

Chloride Ions Penetration and their Role in Sediments Corrosion of Buried Steel Structures in Closed Basin

El-Shamy AM^{1*}, Shehata MF¹, Metwally HIM² and Melegy A²

¹Physical Chemistry Department, Electrochemistry and Corrosion Lab, National Research Centre, Dokki, Giza, Egypt

²Geological Sciences Department, National Research Centre, Dokki, Giza, Egypt

Abstract

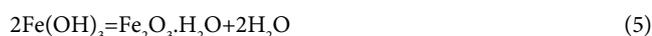
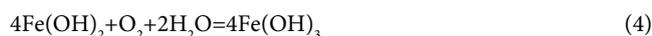
Buried pipelines and steel structures passes or existed through different kind of sediments and saline water. This paper aims at optimize the best conditions to protect the buried pipelines from corrosive environments. Two ponds are selected for this study because of the big difference in chloride contents. The ponds were subjected to chemical, mineralogical, electrochemical analysis and the moisture content is also considered in this study. The results proved that mild steel is probably used in pond 1 at moisture content of 150% but in case of pond 4 mild steel is totally not recommended in all moisture content.

Keywords: Lake Quaroun; Chloride ions penetration; Mild steel; Buried pipelines; Corrosion rate

Introduction

Corrosion overview

The buried pipelines and steel structures of mild steel are affected by many factors which cause deterioration and equipment failure [1,2]. The corrosion phenomenon occurs in all environments and the form of corrosion is depending on six essential factors. The most famous forms of corrosion are general, localized and intergranular corrosion which occurs along the grain boundaries. The steel pipelines in soil could be suffered from all these kinds of corrosion depending on the soil constituents. The general corrosion is already existed in presence of humidity in the soil but the localized attack is affected by presence of chloride and or presence of sulfide ions. The corrosion reaction could be summarized in the following reactions:



It is well known that one of the most important causes of underground corrosion is the presence of anaerobic sulfate-reducing bacteria [3-6]. This kind of bacteria is characterized by splitting acidic metabolites which deteriorate the steel surface through localized attack by sulfide ions. The microbial corrosion is very harmful for the steel structures in both the internal and external surfaces. The internal pitting corrosion comes from the formation of biofilm and the pitting is existed underneath the formed biofilm. To prevent the internal microbially influenced corrosion, non-oxidizing biocides are considered to kill sulfate reducing bacteria [7]. Corrosion inhibitors are also used in parallel addition to protect the steel pipelines from the internal corrosion against severe environment [8,9]. In case of external corrosion the steel pipelines is generally controlled by coating. The cathodic protection is preferred to overcome the defects which could be existed in the coating process, which involves the use of a sacrificial anode such as zinc or aluminum [10,11]. The underground corrosion is affected by presence of chloride and sulfide ions which is concerned with the pitting corrosion. In some cases the oxidation of sulfide ions is used to get rid the sulfide attach on the steel surfaces [12,13].

Geological overview

Lake Quaroun is an inland closed basin located in the western desert between Longitude 30° 20' and 30° 50' E and Latitudes 29° 24' and 29° 31' N, some 80 km southwest of Cairo (Figure 1). Its maximum length is approximately 45 km and the maximum width reaches approximately 9 km with a maximum depth of 9 m, covering an area of 240 km². The lake occupies the deepest part of the Fayium Depression, at 45 m below sea level (b.s.l). Cultivated lands surround the lake from south with steep inclination towards the lake. The slope is estimated to 94 meters per 60 kilometers i.e., 1.57 m/km. Such a steep inclination renders the depression the collective character of the cultivated land wastewater in the Fayium Province [14]. The water level of Lake Quaroun lies below mean sea level; fixation of its level due to irrigation purposes is controlled by the inflow of drainage water subtracted from the calculated evaporation rate. The average water depth in the Lake is about 4 to 7 meters. More than 67% of the Lake area has average depth of 5 to 8 meters. The deepest regions lie to the west of El-Qarn Island (Figure 1).

Location and general characteristics of the studied sediments

In Figure 1 shows the small extrapolation of pond 1, approximately (105,000 m²) in area. This pond provides a conduit for brine from pond 1 to pond 2. This modification entailed no increase in area to ensure a suitable concentration for factory operations. The following is a description of the size and function of each pond. The concentration ponds are four in number, in addition to extrapolation smaller one known as pond 4 used as to complete the salinity calculations needed for mass balance calculations. Description of the functions, dimensions, levels and concentration of each pond are descript in Table 1.

***Corresponding author:** El-Shamy AM, Physical Chemistry Department, Electrochemistry and Corrosion Lab, National Research Centre, El-Bohouth St. 33, Dokki, PO 12622, Giza, Egypt, E-mail: elshamy10@yahoo.com

