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ABSTRACT

The effect of cathodic protection currents on carbon steel corrosion in 0.1N NaCl solution was investigated under flow conditions of rotation velocity range 0-0.262 m/s for a range of temperatures (35-55°C) using rotating cylinder electrode. The corrosion rate was determined using both weight loss method and electrochemical polarization technique (limiting current density). Various values of protection currents were applied to protect carbon steel from corrosion, these were $I_{app.} = I_{corr.}$, $I_{app.} = 2I_{corr.}$, and $I_{app.} = 2.4I_{corr.}$ under stationary and flow conditions. The variation of protection potential with time and rotation velocity at various applied currents was assessed. It was found that the corrosion rate of carbon steel increases with flow velocity and has unstable trend with temperature. The protection currents required vary with temperature and it increases considerably when the rotation velocity was increased. The protection potential decreases appreciably (shifts to more negative) with time and with increasing rotation velocity. Also it shifts to more positive with increasing temperature.

Keywords: cathodic protection, steel, impressed current, flow, salt.

NOMENCLATURE

A: Area (m^2)
D: Diffusivity (m^2/s)
Ec: corrosion Potential (V)
F: Faradays constant (96487 coulomb/equivalent)
i: Current density (A/m^2)
 $I_{app.}$: Applied current (mA)
 $I_{corr.}$: Corrosion current (mA)
 I_L : Limiting current (mA)
K: Mass transfer coefficient (m/s)
M: Molecular weight (g/mole)
N: Molar flux (mole/m.s)
t: time of exposure (hours)
V: Volts
 Δw : weight loss (g)

Subscript

Corr: Corrosion
L: Limiting

Abbreviations

C.R: Corrosion rate ($g/m^2.d$)
PP: Protection percent
SCE: Standard calomel electrode

Greek Letters

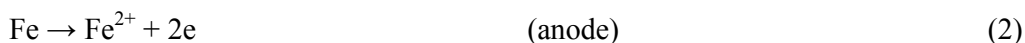
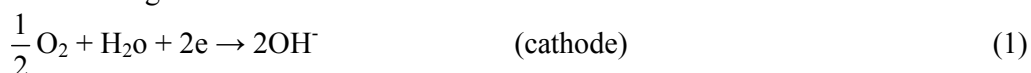
δ : diffusion layer thickness

1. Introduction

There are many methods for corrosion prevention, one mean of controlling is by the use of cathodic protection. Cathodic protection is one of the most common and effective methods for corrosion control of steel in neutral medium. It is an electrochemical technique in which a cathodic protective potential is applied to an engineering structure in order to prevent corrosion from taking place [1]. Cathodic protection can be applied in practice to protect metals such as steel, copper, lead and brass against corrosion in all soils and in almost all aqueous media. The principle of cathodic protection is in connecting an external anode to the material to be protected from corrosion and passing of an electrical DC current. So that all area of the metal surface becomes cathodic and therefore do not corrode. The external anode may be a galvanic (sacrificial) anode or it may be an impressed current anode where the current is impressed from external DC source [2].

Cooling systems suffer many forms of corrosion and failure [3]. Cooling water used removes heat from production processes. The open recirculating cooling water system is one of the most vital units of a petroleum refinery [4]. It often contains some salts that enhance the corrosion of metals. It is well known that the rate controlling step in most natural water corrosion process is the cathodic half reaction. The most important cathodic process in aerated waters is oxygen reduction. 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. The oxygen concentration will vary from a minimum at the surface to bulk concentration at the outside edge of the boundary layer. This results in the concentration gradient and promotes the transport of oxygen [5]. If the corrosion process is under cathodic diffusion control then the flow increases the corrosion rate. This effect generally occurs when an oxidizer is present in small amounts, as is the case for dissolved oxygen in acids or water [6]. The flow of fluid past a structure to be protected, affects the cathodic protection currents and protection potentials, however, higher protection currents are required since the corrosion rate is higher as has been evidenced by numerous studies [7; 8;9; 6,10].

The mechanism of cathodic protection is simply understood by reference to the following reactions.



Making the surface more negative and increase the concentration of electrons which accelerates the rate of cathodic reaction and decreases the rate of the anodic reaction. (i.e the rate of the anodic reaction becomes zero and the whole surface of the metal becomes cathodic [11].

Temperature of the medium governs the solubilities of the corrosive species in the fluid, such as oxygen (O_2), carbon dioxide (CO_2), chlorides, and hydroxides. Temperature increases the rate of almost all the chemical reactions. When the rate determining step is the activation process, the temperature changes have the greatest effect [12]. In open systems, the effect of temperature is complex in that the diffusivity of oxygen increases, but solubility decreases with temperature increase [6, 12, 13].

Several researches showed that the limiting current density increases with the increase in fluid velocity [8,9,10,20,21,22]. Konsowa and El-Shazly [14] found that the rate of zinc consumption used for cathodic protection of copper in saline water increases with increasing solution flow rate and temperature. Also the authors concluded that the cathodic protection is controlled by the rate of diffusion of dissolved oxygen towards the walls of the copper tube. Scantlebury [15] found that in marine environment, mild steel does not corrode under the protection potentials, viz., -780 and -1100 mV. Sami and Ghalib [16] noticed that cathodic protection current density increases with increasing temperature in sea water under stationary conditions.

Experimental data concerning the effect of flow velocity on the cathodic protection are scarce in the open literature, therefore, this work is devoted to study the impressed current cathodic protection of steel structure in 0.1N NaCl solution under flow conditions at various temperatures using weight loss and electrochemical technique (limiting current density) to determine the cathodic protection currents and potentials required to obtain high protection percents.

