

Electrochemical studies on the alternating current corrosion of mild steel under cathodic protection condition in marine environments

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Abstract

Alternating current (AC) corrosion of mild steel in marine environments under cathodic protection (CP) condition was studied. Electrochemical studies at the two protection potentials namely -780 and -1100 mV versus SCE were examined by different techniques. DC polarization study was carried out for mild steel in natural seawater and 18.5 g/L NaCl solution to evolve corrosion current density. The corrosion rate determination, pH of the end experimental solution and surface morphology of the mild steel specimens under the influence of different AC current densities were studied. The amount of leaching of iron into the solution was estimated using inductively coupled plasma spectrometry. All these techniques revealed that AC influences the corrosion of mild steel in the presence of marine environments even though CP was given. Surface micrographs revealed that spreading of red rust products noticed on the mild steel surface. At -780 mV CP, red rusts are visually seen when the AC source was above 10 A/m^2 in both the media but red rusts are appeared after 20 A/m^2 in the case of -1100 mV CP. Weight loss measurements coupled with surface examination and solution analysis is a effective tool to characterize and quantify the AC corrosion of mild steel in marine environments. © 2006 Elsevier Ltd. All rights reserved.

Keywords: AC corrosion; Mild steel; Cathodic protection; Seawater; NaCl

1. Introduction

It is well known that unidirectional currents are involved during the process of metallic corrosion. However, under certain conditions, alternating currents can also cause corrosion although at a lower rate than direct currents. A review of literature reveals that a number of investigations have been carried out on the AC corrosion of Fe, Pb, Cu, Al and other metals [1–3]. Besides the corrosion of metals, the dissolution of metals by AC has also attracted the attention of a number of investigators [4,5]. Kulman [6] and Chin and Venkatesh [7] reported the corrosion of metallic structures due to the presence of stray AC current. AC corrosion mechanism is not yet understood and reliable techniques for the determination of the corrosion risk are not yet available. It becomes more difficult to install metal-

lic pipelines with adequate distance from overhead high-voltage AC power transmission lines and/or AC powered rail transit systems. Pipelines installed with the close proximity to the seashore areas have additional risk of marine corrosion. Several cases of AC corrosion on buried steel pipelines due to induced AC arising from high-voltage AC power transmission lines have been reported [8]. A number of laboratory and field studies have been performed to investigate the influence of AC induced voltage on the corrosion behaviour of corroding systems. Most of the literatures concern the influence of AC in acidic and sulfate solutions [9–14]. However, more researches are essentially needed for the influence of AC on chloride environments, since the marine environments have a high concentration of chloride ions, it is expected that AC may accelerate the corrosion. This investigation is systematic and detailed studies on the influence of AC induced corrosion of mild steel in natural seawater under CP condition. For a comparison, the AC corrosion of mild steel in 18.5 g/L NaCl solution was also carried out under CP condition.

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2. Experimental

2.1. Materials

Mild steel obtained from Dongil Industries Co., Ltd., South Korea, was used and the composition in wt% is follows: C (0.43), Si (0.22), Mn (0.72), P (0.013), S (0.015), Ni (0.05), Cr (0.10), Cu (0.12) and Fe balance. Mild steel specimen of size 2 cm (l) \times 2 cm (b) \times 0.5 cm (thickness) was used and the total exposed surface area was 11 cm² and they were given fine mechanical polishing. Before and after immersion individual specimens were cleaned with 50% (v/v) hydrochloric acid, followed by a rinse with de-ionized water and acetone. Natural seawater (Jinhae sea, South Korea) and AR grade NaCl was used as aggressive solutions. Since the seawater used here contains 18.5 g/L of chloride, studies were also carried out in NaCl solution containing the same amount of chloride. Solutions were purged with nitrogen gas to remove the dissolved oxygen. In order to maintain the uniformity, the chloride level, oxygen level and the temperature are kept constant for both the solutions.

2.2. Methods

2.2.1. DC polarization technique

Potentiodynamic polarization for mild steel in 18.5 g/L NaCl solution and natural seawater environments were carried out

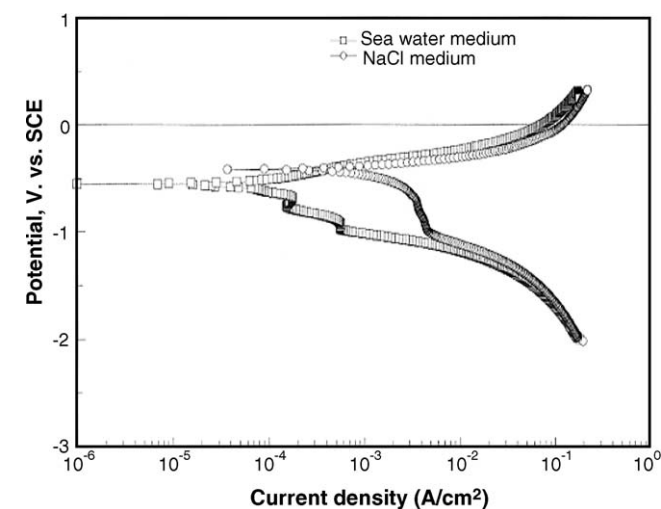


Fig. 1. Potentiodynamic polarization curves for mild steel in NaCl solution and natural seawater.

using three electrode cell assembly. The working electrode was mild steel specimen. Platinum foil and a saturated calomel electrode served as the auxiliary and reference electrodes, respectively. An electrochemical cell consists of 250 mL of test solutions. Both cathodic and anodic polarization curves were recorded potentiodynamically using a Solartron-1480 multi channel electrochemical interphase controlled by computer. Procedures and parameters specified in ASTM standard G5-87 were followed. All the experiments were carried out at a temperature 25 ± 1 °C.

