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# Corrosion of Carbon Steel in Oxygen and NaCl Concentration Cells: the Influence of Solution Temperature and Aeration

**Muayad F. Hamad, Huda D. Abdul Kader, Hussein A. Alabdly, Basim O. Hasan, Israa S. M. Ali**

*Department of Chemical Engineering, Al-Nahrain University, Iraq*

## Abstract

Corrosion rate tests were carried out on carbon steel under concentration cells conditions of oxygen and sodium chloride. The effect of aeration in one compartment on the corrosion rate of both coupled metals was determined. In addition, the effects of time and temperatures on the corrosion rate of both coupled metals and galvanic currents between them were investigated. Corrosion potentials for the whole range of operating conditions under concentration cell conditions were also studied. The results showed that under aeration condition, the formation of concentration cell caused a considerable corrosion rate of the Carbon steel specimens coupled in different concentrations of $O_2$ and NaCl due to the galvanic effect. Aerating one compartment caused a noticeable increase in the corrosion rate of the coupled specimen in the other compartment due to the galvanic effect. Increasing temperature caused unstable trends in the free and galvanic corrosion potentials. Increasing the temperature led to an increase in the corrosion rate for both metals.

*Keywords: carbon steel, corrosion rate, concentration cell, galvanic current, temperature, Sodium chloride*

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## 1- Introduction

The oxygen concentration cell is a case in which corrosion cell is established, due to uneven exposure of the metal to air which influences the corrosion behavior of the metal [1]. This case is sometimes called differential aeration. A complete understanding of differential aeration mechanism is difficult due to the effect of complicated phenomena that may occur at the metal-liquid interface such as: passivity, change of surface morphology, change of local pH. Gruyter et al. [2] stated that for several decades the concepts of differential aeration have not been really proved by experimental works and these may fail in some occasions.

They demonstrated that the differential aeration is not necessarily causing an increased corrosion of the oxygen-starved metal. Additionally, the formation of corrosion product, local fluid motion, the electrochemical potential of the metal can be altered by electrochemical transport through operation of concentration cells [3]. This type of corrosion occurs normally when the metal surface is exposed to two environments of different oxygen concentration such us underground corrosion of pipes, tanks, and other structures. In this case the corrosion is slow but destructive. The common terms for this type of corrosion include crevice corrosion, oxygen screening, and poultice action; the difference in oxygen concentration produces a potential difference and thus causes a current flow.

Anodic and cathodic areas are established on the metal surface forming a corrosion cell. The severity of corrosion is dependent on the different prevailing conditions such as the presence of salt, the temperature, and the moisture or solution nature.

The substantial difference of corrosion rate between anode and cathode expresses an acceleration of corrosion due to the oxygen concentration difference. Typical examples of the corrosion due to the $O_2$ concentration cell are the water-line corrosion (it is observed in steel water tanks partially filled with water) and pitting corrosion ( observed when dust particles or oil drops deposited over the metal surface)) [4], [5].

LaQue [6], [7] found that that the corrosion proceeded along the periphery for copper disk where the transport of oxygen was higher. Conversely, the iron disks spinning under same conditions corroded at the center where there is a decelerated transport of oxygen. The active-passive transition can also appear depending on the electrode area, oxygen amount, and solution electrical conductivity [8], [9].

Oxygen concentration cells can also causes crevice corrosion in some metals such nickel, aluminum, and stainless steels, and other passive metals that are exposed to aqueous environments such as seawater [3]. The aim of the present work is to study the corrosion behavior of carbon steel under the influence of oxygen concentration cells in NaCl solution at different temperatures.

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Corresponding Authors: Name: Basim O. Hasan , Email: basimohasan13@gmail.com

## 2- Experimental Work

Fig. **1** shows a schematic diagram of the experimental set up. It is composed of water bath, Zero-Resistance Ammeter (ZRA), two beakers containing the corrosive NaCl solutions, air pump of a flow rate of 2.5 l/ min, specimens, salt bridge to ensure electrical connect of the two compartments. The apparatus contained also Saturated Calomel Electrode (SCE) and voltmeter to measure the potentials of the specimens under corrosion conditions and digital balance.

Carbon steel coupons of dimensions of 40 × 40 × 0.2 mm were used for corrosion tests. They were polished with 400 and 600 grid emery papers, washed with distilled water, and then rinsed with ethanol for 5 minutes. After that the specimens were allowed to dry and placed in oven at 80 $^oC$ for five minutes for complete drying [10], [11].

