

Corrosion inhibition effect of sodium iodide for mild steel in 1 M hydrochloric acid: Gravimetical and electrochemical studies

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Abstract

The corrosion inhibition behavior of mild steel in 1 M HCl was investigated in the absence and the presence of sodium iodide (NaI) as an inorganic corrosion inhibitor. The influence of temperature, time and inhibitor concentration was studied using weight loss technique and Tafel polarization technique. The results showed that NaI act as a powerful inhibitor and the inhibition efficiency increase with increase in inhibitor concentration, time and temperature. Maximum inhibition efficiency was 96.1% at a higher level of inhibitor concentration, time and temperature. The adsorption of NaI on mild steel surface was found to follow Langmuir adsorption isotherm. The values of the free energy of adsorption were between –20 and –40 kJ/mol that indicate the cooperative adsorption (physical and chemical adsorption). The activation energy of the corrosion process was evaluated using Arrhenius-type equation. Electrochemical studies confirmed the inhibition action of NaI. Corrosion current densities moved to lower value with the addition of NaI. Inhibition efficiency values were in a good agreement with weight loss results. The shift of corrosion potential by iodide anion was lower than 85 mV indicates that it inhibits the anodic reaction and the cathodic reaction, which confirm that sodium iodide act as a mixed – type inhibitor.

Keywords: corrosion, inorganic inhibitor, mild steel, acidic solution.

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1. Introduction

Corrosion can be defined as an irreversible reaction of a material with the environment which usually (but not always) results in the deterioration of a metal by chemical or electrochemical reaction. There are several parameters that influence the rate and nature of the corrosion reactions; these include the material properties, type of environment, temperature, velocity, *etc.* [1, 2]. Corrosion is one of the main concerns in the durability of metallic materials and their structures. Iron and its alloys represent one of the most widely used materials in industrial applications. It can be contacted with different environments,

such as acidic solutions during the process of pickling and acid cleaning [3]. Acids are very aggressive media that strongly damaged the steel structures. Many efforts have been made to develop a corrosion control technique to prolong the life of existing structures and minimize corrosion damages [4]. One of these methods is by using a corrosion inhibitor. Corrosion inhibitor is a substance which, when added in small concentration to an environment effectively reduces the corrosion process [5]. Different mechanisms are suggested to explain the action of these anticorrosion materials. The most common mechanism is the adsorption of inhibitor molecules on the surface of metals, usually, by replacement of water molecules adsorbed on the metal surface [6]. Most of researchers focused on using organic corrosion inhibitors [7], synthesis inhibitors [8], and blends of inhibitors [9]. Limited researches are taking into account the using of inorganic corrosion inhibitors in acidic solutions. The present work is an attempt in the direction of using NaI as an inorganic corrosion inhibitor for mild steel in hydrochloric acid solution.

2. Experimental work

2.1. Weight loss experiments

A corrosive solution of 1 M HCl was prepared by dilution of analytical grade 37% HCl with distilled water. Sodium iodide (NaI) was used as a corrosion inhibitor. Saturated solution of NaI was prepared and added to 1 M HCl in concentrations of 0.5, 1, 1.5, 2, and 2.5 ml/l. Corrosion rates were evaluated at different temperatures (30, 40, 50, and 60°C) and different exposure times (1, 2, 3, and 4 h). Mild steel samples with dimensions of 1×3×0.1 cm were used during the tests. Carbon steel has the following chemical compositions (%wt): 0.1 C, 0.335 Mn, 0.033 Si, 0.0067 S, 0.0056 P, 0.057 Al, 0.0476 Cu, 0.0201 Cr, 0.001 Co, 0.0007 Ti and the balance is Fe. Each sample was completely immersed in a 250 ml conical flask with 1 M HCl for a specific period of time. Weight loss method represents widest and simplest way for measuring corrosion rates. Steel samples were first degreased with benzene and acetone, and then annealed in a vacuum at 600°C for 1 hour, and cooled to room temperature. Samples were abraded in sequence under running tap water using emery paper of grad number 200, 400, 600, 800 and 1000 then washed with running tap water followed by distilled water, dried with clean tissue, immersed in acetone and benzene, kept in desiccators over silica gel bed until use. Mass of each sample was accurately weighed to the fourth decimal of gram and dimensions were measured with a vernier to second decimal of millimeter. After tests, metal samples were cleaned, washed with running tap water followed by distilled water, dried with clean tissue, then immersed in acetone and benzene and dried again. Weight losses in $\text{g m}^{-2} \text{ day}^{-1}$ were determined in the presence and absence of inhibitor. Each test was repeated twice and the average value of corrosion rate was taken.