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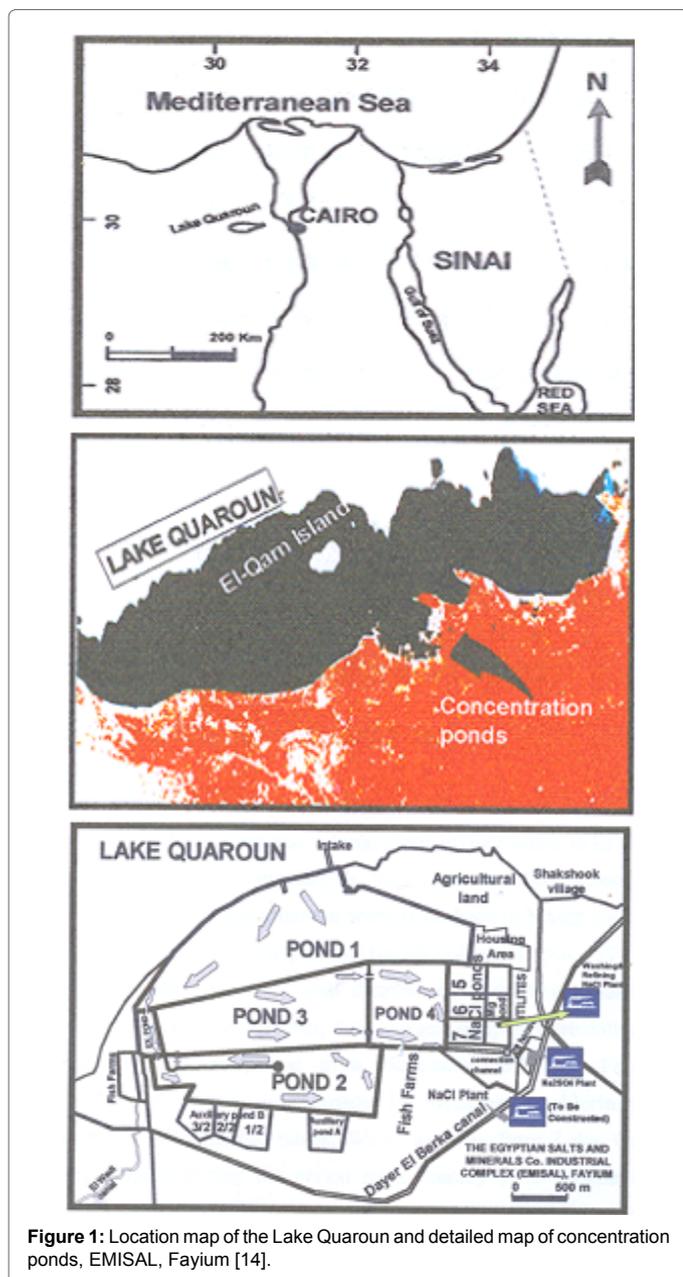


Figure 1: Location map of the Lake Quaroun and detailed map of concentration ponds, EMISAL, Fayium [14].

According to the types of extracted salts, the suitable way for concentration of the brine depends on the solar energy (solar evaporation) which considered as a cheaper method to concentrate salt, four concentration ponds were constructed over an area of about 1200 acres, i.e., about 5 million m². Separating bridges were built with a length of 3 km separating the ponds from Lake Qaroun. The total length of the concentration ponds is about 22 km. the fourth pond is the nearest to the factory, connected through a concentration channel terminates with pumping pit to transfer the brine to the factory. Controlling the flow dynamics of the brines is through the constructed gates between ponds. There are different controlling factors to evaporate brine up to 360 g/l ± 15 g/l, these are:

1. Evaporation rate (i.e., as evaporation rate increases the concentration of salts increase and vice versa).
2. Humidity.

3. Temperature (i.e., on its increase, the concentration of salts increase and vice versa).
4. Wind velocity and direction leading to evapo-transpiration of water molecules.
5. Flow dynamics which signifies the type and speed of brine versus time required concentrating it by solar evaporation.

The pond 1 is relatively the largest pond in area, occupying about 500 acres (nearly 2.1 million m²). Its level is at -44.2 m below sea level (b.s.l). It is adjacent to Lake Quaroun, which receives its water through pumping station and concentrates it to nearly the double of the lake water concentration (i.e., from 36 g/l up to about 70 g/l). The concentration range of this pond helps to eliminate impurities (suspended materials) derived to the pond from the lake water. Its main function is to settle the suspended materials (sand+clay).

The pond 2 occupies about 300 acres (about 1.3 million m²). Its level is at -44.34 m (b.s.l). The following brine from the first pond enters to the second one passing through the extrapolation pond 1 at a concentration less than 80 g/l becoming around 130 g/l at the end of second pond due to the effect of solar evaporation. Through its concentration range, precipitation of carbonates is the main function of this pond.

The pond 3 occupies about 250 acres (about 1403000 m²). Its level is balanced at -45 m (b.s.l). The brine enters this pond from the gate of P 2 with a concentration of about 130 ± 20 g/l. The large of P 3 offers the brine to concentrate up to 250 g/l (max.) through the concentration range of P 3 (from 130 to 250 g/l), calcium sulfate (gypsum) will precipitate in the pond itself.

