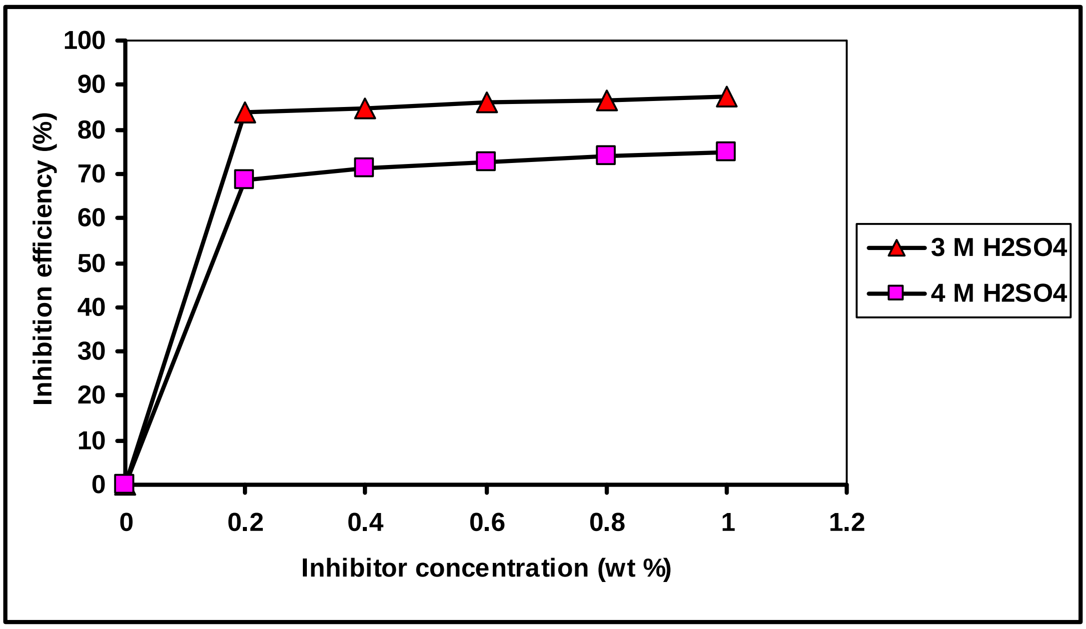


Fig. (8) Variation of inhibition efficiency with polyethylene glycol (PEG 4000) concentrations, at different H_2SO_4 concentrations and at 40 °C.