2. Experimental work

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

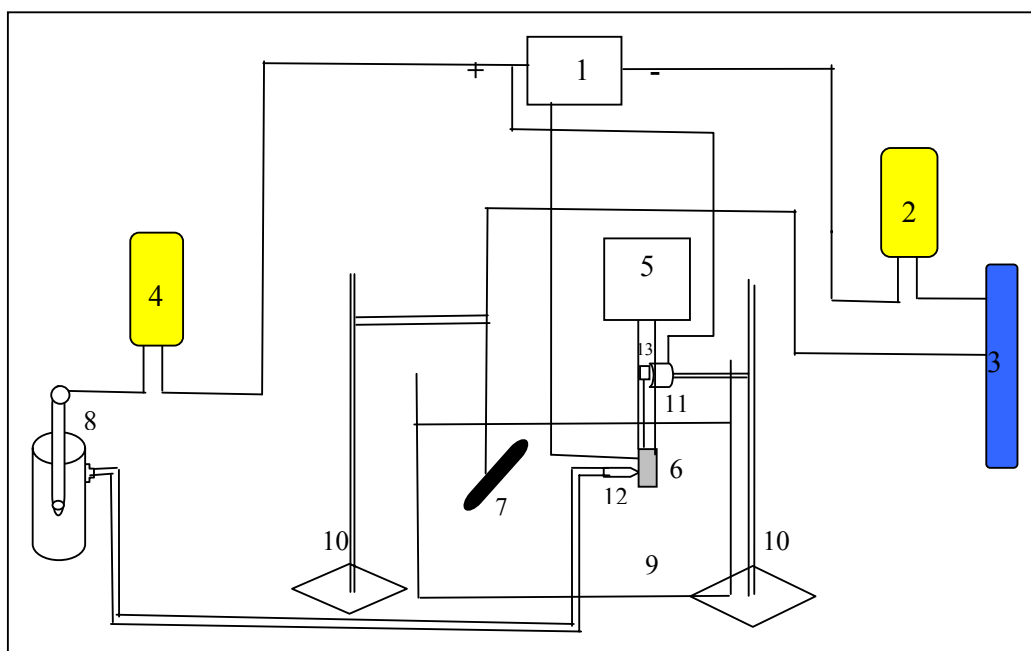


Fig. 1a: Experimental apparatus 1) power supply, 2) ammeter, 3) resistance box, 4) voltmeter, 5) stirrer, 6) working electrode (structure), 7) graphite electrode (anode), 8) calomel electrode (reference electrode), 9) water bath, 10) stand, 11) brush, 12) Luggin capillary, 13) metal for electrical connection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

The experimental apparatus was composed of mechanical agitator to obtain different rotational velocities, water bath to obtain different solution temperatures, carbon steel specimen 30 mm long and 25 mm outside diameter (cathode) attached to the rode of agitator, power supply to apply the required protection potential, digital

ammeter to measure the current, digital voltmeter to measure the potential, variable resistance (rheostat) to control the current flow, graphite electrode as auxiliary electrode (anode) of area approximately 3 times the carbon steel specimen (structure) to ensure the limiting current occur on the cathode rather than anode [17,18], and saturated calomel electrode (SCE) as reference electrode. Digital balance of high accuracy (4 decimal places of gram) to measure the weight loss. The electrical connection between cathode (specimen) and the cell was attained using brush.

In weight loss experiments, before each experiment the specimen was abraded in a sequence of emery papers grades: 120, 200, 320, 400, and 600. It was washed by tap water followed by distilled water, dried with clean tissue, immersed in annular acetone for 5 minutes, rinsed with water and dried with clean tissue. The specimen then was stored in vacuum desiccator over high activity silica gel for 24 h before use. Then it was weighted to nearest 0.1 mg. The carbon steel specimen was immersed in the solution (0.1N NaCl) at a particular temperature to corrode freely before applying the impressed current (cathodic protection) to measure the corrosion rate at stationary condition at different temperature (35, 45 and 55 °C). The corrosion rate was also measured at different speeds (25, 90, 120 and 200 rpm) at constant temperature of 45°C. After each test, the specimen was washed with tap water. Then it was brushed by smooth brush under running tap water to remove the corrosion products, immersed in acetone for 5 minute, washed with distilled water, then dried with paper tissue, and kept for 24 h in desiccator over high activity silica gel. Finally it was weighed using the digital balance. From the weight loss the corrosion rate (CR) was calculated as:

$$CR(gmd) = \frac{\text{weightloss}(g)}{\text{Area}(m^2) \times \text{Time}(d)} \quad (3)$$

Where CR represents corrosion rate in g/m².d. The protection percent (pp) was calculated as:

$$PP\% = \frac{CR_0 - CR}{CR_0} \times 100 \quad (4)$$

CR₀ and CR are the corrosion rate in absence and in presence of impressed current respectively.

In polarization experiments, 12 liter of 0.1N NaCl solution was used. The specimen was connected to -ve terminal of power supply to serve as cathode and graphite to +ve terminal to serve as anode. When the bath reached the required temperature, the specimen was immersed and the electrical circuit was switched on. The power supply was set at 5 V (applied voltage). The specimen (working electrode) was cathodically polarized from a particular potentials (-1.3 to -1.5 V versus SCE) to the corrosion potential (where I_{app.} = 0) by changing the applied current using rheostat. The current was recorded for step changes in potential. Two minutes were allowed for steady state to be reached after each potential increment [19]. The capillary tube was placed at distance 1-2 mm from cathode and connected to calomel electrode to measure the specimen potential. Thus polarization curve can be drawn and the limiting current can be obtained. In area of anode (graphite) immersed in water was 3 times the area of cathode (working electrode) to ensure the limiting current density occurs on cathode. For the case of flow conditions experiments, the specimen was attached to the agitator rod. A brush was used to ensure the electrical

connection between cathode and the -ve terminal of power supply. The corrosion current ($I_{\text{corr}}=I_{\text{Fe}}=I_{\text{O}_2}$) was also obtained from weight loss measurement as follows:

$$N_{\text{Fe}} = (\Delta w / M) / (A \cdot t) \quad \dots\dots(5)$$

where Δw represents weight loss, M is the molecular weight of carbon steel (iron), A is the area in m^2 and t is the time in second, N_{Fe} is the moles of Fe lost per unit area per unit time ($\text{gmole}/\text{m}^2 \cdot \text{s}$). Since:

$$i_{\text{Fe}} = z F N_{\text{Fe}} \quad \dots\dots(6)$$

where i_{Fe} is the corrosion current density in (A/m^2), F is Faradays constant (96487 coulomb/equivalent), z is the number of electron freed by corrosion reaction (for iron $z=2$). Hence

$$i_{\text{Fe}} = 2 \cdot 96487 \cdot N_{\text{Fe}}$$

$$I_{\text{Fe}} = i_{\text{Fe}} \cdot A$$

$$N_{\text{O}_2} = 0.5 N_{\text{Fe}} \quad (\text{according to equations 1 and 2}).$$

$$\text{Since } N_{\text{O}_2} = \frac{i_{\text{O}_2}}{z_{\text{O}_2} \times F}$$

Hence $I_{\text{Fe}}=I_{\text{O}_2}=I_{\text{corr.}}$. Also $I_{\text{O}_2}=I_{\text{L}}$.

