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Inhibition of cold rolled steel corrosion in sulphuric acid solution by 2-mercapto-1-methylimidazole: Time and temperature effects treatments

Benali Omar ^{a,*}, Ouazene Mokhtar ^b

^a *Département de biologie, Faculté des sciences et de la technologie, Université de Saïda, 20000 Saïda, Algeria*

^b *Département de Chimie, Faculté des sciences et de la technologie, Université de Saïda, 20000 Saïda, Algeria*

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Abstract 2-Mercapto-1-methylimidazole (MMI) has been evaluated as a corrosion inhibitor for cold rolled steel in aerated 2 M H₂SO₄ by gravimetric method. The effect of MMI on the corrosion rate was determined at various immersions time and concentrations. The effect of the temperature on the corrosion behaviour with addition of different concentrations of MMI was studied in sulphuric acid medium. The inhibition process is attributed to the formation of an adsorbed film of MMI on the metal surface which protects the metal against corrosion. The protection efficiency increased with increase in inhibitor concentration at various immersions time and decreased with increase in temperature. Adsorption of MMI on the cold rolled steel surface is found to obey the Langmuir adsorption isotherm. Some thermodynamic functions of dissolution and adsorption processes were also determined.

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1. Introduction

The use of chemical inhibitors to decrease the rate of corrosion processes has been the focus of very many efforts within the chemical process industry (Álvarez-Bustamante et al., 2009). In recent years, there is a considerable amount of effort devoted to find novel and efficient corrosion inhibitors and sulphur and/or nitrogen containing molecules have been found to be effective corrosion inhibitors (Larabi et al., 2005, 2006; Ali et al., 2008, 2003; Wahyuningrum et al., 2008; Benmessaoud et al., 2007; Refaey et al., 2006; Kaniskan and Ogretir, 2002). The corrosion inhibition of cold rolled steel is a subject of large-scale technological importance, due to the fundamental economical implications of this material in the industrial medium (Tang et al., 2005, 2006; Mu et al., 2005; Qu et al.,

* Corresponding author. Tel.: +213 662220312.
E-mail addresses: benaliomar@hotmail.com, o_benali_2002@yahoo.fr (B. Omar).

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2009a,b). Recently our research group has shown (Benali et al., 2005, 2007; Larabi et al., 2006) that 2-mercapto-1-methylimidazole (MMI) can be a very useful corrosion inhibitor for steel and copper under acidic conditions. The latter gives rise to a considerable molecular adsorption of the MMI on the metal surface exposed to the H_2SO_4 electrolyte, which has been widely used to evaluate corrosion inhibitive properties of different molecules (Hosseini and Azimi, 2009; Abdel Rehim et al., 2008).

2. Experimental conditions

The inhibitor used is 2-mercapto-1-methylimidazole (Merck).

The molecular structure of the inhibitor is shown in Fig. 1.

A H_2SO_4 solution was prepared from an analytical reagent grade of H_2SO_4 96% and double-distilled water and was used as the corrosion medium in the studies. For the weight loss measurements, the experiments were carried out in a solution of 2 M H_2SO_4 (uninhibited and inhibited) on cold rolled carbon steel containing $\text{C} \leq 0.15\%$, $\text{Mn} \leq 0.50\%$, $\text{P} \leq 0.03\%$, $\text{S} \leq 0.03\%$ and the rest iron. The specimens for weight loss measurements were 15 mm by 15 mm by 1.4 mm. They were polished with emery paper (from 400 to 1000 grade). Each run was carried out in a glass vessel containing 50 ml test solution. A clean, weighed mild steel specimen was completely immersed at an inclined position in the vessel. After different times of immersion in 2 M H_2SO_4 with and without the addition of MMI at different concentrations, the specimen was withdrawn, rinsed with distilled water, washed with acetone, dried and weighed using an analytical balance accurate to 0.1 mg. The weight loss was used to calculate the corrosion rate in milligrams per square centimeter per hour.