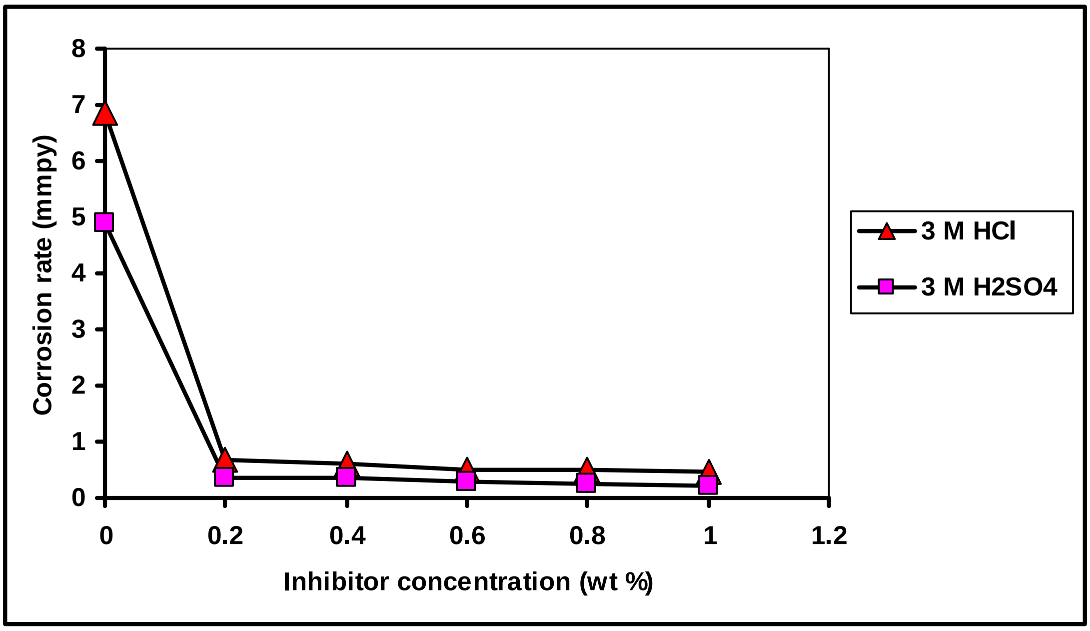


Fig. (9) Variation of corrosion rate with polyethylene glycol (PEG 6000) concentrations in 3 M HCl and 3 M H_2SO_4 solutions at 20 °C.

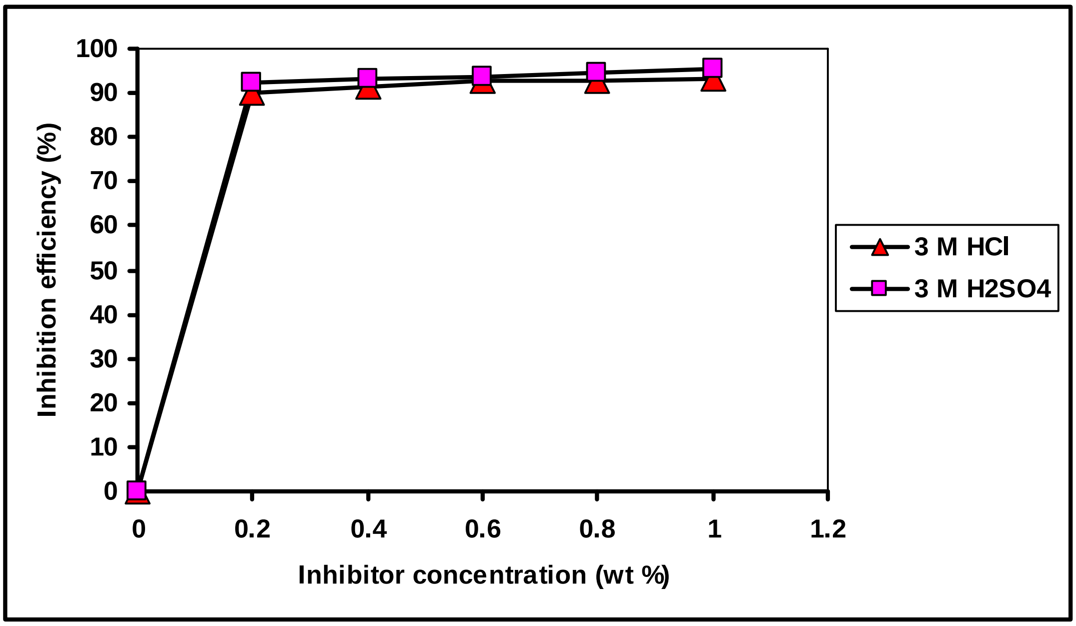


Fig. (10) Variation of inhibition efficiency with polyethylene glycol (PEG 6000) concentrations in 3 M HCl and 3 M H_2SO_4 solutions at 20 °C.

From mentioned **Figure (9)** and **Figure (10),** it was clear that the corrosion rate (R) was decreased with increasing in the concentration of inhibitor, while corrosion inhibition efficiency (IE) increases with increasing in the concentration of the inhibitor, it is attributed to increasing the adsorption of inhibitor on the metal surface (i.e. increasing the thickness of protection layer on the metal surface).

In addition, the maximum corrosion efficiency of PEG 6000 was 93.21 % at 3 M HCl, and 95.25 % at 3 M H_2SO_4, at 1 wt % inhibitor PEG concentration, and at 20 °C respectively. And also it was concluded that the inhibitor was best inhibitor in low carbon steel corrosion in HCl and H_2SO_4 solutions. But when comparing with acids the inhibitor efficiency was best in H_2SO_4 than HCl.

