The Evaluation of Synergistic Effect of Marine Environmental Factors on Corrosion Rate of 1045 Carbon Steel

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Abstract

Seawater is a kind of severely corrosive environments. Although the effect of individual environmental factors on corrosivity of seawater is generally considered, the synergistic effect of those parameters on marine corrosion process is not widely understood. This research evaluates the synergistic effect of environmental parameters on corrosion rate of carbon steel 1045. The considered parameters are salinity, pH, velocity and temperature. This research takes the advantage of both quantitative and qualitative analyses. At the first phase of this paper, quantitative analysis is performed via full two-level factorial ANOVA method to calculate the contribution value of each parameter in changing the quantity of average corrosion rate in both individual and synergistic cases. At the second phase, the qualitative analysis has been used to predict the variation direction of corrosion rate as a result of considered parameters' effect in both individual and synergistic manners. These two complementary analyses result in understanding both the value and quality of variations applied on the reference corrosion rate considering the synergistic action of effective environmental seawater factors. This study offers the more comprehensive understanding of synergistic actions on corrosion rate which has an undeniable advantage in marine application designs.

Keywords: Carbon steel; Synergistic effect; Environmental parameters; Oxidation.

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1. Introduction

Seawater is generally known to be a very corrosive environment as a result of its high conductivity and the penetrating power of the chloride ion through surface films on a metal. Metals corrosion by seawater is a serious problem and countermeasures have been sought by related industries for years. Different kinds of metals have been examined in this environment. Carbon steel is one of the most widely used metals in seawater. Despite its relatively limited corrosion resistance, carbon steel is used in large tonnages in marine applications, specifically in seawater pipelines [1-11].

The corrosion rate of carbon steel in seawater is under cathodic control and varies by the availability of oxygen to the metal surface [7]. In such procedures, which corrosion rate is subject to mass transfer control, availability of oxygen at the metal surface becomes the rate-controlling factor [7]. Every parameter that fascinates the arrival of oxygen to the metal surface accelerates the corrosion rate of carbon steel in seawater. In this article the effect of velocity, temperature, salinity and pH on corrosion rate of carbon steel samples have been investigated. These parameters are of the most well-known affecting parameters on the corrosion rate of metals in marine environment [2, 7].

As velocity causes a mass flow of oxygen to the surface, corrosion is very dependent on flow rate [7]. Although oxygen solubility tends to fall with a rise in temperature, the higher temperature generally tends to increase reaction rate in chemical or electrochemical reactions, like corrosion. On the other hand, increase in temperature accelerates film formation, which decreases the corrosion rate by the restriction of oxygen diffusion to the metal surface [11-14]. The third parameter, salinity has an ambivalent effect on the corrosion rate of carbon steel in marine environment. It has an accelerating effect by the increase in the conductivity of seawater and on the other hand, it can show the reducing function by decreasing the amount of dissolved oxygen [2, 7, 15]. The effect of the last parameter, pH, is closely related to its sharp effect on the formation of protective films on the surface of carbon steel [14].

Since corrosion of materials in the marine environment has been of great importance, it is not surprising that many studies have been made to evaluate the effect of corrosion controlling parameters in marine environments. In addition, lots of corrosion prevention techniques have been considered to control the rate of corrosion such as modified coatings, inhibitors, surface modification, etc [16,17,18]. Before such prevention methods to decrease corrosion rate of metals, prediction of materials' behaviour under the applied conditions is of great importance. In 1985, an experimental study by Sefaja and Malina clearly indicated an influence of oxygen on the kinetics of

the corrosion process, which could be seen as a change of the controlling mechanism as the oxygen concentration was varied [19]. Melchers in 2003, in a study, regarding the mathematical modelling of the diffusion controlled phase in marine immersion corrosion and McNeill in a part of his research, surveyed the effect of temperature on the corrosion rate of carbon steel and showed both increasing and decreasing effect of temperature [20, 21]. In 2004, the effect of water velocity on marine immersion corrosion of carbon steel was demonstrated in laboratory conditions and the researchers found that velocity increases the rate of corrosion nonlinearly [8]. The effects of pH and salinity were investigated and modelled individually in other studies published in 2005 and 2008 [22, 23]. In all these researches, the effect of controlling factors has been studied individually and it is not understood how the simultaneous action of these factors can affect on the rate of corrosion. In 2007, in an attempt to investigate the simultaneous action effects, Meng and Neville considered the erosion–corrosion performance of two stainless steels as a function of simultaneous action of electrochemical and mechanical parameters in marine condition [24] and in 2008 another research evaluated the synergistic effects of two different mechanical parameters on the erosion–corrosion behaviour of aluminium [25]. In 2010 and 2011, Atashin et.al studied the synergistic effects of marine parameters on stainless steels 314 and 316 through two separate researches [26, 27].