The pond 4 occupies about 150 acres (about 600000 m²). Its level is at -45.30 m (b.s.l). The brine enters this pond with concentration of about 250 g/l and it might increases to about 360 g/l at the outlet gate of P 4. The brine becomes suitable for industrial extraction of anhydrous sodium sulfate (thenardite). The brine enters the factory through a connection channel (500 m in length) ends with a pumping station transporting the brine to the factory. The flow dynamics of the brines is controlled through the gates constructed between ponds. The shallowest region lies at the eastern part of the lake and it is subjected to the influence of land drainage carrying considerable amounts of mud, which settle down near the mouth of the drains [15]. However, the amount of mud varies with the flow of water and reflects the variability in the water depth near drain mouths. Salinity (Total Dissolved Salts gram/liter) of Lake Quaroun water has increased with time; it was fresh water about 150 years ago. The annual supply of the water drainage during the last 10 years was insufficient to contaminate the lake water. During the decade, the lake received a greater amount of agriculture

Ponds	Area (m ²)	Max height brine (cm)	Bottom level (m)	Function
Pond (1)	2210000	160	-44.20	Concentration from 36 g/l to 80 g/l Deposition of suspended materials and carbonates
Pond (2)	1052050	164	-44.34	Max. Conc. from 130-160 g/l Deposition of the rest of ALL Carbonates and start of deposition of calcium sulfates
Pond (3)	1403000	230	-45.00	Max. Conc. From 240-260 g/l Calcium sulfate precipitation
Pond (4)	633000	260	-45.30	Max. Conc. 370 g/l Decrease concentration to eliminate exists of any precipitates.

Table 1: The different areas, levels and ponds function.

wastewater, hence its salinity reached 34.9 g/l (April 1999). The salinity in (May 2000) reached 34.9 g/l. The estimated increase in salinity during the last decade is 0.35% to 0.38%/year. The main reason of adding treated wastewater is to maintain lake level at -43 m (b.s.l) and to control salinity. The two main problems are facing Lake Quaroun at present time, they are:

1. The continuous increase in salinity with time.
2. Maintaining a constant level of lake water.

This paper aiming at studying the effect of chloride ions concentration on the corrosion behavior of mild steel and pond 1 and pond 4 are selected for study because of the big difference in chloride content.

In this study the pond 1 and pond 4 are selected for evaluation the chloride ion effect on the mild steel surface. It is clear that the salinity of Lake Quaroun increases with time according to feasibility study carried out by ref. [15]. It is also reported that the salt content in the lake is increasing with time as the drain water contributes annually to the lake a certain amount of dissolved salts. Nearly a volume equal to the drainage water is lost annually through evaporation from the lake while its dissolved salts were leftover in the lake. The average of increasing salinity is about 0.5 g/l annually, where according to ref. [16] the salinity of 40 g/l will be reached in the year (2010). Recent researches added a new scope on the effect of the added wastewaters, where it reveals that toxic heavy metals are present at the bottom sediments [17]. It is obvious that the salinity of Lake Quaroun was raised annually leading to excess in salt concentration over the original salinity of the lake. Precipitations of about 350,000 tons of salts annually were added to the original stored salts in the lake. If salts are not extracted from the lake, the salinity will continue rising annually until it turns to a dead lake with no biota except for certain species living in very high saline water. This will lead to complicate social problems; hence hundreds of people will face socioeconomic problems due to their long lasting activities as fishermen. To-rebalance the environmental changes and hazardous, feasibility study was carried out to extract the salts from the lake. The first studies were done by ref. [18] had led to the foundation of the Egyptian Salts and Mineral Co. (EMISAL) as an industrial complex of three stages to extract firstly sodium sulfate, secondly sodium chloride and magnesium salts as a final stage. This was accomplished through applying solar energy to concentrate the lake water into flowing brine in concentration ponds. The ponds are in consequent manner, where the path flow was starting with 35 g/l to reach 350 g/l at the out let of pond No. 4 directly to the plant to extract sodium sulfate. The four concentrated ponds resemble a semi-closed basin, where water and /or brine are controlled in consequent manner. The salinity of each pond will lead to precipitate the designed salts according to saturation level and solubility products.

Materials and Methods

Materials

Electrodes: Mild steel is used as working electrode with rod shape was subjected for XRF elemental analysis. Table 1 shows the results of elemental analysis and the electrode is stacked in a Teflon holder with exposed area of 0.785 cm². Platinum wire and Cu/CuSO₄ electrodes were used as counter and reference electrodes, respectively. The analysis of the mild steel sample is listed in Table 2.

Methods

Electrochemical measurements: The electrode surface was abraded successively by emery papers of different grades, i.e., 150, 320,

400 and 600 and finely polished with a 2400 polishing paper to obtain uniform mirror like finish, degreased with acetone and washed with distilled water before experiment. The open circuit potential of mild steel electrode was subjected for 30 min in the electrolyte (sediments) in free corrosion conditions before recording the polarization curves. Potentiodynamic polarization curves were measured on the mild steel electrode at different moisture content.

Results and Discussion

Mineral analysis of sediments

XRF measurements of the sediments under study: The selected pond 1 and pond 4 are subjected to chemical analysis and the results are presented in Table 3. The XRF data show that the chloride ion concentration of pond 4 is higher than that of recorded for pond 1 of approximately the double value.

Electrochemical measurements

Open circuit potential: In Figure 2a shows the curves of the open circuit potentials versus time for the mild steel electrode in sediment of pond 1 at different moisture contents starting with zero water content and water contents 10, 20, 30 and 40% for 30 min. The open circuit potential (OCP) variation with time can be measured against Cu/CuSO₄ by determining the voltage difference between immersed mild steel surfaces in sediment of pond 1 at different moisture contents.

It is clearly noticed that the highest value of OCP is obtained at zero moisture content and the OCP values were shifted to more negative potential by increasing the percentage of moisture content. The (OCP) values shifted gradually with percentage of moisture contents from -481.0, -639.0, -706.0, -741.0 and -773.0 at 0.0, 10, 20, 30 and 40% respectively. The results proved that the corrosion rate is directly proportional to the moisture content.