2.2. Polarization experiments

Electrochemical measurements were conducted using a CHI760E electrochemical workstation in a conventional three electrodes corrosion cell. A platinum electrode was used as counter electrode, while mild steel was used as working electrode. Saturated calomel electrode (SCE) was used as a reference electrode. NaI with the concentration of 0.5, 1, 1.5, 2, and 2.5 ml/l was added to the 1 M HCl corrosive solution. Carbon steel samples with chemical compositions (as mentioned above) were used. The mild steel specimens were injected in epoxy resin leaving a working surface area of 1 cm². The electrode potential was measured against the SCE and the polarization curves were recorded at a scanning rate of 1 mV/s with various potential from the detected open circuit potential (OCP), following 1200 seconds of immersion. The anodic and cathodic curves of the linear Tafel region were extrapolated to corrosion potential to achieve the corrosion current densities. Polarization from the lower cathodic potential of –800 mV to anodic potential of –100 mV was evaluated. All experiments were carried out at temperatures of 30, 40, 50 and 60°C.

3. Results and discussion

3.1. Weight loss studies

The outcomes of weight loss data are summarized in Table 1. The weight loss due to corrosion of the mild steel alloy in 1 M hydrochloric acid solution at various temperature and NaI concentrations was evaluated. The corrosion rate value was evaluated *via* the following equation:

$$CR = \frac{\text{weight loss (g)}}{\text{area (m}^2\text{)} \times \text{time (day)}} \quad (1)$$

Corrosion rate (CR) can be used to calculate the percentage of inhibition efficiency (%IE_w) according to Eq. 2:

$$\%IE_w = \frac{CR_0 - CR}{CR_0} \times 100 \quad (2)$$

Where CR and CR₀ are corrosion rates in the presence and absence of NaI, respectively. In the uninhibited and inhibited acid solutions, the corrosion rate of mild steel increases with an increase in temperature. Table 1 shows the effect of inhibitor concentration (C), temperature (T) and time (t) on corrosion rate. It is clear from Table 1 that the rate of mild steel dissolution in 1 M HCl is nearly double for each 10°C rise in temperature. This result was agreed with many values in the literature for the corrosion of steel in acidic solution [10]. It is also obvious that at a specific test temperature, the corrosion rate of steel decreases with an increase in the NaI concentration. Corrosion rate has slightly decreased with time. In the first hour and the absence of inhibitor, the corrosion rate is high, with

time, it decreases may be due to the formation of corrosion products on steel surface. Same behavior was observed in the presence of NaI, but the corrosion rate can be reduced due to both; formation of corrosion products on inhibitor free – metal surface and increasing in the adsorption of NaI molecules on metal surface.

Table 1. Weight loss data for mild steel in 1 M HCl acid without and with different concentrations of NaI at different temperatures and times.

No.	C (ml/l)	T (°C)	Corrosion rate (gmd)			
			1 h	2 h	3 h	4 h
1	0	30	45.011	42.254	38.2176	36.324
2	0.5		20.341	18.736	15.9912	13.317
3	1		18.844	16.519	13.356	12.237
4	1.5		17.038	15.494	12.1272	10.888
5	2		15.3312	13.781	10.939	9.605
6	2.5		13.6224	12.024	10.351	7.8408
7	0	40	74.935	72.842	51.876	48.698
8	0.5		32.208	30.748	18.864	17.635
9	1		27.465	26.107	15.693	14.431
10	1.5		24.069	21.465	13.881	12.604
11	2		22.144	18.098	12.127	9.991
12	2.5		17.054	15.472	9.781	8.265
13	0	50	177.78	109.05	107.913	98.796
14	0.5		57.715	34.166	24.0312	21.528
15	1		48.18	28.687	23.2584	19.696
16	1.5		37.785	22.459	21.9984	17.872
17	2		34.128	20.738	19.6104	16.965
18	2.5		30.626	18.014	16.7304	9.5784
19	0	60	436.87	316.29	295.37	266.57
20	0.5		127.46	81.288	48.127	40.917
21	1		103.96	67.901	32.054	26.301
22	1.5		77.755	55.416	28.468	20.469
23	2		69.456	50.143	27.228	18.696
24	2.5		44.572	30.991	17.282	10.456

Table 2 shows the effect of inhibitor concentration, temperature and time on inhibition efficiency. Inhibition efficiency values were enhanced with increase in the NaI concentration, while %IE increases when increasing the temperature reaching a maximum value of 96.07%. The increase in sodium iodide performance with temperature is probably due to an increase in the adsorption of inhibitor molecules on metal surface. Adsorption represents an important step in studying the inhibitor performance. It assumes that corrosion reactions prevented from occurring over active sites or areas of the metal surface protected by adsorbed NaI molecules [11]. The surface coverage ($\theta = \%IE/100$) is very valuable data when discussing adsorption modes. This formula can be used based on the assumption on the blocking mechanism of inhibitive action of iodide anion. The surface coverage is related to the inhibitor concentration at a constant temperature by the well-known adsorption isotherm relationships that can be evaluated at the equilibrium conditions. Langmuir adsorption isotherm assumes a fixed number of adsorption sites on a mild steel surface, and each site takes only one adsorbed molecule. Linear plots for C/θ versus C can be drawn at different conditions.

