

Experimental Study on Carbon Steel Corrosion and its Inhibition Using Sodium Benzoate Under Different Operating Conditions

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Abstract

Corrosion experiments were carried out to investigate the effect of several operating parameters on the corrosion rate and corrosion potential of carbon steel in turbulent flow conditions in the absence and presence of sodium benzoate inhibitor using electrochemical polarization technique. These parameters were rotational velocity (0 - 1.57 m/s), temperature (30°C – 50°C), and time. The effect of these parameters on the corrosion rate and inhibition efficiency were investigated and discussed. It was found that the corrosion rate represented by limiting current increases considerably with increasing velocity and temperature and that it decreased with time due to the formation of corrosion product layer. The corrosion potential shifted to more positive with increasing temperature and velocity while it shifted to more negative with time. Sodium benzoate gave good inhibition efficiency for the whole investigated range of temperature and velocity. Its efficiency was high on clean surfaces and decreased with time in stationary and flow conditions. No noticeable effect of temperature on the inhibition efficiency was noticed.

Keywords: corrosion rate, polarization curve, sodium benzoate, carbon steel, Inhibition efficiency.

الخلاصة

تم اجراء عدة تجارب على عملية التآكل والتي من خلالها تم استنتاج تأثير بعض العوامل العملية المؤثرة على معدل التآكل وفولتية التآكل للكربون الفولاذي في المجرى المضطرب بغياب او بوجود بنزوات الصوديوم كمانع تآكل وباستخدام تقنيات الكهروكيميائية القطبية. وهذه العوامل هي السرعة الدورانية، درجة الحرارة، والزمن. وتم مناقشة تأثير هذه العوامل على معدل التآكل وكفاءة المانع. لقد وجد بأن معدل التآكل المتمثل بنيار التآكل المحدد يتناقص مع الزمن نتيجة لتكوين طبقة من نواتج التآكل ويزداد مع السرعة ودرجة الحرارة. إن فولتية التآكل تتجه نحو زيادة أكثر مع زيادة درجة الحرارة والسرعة بينما تتناقص بزيادة الزمن. بنزوات الصوديوم تعطي كفاءة جيدة وكفاءتها تكون عالية للسطوح النظيفة وتتناقص مع الزمن في الظروف الساكنة وظروف الجريان مع ملاحظة عدم وجود تأثير لدرجة الحرارة على كفاءة المانع في هذه الظروف.

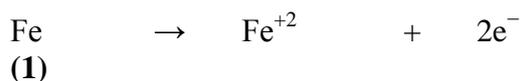
Introduction

Corrosion is defined as the destruction or deterioration of a material because of reaction with its environment [1]. Corrosion in aqueous environment and atmospheric environment is an electrochemical process because corrosion involves the transfer of electrons between a metal

surface and an aqueous electrolyte solution. It results from the overwhelming tendency of metals to react electrochemically with oxygen, water and other substances in the aqueous environment [2].

Carbon steel, the most widely used engineering material, accounts for approximately 85% of the annual steel

production worldwide. Despite its relatively limited corrosion resistance, carbon steel is used in large amounts in marine applications, chemical processing, petroleum production and refining, construction and metal-processing equipment [3]. When carbon steel is immersed in a neutral aqueous environment, the anodic reaction is [4]:-



The cathodic reaction is:



An *inhibitor* is a chemical substance, when added in small concentration to an environment, effectively decreases the corrosion rate. There are several classes of inhibitors:

passivators, organic inhibitors, including slushing compounds and pickling inhibitors, and vapor-phase inhibitors [5]. The concentration of a given inhibitor needed to protect a metal will depend on a number of factors:

such as the composition of the environment, temperature, the velocity of the liquid, the presence or absence of the metal, the internal or external stresses on the metal, composition of the metal, and the presence of any other metal content [6]. Several inhibitors have been used to prevent corrosion of carbon steel in aqueous solution such as chromate molybdate, pertechnetate, nitrate, phosphate, silicates, cations, organic inhibitors, carboxylates and tannins [7].

Increasing temperature affects the corrosion rates by a combination of factors- such as; the common effect of temperature on the reaction kinetics themselves and the higher diffusion

rate of many corrosive by products at increased temperatures. Many corrosive gases have lower solubility in open systems at higher temperatures. As temperatures increase, the resulting decrease in the solubility of the gas causes corrosion rates to go down [5]. In the case of corrosion in neutral solution, the increase of temperature has a favourable effect on the overpotential of oxygen depolarization and the rate of oxygen diffusion, but it leads to a decrease of oxygen solubility [8].

Velocity has a significant effect on corrosion rates. Stagnant or low velocity fluids usually give low general corrosion rates. Corrosion rates generally increase with increasing velocity due to the depolarizing effect on the cathode [9]. Velocity primarily affects corrosion rate through its influence on diffusion phenomena. It has little effect on activation controlled processes. The manner in which velocity affects the limiting diffusion current is a marked function of the physical geometry of the system. In addition, the diffusion process is affected differently by velocity when the flow conditions are laminar as compared to the situation where turbulence exists [4].

When iron or steel is exposed to high temperature water, the rate of corrosion of the metal decreases with exposure time during the early period of exposure. After several hours, the corrosion rate becomes relatively constant at a lower value. During the early period of exposure, while the corrosion rate is decreasing, the oxide film on the surface of the metal grows in thickness. However, the rate at which the film grows decreases with time. The thickness of the oxide film soon reaches a relatively constant value, and thereafter film thickness does not change appreciably with

further exposure. As might be expected, a relatively constant corrosion rate and oxide film thickness are attained at about the same time. This process, referred to as pretreatment or pickling involves careful control of reactor coolant water chemistry and temperature during the pretreatment period.