After measuring $I_{\text{corr.}}$, different values of applied current were impressed to cathode at each temperature and velocity for 4 h to protect the metal and the potential was recorded at every 15 min to obtain the protection potential change with time. The values of applied current that were used to protect the metal at stationary and flow conditions at 45°C were $I_{\text{app}}=I_{\text{corr.}}$, $I_{\text{app}}=2I_{\text{corr.}}$ and $I_{\text{app}}=2.4I_{\text{corr.}}$ to determine the protection percent for different values of rotation velocity at 45°C . The distance between anode and cathode was 300 mm. Each run was carried out twice.

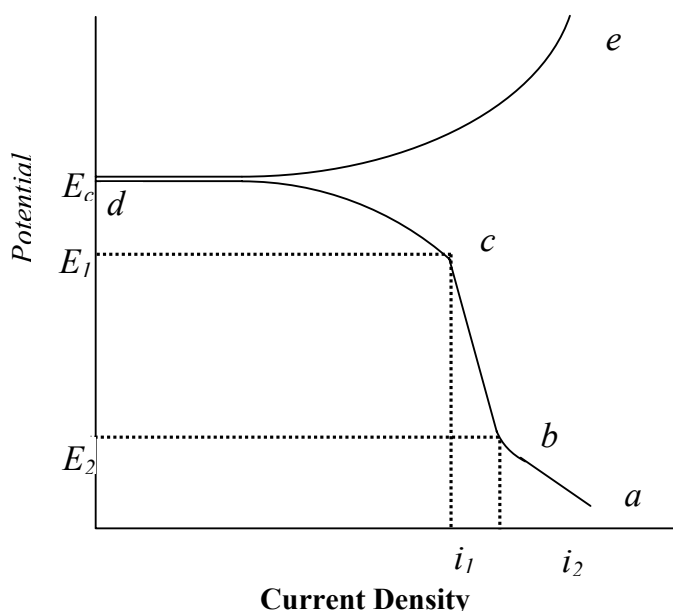


Fig. 1b: Typical Polarization Curve on C. Steel in Water.

I_{corr} was obtained from weight loss measurements for 4 h time of exposure. Since I_{corr} is lower than I_L where the later is for clean surface while I_{corr} for 4 h includes the corrosion product effect that reduces the corrosion rate [20, 21], so applying I_{corr} is not expected to protect the clean surface completely and it is needed to apply higher values. The limiting current plateau is not well defined, thus the method given by Gabe and Makanjola [34] was adopted to obtain the limiting current density values, i.e.:

$$i_L = \frac{i_1 + i_2}{2}$$

where i_1 and i_2 are the currents associated with E_1 and E_2 (Fig. 1b).

3. Results and discussion

Table 1 lists the experimental results for the stationary conditions.

Table (1) protection percent at various temperatures and applied currents ($A_{specimen} = 2355 \text{ mm}^2$).

Temp. (°C)	$I_{app.}$ (mA)	C.R (g/m ² .d)	%PP
35	0	22.549	-
	$I_{corr.}=1.99$	6.543	69.88
	$2I_{corr.}=3.97$	2.786	86.70
	$2.4I_{corr.}=4.76$	0.805	96.38
45	0	25.270	-
	$I_{corr.}=2.41$	5.636	78.22
	$2I_{corr.}=4.83$	3.413	87.1
	$2.4I_{corr.}=5.79$	0.249	99
55	0	24.49	-
	$I_{corr.}=2.29$	4.341	82.3
	$2I_{corr.}=4.59$	2.366	90.62
	$2.4I_{corr.}=5.51$	0	100

Table 2 lists the values of corrosion rate

and protection percent for different rotational velocities at 45°C.

Table(2) Effect of rotation speed on the cathodic protection percent ($A_{specimen}=2355\text{mm}^2$).

Speed (m/s)	$I_{app.}$ (mA)	C.R (g/m ² .d)	PP%
0.033	0	31.310	-
	$I_{corr.}=3.04$	11.301	64.56
	$2I_{corr.}=6.08$	2.689	99.2
0.118	0	59.418	-
	$I_{corr.}=5.65$	23.499	60.59
	$2I_{corr.}=11.3$	3.255	94.498
0.157	0	94.895	-
	$I_{corr.}=8.88$	34.068	62.8
	$2I_{corr.}=17.76$	4.407	95.42
0.262	0	154.440	-
	$I_{corr.}=14.76$	59.931	62.39
	$2I_{corr.}=29.52$	15.348	90.27

The best indication for cathodic protection is by weight loss. Fig. 2 shows the effect of temperature on the corrosion rate as gmd and as A/m^2 at stationary conditions. The results show that increasing temperature leads to unstable trend of corrosion rate and corrosion current. Unstable trend was also noticed by previous workers [20, 9, 21]. From this figure it can be seen that the corrosion rate at $45^\circ C$ is higher than at $35^\circ C$ and the corrosion rate at $55^\circ C$ is slightly lower than at $45^\circ C$ this can ascribed to the lower concentration of O_2 at $55^\circ C$ than $45^\circ C$. Increasing the temperature will increase the rate of oxygen diffusion to the metal surface and decrease the viscosity of water which will aid the oxygen diffusion. All these factors enhance the corrosion rate. On the other hand, increasing temperature decreases the oxygen solubility the factor that restrains the corrosion [12] as shown in Table 3 in the appendix.

3.2. Polarization results

Figures 3 to 6 show the polarization curves at $45^\circ C$ and different speed. It can be seen that as the velocity increases I_L is increased, i.e. at speed of 0.033, 0.118, 0.157 and 0.262 m/s, I_L is 4, 14.5, 19.5 and 22 mA respectively. This can be attributed to the increase in oxygen transport from the bulk of the solution to the metal surface leading to higher I_L [20, 9, 21,20].