Figure 1 is an example of Langmuir adsorption isotherm for adsorption of NaI molecules on the low carbon steel surface at 60 °C. Mathematically, Langmuir adsorption isotherm can be written as:

$$\frac{C}{\theta} = \frac{1}{K_{\text{ads}}} + C \quad (3)$$

Where K_{ads} is the adsorption equilibrium constant, demonstrating the degree of adsorption. The higher the values of K_{ads} , the more the NaI molecules are adsorbed strongly on the surface of metal, while C represents the NaI concentration. Values of K_{ads} and slope of Langmuir adsorption lines are listed in Table 3. The slope of Langmuir lines are near unity, which means that each active site on the metal surface occupies one inhibitor molecule. Values of K_{ads} are increased with time and temperature, which indicate that adsorption enhanced with time and elevation of temperature. The adsorption free energy (ΔG_{ads}) was determined using the following equation [12]:

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

The number 55.5 represents the water concentration in the solution expressed in molar (it converted to ml/l for dimensional consistency), T is the absolute temperature (in K), and R is the gas constant. The ΔG_{ads} values are shown in Table 3. The negative value of ΔG_{ads} ensures the stability of the adsorbed layer on the metal surface and spontaneous process of adsorption. Commonly, the value of ΔG_{ads}^0 up to $-20 \text{ kJ}\cdot\text{mol}^{-1}$ assumes the electrostatic interaction between the charged metal and the charged inhibitor molecules (physical adsorption), while those around $-40 \text{ kJ}\cdot\text{mol}^{-1}$ or higher are related to chemical

adsorption due to transferring or sharing electrons from the molecules to the metal surface that form a coordinate bond [13]. In the present work ΔG_{ads}^0 values were higher than -20 and lower than $-40 \text{ kJ}\cdot\text{mol}^{-1}$. In other words, the adsorption is neither classic physical adsorption nor classic chemical adsorption. It is a complex mixed mode that is the adsorption of inhibitor molecules on the metal surface in current work involves both physical and chemical adsorption and physical adsorption is predominant mode. This hypothesis is confirmed by data obtained from the dependence of temperature for inhibition process [13]. Table 3 also indicates an increase in the absolute value of ΔG_{ads}^0 with an increase in temperatures.

