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# Synergistic Effect of Potassium Iodide on Inhibitive Performance of Propyl Alcohol during Corrosion of Mild Steel in 1.0M HCl

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#### Abstract

The inhibition of mild steel corrosion in 1.0M HCl by 1-propanol and the synergistic effect of potassium iodide (KI) was investigated using weight loss and polarization techniques in the temperature range  $(30 - 50)^{\circ}$ C. A matrix of Doelhert to three factors was used as the experimental design, adopting weight loss results as it permits the use of the response surface methodology which exploited in determination of the synergistic effect as inhibition on the mild steel. The results were confirmed using electrochemical polarization measurements. Experimental results showed that the inhibition efficiency (IE%) increases with increase in concentration of inhibitor and with increasing of temperature. The addition iodide ions to alcohol (inhibitor) enhanced the inhibition efficiency due to synergistic effect. Potentiodynamic polarization studies showed that the studied compound is mixed-type inhibitor causing blocking of active sites on the mild steel surface .The adsorption of the inhibitor and its combination with iodide ions on mild steel surface followed

Langmuir adsorption isotherm via physisorption mechanism, which was proposed based on values of adsorption Gibbs free energy difference  $\Delta G_{ads}$ . Statistically, the multi-variable regression equation describes the behavior of the corrosion inhibition process with high accuracy (correlation coefficient R<sup>2</sup> between 0.974 and 1).

**Key Words**: Acid Corrosion, Synergistic Effect, Mild Steel, Potentiodynamic Polarization, Weight loss Method, Doelhert Design

#### Introduction

Despite mild steel has a relatively high cost; it is widely used in collective amounts in chemical processing, petroleum production and refining. construction and metal processing equipment, and marine applications. These applications usually stimulate corrosion attack on metallic equipment, pipelines and tubes made of iron and its alloys, especially in acidic environments [1].

The study of corrosion inhibition of mild steel in acid solutions have practical importance because that acid solutions are widely used in many industrial processes such as acid pickling of metals, acid cleaning of boilers, scale removal in metallurgy and oil well acidizing. To minimize the metal loss during this process different inhibitors corrosion are used in corrosive solutions. Some organic compounds are found to be effective corrosion inhibitors for many metals

and alloys in acidic media. These act as corrosion inhibitors according to their functional groups (effective groups), which are adsorbed on the metal surface. The adsorption of these organic molecules depends mainly on some physic-chemical properties of the inhibitor such as the functional group, steric factor, aromaticity, electron density at the donor atoms and porbital character of donating electrons [2–4], and the electronic structure of the molecules [5]. The adsorption ability of organic cations is increased by the addition of halide ions to the corrosive media. Interconnecting negatively bridges between the charged metal surface and the cations of inhibitor are formed. Synergism is an effective approach to enhance the inhibition efficiency of the inhibitors to decrease the dosage of organic compounds and to diversify the application of the inhibitors in acidic media [6]. The order of synergism of halide ions is  $\Gamma > Br > Cl [7]$  because it has larger size and easy polarizability, I ions are chemisorbed on the metal surface and provide better synergistic effect [8].

### **Experimental Method**

A mild steel sheet was used as working electrode of 2cm (width), 3cm (length), and 0.1cm (thickness) in weight loss method. Area bout (4.97cm<sup>2</sup>) was used for polarization method. The following composition (Wt %) Si - 0.01, Al - 0.004, Cu -0.055, Mn - 0.346, Mo - 0.002, Cr -0.035, Ni - 0.056, Co - 0.007, P -0.002, S -0.021,C -0.067, and the balance Fe. The aggressive medium (1.0M HCl) was prepared by dilution of analytical grade 37% HCl, and appropriate concentrations of inhibitor were obtained using distilled water.

### Weight Loss Method

The specimens were smoothened by emery papers and then cleaned with tap water, distilled water, benzene and acetone. After that they were dried and weighed on a digital scale. Each of the specimens is designated and its initial weight is noted. After each test, the specimen was washed with running tap water, scrubbed with a brush to remove corrosion products, then washed with tap water followed by distilled water and dried on clean tissue, immersed in benzene, dried, immersed in acetone, dried and left in a desiccators over silica gel until use. The time of immersion in HCl solutions was four hours.