Polyethylene glycol (PEG) in both HCl and H_2SO_4 shows goods inhibitory character. So, inhibition behavior of PEG increases tremendously in H_2SO_4 when compared with HCl [**Figure (9)** and **Figure (10)].** The inhibition efficiency depends on the degree of surface coverage (θ) of low carbon steel (LCS) surface by molecules of inhibitor. The θ of LCS was calculated using equation (3) and the results are given in **Table (4).**

Table (4): The values of the degree of surface coverage (θ) of low carbon steel in 3 M and 4 M HCl, and in 3 M and 4 M H_2SO_4 solutions in absence and in presence of polyethylene glycol (PEG 4000) at different acid temperatures.

Acid temperature (°C)	**Inhibitor Concentration (wt %)**	**Degree of surface coverage**			
		Acid concentration (M)			
		3 M HCl	**4 M HCl**	**3 M H_2SO_4**	**4 M H_2SO_4**
20	Zero	Blank	Blank	Blank	Blank
20	0.2	0.8833	0.7154	0.9024	0.7356
20	0.4	0.8910	0.7311	0.9095	0.7492
20	0.6	0.8995	0.7551	0.9144	0.7744
20	0.8	0.9030	0.7731	0.9230	0.8129
20	1.0	0.9123	0.7930	0.9390	0.8466
40	Zero	Blank	Blank	Blank	Blank
40	0.2	0.8240	0.6312	0.8392	0.6527
40	0.4	0.8310	0.6623	0.8488	0.6798
40	0.6	0.8395	0.6840	0.8593	0.7033
40	0.8	0.8470	0.7135	0.8658	0.7413
40	1.0	0.8544	0.7280	0.8758	0.7483
60	Zero	Blank	Blank	Blank	Blank
60	0.2	0.6569	0.4430	0.6875	0.4912
60	0.4	0.6861	0.5052	0.7190	0.5260
60	0.6	0.7154	0.5388	0.7452	0.5578
60	0.8	0.7395	0.5865	0.7693	0.6055
60	1.0	0.8067	0.6432	0.8324	0.6643

These results clearly show that the θ of low carbon steel increases with increase in inhibitor PEG concentration and PEG average molecular weight, and it is decreased with increasing in acid concentration and acid temperature. Several adsorption isotherms have been employed to characterize inhibitor performance [17, 18].

The values of **θ** were tested graphically for fit to Temkin (**θ** vs log **C**), Langmuir (**C/θ** vs **C**), Flory-Huggins (log **θ**/**C** vs log (**1-θ**), and Frumkin (**θ** vs **C**) isotherms.

Where:

C = inhibitor concentration (wt %).

θ = degree of surface coverage.

Figure (11), and **Figure (12)** show that a Langmuir and Frumkin adsorption isotherms had the best fitting with correlation coefficient of more than 0.99 respectively, and that reveals the inhibitor PEG obeys the mentioned adsorption isotherms.

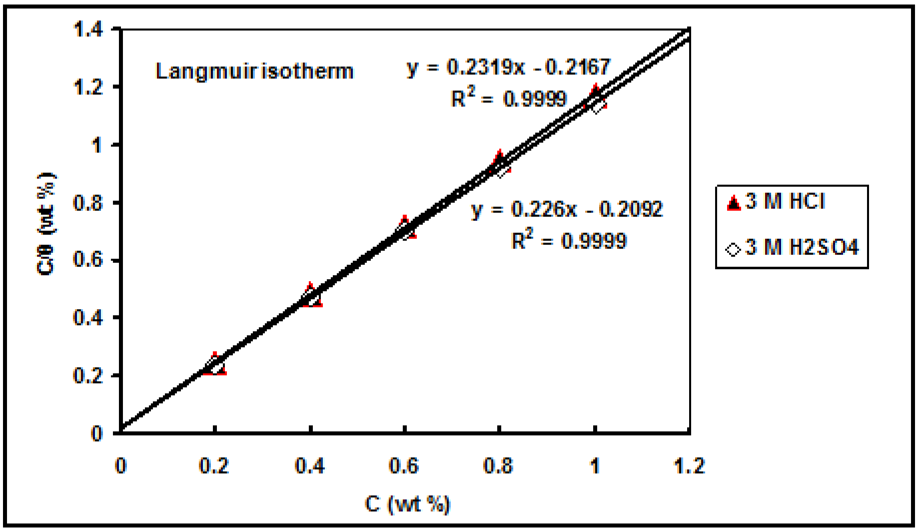


Fig. (11): Langmuir isotherm plot of C/θ against C for PEG 4000, at 40 °C.

