The Effect of Time and Corrosion Products Formation on Corrosion Rate of Carbon Steel Pipe Under Turbulent Flow Conditions

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Abstract

The effect of time (or corrosion products formation) on corrosion rates of carbon steel pipe in aerated 0.1N NaCl solution under turbulent flow conditions is investigated. Tests are conducted using electrochemical polarization technique by determining the limiting current density of oxygen reduction in Reynolds number range of 15000 to 110000 and temperature range of 30 to 60oC. The effect of corrosion products formation on the friction factor is studied and discussed. Corrosion process is analyzed as a mass transfer operation and the mass transfer theory is employed to express the corrosion rate. The results are compared with many proposed models particularly those based on the concept of analogy among momentum, heat, and mass transport. The capability of these models to predict corrosion rates in presence of corrosion products is examined and discussed. It is found that formation of corrosion products with time decreases the corrosion rate (or mass transfer rate) at low Reynolds number and temperature while it increases the corrosion rate at high Re and temperature. It increases momentum transport and this increase depends on temperature, Reynolds number, and corrosion rate. Increasing roughness due to the formation of corrosion products causes overestimation of analogy correlations results by increasing friction factor and decreasing corrosion rate.

Keywords: corrosion, carbon steel, time, corrosion product, friction factor, analogy.

Introduction

The corrosion of carbon steel in neutral environments is of practical importance, therefore it is considered by many studies. It is widely recognized that the corrosion of carbon steel may be accounted for by the following anodic reaction:

$$Fe \rightarrow Fe^{2+} + 2e \qquad (1)$$

and cathodic reaction in presence of oxygen,

$$\frac{1}{2}O_2 + H_2O + 2e \rightarrow 2OH^- \qquad (2)$$
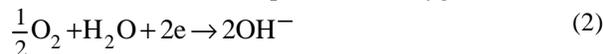
In many corrosion problems, there is strong evidence that the rate of uniform corrosion is controlled by the rate of mass transfer. This is true whether the corrosion fluid remains static or in fast motion with respect to the metal surface. However, molecular diffusion is not the only factor which influences 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 [1]. It is accepted that the corrosion of mild steel in areated water is controlled by the rate of cathodic reduction of oxygen and hence by the oxygen transport from the main stream solution to the reacting surface.

Many studies [1,2,3,4,5] were carried out to express the corrosion process, that is under diffusion control, as a mass transfer operation in terms of mass transfer parameters using modified form of Reynolds analogy. These studies were performed for various metal-environment systems.

Mass transfer theory can be applied to predict corrosion rates for diffusion controlled systems not exhibiting passivity phenomena.

In previous study [6] the effect of Re and temperature on the corrosion rate of carbon steel in aerated 0.1N NaCl
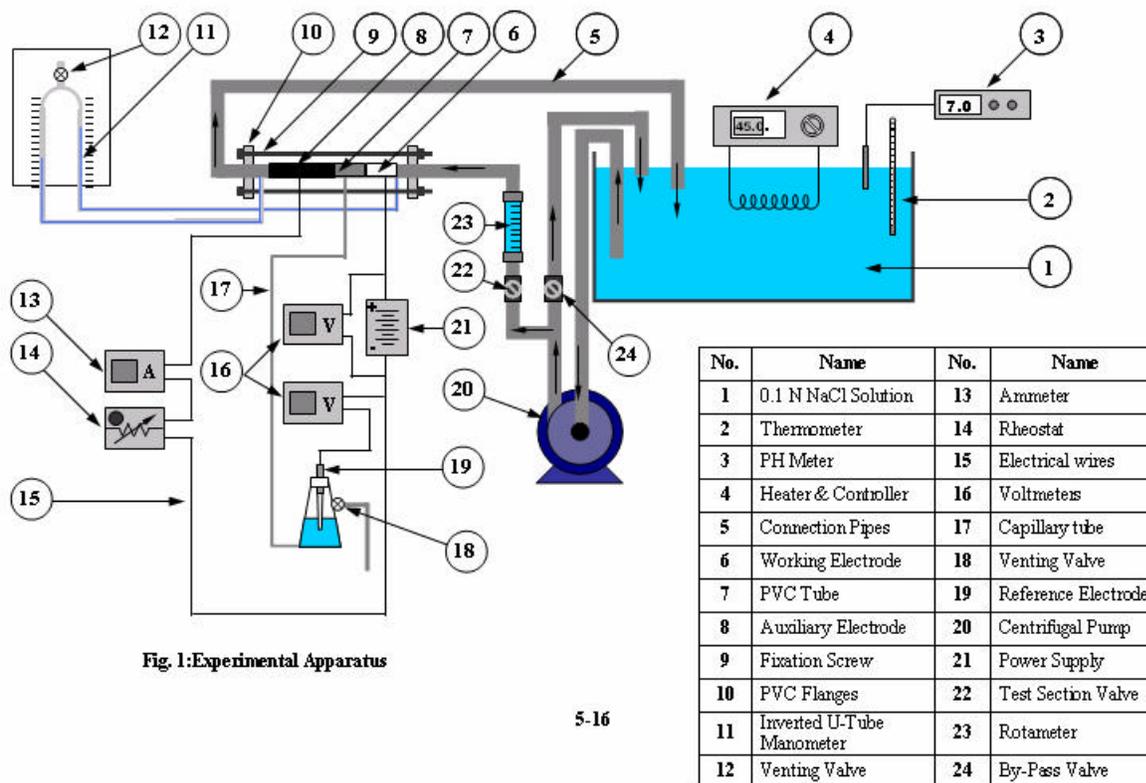
Fig. 1:Experimental Apparatus