## **Polarization Technique**

The electrochemical cell was composed of graphite counter electrode, prepared mild steel specimen as working electrode and saturated calomel electrode (SCE) as a reference electrode. The corrosion potential  $(E_{corr})$  was measured against SCE. The corrosion cell parts were joined to each other, and then connected to potentiostat (TACUSSEL type PRT 10-05, France), ammeter and voltmeter. Starting with cathodic polarization until it reaches the corrosion potentials, then continuing with anodic polarization. The potential was changed (20-25mV) for each step after one minute period the current was recorded.

### **Results and Discussion**

## 1- Weight Loss Method

Corrosion inhibition (IE%) efficiency calculations from weight loss data were performed according to equation:

IE (%) = 
$$\frac{Wo - W}{Wo} * 100$$
 ... (1)

where Wo and W are the weight loss of mild steel in the absence and presence of inhibitor, respectively.

Table (3) display the applied Doehlert matrix design [9]. The maximum inhibition efficiency for 1propanol was obtained with presence 0.367M of alcohol, and 0.04M KI, at 50°C. It is 94% as shown in table (2). It is clear that the addition of KI enhanced the inhibition efficiency and this indicates a synergistic effect between 1-propanol with KI. The formula used for calculation of the number (N) of experiments required is  $(N=n^2+n+no)$ , where (n) is the number of variables and (no) is the number of center points[10]. quadratic Α polynomial model is established to evaluate the effect of the three factors on the response:

Y = b0 + b	1X1 + b2X2	+ b3X3 +
b4X1X2	+ b5X1X3+	b6X2X3 +
$b7X1^2 + b8X_2^2$	$2^2 + b9X3^2$	(2)

## Where:

Y: inhibition efficiency; X1, X2 and X3: independent variables representing of KI. the concentration the alcohol concentration of and of temperature the solution. respectively; b0: constant; b1, b2 and b3: coefficients reflecting the effect of factors X1, X2, and X3; b4, b5, and b6:coefficients reflecting the interaction between two factors X1X2, X1X3and X2X3 ; b7, b8 and b9: coefficients reflecting the influence of quadratic X1,X2, and X3[11].

Table(1) represents the low and high levels factor, the matrix of the Doelhert design and results are shown in table (2).

Table	1,	Factors	and	levels	used	in
Doelhe	ert (	experime	ntal o	design.		

<b>T</b>		
Variable	Low	High
	level	level
KI Conc.(M) X1	0.03 (-1)	0.07(+1)
Propanol Conc.(M) X2	0.1 (-1)	0.5(+1)
Temp.( <sup>0</sup> C) X3	30 (-1)	50(+1)

The centers and the variation steps for the three studied factors are shown in table (3) according to Doelhert experimental design.

Table (2) Center and variation step of parameters.

Factor		Unit	Center	Variatio n step
X1	Temp.	°C	40	10
X2	Alcohol conc.	Μ	0.3	0.2
X3	KI Con.	М	0.05	0.02

The results were analysed using the analysis of variance as appropriate to experimental design used. The regression equations obtained for 1propanol after analysis of variance gives the levels of corrosion inhibition of mild steel as function of the three variables: Temperature (X1), Alcohol concentration and (X2), KI concentration (X3). All terms were included in the following equations:

Where: Y1 is the response, that is corrosion inhibition of mild steel, and X1, X2, and X3 are the real values of the test variables, Temperature (X1), Alcohol concentration (X2), and KI concentration (X3), respectively. Synergistic Effect of Potassium Iodide on Inhibitive Performance of Propyl Alcohol during Corrosion of Mild Steel in 1.0M HCl

### **2-** Polarization Technique

Tafel extrapolation method can be applied in order to obtain representative values of the corrosion rate in solution. 1.0MHCl The polarization profiles of mild steel in 1.0M HCl solution at 30, 40, and 50°C for various alcohol concentrations in the absence and presence of KI are shown in figures (1) through (3). The presence of increasing amounts of of 1-propanol led to a decrease in both the cathodic and anodic current densities.

The addition of alcohol reduced anodic dissolution and also retarded the hydrogen evolution reaction, indicating that alcohol is a mixed-type inhibitor and controls both the anodic and cathodic reactions. The decrease in  $(i_{corr})$  value was due to the adsorption of the inhibitor molecules [12]. The values of electrochemical parameters such as corrosion potential  $(E_{corr})$ , corrosion current density  $(i_{corr})$ , anodic and cathodic slopes ( $\beta_a$  and  $\beta_c$ ), and corresponding efficiencies were calculated from Tafel plots (table(4)). From the polarization curves it was noted that the curves were shifted toward lower current density region without significant change in corrosion potential. The maximum shift obtained (28mV) was another reason proved that the investigated compound behaved as mixed-type of inhibitor.  $\beta_c$  values increased with the increase in concentration of inhibitor compound and the presence of KI[13-16].