2.2.2. Weight loss experiments with and without CP condition

The weight of the mild steel samples was taken before and after immersion using Ohaus Explorer four-digit electronic balance for gravimetric weight loss measurements. The AC signal was applied between the working electrode (mild steel) and the counter electrode (platinum gauge). Solartron-1480 Multistat electrochemical measurement unit coupled with Solartron 1255-B Frequency Response Analyzer and multi media computer was used to supply the required AC waveform. An oscilloscope was used to adjust as well as to monitor the AC signal applied. Experiments were carried out at the two protection potentials are as follows: (i) to find out the actual cathodic DC current (cathodic protection current) at the protection potentials namely -780 and -1100 mV versus SCE in NaCl solution and natural seawater environments in the absence of AC sources and (ii) to find out the corrosion behaviour of mild steel in NaCl solution and natural seawater environments under CP condition in the presence of various AC sources. In the first step, separate experiments were carried out by applying constant protection potential namely -780 and -1100 mV on mild steel working electrode with reference to SCE in NaCl solution and natural seawater environments. The protection current (DC current) at the respective protection potential was monitored for an exposure period of 24 h using a computer controlled data logger. Actually, the current was monitored by using a Keithley 2701 Ethernet Multimeter data acquisition system controlled by computer. So the program was made to record one value at the end of 1 h. Like that 24 readings are recorded for the total exposure period. The values obtained are reasonable and reproducible from the triplicate set of experiments. In the second step, the CP condition was simulated in the electrochemical cell by maintaining the cathodic DC current constant and varied the AC amplitudes at different current densities and fixed frequency of 60 Hz. Tripli-

Table 1
Corrosion rate of mild steel in NaCl and natural seawater with and without AC

Solutions	Corrosion rate (mm/y)									
	-780 mV vs. SCE					-1100 mV vs. SCE				
	No AC	10 (A/m ²)	i_{corr} 1.22 (mA/cm ²)	20 (A/m ²)	100 (A/m ²)	No AC	10 (A/m ²)	i_{corr} 0.94 (mA/cm ²)	20 (A/m ²)	100 (A/m ²)
NaCl	NIL	0.4776	0.4816	0.6256	0.9891	NIL	0.0036	0.0035	0.0156	0.1690
Natural seawater	NIL	0.2649	0.2916	0.5428	0.9806	NIL	0.0034	0.0032	0.0126	0.1520

cate experiments were carried out for each AC source and the average corrosion rate was made in mm/y. Corrosion rate in mm/y was made by using difference in weight losses between initial and final weight of the specimens for the exposure period of 24 h.

2.2.3. Visual examinations

At the end of weight loss measurements the mild steel specimens were taken out and carefully examined for any corrosion products like red rust observed on the surface aided with magnifying lens [15].

2.2.4. pH measurements

An electrochemical cell consists of 250 mL of electrolyte solutions. AC sources were applied on the mild steel working electrode and platinum counter electrode for the exposure period of 24 h. pH was measured using a standard calibrated pH meter (portable ISTEK pH meter (Model 76P)) with a relative accuracy of ± 0.002 . Triplicate measurements were made for each system and the average was given.

2.2.5. Surface examination by optical microscopy and X-ray diffraction

Optical microscopy was used to examine the nature of corrosion on the mild steel surface before and after immersion in various test solutions. The mild steel specimens were immersed in test solutions for the exposure period of 24 h under the influence of AC with and without CP. At the end, specimens were taken out from the solution thoroughly washed with distilled water, cleaned with acetone and dried. Micrographs were recorded for mild steel surface at the different applied AC current densities in NaCl solution and natural seawater environments. The metallographic examination of the specimen was carried out using Olympus-GX 71 make computer controlled microscope. Physical characterization by XRD was carried out on the mild steel surface in the absence and presence of corrosion products. XRD for the corrosion products was done by powder X-ray diffraction technique on a Philips 1830 X-ray diffractometer using Ni filtered Cu K α radiation ($\lambda = 1.5406 \text{ \AA}$) in the 2θ range of $10\text{--}115^\circ$ at a scan rate of $0.01^\circ \text{ s}^{-1}$.

2.2.6. Solution analysis by inductively coupled plasma (ICP) spectrometry

The solution analysis was carried out with an idea to quantify the leaching characteristics of mild steel. The concentration of metal ions present in the test solution was determined after treatment with AC under CP conditions. The solution analysis was carried out using an inductively coupled plasma-atomic emission spectrometry (ICP, Applied Research Laboratory, USA). Triplicate experiments were carried out for each system and the average values are noted.