Then the specimens were placed in a vacuum desiccator over a high activity silica gel until use. After preparing the NaCl solution, the specimens were exposed to the corrosion environment for 2.5 h under specified operating conditions. The experiments included two parts: free corrosion and galvanic corrosion. In free corrosion tests, the weight loss of specimens under different operating conditions of temperature and aeration was determined as well as the potential variation with time. After each test, the specimens were cleaned carefully by water with brushing by plastic brush to remove the corrosion products. Then, the specimens were placed in 5% HCl containing organic inhibitor (Hexamine) to ensure the removal of the corrosion product from the specimen surface, rinsed with water, dried by oven at 80 $^oC$ for 5 min, and kept in a disscator to cool then weighed. The corrosion rate was determined as:

$$\text{corrosion rate (CR)} = \frac{W_1 - W_2}{A * t} \qquad (1)$$

Where, $W_1$= initial weight of the specimen, $W_2$= final weight of the specimen, A= surface area of the coupons, t= time of the experiment.

In concentration cell galvanic corrosion tests, two types of concentration cells were investigated. First type is an oxygen concentration cell in which air is pumped into a compartment of 0.1N NaCl solution containing a CS specimen connected to another compartment containing 0.1N NaCl solution with no aeration.

The metal under aeration conditions ($M_{1A}$) was connected to the negative terminal of the ZRA. Due to air pumping a differential aeration cell was established in which a potential difference causes a galvanic current to flow between the two poles. Zero resistance ammeter (ZRA) was used to measure the galvanic current under different operating conditions. At the same time the weight loss of each specimen in the galvanic couple was determined to calculate the corrosion rate.

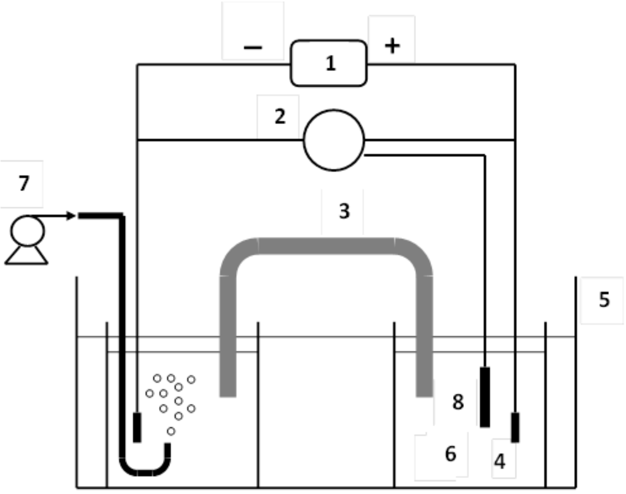

Fig. 1. Schematic diagram of the experimental apparatus, 1. Zero Resistance Ammeter, 2.Voltmeter, 3.Salt bridge, 4.Carbon steel specimens, 5.Water bath, 6.Beakers, 7. Air pump, 8. Calomel electrode

The second type concentration cell investigated was a salt concentration cell. In these experiments, a galvanic cell was established between two compartments of different salt concentration under different temperatures with and without aeration. The galvanic current, galvanic potential and weight loss of each specimen in the couple were determined. Each experiment was carried at least twice.

## 3- Results and Discussion

### 3.1. Oxygen Concentration Cell

#### a. Corrosion Potential

Fig. **2** to Fig. **5** show the electrode potential vs. time at different temperatures: 25, 35, 45, and 55 $^oC$ in the presence and absence of aeration for free corrosion and galvanic coupling in 0.1N NaCl solution. In these figures the “free” means no coupling between the two terminal and “galvanic” means the two concentration cell terminals are coupled. It is evident that the aeration condition has a noticeable effect on the free corrosion potential and galvanic corrosion potential. The aeration leads to an increase in $O_2$ concentration shifting the potential to more positive [10], [12], [13]. This is a general trend, but in some occasions the potential with aeration is seen to be more negative than without aeration. This can be attributed to the formation of corrosion product layer which represent a thermal resistance to the current flow leading to increase the resistance polarization which shifts the potential to more negative as has been evidenced by some previous studies [14], [21].

The figures indicate that the cell potential becomes more negative with time due to the passivation of metal surface by the formation of corrosion product film which leads to a decrease in the surface activity [10], [14].