3. Results and discussion

3.1. Influence of time of immersion on inhibition

The value of corrosion rate was calculated from the following equation (Obot and Obi-Egbedi, 2008):

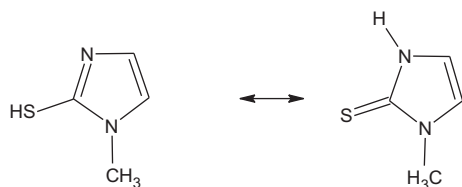


Figure 1 Molecular structures of 2-mercapto-1-methylimidazole.

$$V_{\text{corr}} = (m_1 - m_2) / St \quad (1)$$

where m_1 is the mass of the steel coupon before immersion in (mg), m_2 the mass of the steel coupon after immersion in (mg), S is the total area of the cold rolled steel coupon in (cm^2), t is the corrosion time in hours (h) and V is the corrosion rate in ($\text{mg cm}^{-2} \text{h}^{-1}$). The calculated corrosion rate (V_{corr}) in Eq. (1) is an average corrosion rate.

From the corrosion rate, the percentage inhibition efficiency IE% and the degree of surface coverage (θ) were calculated using Eqs. (2) and (3), respectively (Obot and Obi-Egbedi, 2008):

$$\text{IE}\% = (V_{\text{corr0}} - V_{\text{corr}}) / V_{\text{corr0}} \times 100 \quad (2)$$

$$\theta = (V_{\text{corr0}} - V_{\text{corr}}) / V_{\text{corr0}} \quad (3)$$

where V_{corr0} and V_{corr} are the corrosion rates of the mild steel coupon in 2 M H_2SO_4 in the absence and presence of MMI.

Values of the inhibition efficiency and corrosion rate obtained from the weight loss measurements of cold rolled steel for different concentrations of MMI in 2 M H_2SO_4 at 30 °C after 90, 180, 270 and 360 min of immersion are given in Table 1 and Figs. 2 and 3.

This shows that the inhibition efficiency increases with the increasing inhibitor concentration. At this point, one observes that the optimum concentration of inhibitor required to achieve the efficiency is found to be 2.5×10^{-3} M (inhibition

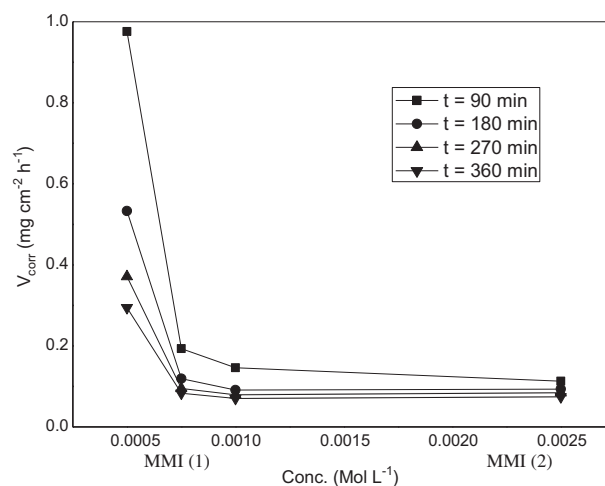


Figure 2 Corrosion rate of cold rolled steel efficiency for different concentrations of MMI in 2 M H_2SO_4 at different immersion times.

Table 1 Corrosion rate of cold rolled steel and inhibition efficiency for different concentrations of MMI for the corrosion of cold rolled steel in 2 M H_2SO_4 at different immersion times.

Concentration (M)	Immersion time							
	90 min		180 min		270 min		360 min	
	V_{corr}	P%	V_{corr}	P%	V_{corr}	P%	V_{corr}	P%
Blanc	6.845	—	6.475	—	6.273	—	5.595	—
5×10^{-4}	0.976	85.74	0.533	91.76	0.371	94.08	0.294	94.74
7.5×10^{-4}	0.193	97.18	0.119	98.27	0.095	98.48	0.083	98.52
10^{-3}	0.146	97.85	0.091	98.59	0.079	98.74	0.070	98.74
2.5×10^{-3}	0.113	98.35	0.093	98.57	0.084	98.66	0.074	98.67
5×10^{-3}	0.173	97.47	0.147	97.73	0.131	97.91	0.124	97.77