3. Results and discussion

3.1. DC polarization of mild steel in NaCl and natural seawater

The typical potentiodynamic polarization curves for mild steel in NaCl solution and natural seawater environments are given in Fig. 1. Mild steel showed no significant active/passive regions immersed in NaCl solution and natural seawater environments but a weak passivation range was observed in NaCl solution may be due to the adsorption of water molecules on the mild steel surface. In general, chloride anions are relatively small and the attacks on the steel surface through diffusion. But the water molecules are bigger than chloride anion and there was a competition between water molecules and chloride anion towards the steel surface. Mild steel is always covered with a passivated surface; the time taken for the rupturing the passive layer depends on the amount of chloride ions near the steel surface. So initially it shows weak passivation range, but in due time the concentration of chloride on the vicinity of the steel increased and the passivity was destroyed. The corrosion current density (i_{corr}) value for mild steel in NaCl solution and natural seawater environment was found to be 1.22 and 0.94 mA/cm^2 , respectively.

3.2. Studies on the corrosion behaviour of mild steel under CP condition in NaCl solution and natural seawater environments in the absence of AC

Mild steel was subjected to perfect CP condition i.e. -780 and -1100 mV versus SCE in both the media. The respective cathodic DC current was measured for the exposure period of 24 h. The cathodic DC current at -780 mV was -0.15 mA for NaCl solution and -0.095 mA for natural seawater. For -1100 mV CP, the currents were -0.37 and -1.21 mA , respectively, for NaCl and natural seawater. In both the cases, as expected the system was in CP condition, no corrosion was noticed. Specimens were examined visually at the end of exposure period and it was found that no rust was formed on the mild steel surface. The pH value remains constant throughout exposure period. All these data indicates that the system was perfectly cathodically protected. It is interesting to note that lower cathodic current was noted for natural seawater than NaCl solutions. This is due to the fact that difference in the composition of additional constituents present in the seawater. Turekian [16] reported the detailed composition natural seawater. Only chloride is taken care of when preparing 18.5 g/L NaCl solutions. But natural seawater contains other additional constituents like Ca, Mg, K, S, Br, etc., and few heavy elements inhibit the corrosion of mild steel and thereby showed a lower cathodic current. These currents are utilized for the further studies with different AC current densities. The protection potential was increased from -780 to -1100 mV versus SCE, the protection current was also increased in both the media. Interestingly this increase was found to be more in natural seawater than NaCl solutions. For example there was a 2.5-fold increase in the cathodic DC current in NaCl solution and 12.7-fold increase in the natural seawater

environments indicating that the higher protection currents are required for natural seawater than NaCl solution.

3.3. AC corrosion of mild steel under CP condition in NaCl solution and natural seawater environments at various AC current densities

3.3.1. Determination of corrosion rate

Table 1 shows the corrosion rate of mild steel in NaCl solution and natural seawater environments with and without AC sources at the two CP potentials. No corrosion of mild steel was noticed

under CP condition in the absence of AC. On the other hand, the corrosion rate of mild steel in NaCl solution was increased with increasing the AC current densities. The lowest corrosion rate obtained at 10 A/m² was 0.4776 mm/y. The corrosion rate at the i_{corr} was 0.4816 mm/y. The other systems namely 20 and 100 A/m² showed 0.6256 and 0.9891 mm/y, respectively. Similarly, in natural seawater environment also AC sources accelerate the corrosion of mild steel but the extent of corrosion rate was found to be less when compared to NaCl solution. Here also, the corrosion rate was increased when increasing the AC sources from 10 to 100 A/m². The corrosion rates are 0.2649 mm/y at

Table 2
pH of the end experimental solutions with and without AC

Solutions	pH									
	–780 mV vs. SCE					–1100 mV vs. SCE				
	No AC	10 (A/m ²)	i_{corr} 1.22 (mA/cm ²)	20 (A/m ²)	100 (A/m ²)	No AC	10 (A/m ²)	i_{corr} 0.94 (mA/cm ²)	20 (A/m ²)	100 (A/m ²)
NaCl	6.60	6.70	6.75	6.80	7.40	6.60	6.72	6.60	6.85	7.50
Natural seawater	8.34	8.35	8.37	8.50	9.00	8.34	8.36	8.34	8.49	9.20