Besides, the formation of corrosion product layer with time prevents the arrival of $O_2$ to the surface which causes a decrease in the potential [14], [15]. The formation of insoluble corrosion products creates a shielding effect [16], [17] and also passivates the metal thermodynamically [18].

It can be seen from Fig. **2** to Fig. **5** that under galvanic coupling, pumping of the air into one compartment leads to increase the potential difference between the two poles of the galvanic cell. This increase in the potential difference enhances the galvanic corrosion attack of the more active pole.

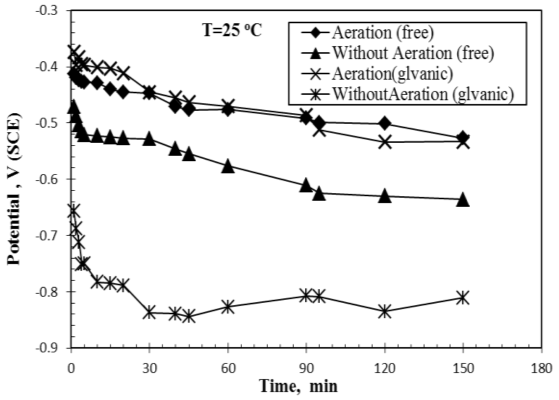

Fig. 2. Free corrosion and galvanic corrosion potentials of CS in the presence and absence of differential aeration in 0.1 N NaCl solutions at 25 °C

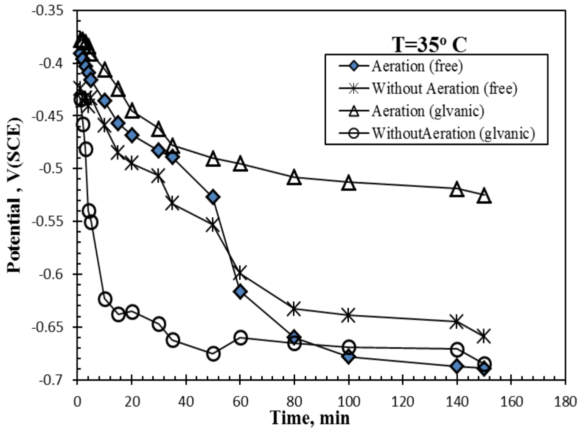

Fig. 3. Free corrosion and galvanic corrosion potentials of CS in presence and absence of differential aeration in 0.1 N NaCl solutions at 35 °C

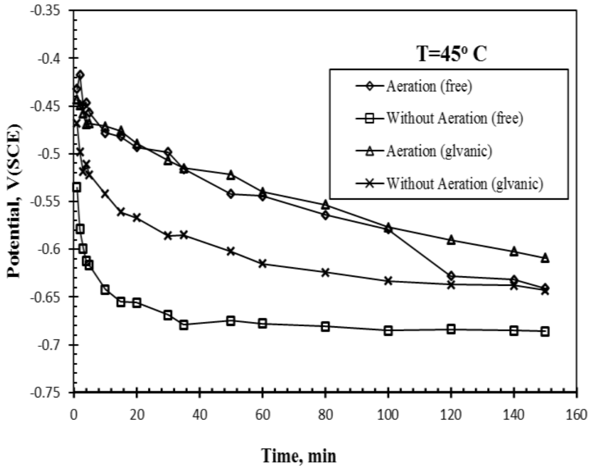

Fig. 4. Free corrosion and galvanic corrosion potentials of CS in presence and absence of differential aeration in 0.1 N NaCl solutions at 45 °C

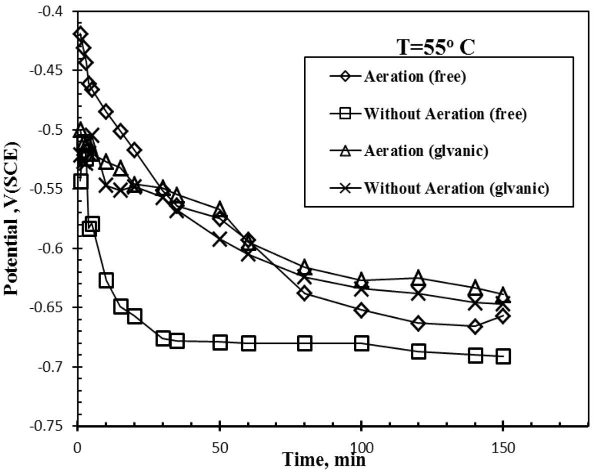

Fig. 5. Free corrosion and galvanic corrosion potentials of CS in presence and absence of differential aeration in 0.1 N NaCl solutions at 55 °C

Fig. **6** shows the free corrosion potential of CS under aeration conditions for different temperatures for non-aerated metal.