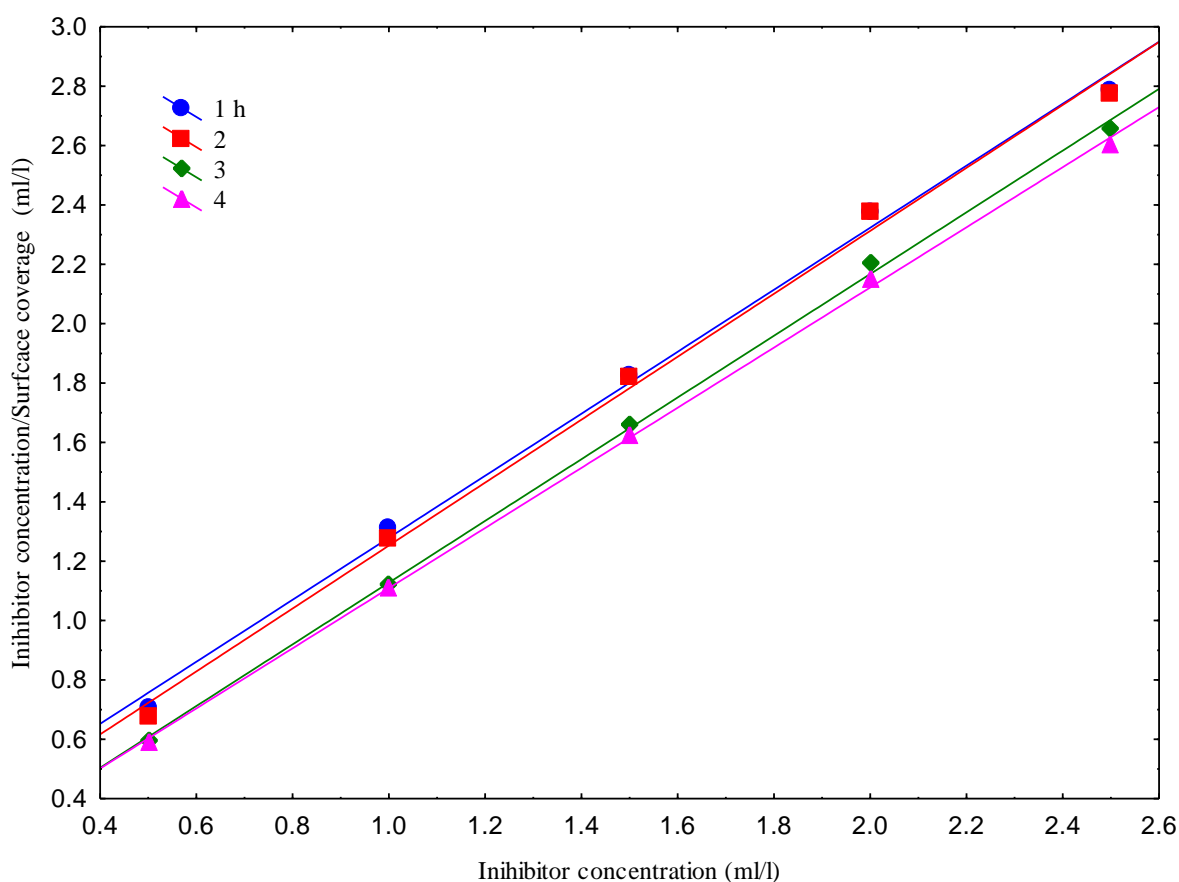


Figure 1. Langmuir adsorption isotherm at 60°C .

Kinetics of inhibitor action can be studied by relating the activation energy (E_a) in the absence and presence of NaI. Activation energy (E_a) for both corrosion and corrosion inhibition of steel in hydrochloric acid at different temperatures were determined using an Arrhenius-type equation [14]:

$$\text{CR} = A \exp\left(-\frac{E_a}{RT}\right) \quad (5)$$

Table 2. Surface coverage data for NaI on mild steel in 1 M HCl acid different temperatures and times.

<i>C</i> (ml/l)	<i>T</i> (°C)	Surface coverage			
		1 h	2 h	3 h	4 h
0.5	30	0.548	0.556	0.581	0.633
1		0.581	0.609	0.651	0.663
1.5		0.621	0.633	0.682	0.70
2		0.659	0.673	0.713	0.735
2.5		0.697	0.715	0.729	0.784
0.5	40	0.571	0.577	0.633	0.638
1		0.633	0.641	0.697	0.703
1.5		0.678	0.705	0.732	0.741
2		0.704	0.751	0.766	0.794
2.5		0.772	0.787	0.811	0.831
0.5	50	0.675	0.686	0.777	0.782
1		0.728	0.736	0.784	0.801
1.5		0.787	0.794	0.796	0.819
2		0.808	0.809	0.818	0.828
2.5		0.827	0.834	0.844	0.903
0.5	60	0.708	0.742	0.837	0.846
1		0.762	0.785	0.891	0.901
1.5		0.822	0.824	0.904	0.923
2		0.841	0.841	0.908	0.929
2.5		0.897	0.902	0.9414	0.961

The linear form of Eq. 5 can be written as:

$$\ln(\text{CR}) = \ln A - \frac{E_a}{RT} \quad (5a)$$

CR is the average corrosion rate, *A* is the constant or frequency factor of Arrhenius equation, and *R* is the universal gas constant. Eq. 5a can be plotted as the natural logarithm of corrosion rate ($\ln(\text{CR})$) against reciprocal of absolute temperature ($1/T$) as shown in Figure 2. Slopes and intercepts of Arrhenius equations lines can be used to estimate the values of activation energy (E_a) and frequency factors (*A*) respectively. Table 4 illustrates the activation energy and frequency factors for the metal corrosion reactions in inhibited

and uninhibited acidic solution. The activation energy in the absence of NaI was higher than the values of inhibited one. The reduction in the value of E_a seems to be unreliable, which may be attributed to an increase in the portion of surface covered by the NaI molecules at elevated temperatures [15]. Another explanation suggested that some types of inhibitors of corrosive acidic solutions alter the reaction kinetics of corrosion process by proposing different reaction routes with lower activation energies [16]. As shown in Table 4, frequency factor values in the presence of NaI were lower than those of the NaI-free solution that benefits for the inhibition of the corrosion rate. These results agree with many literature studies, which state that the rise in frequency factor increases the metal corrosion rate [17].

Table 3. Adsorption parameters for NaI on mild steel in 1 M HCl acid different temperatures and times.