3.2.1. Impressed current results

A metal in an aqueous environment, can be protected by impressing an electric current at its surface, delivered by a DC generator to polarize the potential to a value more negative than the equilibrium potential. Fig.7 shows the corrosion rate versus temperature at different applied currents values. The results in this figure indicate that as the applied current increases, the corrosion rate decreases this means the metal is protected by impressed current. For example the CR at $35^\circ C$ is $22.549 \text{ g/m}^2.d$ before applying protection current. When the applied current equals to corrosion current (1.99 mA) the CR becomes $6.543 \text{ g/m}^2.d$ (71% protection), when the applied current is double the corrosion current (3.97 mA) the CR becomes $2.786 \text{ g/m}^2.d$ (88% protection) , and when the applied current is $2.4 I_{corr}$ the CR becomes $0.805 \text{ g/m}^2.d$ (97% protection).

It is important to determine the amount of current or potential needed to be applied to protect a structure and to make sure that the anode can provide that current uniformly across the structure at a reasonable DC output voltage [Varmani and Clemena 1998]. The effect of applied current on the CR at different temperatures is shown in Fig. 8. It is clear that as the protection current for cathodic protection increases the corrosion rate decreases. The oxidation of iron releases electrons and ferrous ions, which dissolve in the solution surrounding the steel. The electrons are deposited on the steel surface, lowering the potential. In reduction current, electrons released by iron at the anode flow towards higher potential (cathodic) sites, where they combine with water and oxygen molecules to form hydroxyl ions. The corrosion reaction will only continue if there is cathodic reaction to accept released electrons, so these corrosion reactions can be stopped if oxygen and water are not available at the cathodic sites on the steel [23].

The protection percent at three temperatures ($35^\circ C$, $45^\circ C$ and $55^\circ C$) with applied current is shown in Fig. 9. From this figure it can be seen that the protection increases as the temperature increases , for example when applying $i_{app} = i_{corr}$ the protection s are 69.88%, 78.22% and 82.3% for $35^\circ C$, $45^\circ C$ and $55^\circ C$ respectively while when

applying current density equal to $2I_{corr}$, the protection percent are 86.70%, 87.1% and 90.62% for 35°C, 45°C and 55°C respectively, and for higher current ($I_{app.}=2.4I_{corr.}$) it is 96.38%, 99% and 100%. The protection percent increased with increasing impressed current ($I_{app.}$) due to the increased reduction of O_2 at the metal surface leading to decrease the corrosion of metal [11, 30]. Fig. 9 also reveals that the protection percent varies with temperature with unstable trend depending on O_2 solubility and diffusivity [9,10].

3.2.2 Protection potential

Figure 10 shows effect of time on the protection potential of carbon steel pipe immersed in 0.1 N NaCl at different temperatures. The figure shows that the potential decreases with time, where the potential is recorded at each 15 min for an experiment time of four hour long. Precisely the potential becomes rapidly more negative in the first hour, and then the curve reaches asymptotic value at -990 mV, -990 mV and -1070 mV vs SCE which represent the protection potential for 55°C, 45°C and 35°C respectively. Numerous studies have demonstrated that E_c is generally influenced by the oxidizer concentration. Several investigators [22; 24,25,26] showed that decreasing oxygen concentration leads to shift E_c to more negative values. It is evident that E_c falls with increasing temperature this because E_c are affected by both anodic and cathodic process. The anodic (activation controlled) process is simulated by increasing bulk temperature to greater extent than that of cathodic (concentration controlled) process [11,27]. The corrosion potential become more negative with time, because the oxygen level at the steel interface can be depleted over time when subject to constant current density [28]. This figure indicates that at applied current of $I_{app.}=I_{corr.}$ the higher the temperature is the higher the protection potential. The decrease in the potential with time can be ascribed to the fact that at low temperature the depletion of O_2 is higher due to its high reduction on the metal surface leading to decrease the protection potential.

Figure 11 shows that the potential decreases with increasing time at different applied current values at temperature of 35°C. The corrosion potential become more negative with time, because the oxygen level at the steel interface can be depleted over time when subjected to constant current density [28]. It can be seen that as the current increases, the potential becomes more negative because high $I_{app.}$ leads to high O_2 reduction, shifting the potential to more negative [22, 26]. The corrosion potentials for current applied and higher current is approximately similar while for lower current are less negative. The decrease in potential is steep during the 90min for $I_{app.}=1.99$ mA while for $I_{app.}=3.97$ mA and $I_{app.}=4.763$ mA the decrease in potential is steep during 30 min.

Fig.12 shows the effect of rotational velocity on the corrosion rate of carbon steel specimen for different applied currents. It is clear that the corrosion rate decreases with increasing applied current and increases with increasing velocity. The increase in the rate of carbon steel dissolution with increasing solution velocity can be attributed to the decrease in the thickness of the hydrodynamic boundary layer and diffusion layer across which dissolved O_2 diffuses to the metal wall [29, 31] with a consequent increase in the rate of O_2 transfer from bulk solution to the surface. It is to be noticed from Fig. 13 that at high applied current values the effect of velocity on the corrosion rate is lower, however, the corrosion rate increases considerably with velocity when $I_{app.}=0$ while it increases little at $I_{app.}=I_{corr.}$ and $I_{app.}=2I_{corr.}$.

4. Conclusion

The conclusions of this work are:

- 1- Under flow conditions, the required currents to protect steel structure are high. The higher the rotational speed is the higher the cathodic currents required for protection.
- 2- The corrosion rate of carbon steel specimen in 0.1N NaCl solution exhibits unstable trend with temperature.
- 3- Increasing rotational velocity (0-0.262 m/s) leads to increase the corrosion rate from 25.27 g/m².d to 154.44 g/m².d at 45°C.
- 4- The impressed current required for cathodic protection depends on temperature and rotational velocity. Applying $I=2I_{\text{corr}}$ give protection percent between 90 to 99% depending on temperature and velocity.
- 5- The higher the impressed cathodic protection currents is the lower the effect of velocity on the corrosion rate will be.
- 6- The protection potential shifts to more negative values with time where it decreases by 40-68% depending on temperature. Also it shifts to more -ve with increasing velocity.
- 7- The lower the temperature is the more negative protection potential will be.