The effect of simultaneous action of controlling parameters should be considered from two points of view. Firstly, how much is the contribution value of each parameter in changing the quantity of average corrosion rate and secondly, how can the simultaneous action of parameters change variation direction of corrosion rate.

The first phase of this article intended to investigate the "quantitative effect" of each considered parameter in changing the average value of corrosion rate via full two-level factorial ANOVA method. Analysis of variance (ANOVA) is a statistical technique to analyze variation in a response variable measured under conditions defined by discrete factors. Frequently, ANOVA is used to investigate the quantitative value of each parameter's contribution in synergistic analyses. Today, ANOVA is applied as a reliable method, in the vast range of statistically synergistic researches such as applied biology, pharmacy, chemistry and engineering. In the corrosion field mostly, ANOVA has been used to study the interaction of different inhibitors in special medium [28, 29]. In recent years, this approach has also been to investigate the combined effect of mechanical and electrochemical factors on the mechanical corrosion of steels [24, 25].

The second phase of this study, determines the way that corrosion rate varies as a function of individual or synergistic action of parameters. This can be described as "qualitative effect" of parameters.

These two stages (quantitative and qualitative analyses) can provide knowledge that can lead to comprehensive study of carbon steel's corrosion behaviour and also finding the most effective parameters under the real marine condition, in which, the determinant factors act quite simultaneously.

The complete systematic information regarding the effect of simultaneous action of affecting parameters is lacking and this study intends to perform the complete and clear view of combined effect of determinant factors by using both two quantitative and qualitative methods. This kind of investigation is really worthy in industrial designs that there is a great tendency to understand the real behaviour of materials which are supposed to be used in specific applications, under simultaneous action of different parameters.

2. Materials and Methods

2.1. Materials, Solution Preparation and Electrochemical Measurement

Specimens used in the polarization resistance measurements were cut from a carbon steel 1045 cylindrical rod of 12 millimetre diameter. The chemical composition of the samples is (C:0.46 wt%, Mn: 0.65 wt%, Si:0.4 wt%, Ni:0.4 wt% and Fe: margin). Surface preparation was carried out by emery paper up to 1200 grits. Afterward, all the samples were rinsed by distilled water, degreased by ethanol and dried by cool air. The corrosive environment was seawater prepared according to ASTM D1141 [30, 31]. The pH of solutions was adjusted with 0.1 Normal sodium chloride solution [30, 31]. The temperature and fluid velocity of system were fixed by the hot plate HP-840 and Heidolph RZR 2021 respectively. Salinity (the total weight in grams of solid matter dissolved in 1000 grams of water) was set and measured according to the value of solution chlorinity (the total weight in grams of chloride ion in 1000 grams of solution) [7,10, 32, 33]. The exact relation of salinity and chlorinity is as Eq. (1).

Salinity= 1.805 chlorinity +0.03   Eq. (1)

To adjust the specific salinity, the relative value of chlorinity was calculated according to Eq. (1). Then the mentioned chlorinity was determined by changing the amount of NaCl as the main component of seawater, while all the other spices were kept fixed. In order to minimize the error of concentration variation by the formation of deposits, a 1000 ml electrochemical cell was used [34].

Corrosion rate data were obtained using a standard three-electrode arrangement in conjunction with μ Autolab type III/FRA2 electrochemical interface potentiostat- galvanostats and the GPES version 4.9 software program. A saturated calomel electrode and platinum electrode were used as reference and counter electrodes, respectively. Before starting the experiments, the specimens were left in the test solution for about 1 hour to attain a steady state condition. The current-potential curves of carbon steel electrodes in seawater solution were measured in the region of approximately ±100 mV around E_{corr}. The scan rate of applied potential was 0.05 mV/sec [35]. Corrosion rates were calculated via the Tafel extrapolation method [36].

2.2. Experimental Condition

Temperature, velocity, pH and salinity were different variables in this experiment. If each variable has two levels, there will be 2^4 trials [7]. Different values for each variable were as follows: Temperature (23°C and 65°C), velocity (0 rpm and 100 rpm), pH (7.5 and 10.5) and salinity (30 and 40). Three tests were performed for each condition to be sure about the validity of the tests. According to the full two-level factorial experimental analysis (the metho\

considered in this research), each parameter should be applied in both high and low intense conditions [24]. Table 1 illustrates possible conditions considering four parameters in both levels.