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No.	Name	No.	Name
1	0.1 N NaCl Solution	13	Ammeter
2	Thermometer	14	Rheostat
3	PH Meter	15	Electrical wires
4	Heater & Controller	16	Voltmeters
5	Connection Pipes	17	Capillary tube
6	Working Electrode	18	Venting Valve
7	PVC Tube	19	Reference Electrode
8	Auxiliary Electrode	20	Centrifugal Pump
9	Fixation Screw	21	Power Supply
10	PVC Flanges	22	Test Section Valve
11	Inverted U-Tube Manometer	23	Rotameter
12	Venting Valve	24	By-Pass Valve

solution for clean surface, where no corrosion products (t=0), was studied and the capability of analogy models to estimate the corrosion rate was examined. Also the mass transfer theory was employed to express the effect of Re and temperature on the corrosion rate. In present work it is aimed to investigate the effect of time or corrosion products formation on the corrosion rate and on the applicability of the correlations that are based on the concept of analogy among mass, heat, and momentum transfer to examine to what extent it is possible to employ these correlations to predict corrosion rate by determining experimental friction factor at various intervals over the investigated range of Re and temperature.

In general, as the pipe corrosion proceeds, the process becomes more complex due to building up of corrosion products which restrain the oxygen transport to the surface, growth of the surface roughness which increases the momentum and mass transport between surface and solution, changing physico-chemical properties of the corrosion products; and changing mechanics of flow [3].

The most important study concerned the effect of time (or corrosion products formation) on the corrosion amount (weight loss) was that of Mahato et. al. [3,7,8]. They used weight loss method to determine the effect of time on the corrosion amount. It was an excellent practical study since it considered long time of exposure but the shortcoming of this study was the relatively low Re range. In addition to that, the weight loss method for

determining corrosion rate gives the average corrosion rate over the test period and not the instantaneous. Accordingly, it is impossible by weight loss to understand the behavior of the corrosion rate during the formation of corrosion products layer. Therefore, in the present work the limiting current density technique (LCDT) was employed to study the behavior of the corrosion rate during the growth of the corrosion products layer. Mahato et. al. studies [3,7,8] revealed that corrosion products formation generally decreases the corrosion rate. This is not always true as will be discovered in present work.

Experimental Work

The dynamic system for performing the experimental work is illustrated in Fig.1

Pressure Drop Measurements

The pressure drop across the test section was obtained employing an inverted U-tube manometer with static pressure taps located at the edges of the test section in the wall of the extension PVC pipe. The friction factor was obtained from pressure drop measurements for a pipe 20cm long for the entire range of Re, temperature, and time. The solution with a particular Re and temperature was pumped to the test section and the pressure drop

across the latter was taken after 10 minutes from the test run start. This value of pressure drop was considered to be at time zero. As corrosion proceeds, there was a continuous growth of corrosion products layer with time and hence continuous increase in pressure drop and friction factor. Therefore the values of pressure drop were recorded at different time intervals during the test period to obtain friction factor variation with time. Each measurement was performed twice and the average was taken.

Polarization Measurements

The solution is prepared and circulated through by-pass to reach the required temperature by using heater and controller. The test section components (working electrode, counter electrode, reference electrode) were mounted in their positions. Care was taken to insure that there were no discontinuities in the cross section of the pipe immediately up stream and down stream of the electrode. Silicon rubber was used to prevent leakages in the test section. After the solution had reached the desired temperature, it was allowed to pass through the corrosion cell and the flow was adjusted to the desired Re with the aid of two control valves. When the solution reached the test section, the electrical circuit was switched on. The specimen (working electrode) was cathodically polarized from a potential of nearly -1.4 V (vs. SCE) to the corrosion potential (where iapp=0) at a rate of 5-20 mV 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 [9,10]. Thus polarization curve can be drawn and the limiting current density can be obtained. The obtained values of iL represent values for clean surface (t=0), i.e., no corrosion products were formed, since during the polarization experiment no free corrosion occured (except at low currents near the corrosion potential) because the specimen was cathodically protected. To investigate the influence of time on the instantaneous corrosion rate (or mass transfer coefficient), the electrical circuit was switched off and the specimen was allowed to corrode freely under flow of the corrosion solution at a particular Re and temperature. A free corrosion of the specimen continues for 2.5 h during this interval corrosion products form on the surface of the pipe. At the end of 2.5 h the electrical circuit again switched on and the specimen was polarized to -1.4 V (vs. SCE) and polarization was repeated to obtain the complete polarization curve and new iL is determined. This value of iL represents the value at t=2.5 h. At the end of the second polarization measurement the electrical circuit was switched off while the corrosion solution was kept flowing through the specimen for another 2.5 h. During this second interval the specimen will undergo a free corrosion forming additional corrosion products. The specimen was then polarized to -1.4 V (vs. SCE) by switching on the electrical circuit again to repeat the polarization experiment and to obtain new iL at t=5 h. Hence three values of iL were obtained for each test run. Each test run was repeated twice and the average was taken.