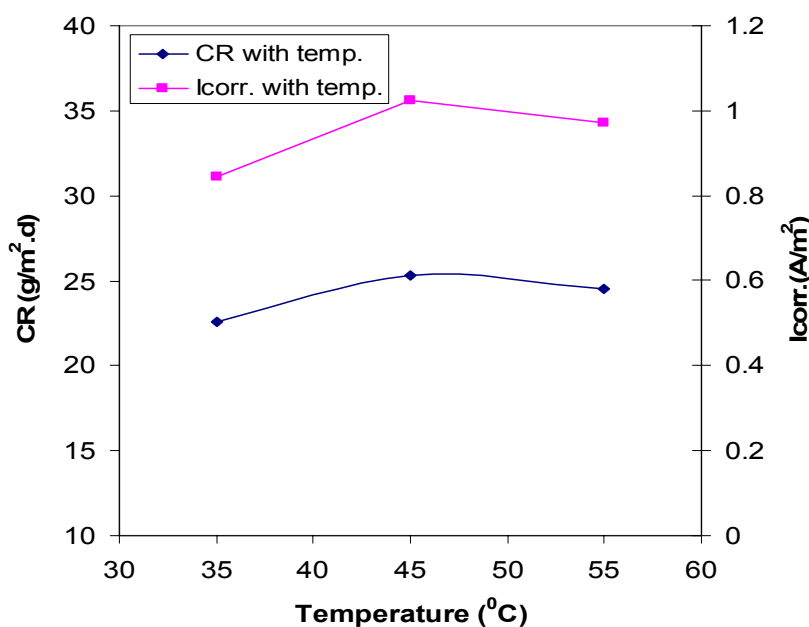


Fig. 2: Corrosion rate and I_{corr} vs. temperature at free corrosion at stationary conditions.

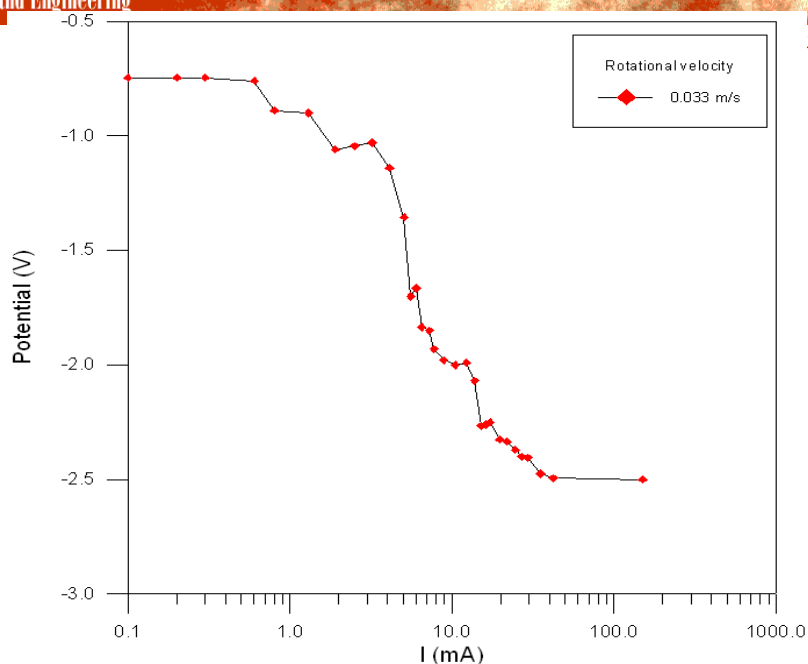


Fig.3: Polarization curve at velocity of 0.033 m/s at temperature=45°C.

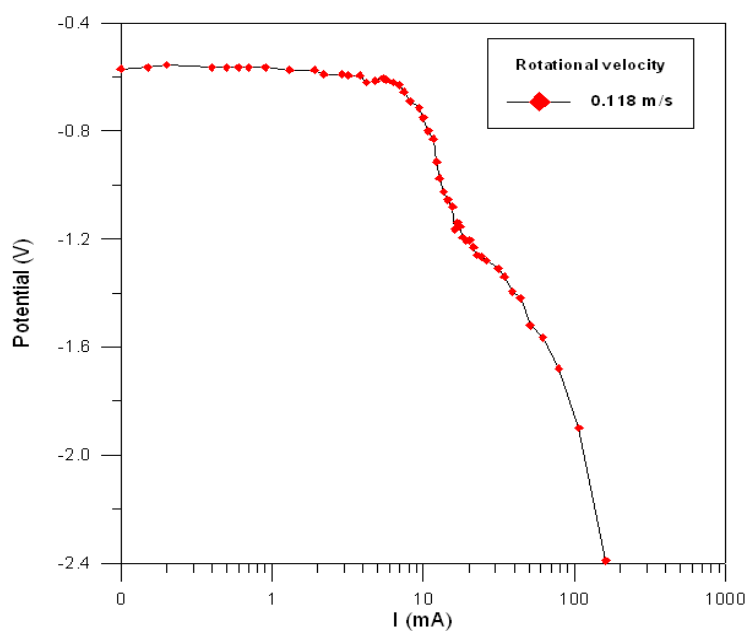


Fig. 4: Polarization curve at velocity of 0.118 m/s at temperature= 45°C.