Several previous studies [10,11,12] showed that the corrosion rate generally decreases with time due to corrosion product formation depending on flow velocity. Slaiman and Husan [12] noticed that at high velocity, the formation of corrosion product leads to increasing the limiting current density (corrosion rate) for carbon steel in water by increasing the turbulence resulted from the increased roughness of the surface.

In many corrosion problems, the rate of uniform corrosion is controlled by the rate of mass transfer. This is true whether the corrosion fluid remains static or in fast motion with respect to the metal surface. However, molecular diffusion is not the only factor which influences the rate of corrosion. In addition, in turbulent fluids, the rate of transport of eddy diffusion appears to participate in the control of the over-all transfer rate. It is only in situations where both anodic and cathodic processes are activation controlled that they will be unaffected by the relative movement between surface and environment [4,13]. The rate of this half reaction is generally limited by the speed at which oxygen can reach the

surface of the metal. This oxygen is transported from the bulk water to the surface across the boundary layer by diffusion [14].

Sodium benzoate has found considerable application as a corrosion inhibitor in low concentrations. Corrosion inhibition has been reported for carbon steel, zinc, copper, copper alloys, soldered joints, aluminum, and aluminum alloys. As a liquid phase inhibitor, low concentrations of sodium benzoate have reportedly been used for corrosion control at a pH as low as 5.5. Movement of the solution, or saturation with oxygen assists inhibition, but a pH below 6 causes breakdown. The most effective inhibition appears to be in the pH range of 6 to 12 [15].

The aim of present work is to study the effect of some operating parameters on the corrosion of carbon steel and its inhibition in 0.1N NaCl solutions by sodium benzoate as inhibitor using electrochemical polarization technique to assess the effect of velocity, temperature, and time on the corrosion rate in the presence and absence of inhibitor.

Experimental Work

Figure 1 shows the experimental apparatus that was used for performing the experimental work.

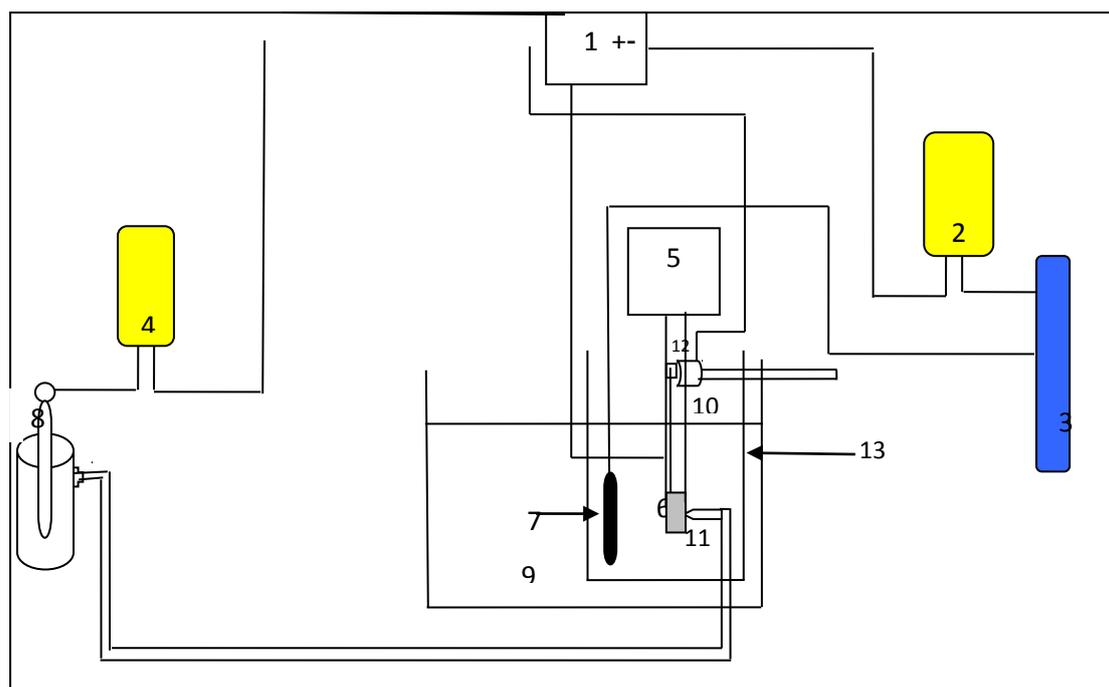


Fig. 1 Experimental apparatus 1- power supply, 2- ammeter, 3- resistance box, 4- voltmeter, 5- stirrer, 6- working electrode (specimen), 7- graphite electrode (anode), 8- calomel electrode (reference electrode), 9- water bath, 10- brush, 11- Luggin capillary, 12- metal for electrical connection with specimen, 13- Beaker containing the solution.

The experimental apparatus was composed of a mechanical agitator to obtain different rotational velocities, a water bath to obtain different solution temperatures, a carbon steel specimen of 30 mm long and 25 mm outside diameter (cathode) attached to the rod of the agitator, power to apply the required protection potential, a digital ammeter to measure the current, a digital voltmeter to measure potential, variable resistance (rheostat) to control the current flow, a graphite electrode as auxiliary electrode (anode), a saturated calomel electrode (SCE) as a reference electrode, and a digital balance of high accuracy. The electrical connection between cathode (specimen) and the cell was attained using brush. Before each experiment the specimen was washed by tap water followed by distilled water, dried with clean tissue, abraded using different grades of emery papers, 500,600,1000 and 1200

immersed in annular acetone for 5 minutes, rinsed with water and dried with clean tissue. Then the specimen was stored in vacuum desiccator over high activity silica gel for 24 h before use. Five liters of 0.1N NaCl solution was used in water bath at different temperatures (30°C, 40°C and 50°C) at stationary conditions. For flow conditions, different rotation speeds (250, 500,750, 1200 rpm) were investigated at constant temperature of 40°C. The specimen was connected to -ve terminal of power supply to serve as a cathode and the graphite to +ve terminal to serve as an anode. When the solution reached the required temperature, the specimen was immersed in the solution and the electrical circuit was switched on immediately. The power supply was set at 5V (applied voltage). The specimen (working electrode) was cathodically polarized from potentials (-1.3 to -1.5 V) to the corrosion

potential (where $i_{app.} = 0$) by changing the applied current using rheostat. The current was recorded for each 5 mV change in potential. Two minutes were allowed for steady state to be reached after each potential increment and the capillary tube was placed at distance 1-2 mm from cathode (specimen) and connected to calomel electrode to measure the specimen potential [16]. Thus polarization curve can be drawn and the limiting current can be obtained. The distance between anode and cathode was 100 mm.