Table 3, Experimental design based on Doehlert matrix in the study of the corrosion inhibition of carbon steel using 1-propanol alcohol and KI synergistic effect, in 1M HCl.

		Coded Variab	ole	Real Variable			Inhibition	
Exp. No.	XI KI conc.	X2 Alcohol conc.	X3 Temp.	XI KI conc.	X2 Alcohol conc.	X3 Temp.	Efficiency (IE%)	
1	+1	0.0	0.0	0.07	0.3	40	85.08	
2	-1	0.0	0.0	0.03	0.3	40	84.03	
3	+0.5	+0.866	0.0	0.06	0.5	40	84.69	
4	-0.5	-0.866	0.0	0.04	0.1	40	89.20	
5	+0.5	-0.866	0.0	0.06	0.1	40	89.70	
6	-0.5	+0.866	0.0	0.04	0.5	40	85.67	
7	+0.5	+0.289	+0.816	0.06	0.367	50	92.49	
8	-0.5	-0.289	-0.816	0.04	0.233	30	72.26	
9	+0.5	-0.289	-0.816	0.06	0.233	30	73.66	
10	0.0	+0.577	-0.816	0.05	0.433	30	80.65	
11	-0.5	+0.289	+0.816	0.04	0.367	50	94.10	
12	0.0	-0.577	+0.816	0.05	0.167	50	93.05	
13	0.0	0.0	0.0	0.05	0.3	40	85.18	
14	0.0	0.0	0.0	0.05	0.3	40	84.15	
15	0.0	0.0	0.0	0.05	0.3	40	89.48	

Inhibitor	Temp.	Ecorr	<i>i</i> <sub>corr</sub>	$\beta_c$	$\beta_a$	IE0/
Conc.(M)	(C)	(mV)	$(mA/cm^2)$	(mV/dec)	(mV/dec)	IE%
1M HCl		-502	0.239	-75.12	64.83	
0.1		-508	0.146	-84.17	58.33	38.91
0.1+0.05M KI		-506	0.0706	-108.6	46.52	70.46
0.3	30	-515	0.099	-94.75	42.33	58.58
0.3+0.05M KI		-497	0.055	-113.9	42.02	76.98
0.5		-500	0.079	-89.46	31.46	66.94
0.5+0.05M KI		-474	0.0444	-183.4	93.61	81.42
1M HCl		-492	0.373	-83.43	48.78	
0.1		-503	0.244	-85.64	49.68	34.58
0.1+0.05M KI		-491	0.051	-85.16	50.53	86.33
0.3	40	-502	0.159	-109.8	54.67	57.37
0.3+0.05M KI		-500	0.0659	-94.93	60.24	82.33
0.5		-498	0.126	-90.6	42.49	66.21
0.5+0.05M KI		-489	0.0676	-75.87	40.19	81.87
1M HCl		-494	0.716	-72.98	48.67	
0.1		-500	0.484	-80.38	34.79	32.40
0.1+0.05M KI		-490	0.0396	-85.2	46.38	94.47
0.3	50	-486	0.367	-83.76	35.43	48.47
0.3+0.05M KI		-503	0.0467	-97.33	55.42	93.47
0.5		-495	0.255	-79.7	29.06	64.39
0.5+0.05M KI		-489	0.0426	-75.87	40.19	94.05

Table 4, Corrosion parameters for mild steel in 1M HCl at different temperatures, different concentrations of 1-Propanol, with and without 0.05M KI



Fig. 1, Tafel polarization curves for corrosion of mild steel in 1M HCl at 30°C in the presence of different concentrations of 1-propanol (P) with and without 0.05M KI.



Fig. 2, Tafel polarization curves for corrosion of mild steel in 1M HCl at  $40^{\circ}$ C in the presence of different concentrations of 1-propanol (P) with and without 0.05M KI.



Fig. 3, Tafel polarization curves for corrosion of mild steel in 1M HCl at 50°C in the presence of different concentrations of 1-propanol (P) with and without 0.05M KI.