In Figure 2b shows the curves of the open circuit potentials versus time for the mild steel electrode in sediment of pond 1 at different moisture contents starting from 40 to 150%. The (OCP) values started

Element	Fe	C	Si	S	P	Mn
% Composition	98.7	0.15	0.08	0.025	0.025	1.02

Table 2: XRF analysis of the mild steel electrode.

Compound Formula	Pond 1 (%)	Pond 4 (%)
Na ₂ O	3.063	7.533
MgO	6.255	3.809
Al ₂ O ₃	6.473	4.428
SiO ₂	21.903	12.559
P ₂ O ₅	0.158	0.080
SO ₃	9.454	22.083
K ₂ O	0.984	1.040
CaO	14.664	12.496
TiO ₂	0.864	0.512
MnO	0.089	0.030
Fe ₂ O ₃	6.194	3.471
CO ₃ O ₄	0.0	0.0
NiO	0.013	0.006
CuO	0.014	0.008
SrO	0.738	0.517
BaO	0.026	0.0
Cl	2.775	5.598
Br	0.0	0.030

Table 3: Chemical analysis of sediment in pond 1 and pond 4.

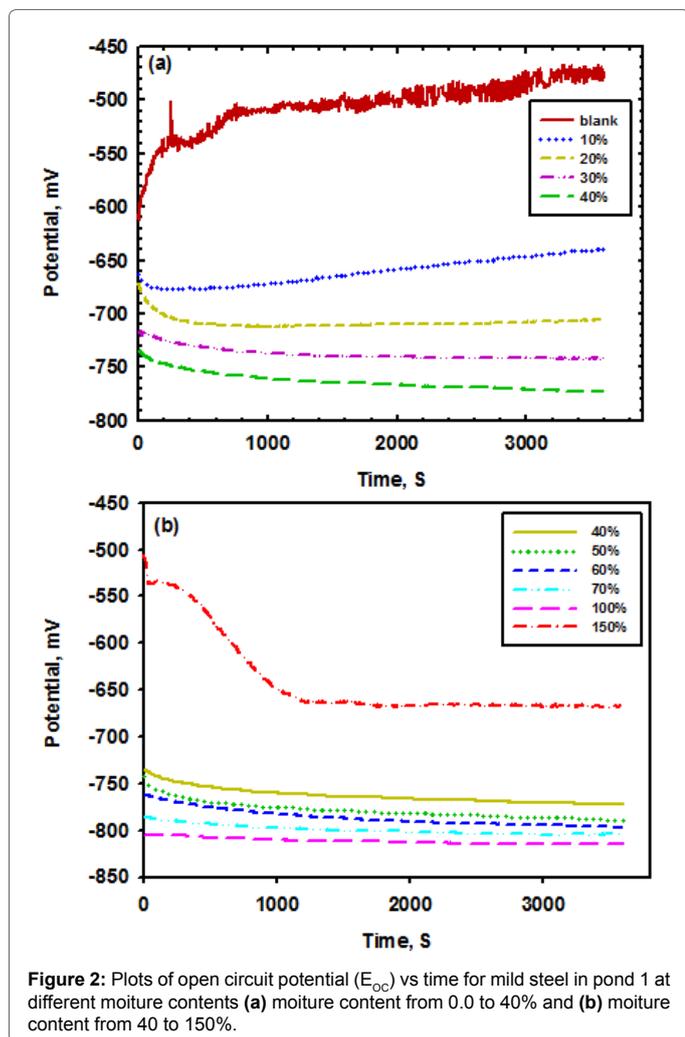


Figure 2: Plots of open circuit potential (E_{oc}) vs time for mild steel in pond 1 at different moisture contents (a) moisture content from 0.0 to 40% and (b) moisture content from 40 to 150%.

from -773.0, -797.0, -798.0 -802.0, -815.0 and -667.0 at 40, 50, 60, 70, 100 and 150% respectively, these data proved that the highest rate of attack on the mild steel surface is observed at 100% water content. The results show decrease in the (OCP) values but with narrow difference compared with the data obtained from Figure 2a. The results of (OCP) values for mild steel vs different moisture content of sediment of pond 1 indicate that the kind of corrosion is general corrosion and the effect of chloride ions is clearly noticed. These results could be also proved that there is another factor which affect on the corrosion behavior especially at the 150% moisture content, this factor is the concentration factor. From mineralogical analysis of sediment of pond 1 we can conclude that the chloride content is constitute the major effect on the corrosion rate from 10 to 100% of moisture content and in case of 150% the dilution factor effect is clearly noticed.

In Figure 3a shows the curves of the open circuit potentials versus time for the mild steel electrode in sediment of pond 4 at different moisture contents starting with zero water content and water contents of 10, 20, 30 and 40%. Figure 3b shows the curves of the open circuit potentials versus time for the mild steel electrode in sediment of pond 4 at different moisture contents starting from 40 to 300%.