Results and Discussion

Friction Factor

Over the years, friction factor (f) has received a wide attention from many authors. Numerous studies have been performed to study the factors influencing friction factor under different conditions, e.g., isothermal and heat transfer conditions [6,10,11,12,13,14,16,22] and roughness conditions [11,14,17,18,19,20,21]. Most of these studies have showed that the main two variables influencing f were Re and surface roughness. In previous work [6] the effect of Re on f was studied and discussed. In present work the effect of time or roughness produced from corrosion products on the friction factor over the whole range of Re and temperature is considered.

Determining f at different time intervals during the corrosion process serves to understand the effect of corrosion products formation on friction factor and calculating the instantaneous corrosion rate (or mass transfer coefficient) from the proposed analogy models that are based on friction factor in order to compare with actual instantaneous corrosion rates obtained from iL. Figures.2 show the variation of friction factor with time for the whole range of Re and temperature. It is clear that f increases with time due to increased roughness of the corroding wall. This increase depends on Re, temperature, and corrosion rate. Figure 3 shows a plot of the relative increase in friction factor (f/fo) with time with Re as a parameter. Observing these figures indicate that at a particular temperature the higher the Re is the higher the increase in f (or f/fo) due to higher corrosion rate. It is evident also that for Re=80000 the maximum value of f/fo at 30oC is 1.3, at 45oC is 1.5, and at 60oC is 1.42, i.e., in spite of the corrosion rate at 60oC is higher than at 45oC , the relative increase in f is lower. This can be attributed to three reasons: (i) increasing temperature leads to decrease the viscosity which plays an important role in decreasing pressure drop [15,23] causing lower f values at a particular time. (ii) increasing temperature affects the nature of corrosion products layer particularly density and porosity [7], i.e, the rough nature of this layer decreases with temperature rise leading to lower pressure drop and hence lower f (or f/fo). (iii) solubility and erosion of corrosion products layer at high temperatures and Re leading to reduce the growth of this layer and hence the increase of f. Fig.4 shows the variation of friction factor with Re for rough (corroded) surface. The

formation of corrosion products with time deviates the relation between f and Re from smooth surface behavior, i.e, the friction factor increases with Re rather than decreases. The higher the smoothness of surface is the higher the decrease of f with Re. Studies for fully rough surface [20, 21, 24, 25] showed that no effect for Re on f at high values of Re and the higher the surface roughness the lower the decrease of f with Re. This independency of the friction factor on Re in fully rough region is interpreted as follows: as Re (or velocity) increases the thickness of the momentum boundary layer decreases and eventually the protrusions penetrate the boundary layer and the behavior deviates from that of smooth pipe [25]. Add to that, as Re increases, the corrosion rate increases resulting in more corrosion products and hence the friction factor increases.
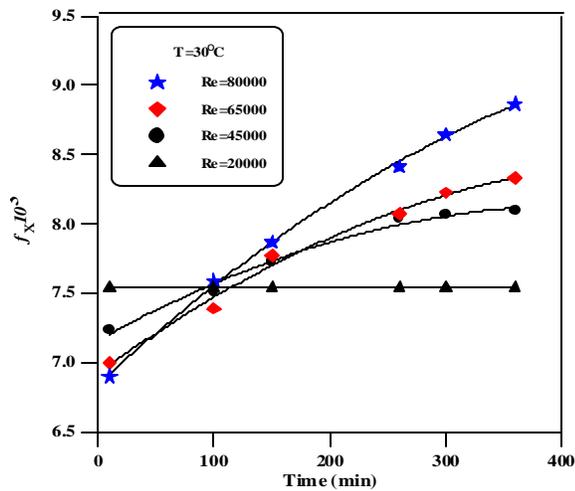
Fig. 2a: Variation of Friction Factor with Time for Various Re at 30°C
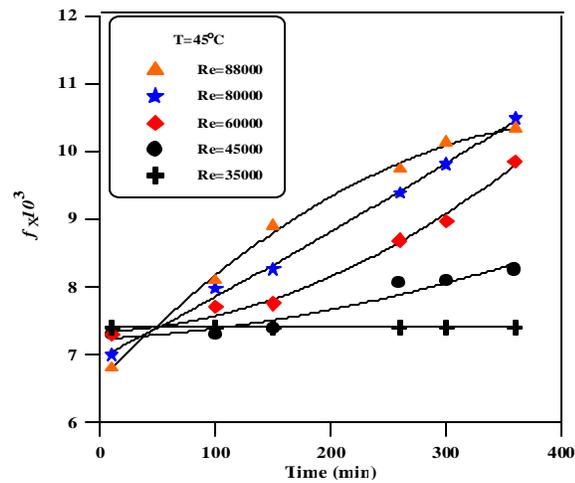
Fig. 2b: Variation of Friction Factor with Time for Various Re at 45oC
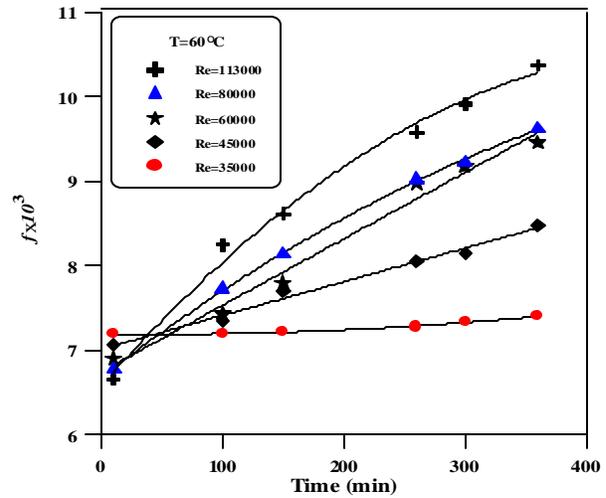
Fig.2c: Variation of Friction Factor with Time for Various Re at 60oC
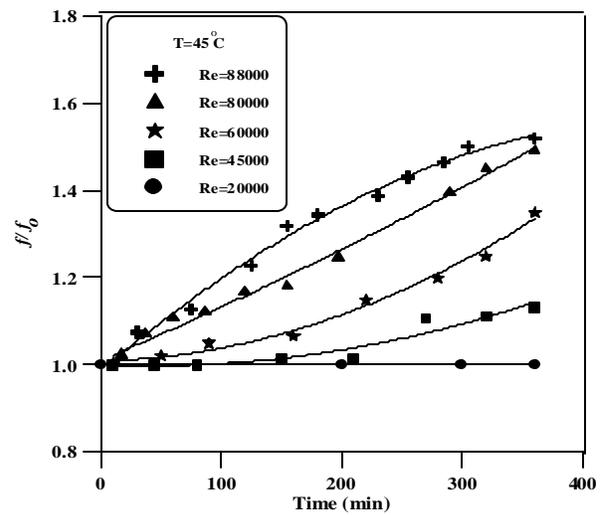
Fig.3: Variation of Relative Friction Factor (f/fo) with Time at 45oC