#### **3-** Corrosion Kinetic Parameters

The temperature effect on the corrosion parameters of mild steel in 1.0 M HCl was studied at 30, 40, and 50 °C. The inhibition mechanism can be concluded by comparing the activation energy values in the presence and absence of the inhibitor. The plots of Arrhenius or transition state were used to calculate the activation energy  $(E_a)$ , activation enthalpy  $(\Delta H_a),$ and activation entropy  $(\Delta S_a)$  for the corrosion of mild steel in 1.0 M HCl with and without inhibitor. The activation energy can be obtained by Arrhenius equation and Arrhenius plot [17]:

$$i_{corr\,v} = A \, exp(-\frac{Ea}{RT}) \qquad \dots (4)$$

Where:  $i_{corr}$  is the corrosion current density (taken from polarization measurements) in mA/cm<sup>2</sup>, A is the electrochemical constant(Arrhenius constant),  $E_a$  is the activation energy in J/ mol, R is the gas constant (8.314 J/ mol. K) and T is temperature in K. Using the logarithm:

$$lni_{corr} = \left(\frac{-Ea}{R}\right)\left(\frac{1}{T}\right) + lnA \qquad \dots (5)$$

The graph of  $lni_{corr}$  against 1000/T gives a straight line with a slope of  $(-E_{a}/R)$ . Figure (4) shows the Arrhenius plots for mild steel in 1.0 M HCl with different concentration of 1-propanol with and without 0.05M KI.(*Ea*) values were calculated and tabulated in tables (5). The transition state equation was used to calculate the  $\Delta H_a$  and  $\Delta S_a$ :

$$i_{corr} = \frac{RT}{hN} \exp(\frac{\Delta Sa}{R}) \exp(\frac{-\Delta Ha}{RT}) \dots (6)$$

Where: N is Avogadro's number  $(6.02 \times 10^{23} \text{ mol}^{-1})$  and h is Planck's constant  $(6.63 \times 10^{-34} \text{m}^2 \text{ kg s}^{-1})[17]$ . The activation energy is a temperature dependent quantity as follows:

$$E_a = \varDelta H_a - T \varDelta S_a \qquad \dots (7)$$

Equation (5) was rearranged to become:

$$ln(\frac{icorr}{T}) = (\frac{-\Delta Ha}{R})(\frac{1}{T}) + [ln(\frac{R}{Nh}) + \frac{\Delta Sa}{R}]...(8)$$

A plot of ln ( $i_{corr}/T$ ) against 1000/T gave a straight line with a slope of (- $\Delta H_{a}/R$ ) and intercept of [ln  $(R/Nh)+(\Delta S_a/R)$ ] as shown in Figures (4) and (5).  $\Delta H_a$  and  $\Delta S_a$  values were calculated and tabulated in table (5). From Table (5) the activation energy  $(E_a)$  increased in the presence of the inhibitor.  $(E_a)$  increasing with the presence of1-propanol indicated a physical (electrostatic) adsorption occurred in the first stage. Both the inhibitors are organic compounds that are easily protonated to give a cationic form in acid medium. The values of  $(E_a)$  were greater than 20 kJ mol<sup>-1</sup> in the presence of alcohol, which indicates that the entire process is controlled by the surface reaction [18]. The values of  $\Delta H_a$  and  $E_a$  were nearly the same and higher in the presence of alcohol. This indicates that the energy barrier of the corrosion reaction increased in the presence of the without inhibitor changing the mechanism of dissolution. The positive values of  $\Delta H_a$  for both corrosion processes with and without the inhibitor show the endothermic nature of the mild steel dissolution process and indicate that the dissolution of mild steel is difficult [19,20]. The increase in  $\Delta S_a$  values in the presence of inhibitor with and without KI indicated that the process of adsorption was spontaneous [16].



Fig.4, Arrhenius plots for mild steel in 1.0 M HCl with different concentrations of 1-propanol at different temperatures.



Fig. 5, Transition state plots for mild steel in 1.0 M HCl with different

concentration of 1-propanol at different temperatures.

Table 5, Corrosion kinetic parameters for mild steel in 1.0 M HCl and different concentration of 1-propanol in the presence and absence of 0.05M KI

Inhibitor	$E_a$	$\Delta H_a$	$\Delta S_a$
Conc.(M)	(kJ/mol)	(kJ/mol)	(kJ/mol.K)
1M HCl	44.54	41.93	-118.87
0.1	48.66	46.07	-109.33
0.3	53.13	50.52	-98.11
0.5	47.55	44.95	-118.15
0.1+0.05M KI	23.55	-26.14	-353.43
0.3+0.05M KI	6.418	-9.02	-215.08
0.5+0.05M KI	19.29	-21.9	-338.96

### 4- Adsorption Isotherm

Adsorption isotherm calculations of 1-propanol on the mild steel surface were performed to investigate the mechanism of corrosion inhibition in the presence and absence of 0.05M KI.