The experiments were repeated by adding 1500 ppm sodium benzoate to the solution as corrosion inhibitor; then the polarization curve was drawn to obtain the limiting current at different temperatures (30, 40 and 50°) at stationary conditions, and at different speeds (250, 750,1200 rpm) at constant temperature of 40°C .

The effect of time on the corrosion rate (I_L) and inhibition efficiency was determined by measuring I_L at various time intervals, ($t= 0$ h),($t= 1.5$ h) and ($t = 3$ h) at $T = 40^\circ\text{C}$ for each value of rotation speed. After determining the polarization curve at $t = 0$ the specimen was allowed to corrode freely by switching off the electrical circuit for 1.5 h and then the polarization curve was obtained.

After obtaining the polarization curve at $t = 1.5$ h, the electrical circuit was switched off again to allow the specimen to corrode freely for another 1.5 h (the total time is 3 h). Then the circuit was switched on and the polarization curve is obtained for $t = 3$ h.

Since the limiting current plateau is not well defined, the limiting current was determined by using Gabe [17] method as in Fig.2.

$$I_L = \frac{I_1 + I_2}{2} \tag{3}$$

Since the solution is neutral ($\text{pH} = 7$), $I_L = I_{corr}$. [4,5].

The inhibition efficiency was calculated as:

$$IE = \frac{(I_o - I)}{I_o} \% \tag{4}$$

Where I_o is the limiting current in absence of inhibitor and I is the limiting current in the presence of the inhibitor.

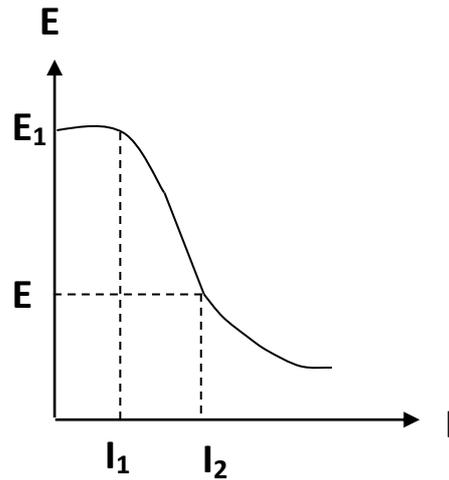


Fig. 2 Determining limiting current

Temperature (°C)	i_L (mA/m ²)	
	Without inhibitor	With 1500 ppm Sodium Benzoate
30	1697.65 3	891.2 68
40	2546.47 9	2122. 066
50	3395.30 5	2546. 479

Table 1, Values of i_L at various temperatures and stationary condition.

Results and Discussions

Table (1) to (3) show the numerical values of experimental results.

Velocity (m/s)	i_L (mA/m ²)					
	t = 0 h		t = 1.5 h		t = 3 h	
	Without inhibitor	With 1500 ppm Sodium Benzoate	Without inhibitor	With 1500 ppm Sodium Benzoate	Without inhibitor	With 1500 ppm Sodium Benzoate
0	8492.569	4246.285	5520.170	3397.028	4670.913	2972.399
0.3271	8067.941	5095.541	6369.427	4246.285	4246.285	3397.028
0.9812	12314.225	6369.427	8917.197	5944.798	8067.941	5520.170
1.57	12738.854	6794.055	9341.826	6369.428	8492.569	5944.798

Table 2, Values of i_L at various velocities and times at T = 40°C .

Without Inhibitor

Effect of Temperature

Figure 3 shows the polarization curve time at stationary conditions and different temperatures (30°C, 40°C and 50°C). From this figure, it can be seen that as the temperature increases the limiting current increases. This can be ascribed to two reasons: first when the temperatures increases, the O₂ diffusivity increases enhancing the corrosion [4,5] and second it decreases the O₂ solubility the factor that leads to decrease the corrosion rate [4]. Also Fig.(3) shows that the corrosion potential shifts to more positive with increasing temperature. The rise in temperature may decrease the equilibrium potential of Fe and H₂ but increase the equilibrium potential of O₂ [18], the increase the diffusion rate of oxygen species by increasing the molecular diffusion coefficient and decreases the oxygen solubility.

Effect of Rotational Velocity

The polarization curves at different rotational speed (0, 0.3271, 0.9812, and 1.57 m/s) at 40°C are shown in Fig.(4). In this figure it is clear that the limiting current increases as the rotational velocity increases due to increase in O₂ diffusion from solution bulk to the surface and decreasing the thickness of diffusion layer on the

metal surface[11,19,20,12]. Evans 1960 [21] indicated that greater turbulence due to high velocities results in more uniform O₂ concentration and more likely initiation of corrosion on a previously uncorroded surface.

Polarization curve for velocity of 0.6542 and 1.3083 m/s are not presented in Fig.(4) to avoid confusion. Also Fig.(4) shows that the E_{corr.} shifts to more positive with increasing velocity due to the increased O₂ transport to the surface. This agrees with previous findings [20,11,22,23].