Fig. **7** shows the variation of free corrosion potential with time for 0.1N NaCl solution at different temperatures with aeration condition. It indicates a considerable more negative in potential with the temperature.

This indicates that the high concentration of $O_2$ leads to a decrease in the effect of temperature on the potential. The figures also indicate that the temperature of 55 °C has no effect.

The high temperature deteriorates the protective properties of the passive film which results in an increase of the passive current density and their destruction at smaller corrosion rate [19].

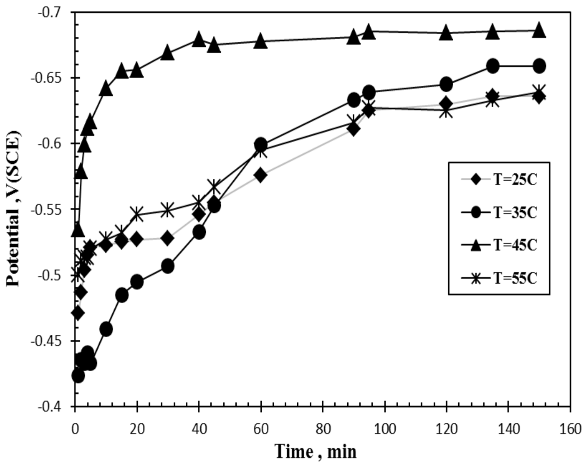

Fig. 6. Potential of free corrosion for different temperatures and without aeration condition in 0.1N NaCl

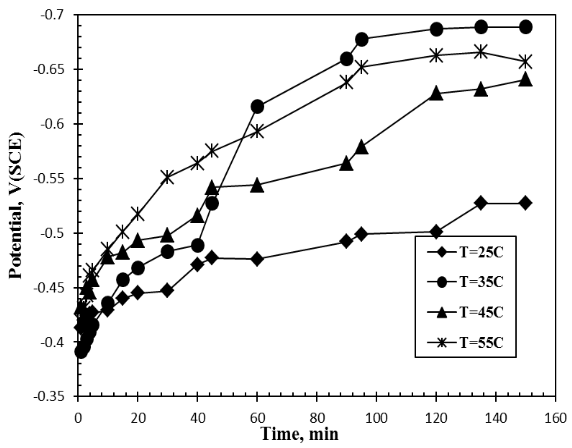

Fig. 7. Potential of free corrosion for different temperatures and aeration condition in 0.1N NaCl

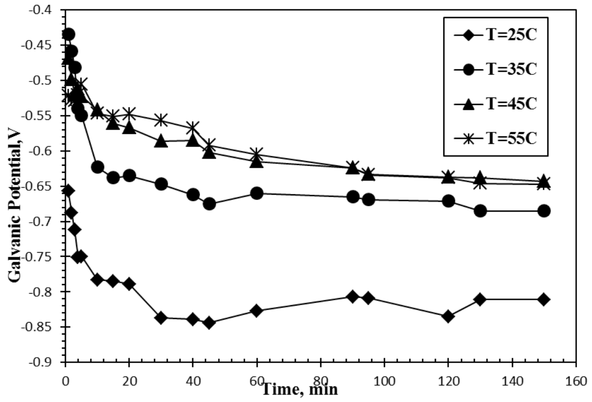

Fig. 8. Galvanic Potential for different temperatures and without aeration condition in 0.1 N NaCl

Fig. **8** shows the potential versus time of CS specimen without aeration (M2) connected to CS under aeration (M1A) 0.1N NaCl solution at different temperatures. It is clear that the galvanic potential shifts to more positive with increasing temperature.

The temperature rise leads to a decrease in the dissolved oxygen concentration which leads to shifting the galvanic potential to more negative.

Fig. **9** shows the effect of temperature on the variation of potential of aerated pole. The same trend of potential was observed.