The (OCP) values started from -602.0, -682.0, -735.0 -762.0, -755.0, -762.0, -782.0, -793.0, -801.0 -804.0, -805 and -807 at zero, 10, 20, 30, 40, 50, 60, 100, 150, 200, 250 and 300% respectively. These data proved

that the corrosion rate is continuously increased with increasing of moisture content. The concentration of chloride ions in sediment of pond 4 is higher that of its content of sediment pond 1 with about double amount 5.598 and 2.775 respectively. This difference in chloride content has the main factor in the obtained behavior of pond 1 and pond 4 and we can notice that the concentration factor is noticed only in pond 1 but in cade of pond 4 there is no effect of concentration factor due to the existed higher concentration of chloride ions.

The values of OCP are shifted towards the negative potential while chloride ion concentration increases by increasing the moisture content. As a result of increasing the chloride ions concentration, the chance of attack of chloride ions to the mild steel surface is also increased and this phenomenon is clearly noticed from the corrosion rate results. The rate of corrosion is affected by the oxygen depolarization which controls the range of sodium chloride concentration.

In Figure 4a shows the open circuit potential values for mild steel in pond 1 at different moisture contents. The results proved that mild steel is highly affected by the chloride ions in case of moisture content from 10 to 100% but it may be used in case of moisture content more than 100%. The big shift in potential in moisture content from 10 to 40% indicate to the kind of corrosion is general corrosion. On the other hand the narrow difference in potential in case of moisture content from 40 to 100% indicate that the kind of corrosion is localized corrosion due to the attack of chloride ions occurs at small areas (anodic areas) and the

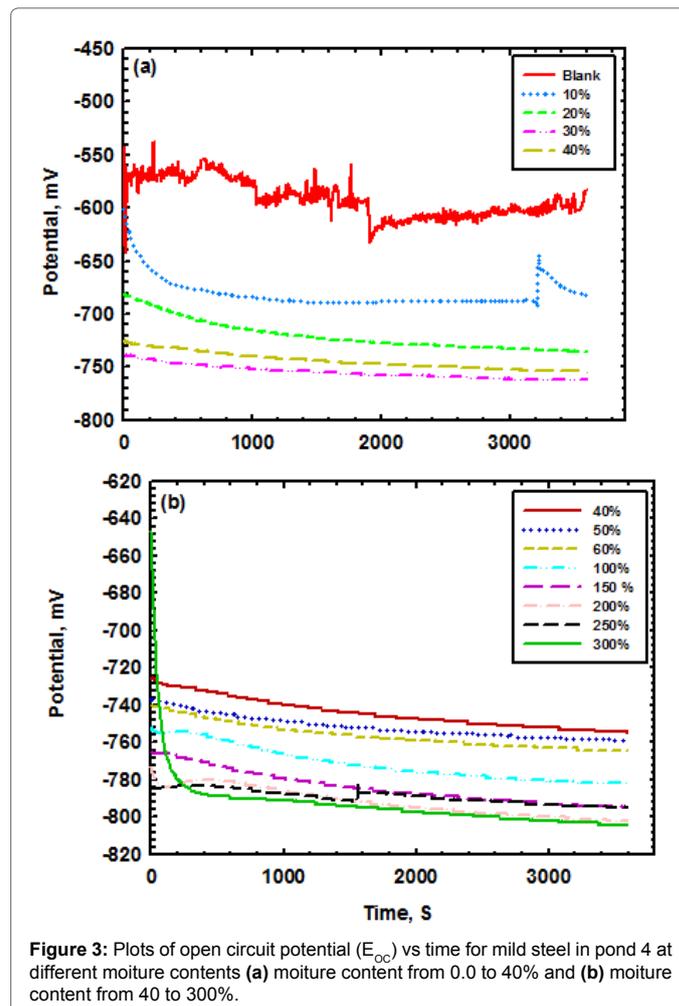


Figure 3: Plots of open circuit potential (E_{oc}) vs time for mild steel in pond 4 at different moisture contents (a) moisture content from 0.0 to 40% and (b) moisture content from 40 to 300%.

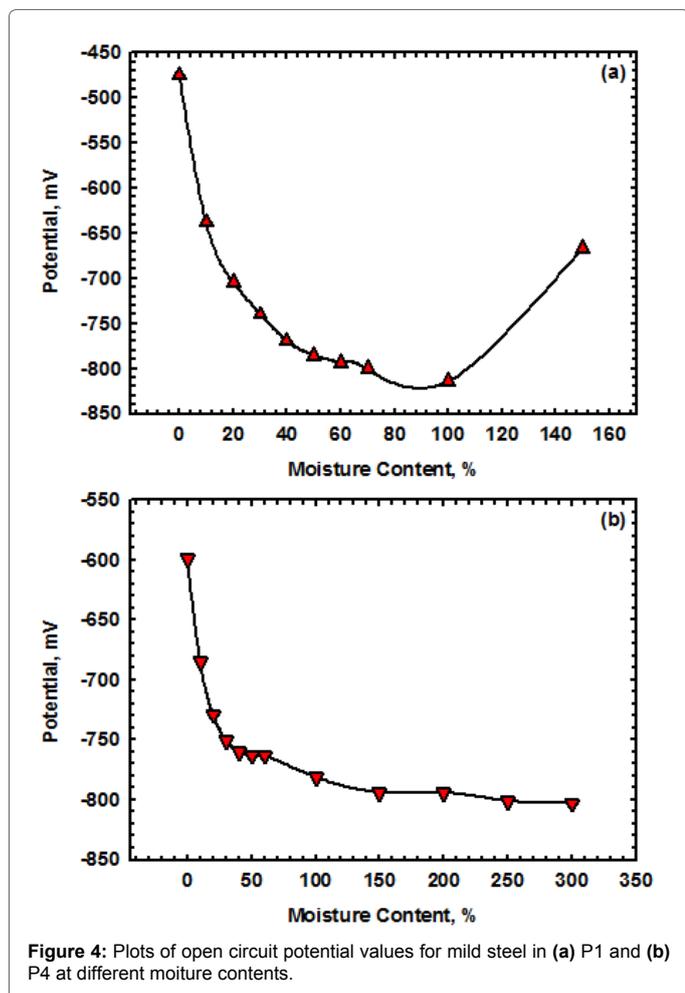


Figure 4: Plots of open circuit potential values for mild steel in (a) P1 and (b) P4 at different moisture contents.