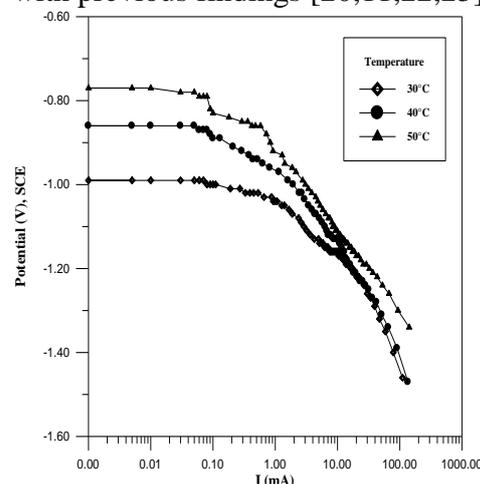


Fig. 3 Polarization curve at stationary conditions at different temperatures

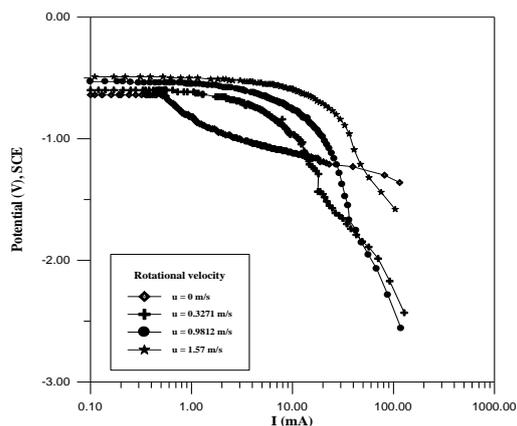


Fig. 4 Polarization curve at T=40°C at different rotational velocity

Effect of time

Over the years, some studies investigated the effect of carbon steel corrosion products on the corrosion rate [10,11,12]. Most of these studies used the weight loss method and not the electrochemical technique (LCD). In fact, the loss in weight gives the average weight loss over the exposure period and not the instantaneous. In the present work the LCD technique was used to determine the instantaneous corrosion rate (I_L) at various times of exposure. Figures 5 through 7 show the polarization curves for different rotational velocities. The figures show that for all velocities, i_L decreases with time. The corrosion potential decreases (becomes more negative) with increasing time. This decrease is ascribed to the formation of corrosion product layer with time which leads to the decrease of the arrival of O_2 to the metal surface [10,11,12]. Also the surface activity decreases with time [10]. It is evident from Figs 5 to 7 that the corrosion potential shifts to more negative with time. This decrease in E_{corr} is due to the decrease in O_2 concentration near the surface due to the formation of corrosion product layer [10,20]. It was found that E_{corr} decreases with time and with the decrease in O_2 concentration.

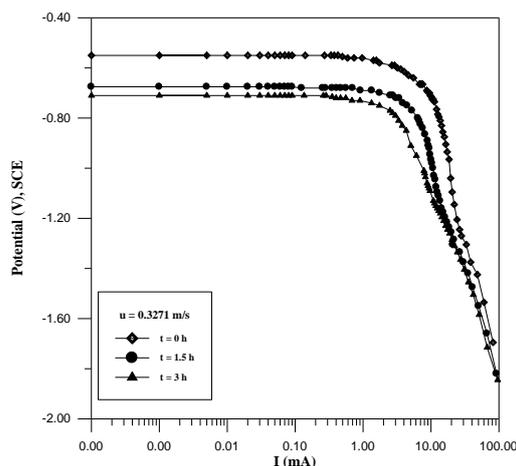


Fig. 5 Polarization curve at T= 40°C and u= 0.3271 m/s for different times

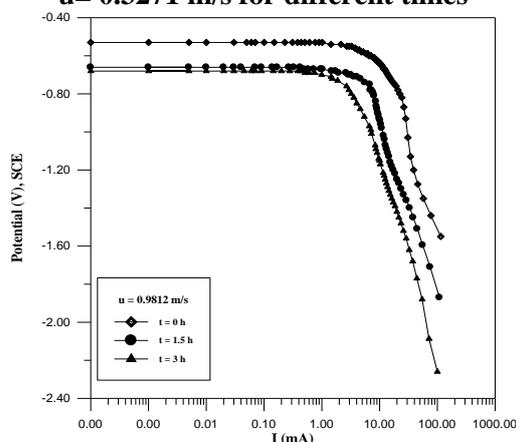


Fig. 6 Polarization curve at T=40°C and u=0.9812 m/s for different times

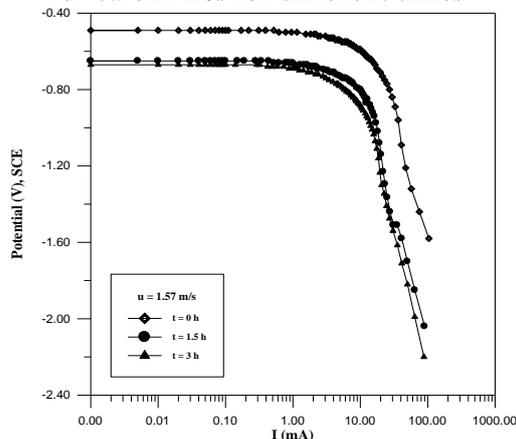


Fig. 7 Polarization at T=40°C and u= 1.57 m/s for different times

Addition of Inhibitor (1500 ppm sodium benzoate)

The effect of sodium benzoate inhibitor on corrosion behavior of carbon steel in chloride neutral media is presented in figures 8 through 25. It can be seen from all figures that the

limiting current decreases with the addition of the inhibitor.

Effect of Temperature

Figures 8 through 10 show the polarization curve at stationary condition and different temperatures (30°C, 40°C and 50°C) in absence and presence of sodium benzoate.