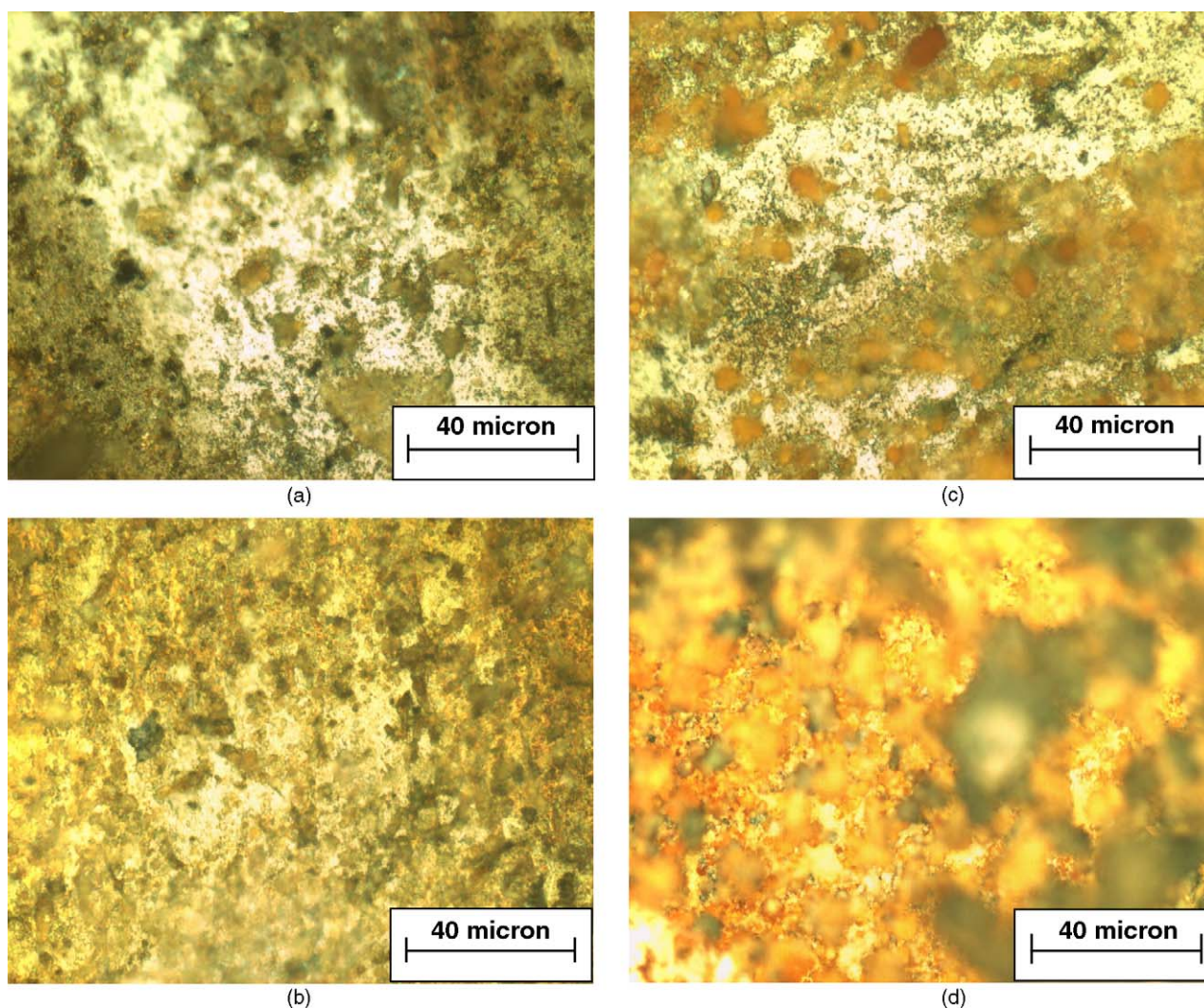


Fig. 2. Micrographs of mild steel in NaCl solution under –780 mV CP condition: (a) 10 A/m², (b) i_{corr} , (c) 20 A/m² and (d) 100 A/m².

lower AC sources and 0.9806 mm/y at the higher AC sources. Here it concluded that AC sources accelerate the corrosion at all the current densities even though mild steel was given perfect cathodic protection in marine environments.

The corrosion rate of mild steel in NaCl solution and natural seawater environments at -1100 mV CP is given in Table 1. Here also, no corrosion of mild steel under CP condition in the absence of AC. But the corrosion rate was increased with different AC current densities even though the system was under CP condition. For NaCl solution, the lowest corrosion rate was obtained at the lowest AC source namely 10 A/m^2 was 0.0036 mm/y . The corrosion rate at the i_{corr} was 0.0035 mm/y . The other systems namely 20 and 100 A/m^2 showed 0.0156 and 0.1690 mm/y , respectively. In natural seawater environments, the lowest corrosion rate at i_{corr} was 0.0032 mm/y . Further the corrosion rates are 0.0034 mm/y (10 A/m^2), 0.0126 mm/y (20 A/m^2) and 0.1520 mm/y (100 A/m^2), respectively. Here it is concluded that the protection potential was increased from -780 to -1100 mV versus SCE there was a decrease in the corrosion rate was observed in both the media. For example in the case of NaCl solution the corrosion rates in mm/y at the two

protection potential criterion in increasing order are as follows:

	10 (A/m ²)	>	20 (A/m ²)	>	100 (A/m ²)
-780 mV	0.4776	>	0.6256	>	0.9891
-1100 mV	0.0036	>	0.0156	>	0.1690

Here it sees that when the protection potential criterion was increased, the corrosion rate was decreased in NaCl solutions. Similarly in natural seawater environments, the corrosion rate (mm/y) at the various AC sources follows the order:

	10 (A/m ²)	>	20 (A/m ²)	>	100 (A/m ²)
-780 mV	0.2649	>	0.5428	>	0.9806
-1100 mV	0.0034	>	0.0126	>	0.1520

Here also when the protection potential was increased to -1100 mV versus SCE, the maximum decrease in the corrosion rate was observed.