Time (h)	T (°C)	K_{ads} (l/ml)	Slope	ΔG_{ads} (kJ/mol)
1	30	2.985	1.331	-20.154
2		3.136	1.304	-20.278
3		4.095	1.281	-20.951
4		2.726	1.202	-19.925
1	40	2.824	1.196	-20.675
2		2.771	1.144	-20.621
3		3.816	1.153	-21.458
4		3.496	1.109	-21.231
1	50	4.926	1.132	-22.829
2		5.157	1.129	-22.952
3		9.901	1.161	-24.704
4		6.518	1.0848	-23.581
1	60	4.255	1.044	-23.131
2		5.194	1.0601	-23.683
3		11.363	1.039	-25.851
4		10.384	1.0128	-25.601

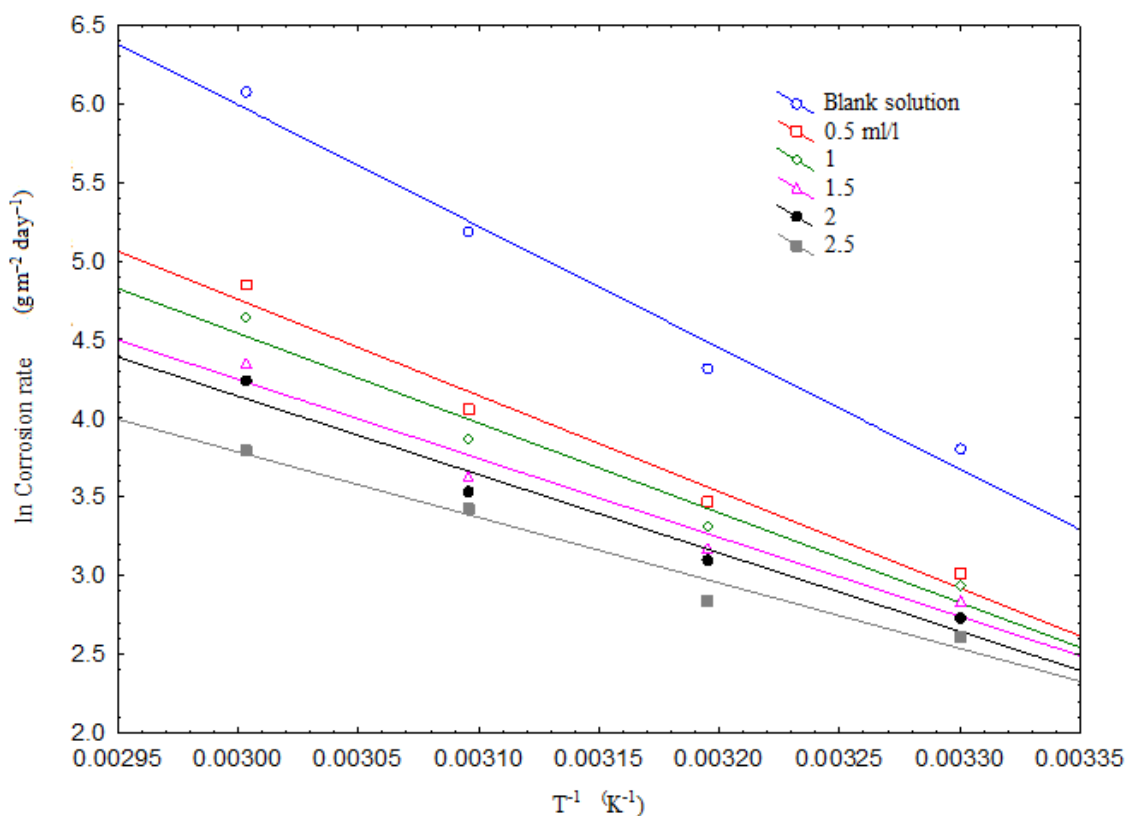


Figure 2. Arrhenius plots for corrosion of mild steel in absence and presence of NaI in 1 M HCl acid at average value of corrosion rate.

Table 4. Kinetics parameters for corrosion of mild steel in absence and presence of NaI in 1 M HCl acid at average value of corrosion rate.