This set of tests was performed to investigate the synergistic effect of salinity, pH, temperature and velocity. Both qualitative and quantitative analyses were carried out on the results. Test conditions were determined according to Table 1. The values of C_1, C_2,..C_n are related to corrosion rates for condition 1, 2, ..., n. In this table, "1" and "2" stand for the lower and higher levels of corresponding parameters, respectively. The parameters are defined as follows:

(S: Salinity); (T: Temperature); (V: Velocity); (p: pH); (C: Corrosion rate).

3. Results

The polarization curves were used to calculate the value of corrosion rates. The polarization curves for all 16 trials, as are defined in Table 1, have been shown in Fig. 1 to Fig. 4. In all the tests, corrosion rate has been calculated in mm/yr through Tafel extrapolation method and corresponding values are listed in Table 2.

4. Discussion

4.1. Quantitative Analysis

This analysis was performed by ANOVA method which is one of the most common methods to explain the individual and combined effect of different parameters [37]. In this research the effect of each parameter on corrosion rate was calculated both individually and synergistically and presented as contribution percentages. The analysis' approach is described in the following order.

4.1.1. Average Effect of Parameters in Low and High Levels

As the first step of quantitative analysis, the average effect of parameters was computed at high and low levels. Average values and their calculation method have been presented in Table 3.

C_{ln}: The average effect of parameter, n, in low level

C_{hn}: The average effect of parameter, n, in high level

As an example, C_{lV} (the average effect of velocity in low level), corresponds to the average value of corrosion rate in the conditions that velocity is applied at lower level. These conditions, according to Table 1, are: 1, 2, 5, 6, 9, 10, 13 and 14.

So, C_{lV}, as the average corrosion rate in these conditions, can be calculated as Eq. (2) [24, 26, 27].

$$C_{lV} = 1/8\ (\ C_1 + C_2 + C_5 + C_6 + C_9 + C_{10} + C_{13} + C_{14}\ ) \qquad \text{Eq. (2)}$$

4.1.2. Sums of Squares

In this stage, that is the second stage of ANOVA analysis, sums of squares for all the individual parameters, as well as their interactions, have been calculated. The number of total possible interactions can be calculated as Eq. (3) [24, 26, 27].

$$\sum_{k=1}^{n}\binom{n}{k} \qquad \text{Eq. (3)}$$

n: Total number of considered parameters.

Since we have taken four parameters (n = 4), the total number of possible interactions would be 15. The values for sums of squares and their calculating equations are presented as Table 4.

SS_n : sums of squares, corresponding to the parameter "n".

C_g: The average value of corrosion rate, in all of 16 trials.

In the cases, which "n" represents the complex index, it stands for the sums of squares, corresponding to the combined action of parameters that have been presented in the index. For example SS_{Vp} stands for the sums of squares, corresponding to the combined action of velocity (V) and pH (p) [24, 26, 27].

4.1.3. Contribution Percentage

In the last step, the contribution percentages of parameters on variation of the average value of corrosion rate have been calculated, in both individually and synergistically manners.

The calculation formula is shown in Eq. (4) [24, 26, 27].

$$K (\%) = SS_K / (SS_V + SS_S + SS_P + SS_T + SS_{VS} + SS_{PS} + SS_{VP} + SS_{ST}$$
$$+ SS_{PT} + SS_{VT} + SS_{SVT} + SS_{VPT} + SS_{PST} + SS_{VSP} + SS_{VPST}) \times 100\% \qquad \text{Eq. (4)}$$

"K" is the parameter, that its contribution percentage is being calculated.

For example "SVT%", stands for the contribution percentage, resulting from the synergistically action of salinity (S), velocity (V) and temperature (T). The results of contribution percentage of parameters have been gathered in Table 5.

4.1.4. Quantitative Analysis of Results

According to the result of Table 5, as we consider the individual effect of parameter, we would come up to the following results:

- The individual effect of parameters, are all less than 6%. This shows the pronounced effect of synergistically action of parameters, compared to the individually acting effect of them.

- Among all the individual effects, pH and temperature have the least contribution percentages. Also, considering the dual action of parameters, the factor "pT" which corresponds to the combined action of pH and temperature has the least contribution. So, we can conclude that, pH and temperature have the least effect on varying the average rate of corrosion rate, compare to the salinity and velocity.