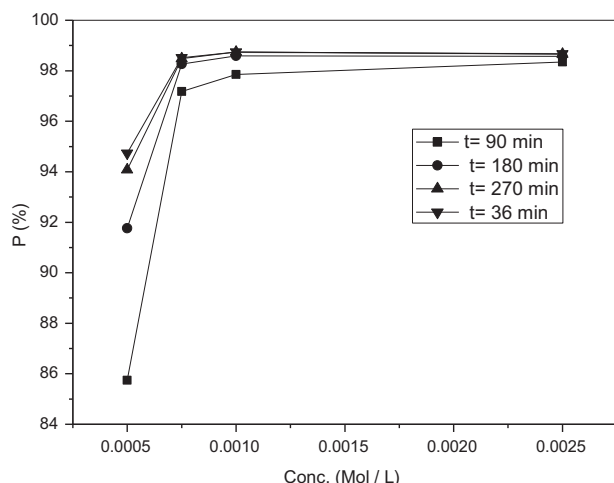
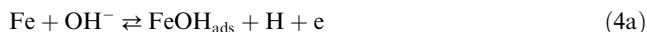


Figure 3 The inhibition efficiency for different concentrations of MMI for the corrosion of cold rolled steel in 2 M H₂SO₄ at different immersion times.

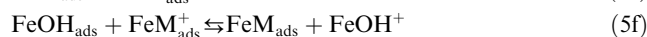
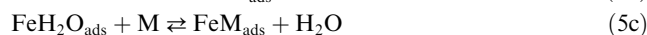
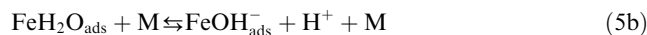
efficiency $\geq 98\%$ for this optimal concentration). The increase in the inhibition efficiency may be due to the adsorption of MMI to form an adherent film on the metal surface and suggests that the coverage of the metal surface with this film decreases the double layer thickness. The augmentation in the inhibition efficiency values may be due to the gradual replacement of water molecules by the adsorption of the MMI molecules on the metal surface, decreasing the extent of dissolution reaction (Benali et al., 2005; Bentiss, 2002). The inhibition of corrosion of mild steel by MMI can be explained in terms of adsorption on the metal surface. This compound can be adsorbed on the metal surface by the interaction between lone pairs of electrons of nitrogen and sulphur atoms of the inhibitor and the metal surface or via interaction of MMI with already adsorbed sulphate ions (Benali et al., 2005; Hackerman, 1966). This process is facilitated by the presence of vacant orbitals of low energy in iron atom, as observed in the transition group metals.

A number of mechanistic studies on the anodic dissolution of Fe in acidic sulphate solutions have been undertaken, and the hydroxyl accelerated mechanism proposed initially by Bockris and Drazic (1962) and reported by Oguzie (2004) and Obot and Obi-Egbedi (2010) has gained overwhelming acceptance:



It has been suggested that anions such as Cl⁻, I⁻, SO₄²⁻ and S²⁻ may also participate in forming reaction intermediates on the corroding metal surface, which either inhibit or stimulate corrosion (Hurlen et al., 1984). It is important to recognize that the suppression or stimulation of the dissolution process is initiated by the specific adsorption of the anion on the metal surface. A representative plot of inhibition efficiency against immersion time for cold rolled steel in 2 M H₂SO₄ in the absence and presence of different concentrations of MMI at 30 °C is presented in Fig. 3.

According to the Bockris mechanism outlined earlier, Fe electro-dissolution in acidic sulphate solutions depends primarily on the adsorbed intermediate FeOH_{ads}. Ashassi-Sorkhabi and Nabavi-Amri (2000) proposed the following mechanism involving two adsorbed intermediates to account for the retardation of Fe anodic dissolution in the presence of an inhibitor:



where M represents the inhibitor species.

Considering the inhomogeneous nature of metallic surfaces resulting from the existence of lattice defects and dislocations, a corroding metal surface is generally characterized by multiple adsorption sites having activation energies and heats of adsorption. Inhibitor molecules may thus be adsorbed more readily at surface active sites having suitable adsorption enthalpies. According to the detailed mechanism above, displacement of some adsorbed water molecules on the metal surface by inhibitor species to yield the adsorbed intermediate FeM_{ads} (Eq. (5c)) reduces the amount of the species FeOH_{ads}⁻ available for the rate-determining steps and consequently retards Fe anodic dissolution (Obot and Obi-Egbedi, 2010).

Table 2 The influence of temperature on corrosion rate parameters and inhibition efficiency for cold rolled steel electrode immersed in 2 M H₂SO₄ and different concentrations of MMI.