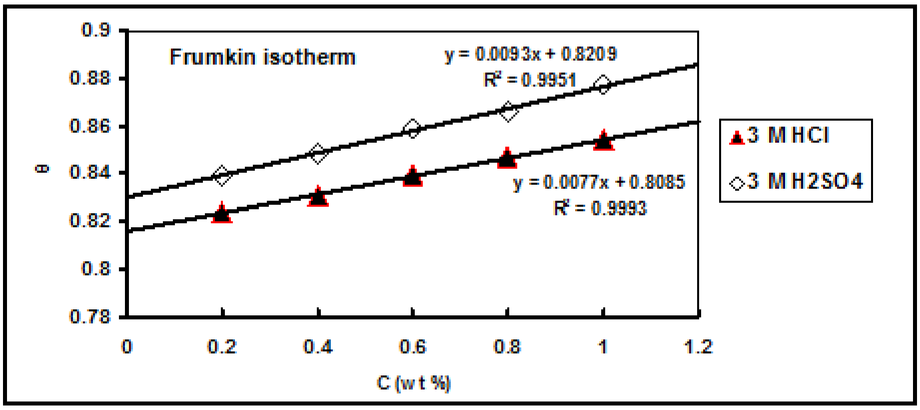
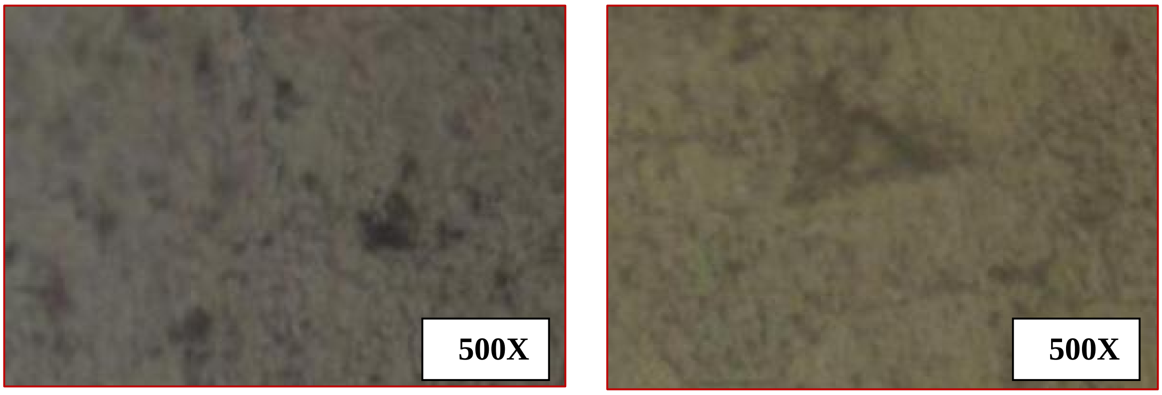


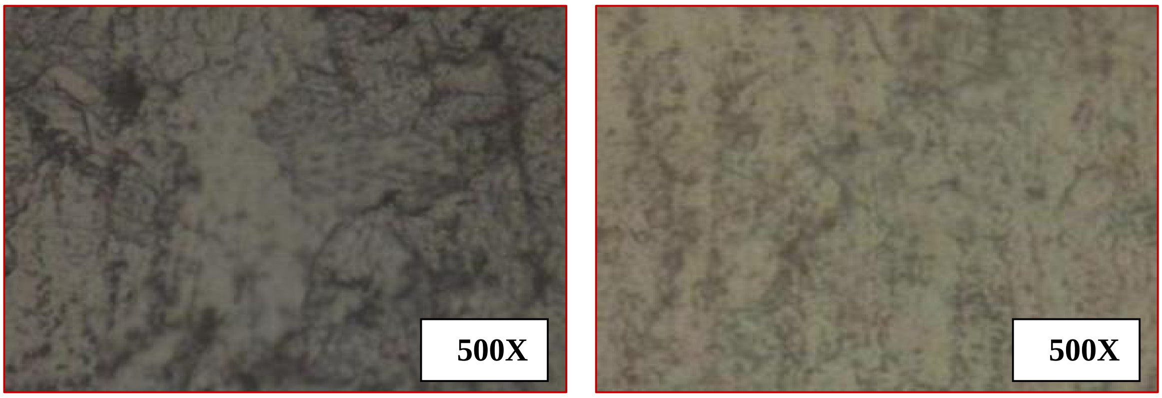
Fig.(12) Frumkin isotherm plot of θ against C for PEG 4000, at 40 °C.