- Considering the ternary action of factors, the parameter "SVT" (synergistically action of salinity, velocity and temperature), "VpT" (synergistically action of velocity, pH and temperature) and "pST" (synergistically action of pH, salinity and temperature), have the largest values of contribution.

- The largest value of contribution belongs to "VpST", that is as a result of combining action of all four parameters.

4.2. Qualitative Analysis of Results

In spite of the undeniable importance of quantitative analysis in clarifying the contribution percentage of parameters on changing the average rate of corrosion, its weakness in defining the direction of this variation is still a big limit. As the quantitative analysis takes the advantage of ANOVA and considers the sums of squares in the calculation, all the variations in the corrosion rate has been applied with the power of two in the calculations and the calculation procedure is not really capable of predicting the direction of this variation.

So, we certainly need another method of data analysing to show the way that each parameter can affect on the rate of reference corrosion, as well as the interaction of parameters. In other word, we want to answer the question that, if the increase in considered parameter can lead to the increase in the rate of corrosion or not. This aspect, it's the real reason of applying the qualitative analysis, which has been done as a complementary method to cover the disadvantages of quantitative analysis.

4.2.1. Method of Qualitative Analysis

In order to perform the qualitative analysis, a 16 x 16 matrix has been designed that all the effects of parameters in both individual and combined manner have been taken into consideration [26, 27]. In this analysis the corrosion rate in trial 1, has been considered as a reference. The experimental condition of trial 1 has been defined in Table 1. Since all the parameters has been applied at their low level in the trial 1, the comparison of other corrosion rates with this reference corrosion rate, can provide the possibility of understanding the effect of increasing each parameters and also their interactions as is described in detail, in the following parts.

In order to calculate the arrays in the first column of matrix, corrosion rates in all the trials have been compared with the reference corrosion rate (corrosion rate of trial 1) [26, 27].

In each array, the corrosion rate of trials, mentioned in the indices of array, have been compared together. For example, in the array "$A_{1,2}$" the corrosion rate of trials 1 and 2 have been compared. Sine these trials' conditions, as is defined in the Table 1, differ only on the level of temperature, this comparison, results to the configuration of temperature effect on the corrosion rate. As the corrosion rate in the trial 2 (C_2) is larger than that of in trial 1 (C_1) we have concluded that, temperature has an increasing effect on the corrosion rate. All the other arrays in the first column

have been calculated with the same procedure. In all cases, ">0" represents the increasing effect of parameter on the corrosion rate, while "<0" represents the decreasing effect of parameter on the corrosion rate and the combined formats, such as "pT", represents the effect of simultaneous increase in the mentioned parameters (in this case: simultaneous increase in both pH and temperature).

At the next stage of this analysis the values of $|C_y - C_1|$ and $|C_x - C_1|$ have been calculated and compared. The result of this comparison has been presented as $A_{x,y}$.

C_x : Corrosion rate in trial "x"

C_y: Corrosion rate in trial "y"

For example, in the calculation of $A_{3,2}$, $|C_2 - C_1|$ and $|C_3 - C_1|$ have been compared. As it was described before, $|C_2 - C_1|$ is related to $A_{2,1}$ and shows the effect of temperature. In the same way, $|C_3 - C_1|$ shows the effect of velocity. Since $|C_2 - C_1| > |C_3 - C_1|$, the effect of temperature on variation of corrosion rate is larger than velocity and is shown as "T>V".

To analyse other arrays and investigate the effect of environmental parameters and their effects on each other, three groups of arrays were defined. First group consists of arrays that show the synergistic effect of parameters on corrosion rate. For example, if we consider T<SVT, it can be calculated according to what explained before. Conditions 1 and 8 are different in the level of salinity, velocity and temperature and $|C_8-C_1|$ shows the effect of "SVT" which is the synergistic effect of salinity, velocity and temperature. Conditions 1 and 2 are different in level of temperature and $|C_2-C_1|$ shows the effect of temperature. If $|C_8-C_1|>|C_2-C_1|$, then T<SVT [24, 25]. According to this result, temperature has less effect on corrosion rate compared to overall salinity, velocity and temperature. It means that in this case the synergistic effect of parameters has increased corrosion rate. Arrays which belong to this group have been specified by a circle.

The second group consists of arrays which show the decreasing effect of parameters on corrosion rate when they act together. If we consider pST<T, it means that salinity, temperature and pH when combined have less effect on corrosion rate in comparison with temperature individually. In this case synergistic effect has reduced corrosion rate. Arrays belong to this group are defined with a triangle sign.