around this area is called (cathodic area) or protected area and so the corrosion rate is decreased which is clearly noticed at 150% moisture content. Mild steel could be used for steel structure in this pond but at moisture content of 150%.

In Figure 4b shows the open circuit potential values for mild steel in pond 4 at different moisture contents. The results proved that the chloride concentration plays an important role in this behavior. The corrosion rate is increased continuously at all moisture contents which prove that the mild steel is not suitable for use in this pond. The same behavior of kinds of corrosion is obtained. Material selection is very important to combat this severe condition of chloride ion concentration by using another kind of steel like stainless steel.

Potentiodynamic polarization results: In Figure 5a shows the potentiodynamic polarization curves of mild steel in pond 1 at different moisture contents from zero to 40%. The results show that the chloride ions in the sediment of pond 1 increase the corrosion rate gradually from 10 to 40%. The values of current density compared with the moisture contents show role of chloride ions in corrosion of mild steel. The values are 2.8×10^{-5} , 0.2, 2.9, 18.3 and 32.4 mA/cm^2 for 0.0, 10, 20, 30 and 40% moisture content respectively. These big difference in current density proved that the present corrosion is general corrosion.

In Figure 5b shows the potentiodynamic polarization curves of mild steel in pond 1 at different moisture contents from 40 to 150%. The results show that the corrosion rate is increased with narrow

difference from 50 to 100% of moisture content and decreased again at moisture content of 150%. The current density values started from 35.2, 39.2, 41.8, 60.7 and 0.17 mA/cm^2 for 50, 60, 70, 100 and 150% moisture content respectively. The values of current density proved that the form of corrosion became localized corrosion. Potentiodynamic polarization result of current density and potential of corrosion processes, which were taken places in this work are presented in Table 4.

In Figure 6a shows the potentiodynamic polarization curves of mild steel in pond 4 at different moisture contents from 0.0 to 40%. The chloride content in the sediment of pond 4 is about the double value of the sediments in pond 1. The results of current density illustrate that increase the corrosion rate gradually from 10 to 40% with big difference but from 50 to 300% still increase with narrow values and the results are presented in Figure 6b. The values are 7.8×10^{-5} , 0.4, 16.8, 28.8 and 36.7 mA/cm^2 for 0.0, 10, 20, 30 and 40% moisture content respectively. Potentiodynamic polarization results of current density and potential of corrosion processes, which were taken places in this work, are presented in Table 5.

On the other hand, the increasing of moisture content increase the conductivity, hence additional anodes and cathodes can operate much farther removed one from the other. The formed ferrous hydroxide $\text{Fe}(\text{OH})_2$ at anodes does not provide a protective barrier layer on the metal surface and react with water in the presence of oxygen to form $\text{Fe}(\text{OH})_3$ away from the metal surface [19].

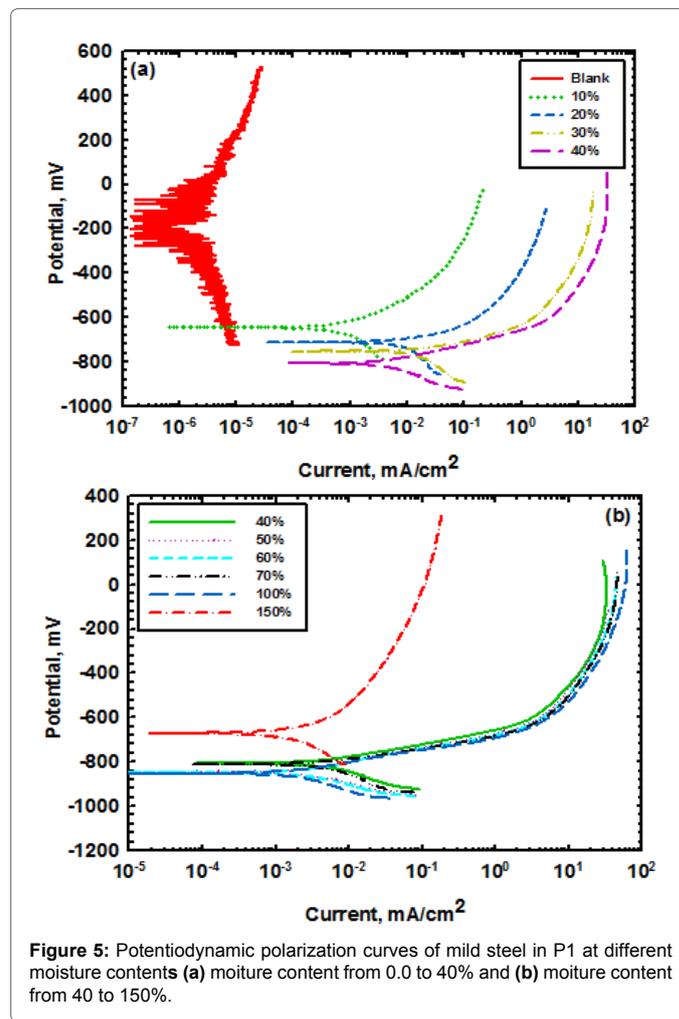


Figure 5: Potentiodynamic polarization curves of mild steel in P1 at different moisture contents (a) moisture content from 0.0 to 40% and (b) moisture content from 40 to 150%.