From weight loss experiments, it was observed that, cathodically protected specimen at the cathodic potential of -780 and -1100 mV showed no corrosion rate, but the corrosion rate

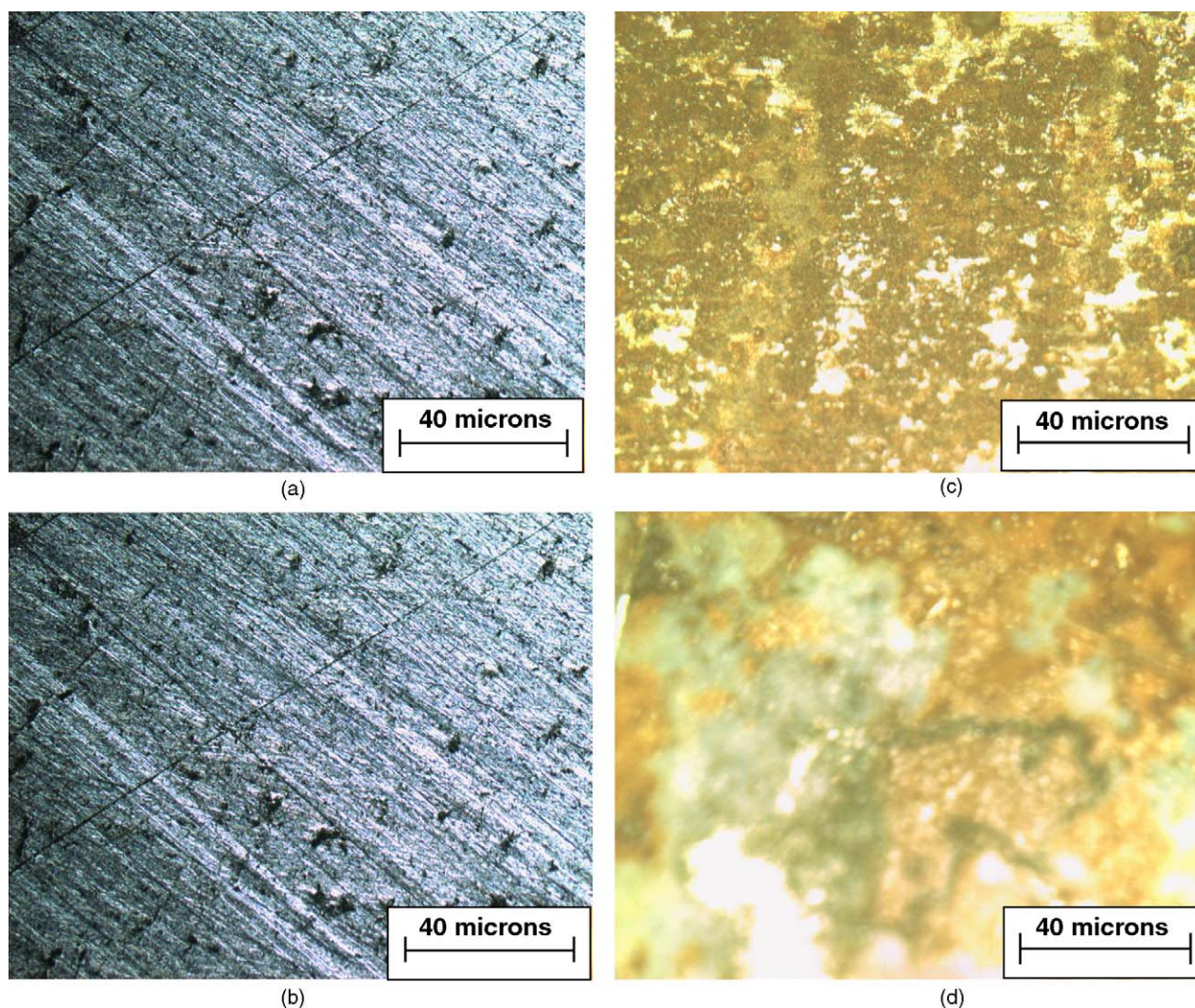


Fig. 3. Micrographs of mild steel in natural seawater under -780 mV CP condition: (a) 10 A/m^2 , (b) i_{corr} , (c) 20 A/m^2 and (d) 100 A/m^2 .

increased abruptly at higher AC current densities. The former was because anodic AC voltage exceeds the corrosion potential when the AC current density was applied. This could be interpreted that AC current flow causing charge transfer still being existed overwhelming cathodic DC current. The higher magnitude of corrosion rate of mild steel in both the media at higher current densities may be due to the destabilization of the passive layer on the mild steel surface. The growth of the passive layer occurs as a result of electrochemical reaction and this passive layer is tends to depassivate at the higher current densities and accelerate the corrosion of mild steel.

3.3.2. Visual examinations

Visual observations showed that no corrosion product was obtained in the absence of AC sources under CP condition. More red rust products are seen on the mild steel surface in both the media at higher AC current densities even though mild steel is under CP condition. Mostly, red rusts formed at the one place rather spread towards the entire surface. At higher AC source, more than 90% area showed red rust products.