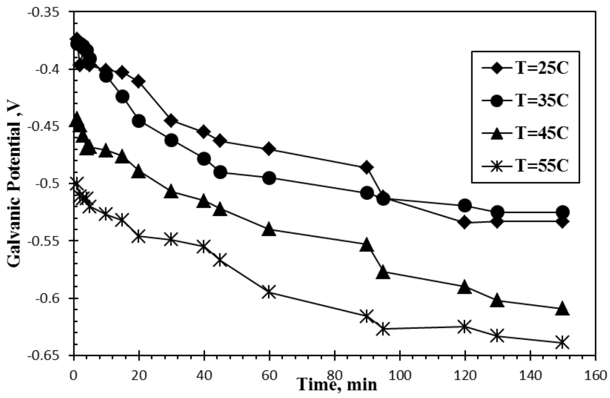

Fig. 9. Galvanic Potential at different temperatures and aeration condition in 0.1N NaCl

b. Corrosion Rates

Fig. **10** shows the corrosion rate of CS metal versus temperature at different conditions. It can be seen that the corrosion rate increases with increasing temperature for both free and galvanic corrosion with and without aeration.

The CR in the presence of air pumping (aeration) is much higher than the case of without aeration.

This is ascribed to the increased oxygen concentration in the solution which causes severe attack to the metal.

It can be seen that the galvanic coupling causes an increase in the CR for the metals in the non-aerated pole.

This is interpreted as follows: the aeration of one pole of the couple leads to increase the concentration of $O_2$ at this pole.

This causes an increase in the cathodic reduction of oxygen at this metal which leads to increase the charge transfer through cell leading to an increased anodic reaction at the other pole. This means an increased corrosion rate of the other pole.

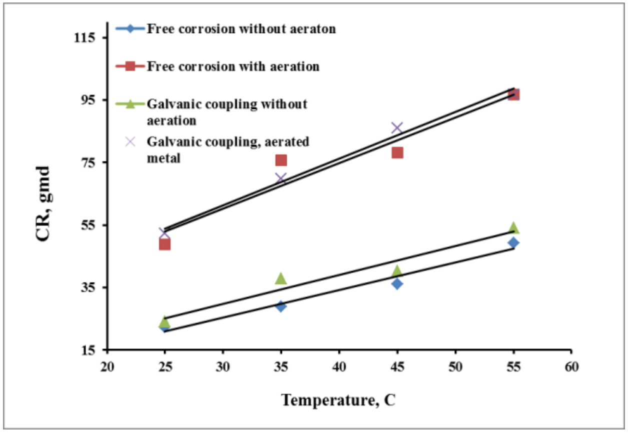

Fig. 10. Corrosion Rates of Carbon Steel under different conditions of 0.1N NaCl

Fig. **11** shows the variation of galvanic current ($I_g$) with time at different temperatures. The figure reveals that the galvanic current of differential aeration cell increases with time reaching to steady asymptotic value after some 20 min. For 35 $^oC$ and 45 $^oC$, there is a slight decreasing Ig after first few minutes. This decrease is attributed to metal passivation due to the formation of a protective film which grows with time [20]. Fig. **11** also shows that the effect of temperature on Ig is not systematic. Maximum current occurs at 35$^o$C followed by 25 $^o$C, 55$^o$C and 45$^o$C. The effect of temperature on the galvanic current and on the corrosion rate is complicated by affecting two important parameters working in a conflicting way: the oxygen diffusivity and solubility. For the current system during the galvanic attack there is four reactions occurring simultaneously on the two metals of galvanic cell. These reactions are $O_2$ reduction reaction on each metal (M1 and M2) which is cathodic reaction:

$$O_2 + H_2O + 4e \rightarrow 4(OH^-) \qquad (2)$$

and anodic reaction of each metal:

$$Fe \rightarrow Fe^{+2} + 2e \qquad (3)$$

It can also be noticed that for 45 $^o$C and 55 $^o$C the galvanic current is negative often. This is due to the polarity reverse at these two temperatures. At 25 $^o$C, the solubility of $O_2$ in the solution is high about (7 ppm) [21].

With increasing temperature the solubility of $O_2$ decreases and thus the concentration of $O_2$ in the solution decrease leading to a decrease in the cathodic currents of $O_2$ reduction reaction.

The sharp decrease in the $O_2$ concentration causes a change in the polarity leading to change the change the electrode behaviors from cathode to anode and vice versa. The same trend was observed by [22], [23].