It is clear that the sodium benzoate leads to decreasing the corrosion rate considerably. The figures show that the temperatures have no noticeable effect on the inhibitor efficiency. It is noticed that the inhibitor (sodium benzoate) shifts the corrosion potential to be more active indicating that it is a cathodic type inhibitor. Cathodic inhibitors reduce corrosion by retarding individual stages of the cathode reaction: ionization of oxygen, diffusion of oxygen to the cathode, and discharge of hydrogen ions which naturally cannot lead to local corrosion. In the presence of sodium benzoate as cathodic inhibitors. A considerable part of the electrode remains inert with regard to the cathodic process, and this reduces the corrosion current sharply. [4,24].

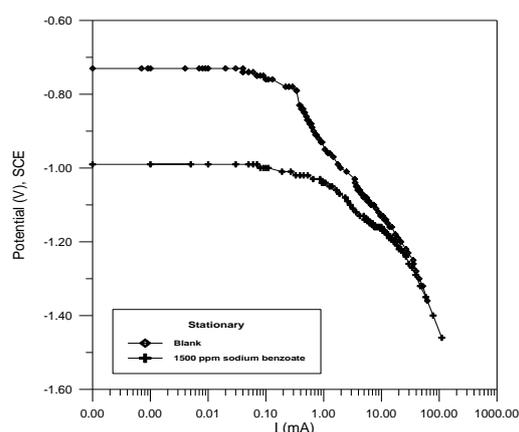


Fig. 8 Polarization curve at 30°C in absence and presence of sodium benzoate inhibitor

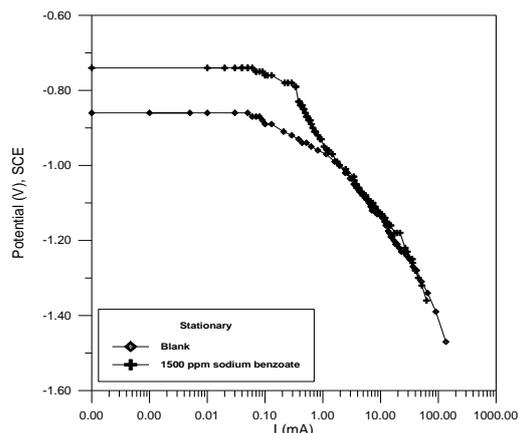


Fig. 9 Polarization curve at 40°C in absence and presence of sodium benzoate inhibitor

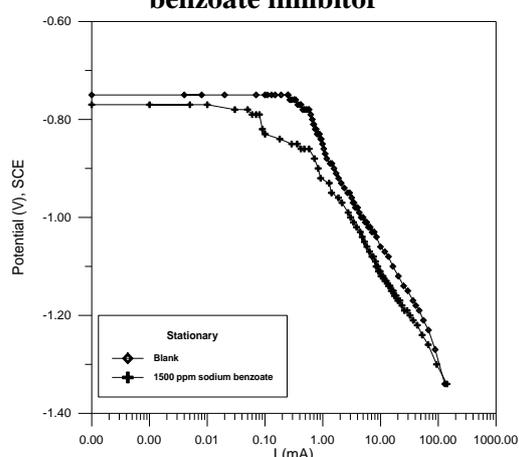


Fig. 10 Polarization curve at 50°C in absence and presence of sodium benzoate inhibitor

Effect of Rotational Velocity

Figure 11 shows the effect of rotational velocity on the i_L in presence and absence of sodium benzoate. When the electrode rotation velocity increased, the cathodic current increased. The increase of cathodic current with the electrode rotation velocity is explained by an increase of the oxygen supply to the metal surface [25]. Fig.(11) reveals that the presence of 1500 sodium benzoate lead to considerable decrease in i_L (or the corrosion) rate. Table (3) lists values of IE with velocity but it is generally decreases with increasing velocity. This may be attributed to the removal of inhibitor layer due to high shear

forces near the surface as evidenced by numerous studies [26,27,28].

Effect of Time on the Inhibitor Effect

The limiting current was measured at various times in the presence of the corrosion inhibitor (sodium benzoate), to determine the effect of time on the inhibition efficiency. Figures 12 to 23 show the polarization curves at various times for various velocities in the presence and absence of corrosion inhibitor at 40°C. Figures 12 to 15 show the polarization curves at stationary conditions. It is clear that the presence of the inhibitor leads to decreasing I_L (or corrosion rate) at different times (0,1.5, 3h). The same behavior is clear in figures 15 to 22 for flow conditions. This indicates that sodium benzoate function even in the presence of corrosion product.

It is clear that the presence of inhibitor leads to decreasing the corrosion rate with inhibition percent (%IE)for all time . From table (3), the IE decreases with increasing time. This is due to the fact that the inhibiting effect of soduim benzoate is due to the occlusion of the metal surface and not adsorption because the inhibitor is a viscofier and thickner. The inhibitor could thus form a layer on the surface, restricting ingress of the corrodent. The efficiency decreases with increasing the time because the inhibitor layer starts to remove with increasing turbulence due to the formation of corrosion products [29].

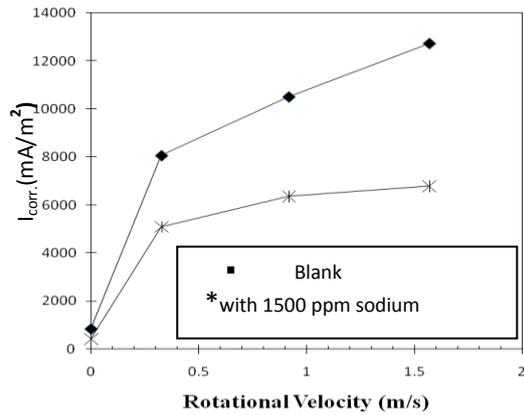


Fig. 11 i_L vs. rotational velocity in absence and presence of sodium benzoate inhibitor