It is not possible to correlate corrosion amount with friction factor increase for two reasons. Firstly, at low Re no pronounced increase in friction factor with time (Figs.2) for all temperatures despite of corrosion (as indicated by polarization measurements). Secondly, erosion at high Re and temperature restricts the growth of product layer and consequently the increase in f.

Corrosion Rate

Studying the limiting current density behavior with time serves to understand the influence of Re, temperature, and corrosion products formation on the instantaneous corrosion rate. The effect of time is due to
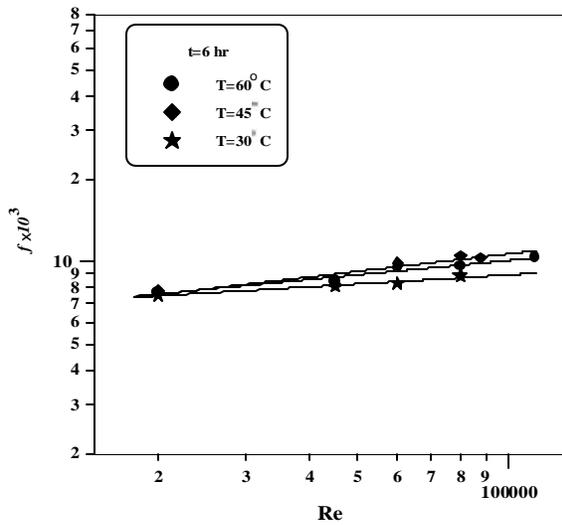
Fig.4:Variation of Friction Factor with Re for Rough Surface (t=5 h).

the continuous growth of the corrosion products layer which affects the transport of O2 to metal surface and the activity of the surface and hence the corrosion rate (or mass transport rate) [1,7]. Figures.5 show the polarization curves for Re=20000 at different intervals for three temperatures. Figures.6 show the variation of iL with time at the three temperatures with Re as a parameter.

The figures indicate that the formation of corrosion products with time may increase or decre iL, depending on the values of Re and temperature. At 30oC, Fig.6a shows that for all Re, iL decreases with time. At 45oC, Fig.6b shows that for low Re, iL slightly decreases with time. As Re increases, the decrease in iL with time gradually vanishes. At high Re, a slight increase of iL with time can be observed. Also it can be noted that at moderate Re, the iL decreases with time and then increases. At 60oC, Fig.6c indicates that for low Re, there is a slight decrease in iL with time. At moderate Re, iL remains nearly unchanged, and at high Re, iL appreciably increases with time. In fact, this is surprising finding where the previous studies [3,7,8] indicated that the corrosion rate always decreases with time. To interpret this phenomena; Mahato et. al. [3] stated that the corrosion process consists of four steps: (i) diffusion of O2 to the solid-liquid interface through a viscous sub-layer; (ii) diffusion of dissolved O2 through the corrosion products layer (iii) reaction at the surface; (iv) precipitation of corrosion products at the solid surface.