(a) Specimen before immersion in acid



(b) Specimen after immersion in H_2SO_4

(c) Specimen after immersion in H_2SO_4 with Inhibitor



(d) Specimen after immersion in HCl

(e) Specimen after immersion in HCl with inhibitor

Fig.(13): Effect of sulfuric acid and hydrochloric acid on the microstructure of the low carbon steel specimens in absence and in presence of inhibitor at T_{acid} = 60 °C, C_{acid} = 3 M, $C_{inhibitor}$ = 0.8 wt %, and $M_{inhibitor}$ = 1500 g mol^{-1}.

In order to evaluate the conditions of the low carbon steel (LCS) surfaces in contact with 3 M H_2SO_4 and 3 M HCl solutions, the superficial analysis was carried out.

Figure (13) represents the micrographs of the specimens that reveal the morphology of the uncorrded and corroded surfaces of low carbon steel specimens after cleaning. The micrographs of the as-received LCS specimen is shown in **Figure (13 a),** and in presence of 3 M H_2SO_4, and 3 M HCl solutions are shown in **Figure (13 b**), and in **Figure (13 d)** respectively.

The influence of the inhibitor PEG addition (0.8 wt %) separately on the microstructure of LCS is given in **Figure (13 c**) and **Figure (13 e). Figure (13 a)** shows that the microstructure of the specimen is not clear, because the specimen is not react with any corrosive environment. **Figure (13 b**) shows clear grain boundary of steel specimen immersed in 3 M H_2SO_4, which mean that etching has been occurred, due to reaction between the acid and metal surface (corrosion), while the **Figure (13 c)** shows less grain boundary, because of reaction between the inhibited acid and the specimen at same conditions. **Figure (13 d**) shows clearly the grain boundary of the steel specimen, attributed to the higher activity of 3 M HCl than 3 M H_2SO_4 as in **Figure (13 b),** while **Figure (13 e)** shows less grain boundary.

The micrographs analysis has shown that inhibition of corrosion by PEG is due to formation of layer on the LCS surface. The light colour regions of the microstructure of metal specimens are ferrite phase, while the dark regions are pearlite phase.

It can be observed from the mentioned Figures that pitting corrosion does not occurred and solid particles do not appear on surface. The surface roughness of the LCS surface appears lower with addition of the inhibitor than that without. The roughness is found to be uniform after treatment with acidic solution which contains inhibitor.

4. Conclusions :

1. Inhibitor PEG was found to be effective inhibitor for low carbon steel in (3 and 4 M) HCl and (3 and 4 M) H_2SO_4 solutions medium.

2. The adsorption of the inhibitor PEG was confirmed to follow the Langmuir and Frumkin isotherms.

3. Corrosion inhibition efficiency of PEG increases with increase in inhibitor concentration and its average molecular weight, and increased by increasing acid temperature and acid concentration. For the comparative studies, it was concluded that the inhibitor efficiency is better in H_2SO_4 than HCl, attributed to H_2SO_4 is dibasic acid, so it stimulated the corrosion rate of low carbon steel.

4. Inhibition efficiency of LCS in HCl and H_2SO_4 solutions by PEG is under anodic control.

5. The maximum corrosion inhibition efficiency of PEG was 93.21 % in 3 M HCl and 95.25 % in 3 M H_2SO_4 solutions respectively at room temperature of 20 °C and at PEG concentration of 1 wt %, and its average molecular weight of 6000 g mol^{-1} for 2 hour of immersion time.

6. The microstructure examination showed that there was improvement in the surface morphology of the as- corroded inhibited low carbon steel compared to uninhibited specimen.

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