Moisture content	OCP (mV) Vs. Cu/CuSO ₄	E _{corr} (mV) Vs. Cu/CuSO ₄	b _a (mV dec ⁻¹)	b _c (mV dec ⁻¹)	I _{corr} (mA cm ⁻²)	C.R, mpy
Blank	-481	-200	31.7	-288	0.004	Approx. zero
10	-639	-645	35	-33.7	0.142	0.0031
20	-706	-710	31.5	-34.9	2.42	0.0528
30	-741	-753	25.5	-32.97	6.4	0.1395
40	-773	-808	29.81	-38.56	7.97	0.1737
50	-797	-845	32.27	-32.68	11.8	0.2579
60	-798	-850	32.73	-29.9	9.12	0.1988
70	-802	-810	30.08	-33.08	4.075	0.0888
100	-815	-855	32.9	-30.76	3.825	0.0834
150	-667	-672	34.99	-33.65	0.30	0.0065

Table 4: Potentiodynamic polarization results of pond 1.

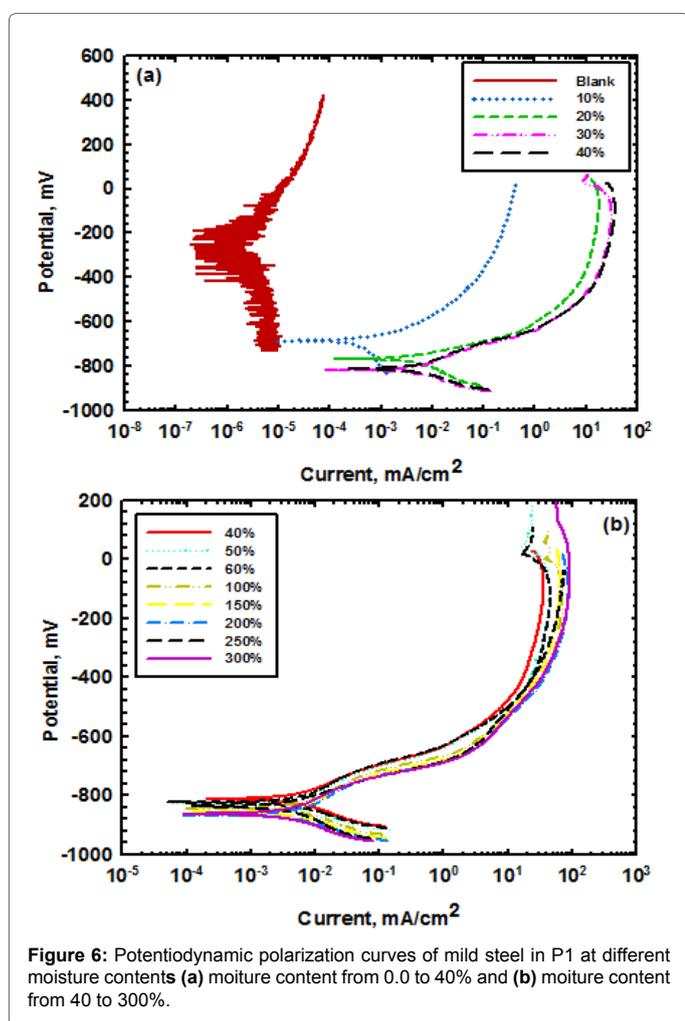
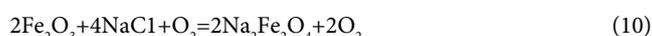


Figure 6: Potentiodynamic polarization curves of mild steel in P1 at different moisture contents (a) moisture content from 0.0 to 40% and (b) moisture content from 40 to 300%.

Hence mild steel is corrodes more rapidly in case of increasing moisture content because of dilution factor of chloride ions which facilitate the movement of dissolved oxygen to reach the cathodic areas [20]. This phenomenon is agreed with the results of Figure 6b and the corrosion rate none stopped at certain concentration because of higher concentration of chloride ions in pond 4 compared with pond 1. The results also proved that the anodic potential stimulates the migration of the chloride ion preferably to the defect sites of the surface which leads pitting formation [21].

In Figure 7a shows the relationship between the current density against moisture contents for mild steel in pond 1. These findings were agreed with the obtained results from Figures 2a, 4a and 5a, the suddenly rise in current density indicates to the general corrosion but at the 150% moisture content the current density is decreased due to the initiation of pitting corrosion.

Generally the higher content of chloride ions enhances the passivation current density, which is a criterion of anodic dissolution of the mild steel in passive state. These findings could be attributed to the adsorption of chloride ions on the oxide surface to form ferrite see the following equations [22,23].