As corrosion proceeds the amount of corrosion products increases for the whole investigated range of temperature and Re which can be concluded from pressure drop (or friction factor) measurements (Figs.2). At 30oC, the growth of corrosion products layer will reduce the amount of O2 arriving to the surface and

therefore iL will be decreased. At 45oC and 60oC, since the corrosion rate is higher than at 30oC, the thickness of corrosion products layer will be greater for same Re. During the initial period the corrosion products are developing within the viscous sub-layer. As the thickness of corrosion products increases with time, the protrusions height increases until it becomes greater than the thickness of viscous sub-layer (db) and hence at high Re the turbulence at the peaks of these protrusions increases leading to decrease the thickness of diffusion sub-layer (dd) which is a fraction of viscous sub-layer and represents the main resistance to mass transfer [3,20,22,26,27,28,29]. Therefore as Re increases, the turbulence around and inside the corrosion products layer increases leading to decrease db and dd and more O2 arriving to the surface and hence higher corrosion rate. The eddy mass transfer resulting from extreme surface roughness will have an important role in the mass transfer of reactant from the main liquid bulk to the solid surface [7]. Evans [30] indicated the greater turbulence due to high velocities results in more uniform O2 concentration. This shows how the corrosion products increase the corrosion rate rather than decrease it (for the investigated range of time). Also the rise in temperature may contribute to increase the porosity (or decrease the density) of corrosion products layer and that facilitates the passage of O2 toward the surface [7]. This is why the effect of corrosion products in reducing the corrosion rate decreases with increasing temperature at a particular Re. Dawnson and Trass [17] and Berger and Hau [31] found that the surface roughness enhances the mass transfer rate. Kandlikar et. al. [14] and Oziski [32] ascribed the enhancement of heat transfer rate in rough surfaces to the disturbance of viscous sub-layer by the protrusions. It is to be remembered that the roughness produced from the accumulation of corrosion products is different from the roughness that is an inherent property of the metal wall.
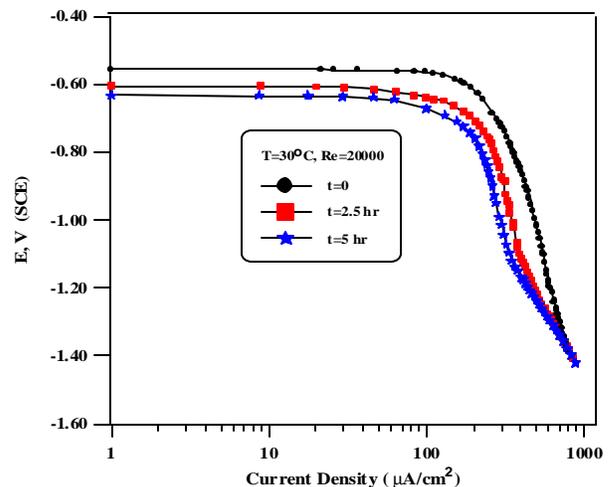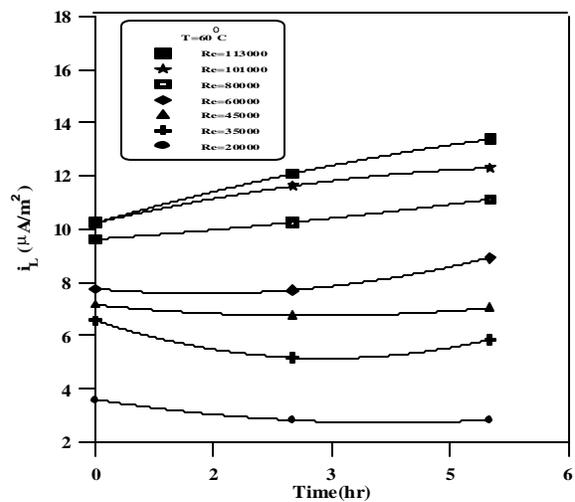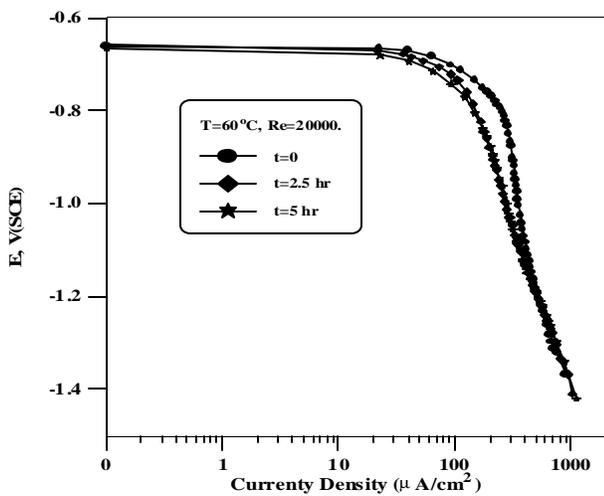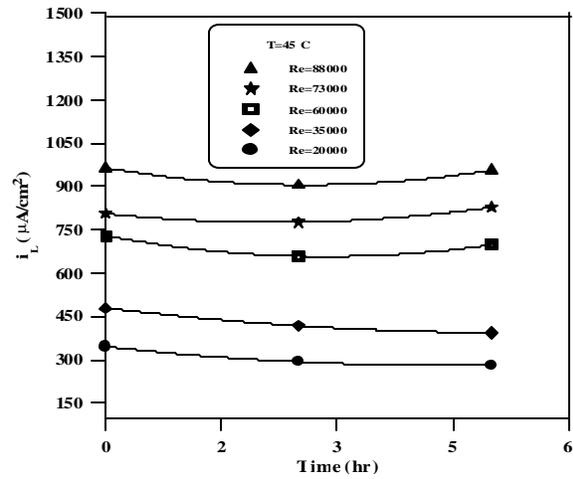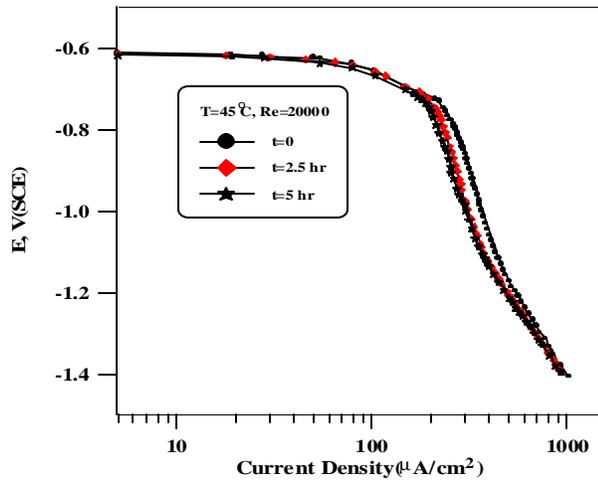
Figs.5: Cathodic Polarization Curves Showing the Effect of Time