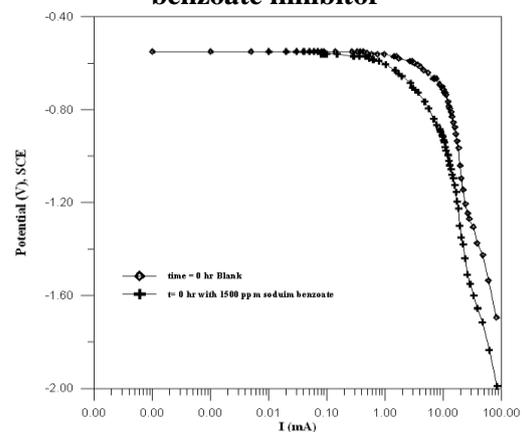


Fig. 12 Polarization at time =0 h in absence and presence of sodium benzoate inhibitor at 40°C

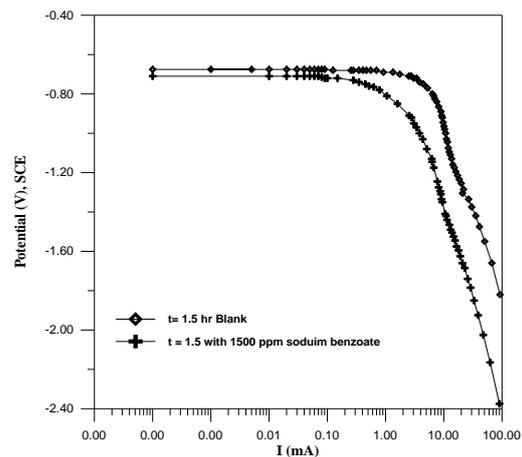


Fig. 13 Polarization at time =1.5 h in absence and presence of sodium benzoate inhibitor at 40°C

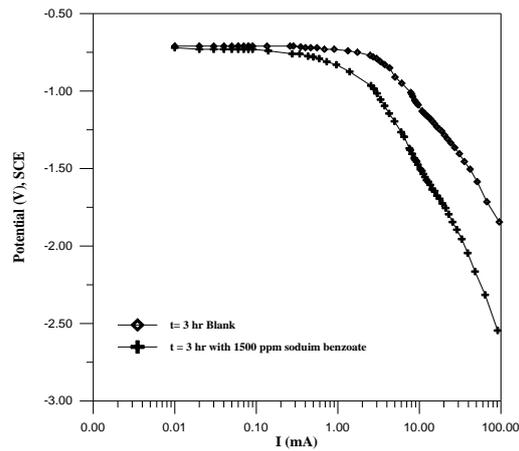


Fig. 14 Polarization at time =3 h in absence and presence of sodium benzoate inhibitor at 40°C

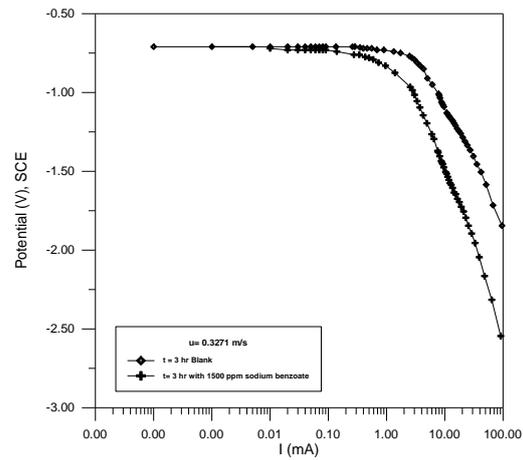


Fig. 17 Polarization at time =3 h and $u=0.3271$ m/s in absence and presence of sodium benzoate inhibitor at 40°C

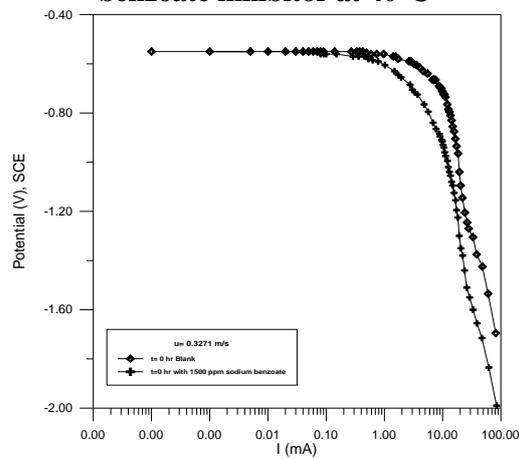


Fig. 15 Polarization at time =0 h and $u=0.3271$ m/s in absence and presence of sodium benzoate inhibitor at 40°C

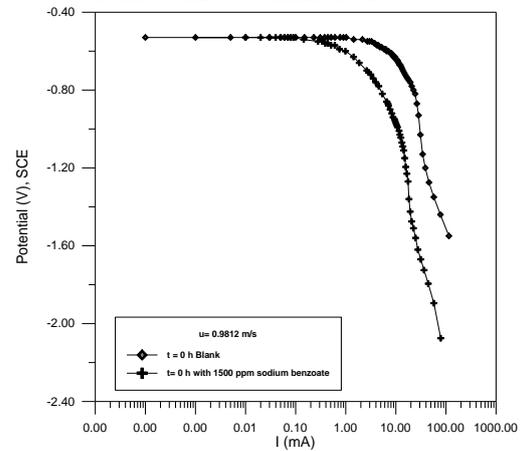


Fig. 18 Polarization at time =0 h and $u=0.9812$ m/s in absence and presence of sodium benzoate inhibitor at 40°C

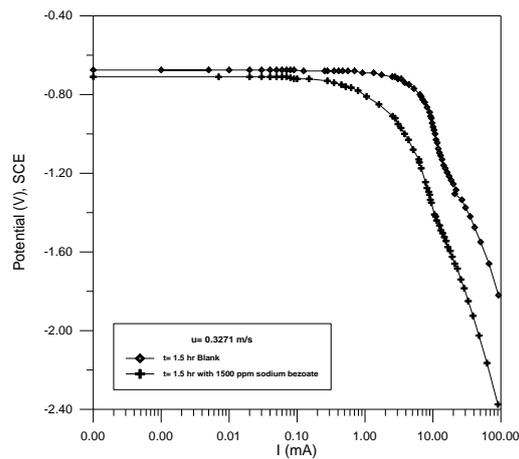


Fig. 16 Polarization at time =1.5 h and $u=0.3271$ m/s in absence and presence of sodium benzoate inhibitor at 40°C