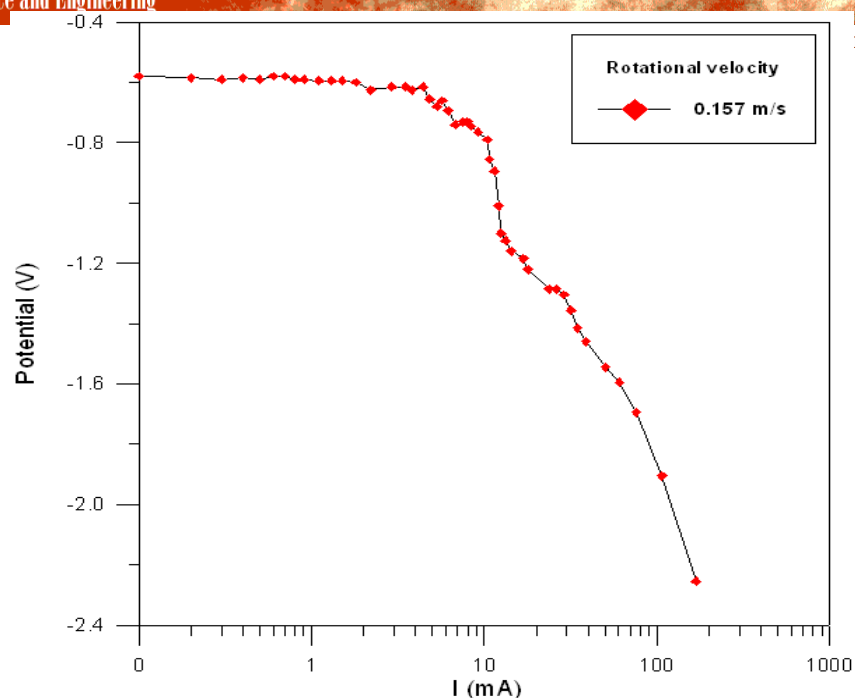


Fig.5: Polarization curve at velocity of 0.157m/s at temperature=45°C.

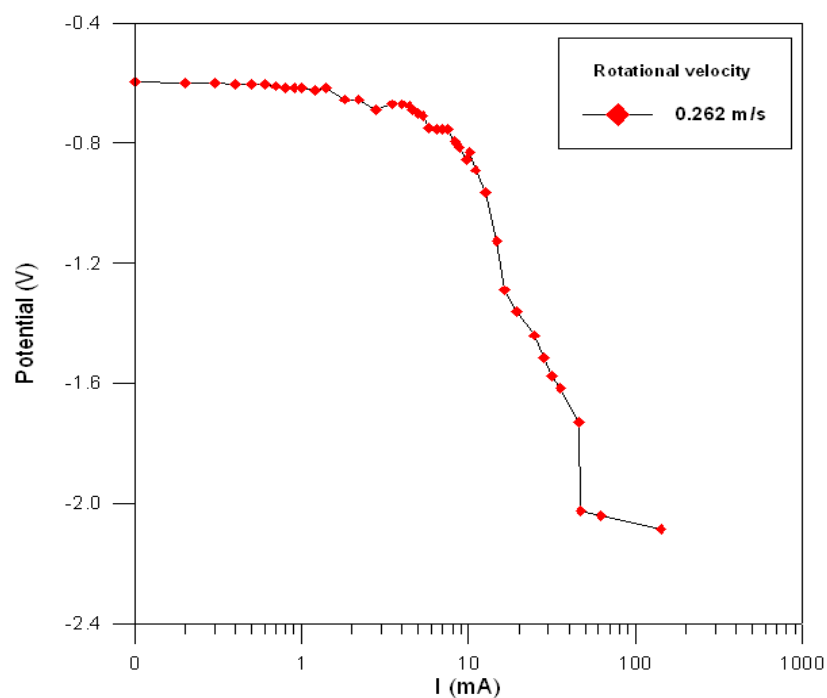


Fig.6: Polarization curve at velocity of 0.262 m/s at temperature=45°C.

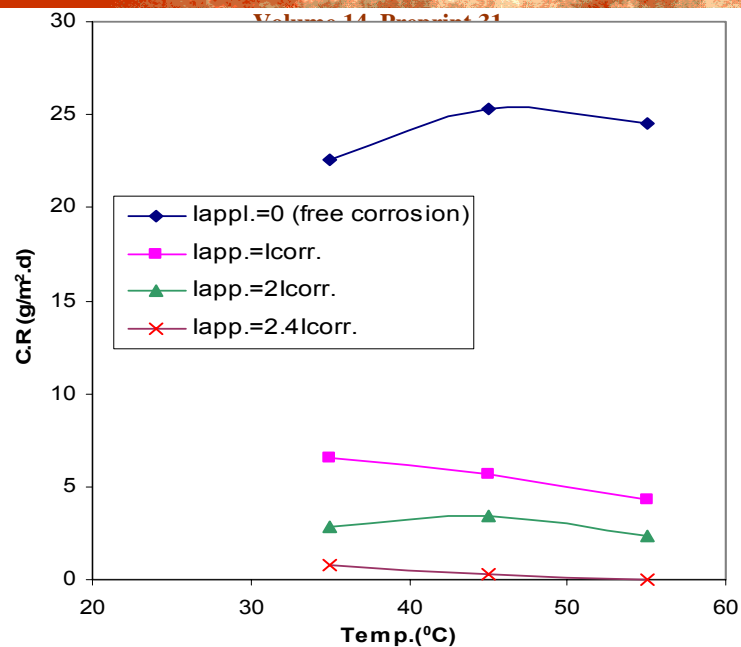


Fig. 7: CR vs. temperature at stationary conditions for different values of applied current.

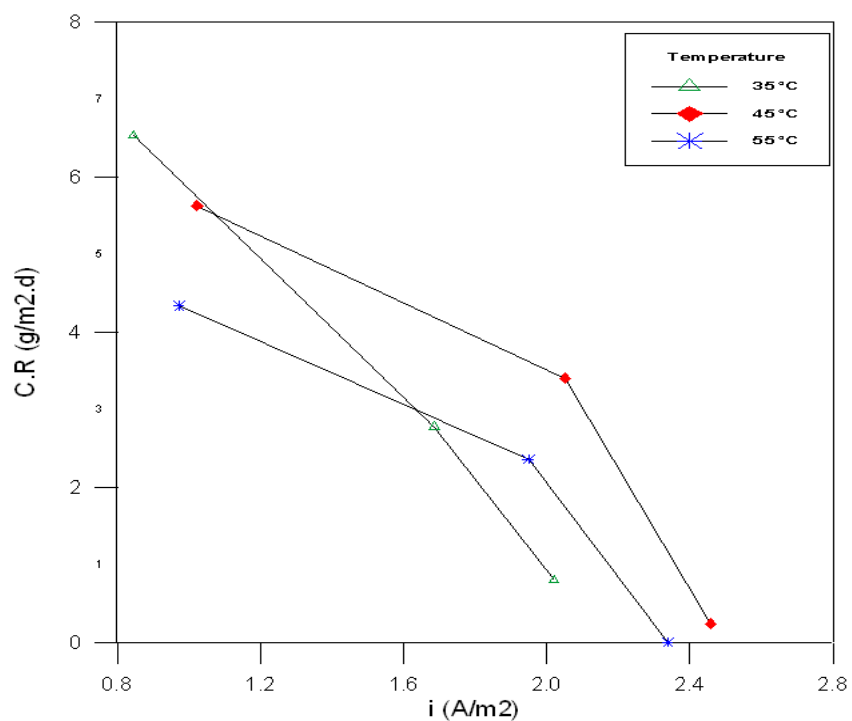


Fig.8: CR vs. i_{applied} at stationary conditions for various temperatures.