3.3.3. pH measurements

The pH values of the experimental solution at the end of the exposure period of 24 h at the two CP potentials are given in Table 2. The pH of the plain NaCl and natural seawater used was 6.6 and 8.34, respectively. In both the cases, a small increase in the pH values with respect to the different AC current densities. It reveals that the degree of alkalization is increased in the solutions. The degree of alkalization of the solutions close to a metal is believed to be playing a major role in the corrosion process. The alkalization arises from influence of either the anodic or cathodic DC current along with AC sources which electrochemically reduces water into OH^- .



or



The accumulation of OH^- and consequent pH change would be the result if a mass balance for OH^- in the solution. The combination of elevated pH and a vibration of the DC potential

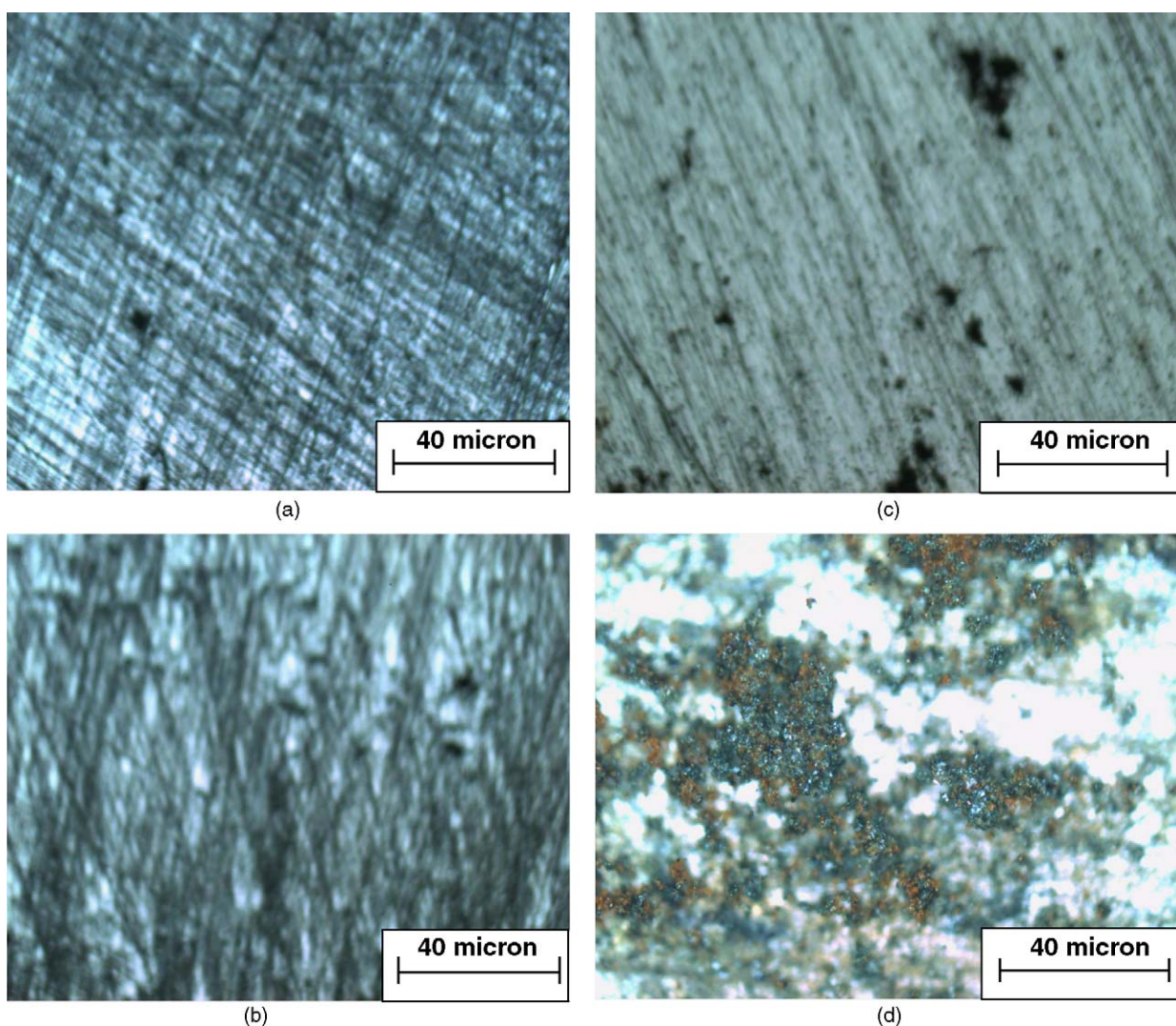


Fig. 4. Micrographs of mild steel in NaCl solution under -1100 mV CP condition: (a) 10 A/m^2 , (b) i_{corr} , (c) 20 A/m^2 and (d) 100 A/m^2 .

within this area caused by the AC may induce corrosion either by a destabilization of the passive layer or in the case of extreme alkalization by entering the general corrosion area (HFeO_2^- stabilization) at high pH. The AC also indirectly contributes to the alkalization process. When AC imposed on a coupon placed in a chloride solution will lead to a depolarization of the electrochemical kinetics and AC accelerates the DC current and consequently the rate by which OH^- is produced under cathodic polarization. In general production of OH^- ions would increase the conductivity of the solution adjacent to the metal.