C (M)	Temp. (K)	Immersion time			
		90 min		360 min	
		<i>V</i> _{corr}	P%	<i>V</i> _{corr}	P%
Blank	303	6.845	—	5.595	—
	313	9.585	—	8.349	—
	323	11.557	—	12.58	—
	333	20.43	—	25.36	—
5 × 10 ⁻⁴	303	0.976	85.74	0.294	94.74
	313	4.435	53.73	2.43	70.89
	323	16.19	-40.08	17.98	-42.92
	333	36.09	-76.65	34.41	-35.68
7.5 × 10 ⁻⁴	303	0.193	97.18	0.083	98.52
	313	0.889	90.72	0.331	96.03
	323	1.27	89.01	0.805	93.60
	333	3.86	81.10	2.95	88.36
10 ⁻³	303	0.146	97.85	0.070	98.74
	313	0.349	96.35	0.138	98.34
	323	0.360	96.88	0.241	98.08
	333	2.44	88.05	1.136	95.52
2.5 × 10 ⁻³	303	0.113	98.35	0.074	98.67
	313	0.245	97.44	0.117	98.59
	323	0.236	97.95	0.187	98.51
	333	0.44	97.84	0.336	98.67

3.2. Effect of temperature

To investigate the mechanism of inhibition and to calculate the activation energies of the corrosion process, weight loss measurements were taken at various temperatures in the absence and the presence of different concentrations of MMI at different time immersions. Corresponding data are given in Table 2, Figs. 4 and 5.

In the studied temperature range (30–60 °C) the corrosion rate increases with increasing temperature both in uninhibited and inhibited solutions and the values of the inhibition efficiency of MMI are nearly constant for the optimal concentration. Thus, the corrosion rate of cold rolled steel increases more rapidly with temperature in the absence of the inhibitor. For the lower concentration (5×10^{-4}) differently to temperature 30 and 40 °C the inhibitor stimulates the corrosion of cold rolled steel for 50 and 60 °C.

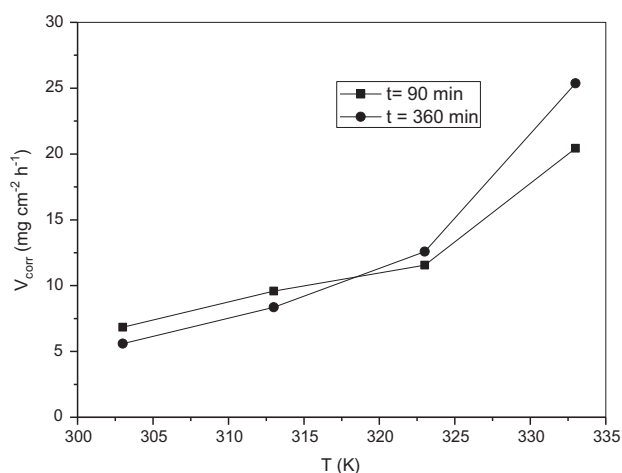


Figure 4 Effect of temperature on the corrosion rate of cold rolled steel in 2 M H_2SO_4 at immersion times: $t = 90$ and $T = 360$ min.

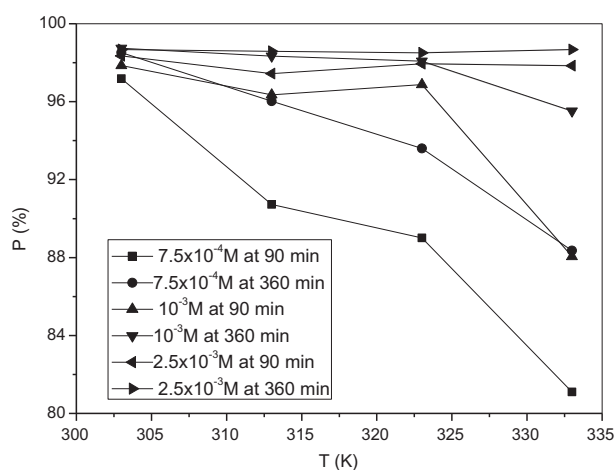


Figure 5 Effect of temperature on the corrosion rate of MMI of corrosion of cold rolled steel in 2 M H_2SO_4 at immersion times: $t = 90$ and $t = 360$ min.

The activation parameters for the corrosion process were calculated from Arrhenius type plot according to the following equation:

$$\ln V_{\text{corr}} = -\frac{E_a}{RT} + \log A \quad (6)$$

where E_a is the apparent activation energy, A the pre-exponential factor, R the universal gas constant and T the absolute temperature.