Since the aerated chamber is connected to the positive terminal of ZRA, the positive current means that the electrons flow from the aerated pole (M1A) to the non-aerated (M2).

When increasing the temperature and with the reduction in $O_2$ concentration, the pumping of air may cause passivation to the aerated pole. In this case, the aerated metal is thought to be passivated and thus become cathode.

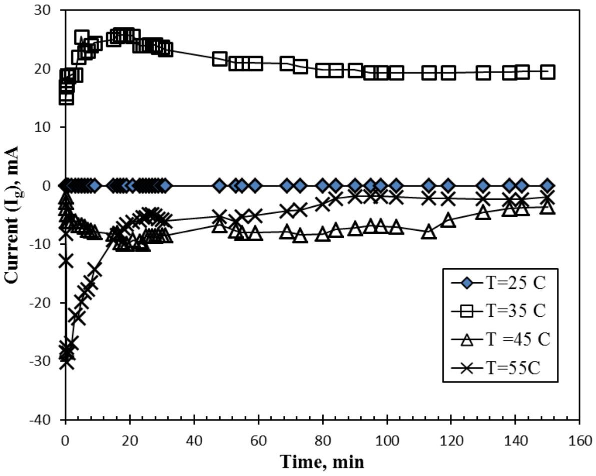

Fig. 11. Galvanic Current of 0.1 N NaCl solutions, first spacemen under aeration Condition is coupled to a spacemen without aeration condition

### 3.2. Salt Concentration Cells

The effect of concentration of NaCl on the potential with time at 25$^o$C is presented in Fig. **12**.

The figure indicates that the effect of salt concentration in free condition is higher than that in coupling condition; the corrosion potential is shifted towards more negative values with increasing sodium chloride concentrations.

It is evident that the free corrosion potential and galvanic potential decrease with time. In addition, the free corrosion potential is higher than the galvanic potential. This agrees with previous workers [23], [24].

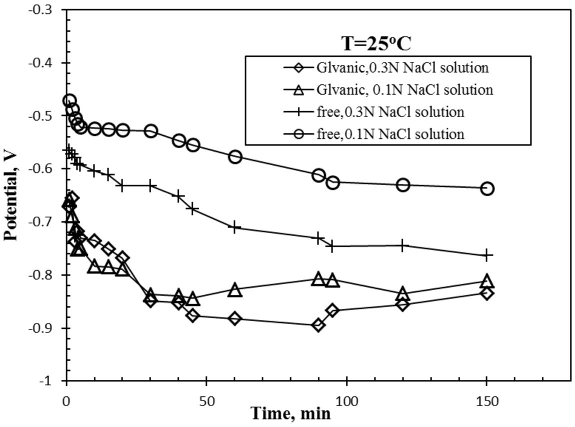

Fig. 12. Galvanic and free corrosion for different concentration of NaCl in without aeration condition

Fig. **13** shows the galvanic current versus time in 0.1N coupled to 0.3N NaCl solutions at different temperatures for the case of non- aeration. It indicates that the value of galvanic current at T=25$^o$C is greater than the value of the galvanic current at T=45$^o$C. This is due to the higher concentration of $O_2$ at 45$^o$C than at 55$^o$C. This leads to increase the $O_2$ cathodic reduction reactions and thus Ig increases.

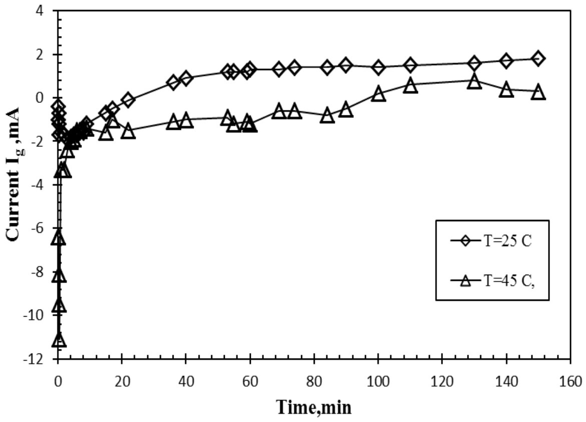

Fig. 13. Galvanic current of 0.1 N coupled to 0.3 N NaCl solutions at 25 and 45$^o$C without aeration condition

Table **1** indicates that the values of corrosion rate for aeration conditions are greater than the corrosion rate in case of without aeration since the amount of oxygen arriving to the surface is higher.