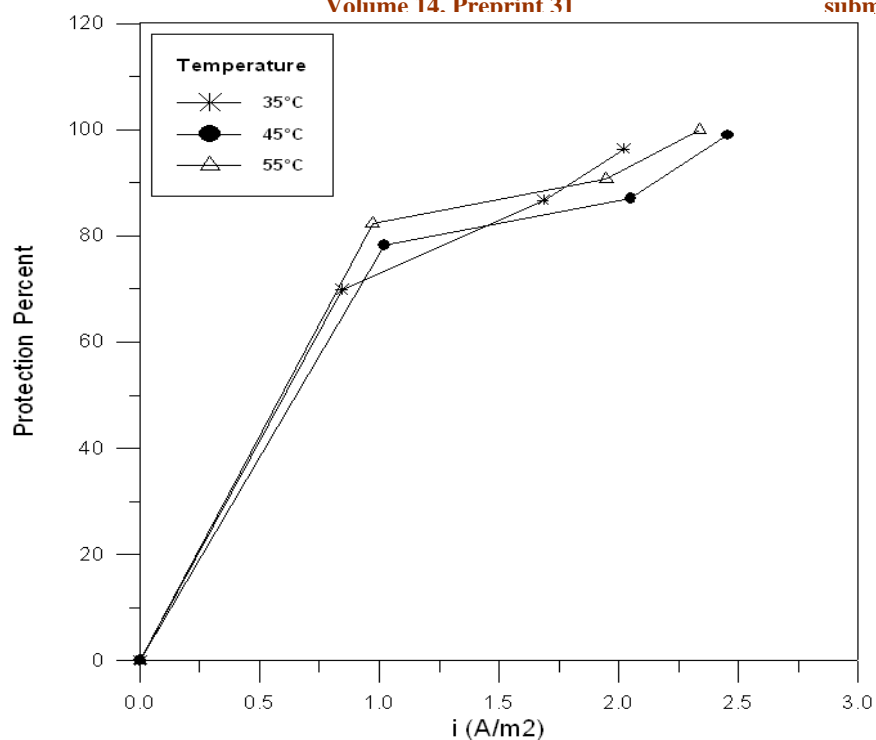


Fig. 9: i_{app} vs. protection percent at different temp. at stationary conditions

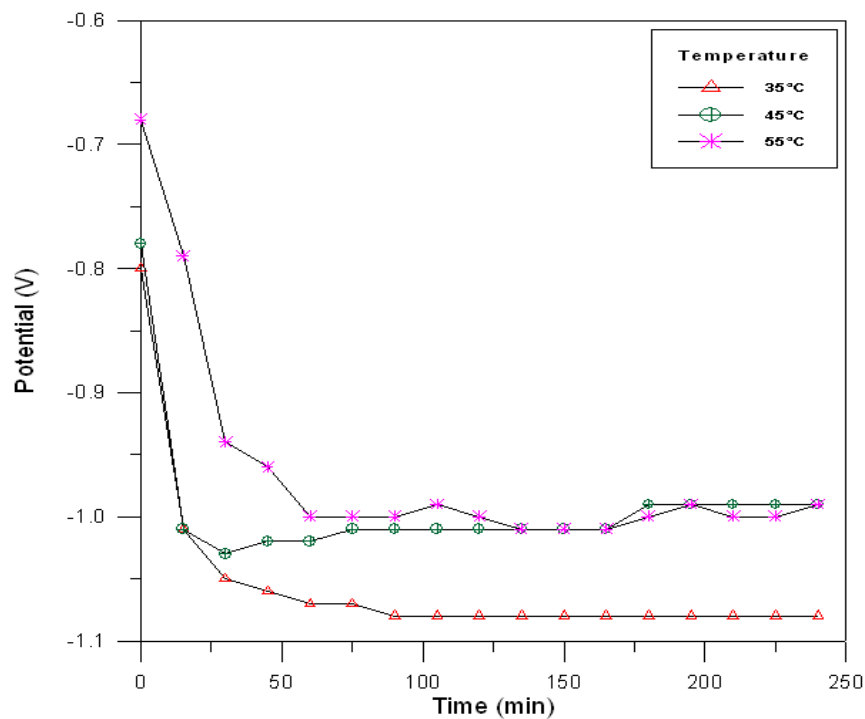


Fig. 10: Potential Vs. time at $I_{app}=I_{corr}$ at stationary conditions.

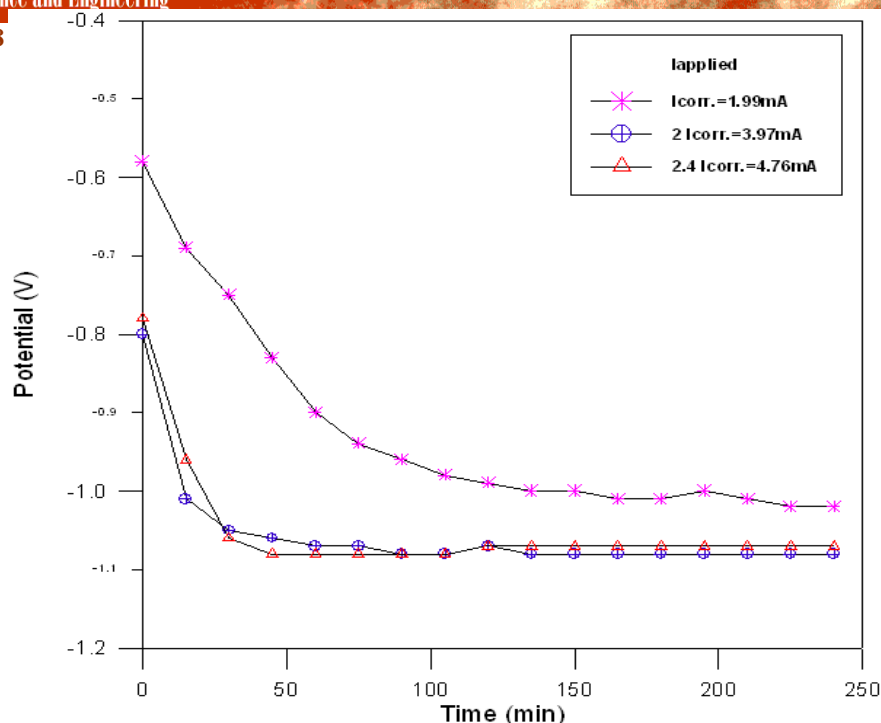


Fig.11 Protection potential vs. time at 35°C for different applied currents.