The variations of logarithm of the corrosion rate of carbon steel ($\log V_{\text{corr}}$) in sulphuric acid containing various concentrations of MMI used with reciprocal of the absolute temperature are presented in Fig. 6. Straight lines with coefficients of correlation (c.c.) higher to 0.98 are obtained.

The calculated values of E_a and pre-exponential factor A for two different immersion times are recorded in Table 3. The values of E_a and pre-exponential factor A increased with increase in MMI concentration and the extent of the increase is very important at 7.5×10^{-4} M. For the present study, the values of E_a and pre-exponential factor A in the presence of MMI are higher than those values in its absence. In the literature, the lower activation energy value of corrosion process in the presence of inhibitor (rather than the absence of inhibitor) is attributed to its chemisorption, while it is found to be oppo-

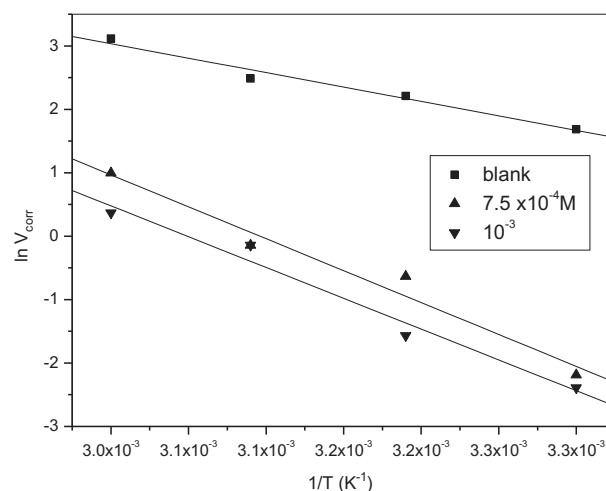


Figure 6 $\ln V_{\text{corr}}$ vs $1/T$ for cold rolled steel dissolution in 2 M H_2SO_4 in the absence and the presence of 7.5×10^{-4} M and 10^{-3} M concentrations of MMI at immersion time: $t = 180$ min.

Table 3 The apparent activation energy E_a and pre-exponential factor A of dissolution of cold rolled steel in 2 M H_2SO_4 in the absence and presence of 7.5×10^{-4} and 10^{-3} of MMI.

Immersion time (min)	Concentration (mol/L)	Pre-exponential factor A	E_a (kJ/mol)
90	Blank	6.15×10^5	28.85
	7.5×10^{-4}	1.09×10^{12}	73.66
	10^{-3}	1.07×10^{12}	69.95
360	Blank	5.94×10^7	40.95
	7.5×10^{-4}	3.71×10^{15}	96.54
	10^{-3}	3.19×10^{11}	73.90

site in the case with physical adsorption (Avci, 2008; Oquzie, 2006). In our case MMI showed the presence of physical adsorption (physiosorption) of MMI onto cold rolled steel surface (Refaey et al., 2006; Fouda, 2007; Morad and Kamal El-Dean, 2006). In addition, the higher the activation energy, the lower the corrosion current density, that indicates the inhibition of corrosion process. The lower the corrosion rate, the less dense the electron transfer in oxidation–reduction process (corrosion), therefore the corrosion reaction rate became slower (Wahyuningrum et al., 2008). On the other hand, we remark that the values of E_a and pre-exponential factor A increase with increase of the immersion time.

3.3. Adsorption isotherm

The inhibitor efficiency depends on the type and number of active sites at the metal surface, the charge density, the molecular size of the inhibitor, the metal–inhibitor interaction, and the metallic complex formation. The adsorption isotherm can give information on the metal–inhibitor interaction. The adsorption isotherm can be derived from the curve surface coverage against inhibitor concentration. Surface coverage was estimated as Eq. (3). The θ values for different inhibitor concentrations at different immersion times were tested by fitting to various isotherms. By far the best fit was obtained with the Langmuir isotherm. According to this isotherm θ is related to concentration inhibitor C via

$$\frac{C}{\theta} = \frac{1}{K} + C \quad (7)$$

with

$$K = \frac{1}{55.5} \exp \left(-\frac{\Delta G_{\text{ads}}^0}{RT} \right) \quad (8)$$

where K is the adsorptive equilibrium constant and ΔG_{ads}^0 the free energy of adsorption. It was found that Fig. 7 (plot of $\frac{\theta}{C}$ versus C) gives straight lines with slope equal or nearly equal to 1.00 for two immersion times (90 and 360 min) at temperature 30 °C and intercepts provide the calculation of the values of K are given in Table 2. The obtained results indicate that the adsorption of the compound under consideration on cold rolled steel / acidic solution interface at all temperatures (30, 40, 50 and 60 °C) follows the Langmuir adsorption isotherm.