3.3.4. Surface micrographs and XRD

The optical micrographs for mild steel in NaCl solution and natural seawater at the protection potential -780 mV with varied AC sources are shown in Figs. 2 and 3. Micrographs showed that the surface of the mild steel was not much affected at 10 A/m^2 . But mild steel showed few red rust products at the i_{corr} and more red rust products at 20 and 100 A/m^2 . Especially at 100 A/m^2 almost 90% areas suffered and showed red rust products. Red rusts formed at one place and then simply spreading or loosening

to the nearby surfaces. Similarly in natural seawater micrograph showed no evidence of corrosion at 10 A/m^2 and i_{corr} , but showed considerable red rust products on the surface at 20 and 100 A/m^2 . The corrosion was found to be uniform throughout the mild steel surface. The optical micrographs for mild steel in NaCl solution and natural seawater at the protection potential -1100 mV with varied AC sources are shown in Figs. 4 and 5. In this case, red rusts are visually seen after 20 A/m^2 only in both the media. A definite correlation was observed between AC corrosion immersion studies and surface morphology studies in both the media. The changes in the surface morphology of mild steel by the AC source is indicating that AC source have a strong influence on the AC current density. Recently Goidanich et al. reported that the AC affects the kinetic parameters such as Tafel slopes and exchange current densities for carbon steel, copper and zinc in 1 M sulphate solutions [17]. The initiation of red rusts at 20 A/m^2 and above is due to the metal solution interface could be altered by the AC signal and thereby affecting the corrosion kinetics. Most probably both reversibility and Faradaic rectification aspects have to be considered to under-

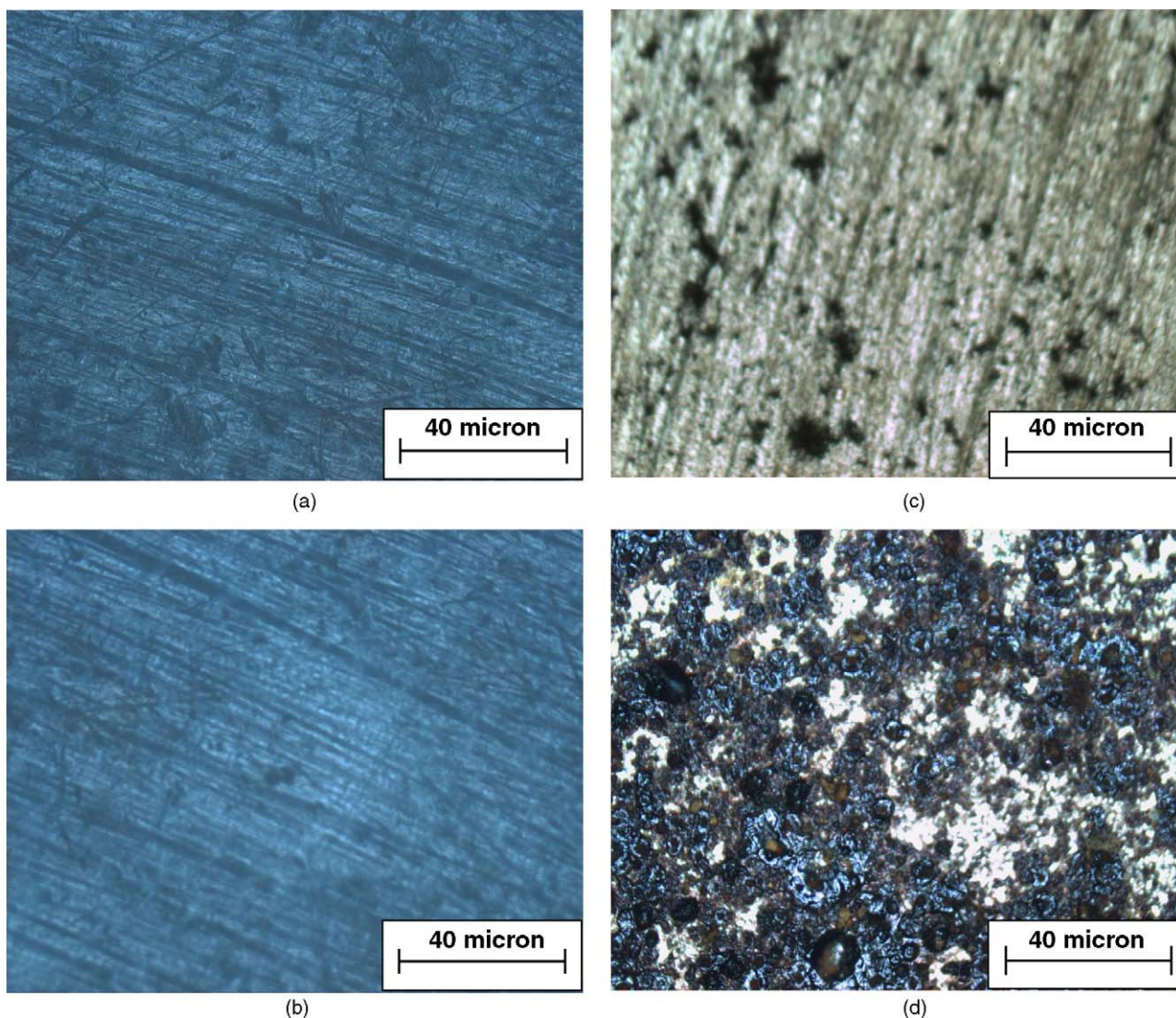


Fig. 5. Micrographs of mild steel in natural seawater under -1100 mV CP condition: (a) 10 A/m^2 , (b) i_{corr} , (c) 20 A/m^2 and (d) 100 A/m^2 .