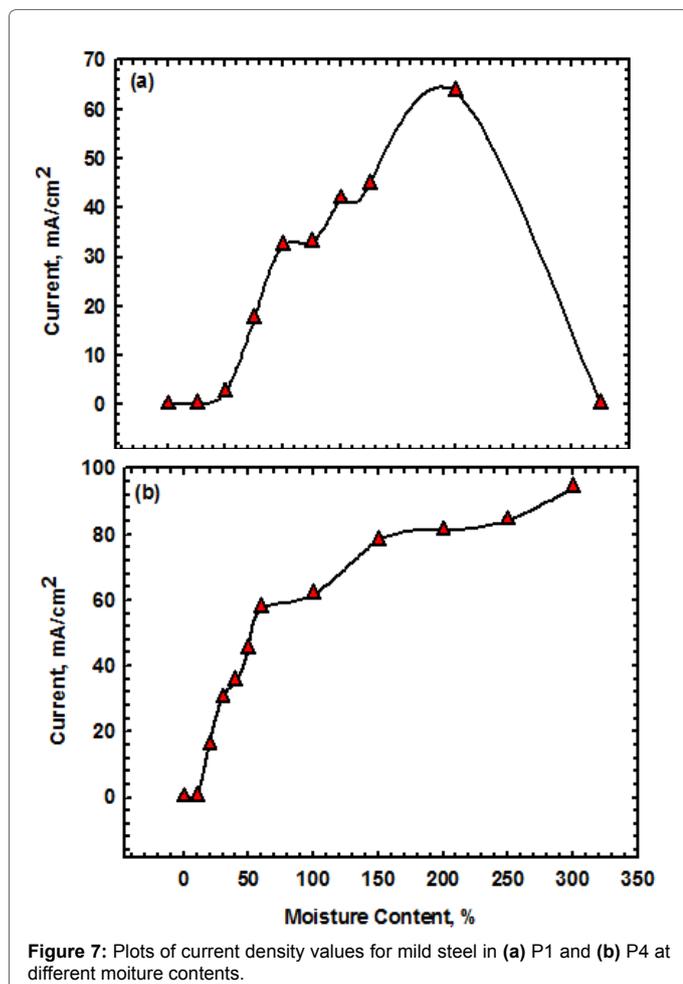
The equation 10 shows that the thinning of the passive film as a result of adsorbed chloride ions, which tend to enhance the dissolution of the oxide film and making the corrosion in continuous process with time. This behavior is clearly noticed from the gained results in Figure 7b the current density begins to rise suddenly, indicating passivity breakdown and initiation of pitting attack [24].

Effect of pH

The obtained pH values with different moisture content are in the range of 6.6 to 7.8 i.e., these values of pH are in neutral range, and the effect of these values of pH are assumed to be ranged from non corrosive to moderate corrosive media. These results could be concluded that the steel pipelines showed no significant effects of pH on the corrosion rate values. It is well known that in pH range from 5 to 10, the corrosion current is quickly decreases because of the oxygen diffusion rate through the passive film was likely to be lower than the corrosion reaction rate and this region is called stable region [25]. In this range of pH the corrosion rate can be expressed simply in terms of the existed amount of hydroxyl group and deposition of insoluble ferric hydroxide tends to stifle the corrosion attack. In case of pH 4.5, acid corrosion is initiated, overwhelming the oxygen control so that the corrosion potentials of carbon steel were not dependent on the cation type (Na⁺) [26].

Inhibitor concentration (ppm)	OCP (mV) Vs. Cu/CuSO ₄	E _{corr} (mV) Vs. Cu/CuSO ₄	b _a (mV dec ⁻¹)	b _c (mV dec ⁻¹)	I _{corr} (mA cm ⁻²)	C.R, mpy
Blank	-602	-250	24.0	-262	0.006	Approx. zero
10	-682	-635	26	-46	0.11	0.0024
20	-735	-768	29	-35	7.02	0.153
30	-762	-780	31	-34	10.91	0.238
40	-755	-812	38	-30	21.3	0.464
50	-762	-827	36	-32	35.3	0.77
60	-782	-823	37	-29	35.7	0.78
100	-793	-843	32	-32	45	0.98
150	-801	-856	30	-33	42	0.94
200	-804	-865	32	-30	53	1.16
250	-805	-873	34	-31	55	1.65
300	-807	-879	37	-34	57	1.71

Table 5: Potentiodynamic polarization results of pond 4.



Conclusion

Concentration of chloride ions has been affected by the moisture content sediments under study and the major effect on corrosion rate is the concentration factor of chloride ions. In pond 1 and pond 4 the corrosion rate increases when the moisture content increases due to the concentration factor of chloride ions which increase the chance of attack on the mild steel surface. This behavior is continuously noticed but in pond 1 it stopped when the moisture content reaches to 150%. In pond 1 the kind of corrosion is general corrosion especially in the moisture content from 10 to 40% due to the big change in corrosion rate values but from 40 to 100% is localized corrosion. The corrosion rate is decreased at 150% due to the chloride concentration in pond 1 is less than in pond 4. The material selection is very important in this environment of chloride ions. Mild steel is recommended in pond 1 especially in moisture content 150%. In pond 4 due to the higher concentration of chloride ions the mild steel is totally not recommended but it is recommended to use stainless steel in all percentages of moisture content.

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