In addition, for galvanic coupling (concentration cell), at 25 $^o$C the corrosion rate for the aerated pole ($M_1$ in 0.1N NaCl) is lower than that of aerated pole (M2). This can be interpreted as follows: the aeration of M1 causes a shift in the potential of M1 to more positive which also increases the potential of M2 because M2 is galvanically connected to M1. Since the solution at M2 is of higher electrical conductivity due to the higher salts concentration, the corrosion rate becomes high for $M_2$.

Table 1. Corrosion rate in (gmd) for 0.1 and 0.3N NaCl solution

| Temperature, $^o$C | Galvanic Corrosion | | | |
|---|---|---|---|---|
| | Aeration | | Without Aeration | |
| | 0.1N (M1) | 0.3 N (M2) | 0.1N (M1) | 0.3N (M2) |
| 25 | 55.8 | 63.7 | 28.2 | 20.4 |

Fig. **14** shows the variation of corrosion rate with temperature for free and galvanic condition. The figure reveals that the increase in temperature causes an increase in the rate of corrosion in free corrosion conditions. This is in agreement with previous works [25], [26]. When the temperature increases, the solution viscosity decreases leading to an increase in the oxygen diffusion to the metal surface which enhances the corrosion.

However, the increased temperature causes a decrease in the $O_2$ solubility due to the escape of $O_2$ to the atmosphere the factor that decreases the corrosion rate [10], [27].

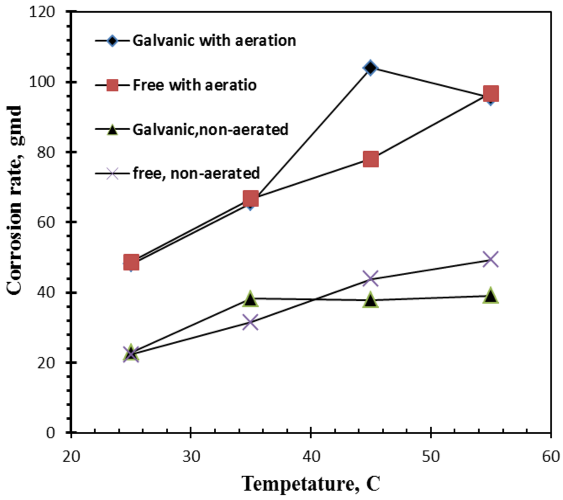

Fig. 14. Corrosion rate versus temperature for aeration and non-aeration conditions in 0.1N NaCl solution

Fig. **15** presents the galvanic current $I_g$ with time for aeration and without aeration conditions when coupling CS specimen 1 (M1) (0.1N NaCl solution) and specimen 2 (M2) (0.3N NaCl solution). It shows that the absolute value of the current in the case of aeration condition is greater than the case of without aeration condition.

The negative values of the current indicate that the current (the electrons) is flowing from the specimen connected to the +ve terminal of the ammeter to the other specimen connected to –ve terminal.

That means the current is flowing from 0.1N NaCl solution to the 0.3N NaCl solution. This is due to the shift of the potential of the aerated specimen to more positive with increases the driving force for galvanic corrosion of non- aerated metal and at the same time the corrosion rate of the aerated metal is also increased. Before coupling, the CR of aerated specimen in 0.3N NaCl solution was 61.8 gmd and without aeration was 47.2 gmd. When coupling the two specimens with one in aerated 0.3 N NaCl solution and the other in non -aerated 0.1N NaCl solution, the aeration of 0.3N NaCl solution increases the galvanic attack of the specimen in 0.1N NaCl solution and increases the corrosion of the specimen in 0.3N NaCl solution as shown in Table **1** for 25 $^o$C. The increased current with the aeration is due to several reactions occurring simultaneously on the galvanically coupled specimens.

These reactions are: carbon steel dissolution in both solution (anodic reactions) and oxygen reduction reactions on both metals (cathodic reactions). At 25 $^o$C, when coupling both specimens in 0.1N and 0.3N NaCl solution, the specimen in 0.1N solution is more anodic because its CR is higher as shown in Table **1**.

This is because the anodic dissolution of specimen 1 is high and thus, the cathodic reduction reaction of $O_2$ on specimen 2 is high. So, specimen 1 is anode and specimen 2 is cathode. When aerating specimen 2, the anodic dissolution of specimen 2 increases and it becomes anode and thus its CR is higher as indicated in Table **1**.