<i>C</i> (ml/l)	<i>A</i> (gmd)	<i>E_a</i> (kJ/mol)
Blank	4.6×10^{12}	64.19
0.5	1.1×10^{10}	50.86
1	2.5×10^9	47.45
1.5	2.4×10^8	41.76
2	1.9×10^8	41.43
2.5	1.2×10^7	34.64

3.2. Electrochemical studies

Figure 3 (A–D) shows polarization plots of mild steel in 1 M HCl in the presence and absence of different levels of NaI at various temperatures. It is clear that the inhibitor

significantly reduces the current density of the metal. Polarization parameters in the presence and absence of NaI are given in Table 5. Table 5 and Figure 3 showed that NaI molecules shift current density towards lower lesser value without affecting the common behavior of Tafel curves, which indicate that NaI molecules retard steel corrosion by blocking the active sites on metal surface [18]. This blocking of active sites may be attributed due to adsorption of NaI. Observation of polarization curves shows that as compared to the Tafel slopes in the absence of NaI, in the inhibited solutions; both anodic and cathodic Tafel slopes values are less affected with the presence of inhibitor which indicates mixed type inhibitor and no change in the steel corrosion mechanism in the presence of the inhibitor [19]. A shift in the corrosion potential (E_{corr}) is another important criterion that gives information about the type of inhibitors. The positive shift of E_{corr} by iodide anion (up to 85 mV) indicates that it preferentially inhibits the anodic reaction. Since, according to voltammetric data, the cathodic reaction is also inhibited, it is more correct to consider this additive as a mixed type inhibitor [20] that predominantly inhibits the anodic reaction. These results agree with weight loss studies. In other words, the NaI molecules are adsorbed in both the anodic and cathodic sites, which reduce the main anodic reaction of the steel dissolution, as well as cathodic reactions in acidic media (oxygen reduction and hydrogen evolution).

The values inhibition efficiencies, according to the polarization measurements (%IE_p) can be calculated using Eq. 5, and listed in Table 1.

$$\% \text{IE}_p = \frac{i_{\text{corr}}^0 - i_{\text{corr}}}{i_{\text{corr}}^0} \times 100 \quad (5)$$

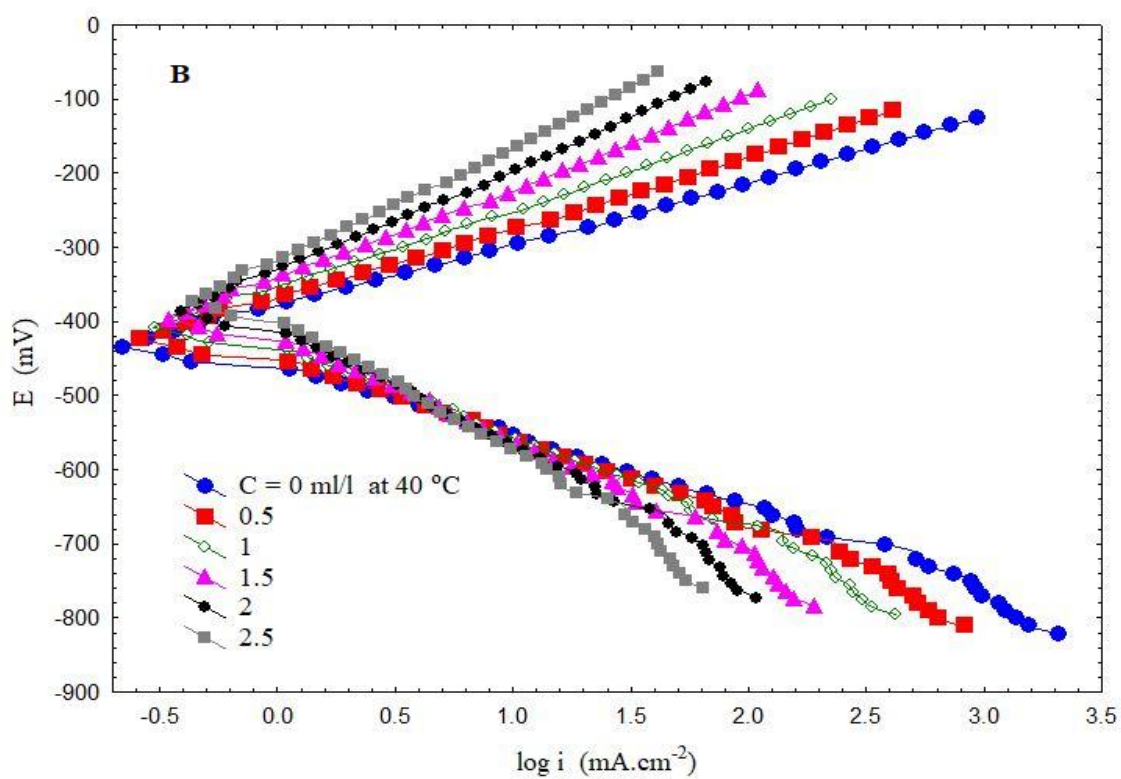
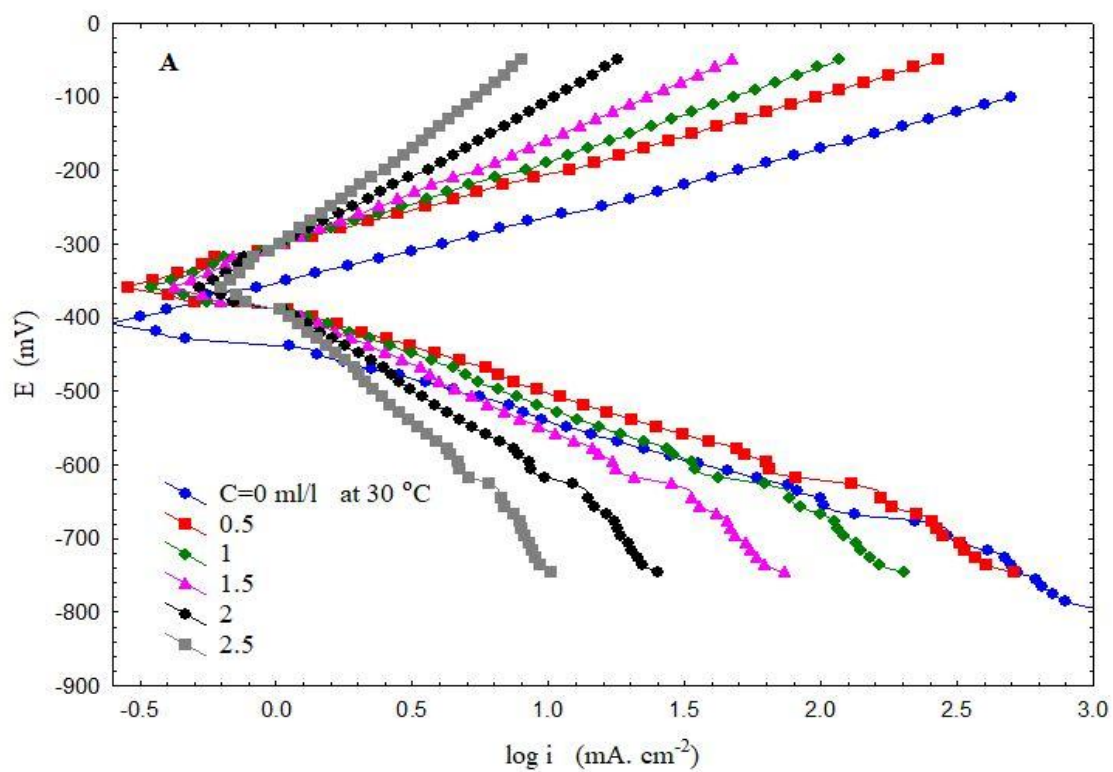
Where i_{corr}^0 and i_{corr} are corrosion current densities in the absence and presence of NaI, respectively. Inhibition efficiency increases with the NaI concentration due to an increase in the coverage of the steel surface by the adsorption of the inhibitor. Inhibition efficiencies from both, weight loss and polarization studies are comparable.