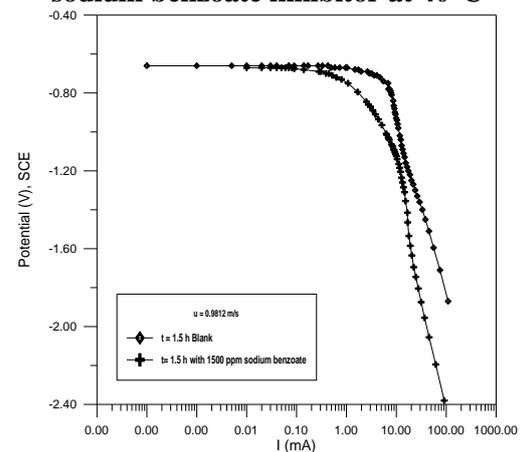


Fig.19 Polarization at time =1.5 h and $u=0.9812$ m/s in absence and presence of sodium benzoate inhibitor at 40°C

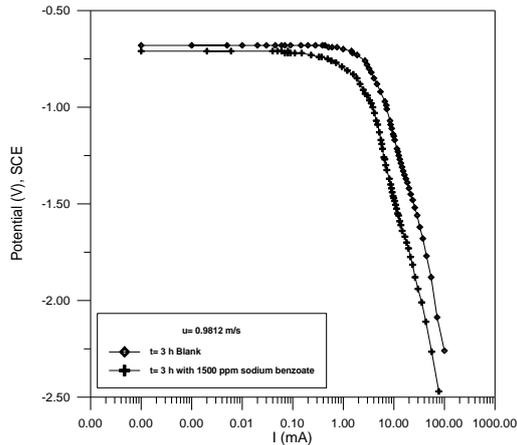


Fig. 20 Polarization at time =3 h and $u=0.9812$ m/s in absence and presence of sodium benzoate inhibitor at 40°C

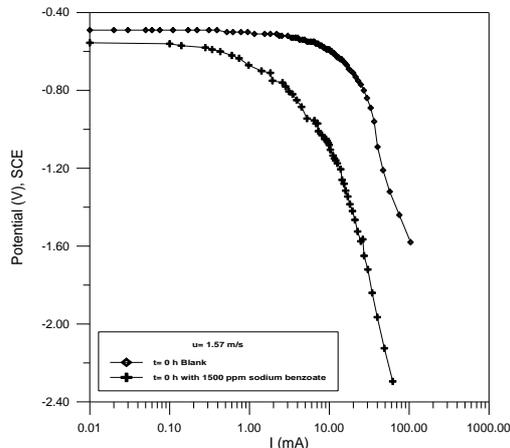


Fig. 21 Polarization at time =0 h and $u=1.57$ m/s in absence and presence of sodium benzoate inhibitor at 40°C

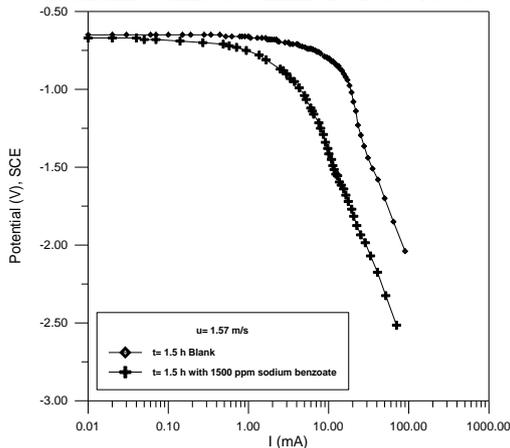


Fig. 22 Polarization at time =1.5 h and $u=1.57$ m/s in absence and presence of sodium benzoate inhibitor at 40°C

Figure 24 shows the effect of temperature on i_L (or corrosion rate) in the absence and in the presence of inhibitor. The current density in the absence of sodium Benzoate is higher

than that of the presence of it for all temperatures. It is evident from Fig. (24) that the temperature has no effect on the inhibition efficiency where the inhibitor still functions even at high temperature. In Fig. (24) the $i_{corr.}$ for the presence of inhibitor is lower than that in the blank. The corrosion current density in cooling water decreased considerably in the presence of inhibitor for oxygen reduction as a result of cathodic inhibitor. Table (3) shows the numerical values of percentage inhibition efficiency of experimental results.

Table (3) Percentage of Inhibition Efficiency of 1500 ppm sodium benzoate

Velocit y (m/s)	% Inhibition efficiency		
	t = 0 h	t = 0 h	t = 0 h
0	50	50	50
0.3271	36.84	36.84	36.84
0.9812	48.28	48.28	48.28
1.57	46.67	46.67	46.67

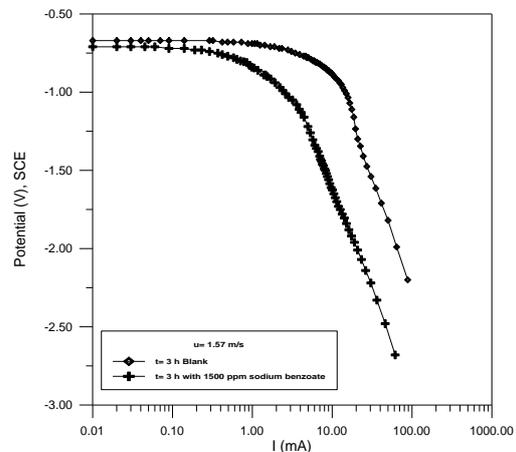


Fig. 23 Polarization at time =3 h and $u=1.57$ m/s in absence and presence of sodium benzoate inhibitor at 40°C