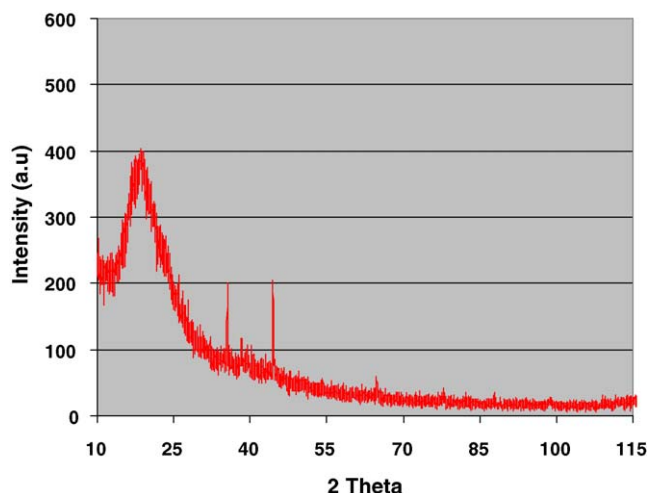


Fig. 6. XRD patterns of corrosion products from mild steel surface due to AC corrosion.

stand the AC induced corrosion [18]. Here the irreversibility of the chemical reactions occurring at the interface causes a change in the double layer compositions and a modification of the metal surface.

The corrosion product is subjected to physical characterization by XRD method and is given in Fig. 6. No XRD response for the uncorroded systems due to the absence of corrosion products. But for the systems showed rust products responds the XRD and showed a sharp and highly intense XRD peaks at 35.43 and 44.55 indicating the oxidation products of iron, which is matching with JCPDS.

3.3.5. Solution analysis by inductively coupled plasma spectrometry

Figs. 7 and 8 show the concentration of iron in test solution at -780 and -1100 mV protection potentials, respectively. In both the cases, concentration of iron is increased while increasing the AC current densities. The concentration of Fe in test solutions upto 10 A/m² is comparatively smaller than higher AC current densities. The concentration of iron in NaCl solution is more than the natural seawater medium. The same observation was noticed in weight loss measurement also. So a definite cor-

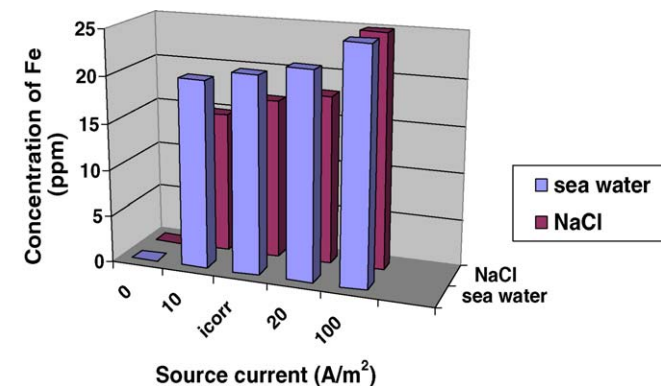


Fig. 7. Concentration of iron in NaCl and natural seawater at the -780 mV potential.

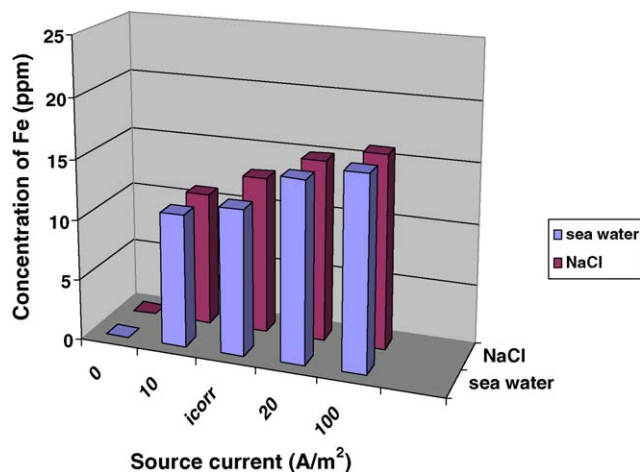


Fig. 8. Concentration of iron in NaCl and natural seawater at the -1100 mV potential.

relation was observed between weight loss measurements and solution analysis studies. All these data clearly indicated that AC sources having definite influence on the corrosion of mild steel in natural sea water under CP condition. Most probably, both reversibility and faradaic rectification aspects have to be considered to understand the AC induced corrosion. The irreversibility of the chemical reactions occurring at the interface causes a change on the double layer composition and a modification of the metal surface.

4. Conclusions

The following broad conclusions can be draw from the present investigation:

- (1) Mild steel does not corrode under the protection potentials, viz., -780 and -1100 mV versus SCE in the absence of AC sources. This is to be observed in both NaCl solution and natural seawater environments. But the AC sources able to accelerate the corrosion of mild steel even though they are cathodically protected in both the media.
- (2) The protection potential criterion was increased to -1100 mV versus SCE, very negligible corrosion rates was obtained upto the AC sources of 20 A/m², after that increase in the corrosion rate was obtained at the 100 A/m².
- (3) The degree of alkalization of the solution is increased with the different applied AC current densities in both the media.
- (4) Surface micrographs revealed that spreading of red rust products noticed on the mild steel surface. In the case of -780 mV protection potential, red rusts are visually seen when the AC source was increased after 10 A/m² in both the media. On the other hand, red rusts are seen only after 20 A/m² in the case of -1100 mV protection potential criterion.
- (5) A good agreement was observed between weight loss measurements coupled with surface morphological studies and solution analysis.

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