Fig. 6: Variation of iL with Time for Different Re,(a) 30oC (b) 45oC, (c) 60oC
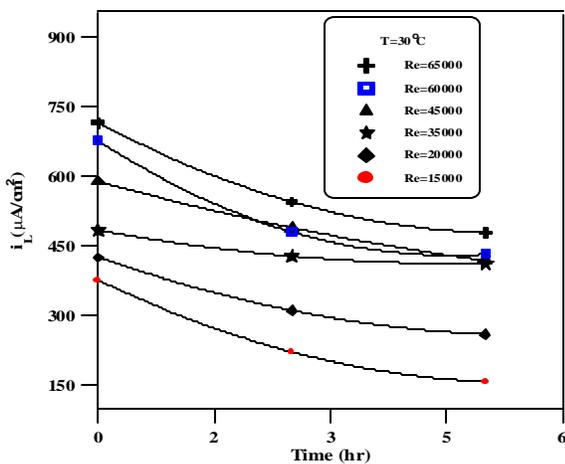
Mass Transfer

The formation of corrosion products with time influences the mass transfer rate of oxygen from the bulk to the metal surface. The limiting current density technique permits the estimation of the instantaneous value of experimental k and consequent dimensionless groups (Sh) via determining iL. The determination of K+ requires the determination of friction factor.

Figs.7 shows the variation of Sh with Re at various temperatures with time as a parameter. These figures indicate that at 30oC and 60oC time effect on Sh is more than at 45oC. At 30oC, the formation of corrosion products causes a significant decrease in Sh for all Re range. At 60oC, the time effect varies with Re, i.e., at low Re, the Sh decreases with time via deposition of corrosion products. As Re increases at this temperature

the effect of time on Sh decreases. At high Re, Sh significantly increases with time, i.e., the corrosion rate (or k) at time 5 h is higher than at time zero (clean surface). At 45oC, slight decrease in mass transfer rate (or Sh) with time occurs at low Re and less or negligible at high Re values.

Also it is found that the formation of corrosion products increases the dependence of Sh on Re for all temperatures. The following correlations are obtained by best fit method for rough surface at t=5 h:

$Sh=0.279\ Re^{0.676}$ C.C=0.88 T=30°C 15000 < Re < 80000   (3a)
$Sh=0.085\ Re^{0.807}$ C.C=0.99 T=45°C 15000 < Re < 88000   (3b)
$Sh=0.073\ Re^{0.828}$ C.C=0.98 T=60°C 20000 < Re < 113000   (3c)

Comparing Eqs.(3) for rough surface (t>0) with that obtained for clean surface (t=0) in previous study [6], i.e,

$Sh=5.588\ Re^{0.434}$ C.C=0.97 T=30°C 15000 < Re < 80000   (4a)
$Sh=0.375\ Re^{0.677}$ C.C=0.99 T=45°C 15000 < Re < 88000   (4b)
$Sh=2.799\ Re^{0.514}$ C.C=0.97 T=60°C 20000 < Re < 113000   (4c)

indicates that the presence of corrosion products appreciably increases the dependence of Sh (or corrosion rate) on Re. At 30oC the increased roughness increases the effect of Re on Sh but decreases the value of Sh at a particular Re. The maximum increase occurs at 60oC where the increased roughness considerably increases the effect of Re on Sh. This increase in the effect of Re on Sh is attributed to the increased turbulence as has been found by other studies concerning momentum, heat, and mass transfer on rough surfaces [8,14,22,25].