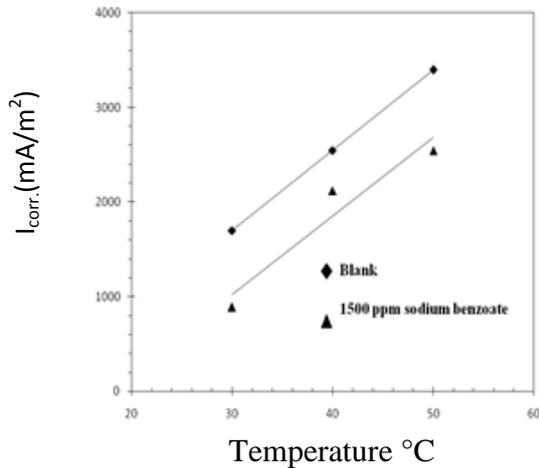


Fig. 24 i_L vs. temperature in absence and presence of sodium benzoate

Mass Transfer

Mass transfer coefficient can be estimated from data of polarization curves according to equation

$$k = \frac{i_L}{ZF\Delta C} \quad (5)$$

Where k is the mass transfer coefficient, and ΔC is the oxygen concentration difference between the bulk and surface in solution $\Delta C = C_b - C_s$, $z=4$ for oxygen reduction, and i_L is the limiting current density for oxygen reduction on Fe at $i=i_L$, $C_s=0$, C_b from table(4) = 0.1372 mole /m³ at 40°C.

Table (4) Values of oxygen Diffusivity and solubility at different temperatures [30,31]

T (°C)	Solubility (mg/l) 0.1 NaCl	D_o^* 10 ⁹ (m ² /s) 0.1 NaCl	C_b (mole/m ³)
30	7.5	2.374	0.2344
40	4.389	3.010	0.1472
50	5.399	3.562	0.1687

Figure 25 shows the variation of mass transfer coefficient with Re at the absence and presence of inhibitor. From this Fig., it can be seen that as Re increase, k increase because the convective mass transport of O₂ will increase. Increasing turbulence leads to

decreasing the thickness of viscous sub-layer and the diffusion layer that represents the main resistance to momentum and mass transport, respectively [32,33]. Hence, the O₂ concentration at the surface will increase leading to increasing k .

The effect of Reynolds number on thickness of different sub-layer is shown in Fig. (26) in the presence and absence of inhibitor. It can be seen that as the Reynolds number increases, the thickness will decrease. This effect of Re is due to the increased turbulence and the penetration of eddies through this layer causing a decrease in its thickness [34,35].

$$\delta = \frac{D}{k} \quad (6)$$

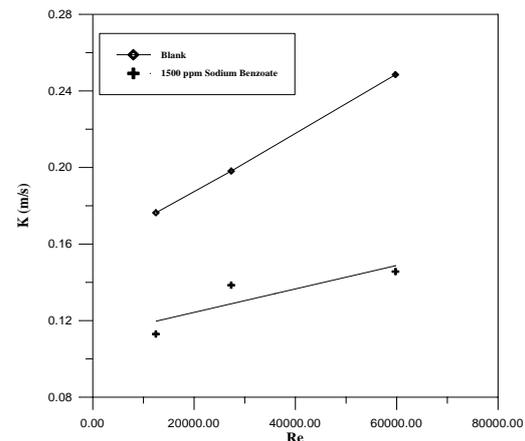


Fig. 25 Effect of Re on mass transfer coefficient

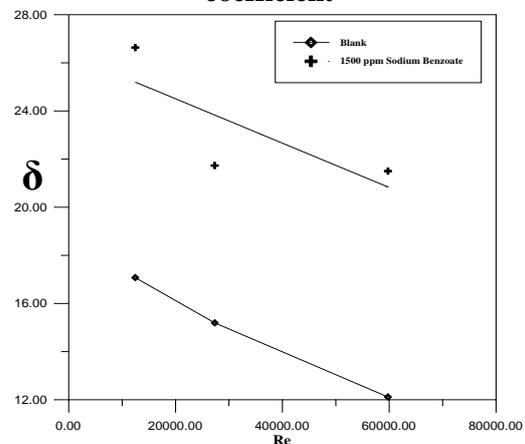


Fig. 26 Effect of Re on thickness

CONCLUSIONS

For the present work, the followings can be concluded :-

- 1- Increasing temperature leads to increase the limiting current from 1697.653 mA/m² at 30°C to 3395.305 mA/m² at 50°C in the absence in inhibitor.
- 2- Increasing temperature shifts the corrosion potential to more positive. In 30°C, the potential is -0.99 V while in 40°C and 50°C the calculated are -0.86 V and -0.77 V respectively.
- 3- Increasing rotational velocity leads to increase the corrosion rate considerably which represent by limiting current from 8067.971 mA/m² at 0.3271 m/s to 12738.854 mA/m² at 1.57 m/s in the absence of inhibitor. Also it shifts the corrosion potential to more positive from -0.6 V at 0.3271 m/s to -0.5 V at 1.57 m/s.
- 4- The corrosion rate (or i_L) decreases considerably with time due to the corrosion products formation. It decreases from 8492.569 mA/m² at time zero to 4670.913 mA/m² after 3h at 40°C and stationary conditions. Also the corrosion potential shifts to more negative, it become -0.76 V after 3 h while in zero time have been -0.57 V at rotational velocity 0.3271 m/s.
- 5- The inhibition efficiency of Sodium benzoate decreased considerably with time and slightly with rotational velocity. It decreased from 50% at time zero to 36.36% after 3 h at stationary conditions. While with rotational velocity it is 36.84%, 48.67% at velocities 0.3271 m/s, 0.9812 m/s and 1.57 m/s, respectively.

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