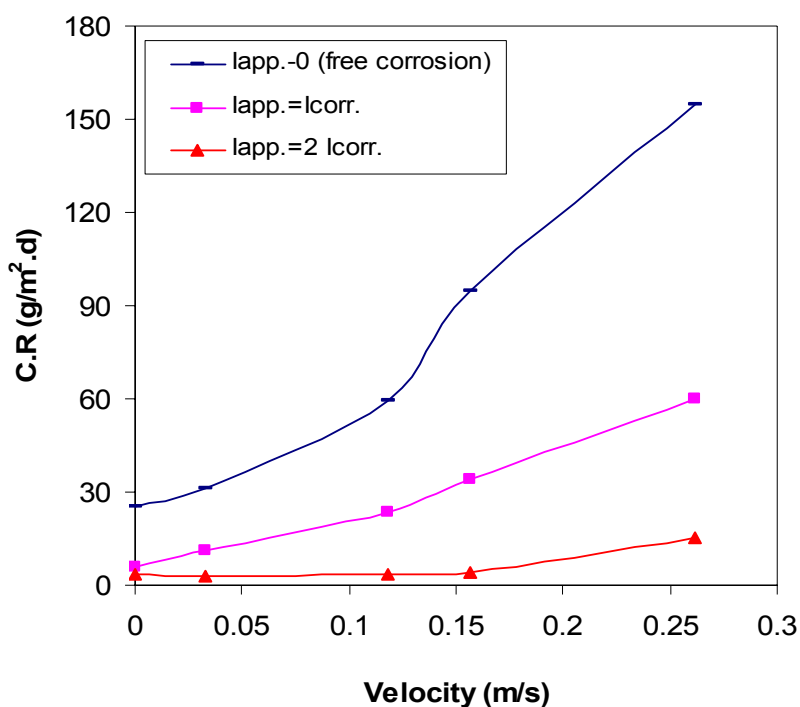


Fig.12 Effect of applied current on the corrosion rate at 45°C.

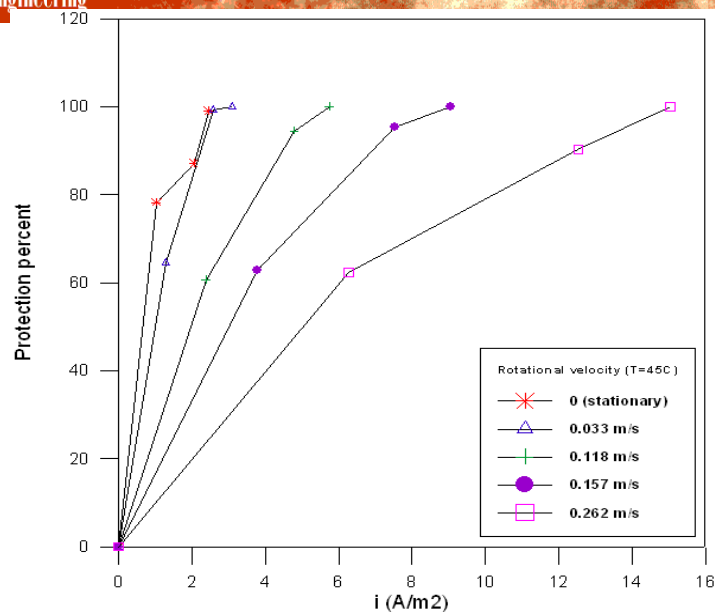


Fig.13: i_{app} vs. PP% at different speed and constant temp. 45°C

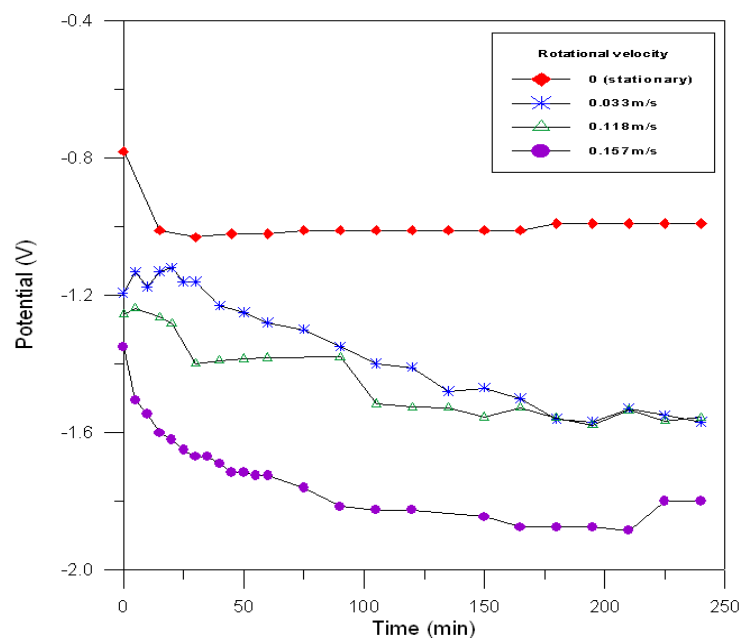


Fig 14 Potential vs. time at $I_{app} = 2I_{corr}$ at temperature=45°C.

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APPENDIX

Table 3: Values of Oxygen Diffusivity and solubility at different temperatures (32, 33)

T(°C)	$10^9 \text{ (m}^2\text{/s)} \times D$	Solubility (mg/l)
30	2.374	7.5
40	3.010	4.389
50	3.562	5.399
60	4.834	4.49