4. Conclusion

Sodium iodide acts as an excellent corrosion inhibitor for the corrosion of mild steel in 1 M HCl acid. Inhibition efficiency increases with inhibitor concentration and maximum inhibition efficiency for NaI was found to be 96.1% at 60°C and 2.5 ml/l NaI. The adsorption of different concentrations of NaI on the steel surface was, according to Langmuir adsorption isotherm. The negative sign of the free energy of adsorption indicates that the adsorption of NaI on the metal surface was a spontaneous process. Based on NaI effect on electrode processes; it is a mixed type inhibitor with predominant inhibition of the anodic reaction. It undergoes cooperative adsorption on steel, *i.e.*, it has indications of both physical and chemical adsorption.

Table 5. Polarization data for mild steel in 1 M HCl acid without and with different concentrations of NaI at different temperatures and times. Potential measured against standard calomel electrode (SCE).

No.	<i>C</i> (ml/l)	<i>T</i> (°C)	i_{corr} (mA/cm ²)	E_{corr} (mV)	b_a (mV/dec)	$-b_c$ (mV/dec)	%IE _P
1	0	30	3.16	-439	85	123	–
2	0.5		1.16	-402	88	125	63.3
3	1		1.06	-389	90	130	66.3
4	1.5		0.95	-410	86	135	70.1
5	2		0.84	-422	75	122	73.6
6	2.5		0.68	-415	80	140	78.4
7	0	40	4.24	-464	87	126	–
8	0.5		1.53	-454	90	128	63.8
9	1		1.26	-439	92	133	70.4
10	1.5		1.1	-427	88	138	74.1
11	2		0.87	-416	77	125	79.5
12	2.5		0.72	-403	82	143	83.1
13	0	50	8.59	-474	88	130	–
14	0.5		1.87	-459	91	132	78.2
15	1		1.71	-448	93	137	80.1
16	1.5		1.55	-436	89	142	81.9
17	2		1.48	-423	78	129	82.8
18	2.5		0.83	-424	83	147	90.3
19	0	60	23.19	-482	82	128	–
20	0.5		3.56	-472	85	130	84.7
21	1		2.29	-461	87	135	90.1
22	1.5		1.78	-451	83	140	92.3
23	2		1.63	-451	72	127	92.9
24	2.5		0.91	-439	77	145	96.1