Under the aeration conditions the current became rapidly more negative in the minutes, and then the curve converged to slower rate, that is because of the formation of the $OH^-$ ions in a high rate and grouping on the electrodes [28].

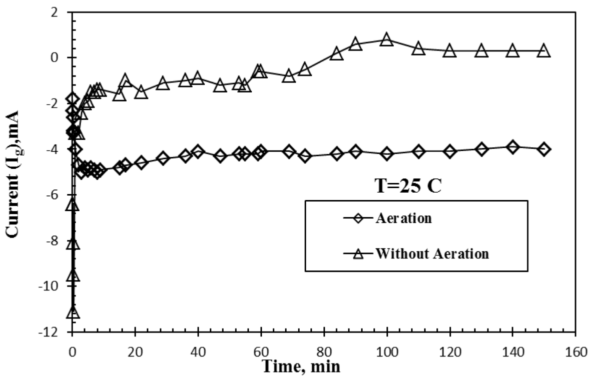

Fig. 15. Galvanic current of Coupling 0.1 N and 0.3N NaCl solutions without aeration and with 0.3N NaCl solution aerated

## 4- Conclusions

The following conclusions are drawn from the experimental results:

1- For oxygen concentration cell, the cell potential for both poles of the cell decreases with time and increases when the oxygen concentration is increased by aeration.
2- The aeration of one-pole of the couple leads to increase the CR in the non-aerated pole.
3- The free corrosion potential and the potentials of both poles were found to be decreased (becomes more negative) with temperature increase
4- The galvanic current for the oxygen cell increases with time reaching to steady asymptotic value after about 20 min.
5- The absolute value of galvanic current (Ig) at the different concentrations of NaCl in two compartments of the concentration cell under aeration condition is much higher than the non-aerated condition.

## Nomenclature

| Symbols | Description | Unit |
|---|---|---|
| A | Surface area of specimen | $m^2$ |
| CR | Corrosion rate | $g/m^2$.day |
| t | time of the experiment | min |
| $W_1$ | initial weight of the specimen | g |
| $W_2$ | Final weight of the specimen | g |
| $I_g$ | Galvanic current | Ampere |

## Abbreviations

| | |
|---|---|
| CS | Carbon steel |
| gmd | Gram per square meter per day |
| SCE | Standard Calomel Electrode |
| ZRA | Zero Resistance Ammeter |

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# تاكل الفولاذ الكاربوني في خلايا تركيز الاوكسجين وكلوريد الصوديوم :تاثير درجة حرارة المحلول والتهوية

**مؤيد حمد, هدى عبدالقادر, حسين العبدلي, باسم عبيد و اسراء علي**

*قسم الهندسة الكيمياوية, جامعة النهرين*

## الخلاصة

اجريت عملية قياس معدل التاكل للحديد الفولاذي تحت ظروف خلايا التركيز للاوكسجين وملح كلوريد الصوديوم. وتم دراسة تاثير عامل التهوية لاحد اجزاء الخلية على معدل التاكل لكلا المعدنين المربوطين بالاضافة الى ذلك تم دراسة تاثير الوقت ودرجات الحرارة على معدل التاكل لكلا المعدنين المربوطين وايضا تم قياس التيارات الكلفانية بينهما .تم قياس ودراسة جهود التاكل للمدى الكلي للظروف التشغيلية لظروف خلية التركيز.اظهرت النتائج انه في حالة استخدام التهوية فان تكُون خلية التركيز يسبب زيادة معدل التاكل للعينات المربوطة وفي تراكيز مختلفة للاوكسجين وملح كلوريد الصوديوم بسبب التاثير الكلفاني. استخدام التهوية لاحد اجزاء الخلية يسبب زيادة ملحوظة في معدل التاكل للعينة المربوطة في الطرف الاخر من الخلية نتيجة التاثير الكلفاني. زيادة درجة الحرارة يؤدي الى اتجاه غير مستقر في جهود التاكل الكلفانية والحرة وان زيادة درجة الحرارة يؤدي الى زيادة معدل التاكل الكلي للمعدنين .

الكلمات الدالة: الفولاذ الكاربوني، معدل التآكل، خلية التركيز، التيار الكلفاني، درجة الحرارة، كلوريد