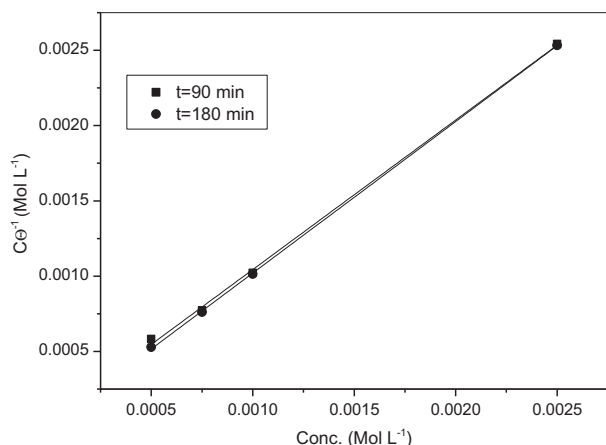


Figure 7 Langmuir adsorption plots for cold rolled steel in 2 M H_2SO_4 containing different concentrations of MMI at $T = 30^\circ\text{C}$.

Table 4 Some parameters of the linear regression between C/θ and C for two immersion times and different temperatures.

Temperature (°C)	Immersion time (min)	K	Slope	ΔG_{ads} (kJ/mol)
30	90	4×10^4	1.00	−36.81
	360	7.9×10^4	1.01	−38.54
40	90	5.2×10^3	0.98	−32.76
	360	1.3×10^4	1.00	−35.11
50	90	3.7×10^4	1.02	−39.05
	360	8.9×10^4	1.02	−41.39
60	90	8.2×10^3	1.00	−36.09
	360	2.7×10^4	1.02	−39.42

Inspection of the data of the Table 4 shows the following adsorption characteristics of MMI in 2 M H_2SO_4 :

- Values of slopes are more than unity indicating that each molecule of the inhibitor is attached to one active site of the steel surface.
- Globally, the value of the equilibrium constant K , decreases with temperature (for the two immersion times) suggesting that this inhibitor is physically adsorbed on the metal surface (Abd el rehim et al., 2001) and desorption process enhances with elevating the temperature.
- All values of negatives ΔG_{ads} show that the adsorption of MMI is a spontaneous process under the experimental conditions described (Tang et al., 2006; Fouda et al., 1989) and also the strong interaction between inhibitor molecules and the metal surface (Kertit and Hammouti, 1996).
- The value of ΔG_{ads} is less than 40 kJ/mol is commonly interpreted with the presence of physical adsorption by the formation of an adsorptive film with an electrostatic character (Moretti et al., 2004; Abiola and Oforka, 2004). These results are in good agreement with those obtained preliminary with the apparent activation energy.
- On the other hand, the calculated $-\Delta G_{\text{ads}}$ values in the range 30–40 kJ/mol indicate, therefore, that the adsorption mechanism of the MMI on cold rolled steel in 2 M H_2SO_4 solution was both electrostatic-adsorption and chemisorption (Duan and Tao, 1990; Bransoi et al., 2001). MMI possess aromatic π -electrons as well as unshared electrons pairs on the nitrogen and sulphur atoms, which can interact with d-orbitals of iron to provide a protective film (Bentiss, 2002; Kertit and Hammouti, 1996).

4. Conclusion

In this study, the inhibition property of 2-mercapto-1-methylimidazole (MMI) was tested by using weight loss measurements. According to the results, 2-mercapto-1-methylimidazole (MMI) is a good inhibitor in 2 M H_2SO_4 . The optimum concentration of the inhibitor is 2.5×10^{-3} M for an inhibition efficiency of $\geq 98\%$. The protection efficiency increased with increasing inhibitor concentration, but decreased slightly with the rise of temperature. The calculated activation energy value indicated that adsorbed inhibitor molecules create a physical barrier to charge, and mass transfer for metal dissolution and hydrogen reduction reaction. Variation of corrosion rate with exposure time, and absence and presence

of inhibitor molecules were also investigated in this study because immersion is also important for the corrosion process. Adsorption of the inhibitor molecule onto mild steel surface obeys the Langmuir isotherm.

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