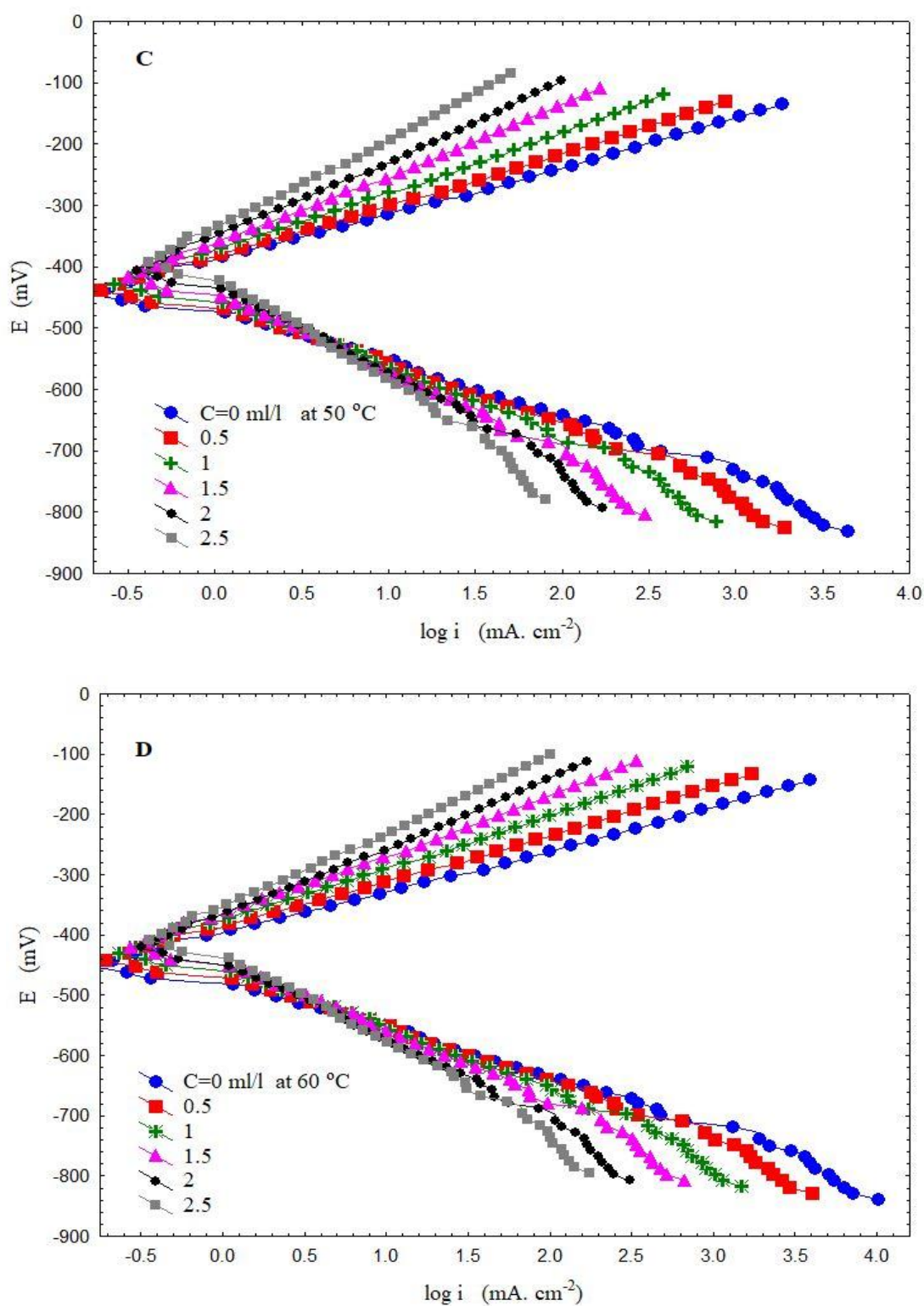


Figure 3. Polarization curves for corrosion of low carbon steel in uninhibited and inhibited acidic solution.

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