It should be noted that this type of roughness (i.e corrosion products) generally decreases the mass transfer rate (except at high Re and temperature) but it increases the effect of Re on mass transfer rate for all temperatures. This is a difference point between this type of roughness and the roughness of a normal metal, where most of heat and mass transfer studies [14,17,18,19,20,21,24,31] showed that the latter increases the heat or mass transfer rates always.
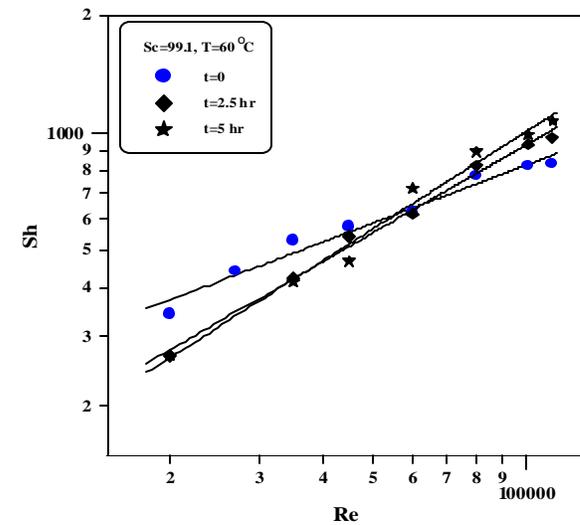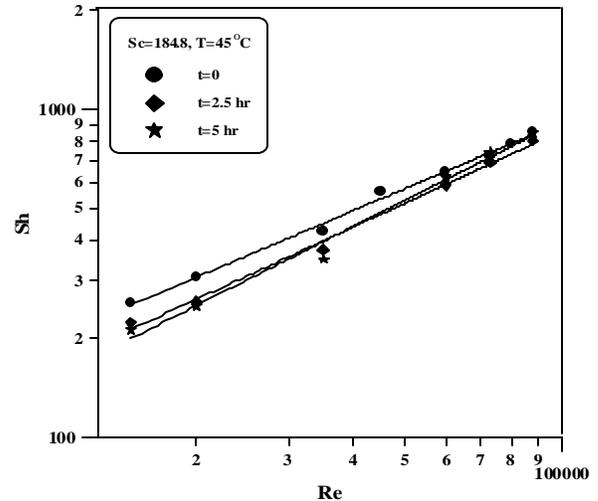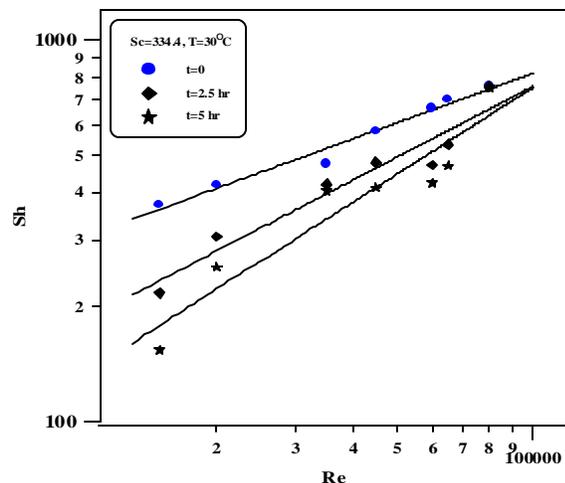
Figs.7: Variation of Sh with Re at Various Intervals