Totally, isotherms adsorption provides data about the interaction among the adsorbed molecules themselves and their interactions with the electrode surface [21]. The fraction coverage  $\theta$  is calculated using the following equation:

$$\theta = 1 - \frac{I_{add}}{I_{free}} \qquad \dots (9)$$

Where:  $I_{free}$  and  $I_{add}$  are the corrosion current densities in the absence and presence of the additive compounds.

In order to study the different types of adsorption isotherm models, the Temkin, Frumkin and Langmuir models have been plotted. Synergistic Effect of Potassium Iodide on Inhibitive Performance of Propyl Alcohol during Corrosion of Mild Steel in 1.0M HCl

According to the R<sup>2</sup>values obtained linear from the graph. it was confirmed that the adsorption of the two compounds on a mild steel best fitted the Langmuir surface adsorption isotherm (which assumes that the metal surface contains a fixed number of adsorption sites and each one holds one adsorbed species) model which is represented by equation (9):

$$\frac{\text{Cinh}}{\theta} = \frac{1}{\text{Kads}} + \text{Cinh} \qquad \dots (9)$$

where: Cinh is the concentration of inhibitor and Kads is the adsorption constant which is related to the standard free energy adsorption,  $\Delta G^{\circ}_{ads}$ , as follows [22]:

$$\Delta G^{\circ}_{ads} = -RT \ln(55.5K_{ads}) \qquad \dots (10)$$

The value of 55.5 is the molar concentration of water in the solution.

The plot of Cinh/ $\theta$  against Cinh in figures (6) through (8) yields a straight line and this supported the assumption that the adsorption of the each applied inhibitor in the presence and absence of KI on a mild steel surface in 1.0 MHCl solution obeys the Langmuir adsorption isotherm.

The increasing value of  $\Delta G^{\circ}_{ads}$  and Kads in the presence of KI reflects the increasing adsorption capability and the negative value of  $\Delta G^{\circ}_{ads}$  indicates spontaneous adsorption of inhibitor molecule on the metal surface[21].

It is well known that large values of  $K_{ads}$  mean better inhibition efficiency of the inhibitor, i.e., strong electrostatic interaction between the negatively charged metal surface and the positively charged entities of the adsorbing inhibitor molecules.  $\Delta G^{\circ}_{ads}$  and Kads values are tabulated in table (6):



Fig. 6, Adsorption isotherms for mild steel in1.0 M HCl in different concentrations of 1-propanol alcohol at 30°C in the presence and absence of 0.05M KI



Fig. 7, Adsorption isotherms for mild steel in 1.0 M HCl in different concentrations of 1-propanol alcohol at 40°C in the presence and absence of 0.05M KI



Fig. 8, Adsorption isotherms for mild steel in 1.0 M HCl in different concentrations of 1-propanol alcohol at 50°C in the presence and absence of 0.05M KI

Table 6, Corrosion parameters for mild						
steel	in	1.0	Μ	HCl	and	1-propanol
alcoh	ol i	n the	e pr	esence	e and	absence of
0.05N	ИK	I.				

Inhibitor	Temp.°C	Kads	∆G°ads (KJ/mol)
	30	7.194	-15.09
1-propanol	40	5.618	-14.94
	50	4.61	-14.89
1-propanol +KI	30	37.04	-19.22
	40	166.67	-23.77
	50	14285.7	-36.48

### Conclusions

- 1- A 1-propanol acts as moderate inhibitor for mild steel in 1.0 MHCl.
- 2- Potentiodynamic polarization and weight loss studies showed that the 1-propanol is mixed-type inhibitors.
- 3- The inhibition efficiency increases with increase in concentration of both inhibitor and with increasing of temperature.
- 4- The addition of iodide ions to 1propanol (inhibitor) enhanced the inhibition efficiency due to synergistic effect.
- 5- The adsorption of inhibitor (1propanol) alone and in combination with iodide ions obeys Langmiur adsorption isotherm.
- 6- The thermodynamic parameters calculated from the adsorption isotherm showed that physisorption is involved in the inhibition process, which was proposed based on values of  $\Delta G^{\circ}$  ads.
- 7- Statistically, the multi-variable regression equations describes the behavior of the corrosion inhibition process with high accuracy (correlation coefficient R<sup>2</sup>between 0.984 and 1).

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