Comparison with Proposed Models

Corrosion products formation affects the capability of correlations based on the analogy concept to estimate the corrosion rate, i.e., the presence of corrosion products increases the difference between the experimental corrosion rate and that obtained from analogy models (many analogy models have been presented in previous paper [6]) due to two causes, first is the change in experimental Sh, and second is the increase in analogy Sh via increasing friction factor.Figs.8 show a comparison between experimental Sh and that obtained from analogies for t=5 h, i.e., in the presence of corrosion products at all temperatures. At 30oC, it is evident from Fig.8a that the time causes some difference between Sh predicted by Von Karmn [33] analogy (Eq. 5) and experimental Sh. This difference increases as Re increases. This is due to two reasons, (i) the decrease of experimental
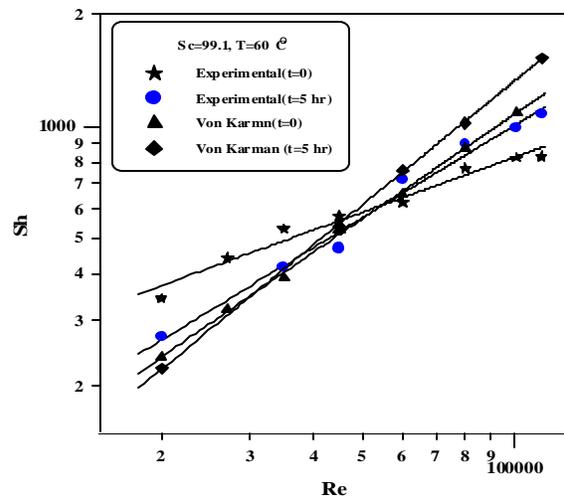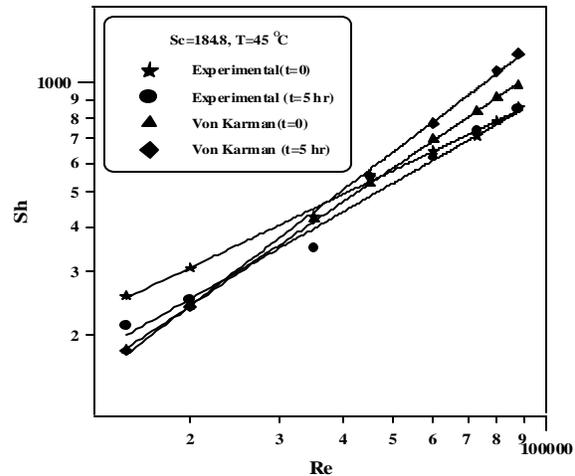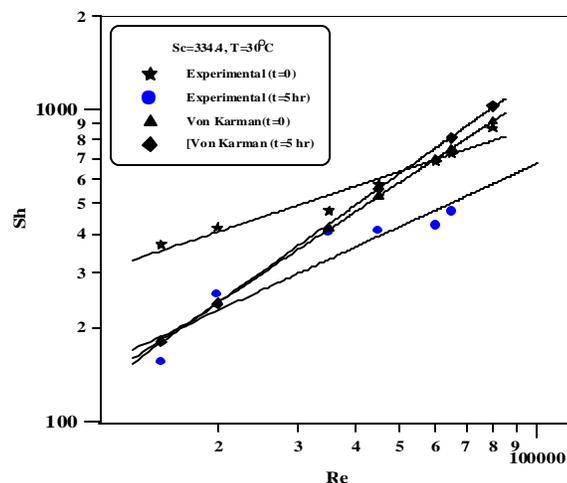
corrosion rate by the formed corrosion products, (ii) the increase in the corrosion rate estimated from analogy correlations due to increasing friction factor (f) via corrosion products. At this temperature the difference reaches 90% at high Re. Accordingly, at 30oC overestimation is expected from the analogy models. At 45oC and 60oC, Fig.8b and Fig.8c reveal that the presence of corrosion products increases the difference between experimental Sh and analogy Sh especially at high Re due to high friction factor increase (f/fo). The difference reaches 40% at high Re. At 60oC, the presence of corrosion products increases the experimental Sh at high Re, thus the difference is mainly due to the increased f. Generally at low Re the difference between analogy predictions and actual corrosion rates is mainly due to the decrease in experimental values induced by the formation of corrosion products, because the increase in f is slight, while at high Re the difference is mainly due to the increase in friction factor, since the effect of corrosion products on actual corrosion rate at high Re is almost low as has been demonstrated.

$$Sh = \frac{(f/2)\,Re\,Sc}{1+5\sqrt{f/2}\left[Sc-1+\ln(1+\frac{5}{6}(Sc-1))\right]} \qquad (5)$$

To reduce the deviation of the corrosion rates estimated using analogy correlations from actual corrosion rates, the analogy correlations should be based on the initial friction factor, fo (t=0). In other word, the change in friction factor is neglected and the analogy correlation is always based on the initial friction factor. By this way it is possible to reduce the increase in analogy results by the following percentage:
error reduction=

$$error\ reduction = \frac{f^{n}-f_{o}^{\,n}}{f_{o}^{\,n}}\times100 = ((\frac{f}{f_o})^{n}-1)\times100 \qquad (6)$$

where n is the dependence of a particular analogy correlation on f . If n=1, when the increase in f is f/fo=1.5, the error reduction is 50%, and so on.

Figs.8: Comparison of Analogy Models and Experimental Results in Presence of Corrosion Products

Conclusions

1. Corrosion products formation with time decreases the corrosion rates at low Re and temperature, while it increases the corrosion rates at high Re and temperature. Also the formation of corrosion products increases the corrosion rate (or mass transfer rate) dependence on Re for all temperatures.

2. Friction factor increases with time due to the formation of corrosion products for all Re and temperatures. The higher the Re is the higher the increase in friction factor with the increased corrosion amount.

3. Overestimation is obtained from analogy correlations with the formation of corrosion products, by increasing friction factor and decreasing experimental k, particularly at low Re and temperature. Basing analogy correlations on the initial friction factor for clean surface reduces this overestimation.

Nomenclature

f	*Friction factor.*
f_o	*Friction factor for clean surface*
i	*Current density, A/m^2.*
i_L	*Limiting current density, A/m^2.*
k	*Mass Transfer Coefficient, m/s.*
Re	*Reynolds number*
Sh	*Sherwood number= kd/D.*
St	*Stanton number= k/u*
u^*	*Friction velocity, m/s.*
μ	*Viscosity, $kg/m.s^2$.*
?	*Kinematic viscosity , m^2/s.*
d_b	*Viscous sub-layer thickness, μm.*
d_d	*Diffusion layer thickness, μm.*
C.C.	*Correlation coefficient*

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