The third group show the arrays that indicate the venial effect of combination of parameters on corrosion rate. As an instance, p ≈ pS means that the combination of salinity and pH has approximately similar effect on corrosion rate in comparison with pH itself. Arrays in this group have been shown with a square sign.

The changes applied in these parameters are not comparable because they have different units. This analysis is not able to suggest a value for the effect of parameters on corrosion rate and is

just used for prediction of corrosion rate change under different conditions and evaluation of synergistic effect of parameters.

4.2.2. Qualitative Analysis of Results

The mentioned qualitative matrix, for the samples, has been presented in Table 6. According to the results in the first column of Table 6, pH is the only single parameter that has decreased the rate of corrosion (p<0). It means that, increasing the pH, decreases corrosion rate whereas other parameters increase corrosion rate, individually. In this steel, pH increment improves corrosion resistance by formation of protective layers. As seen in Eq. (5), the higher the pH, less corrosion rate is expected.

Surface layers influence limiting current density and consequently corrosion rate. This effect is shown in Eq. (5) [2].

$$i_L = z\,F\,(C_B / (\delta/D_{O2}) + (t_B/P_B)) \hspace{3cm} \text{Eq. (5)}$$

i_L: Limiting current density; D_{O2}: Diffusion coefficient of oxygen; Z: Fluid viscosity; F: Faraday number; C_B: Oxygen concentration in solution; δ: Diffusive layer thickness; P_B: Oxygen permeability; t_B: Thickness of scale layer.

By increasing pH, surface layers are formed and corrosion rate decreases. This surface layers are not conductive therefore, cathodic reaction is not performed on the surface and oxygen atoms have to pass this layers to reach the metal surface. In fact, surface layers act like a barrier against oxygen diffusion and lead to decreasing the corrosion rate. The effect of other parameters can be explained according to previous theories. Temperature factor enhancement helps corrosion rate increase. Temperature increases the electrochemical reactions rate.

Corrosion rate of carbon steels in seawater is controlled by cathodic reaction (Oxygen reduction) rate. The effect of temperature on corrosion rate can be adjusted by considering its effect on limiting current density. Eq. (6) shows effective parameters on limiting current density on a surface without precipitate layers [2].

$$i_L = D_{O2}\,z\,F\,(C_B/\delta) \hspace{4cm} \text{Eq. (6)}$$

Z: Fluid viscosity; C_B: Oxygen concentration in solution; δ: Diffusive layer thickness.

Oxygen diffusion coefficient is under the effect of temperature in this case according to Eq. (6) [2].

$$D_{O2} = A \exp(-Q/RT) \hspace{4cm} \text{Eq. (7)}$$

A, Q: Is supposed to be constant in aqueous environment; R: Universal gas constant; T: Temperature (Kelvin)

According to this equation, oxygen diffusion coefficient is proportional to temperature and increases by the temperature. Diffusion layer thickness is dependent on temperature as well and by increasing the temperature, the thickness is decreased. Simultaneous effect of temperature on oxygen diffusion coefficient and diffusion layer thickness will result in corrosion rate increment according to Eq. (6).

In this steel, higher fluid velocities will accelerate corrosion rate as a result of reaching more oxygen to cathode surface. From another point of view, higher fluid velocity can remove surface precipitates. In a phenomenon that is controlled by diffusion, the relationship between corrosion rate and fluid velocity is introduced by Eq. (8) [2].

$$i_{corr} = i_L \propto V^{0.5} \hspace{4cm} \text{Eq. (8)}$$

icorr: Corrosion rate; V: Linear fluid velocity

According to the Eq. (8), by increasing the fluid velocity, limiting current density and consequently corrosion rate will increase.

Last parameter is salinity. In this case, increasing of salinity prevents precipitates layers to be formed and increases solution's conductivity which causes the increment of corrosion. All the arrays that show the simultaneous effect of pH with temperature or velocity (pT,pV, VpT, pST, VpS, VpST), will accelerate the rate of corrosion. It shows that although pH increment decreases corrosion rate, its synergistic effect with temperature and fluid velocity will result in corrosion rate growth. In other word, temperature and velocity have been able to dominate the effect of pH.

Another example for synergistic effect of parameters is decelerating role of salinity and pH together according to array $A_{13,1}$ (pS<0). Salinity increment accelerates corrosion rate according to array $A_{5,1}$ (S>0) but for pH, it will result in corrosion rate decrement. If pH and salinity be considered together, the dominant effect will be pH and will reduce corrosion rate. Therefore, in contrast with fluid velocity and temperature, salinity is not able to conquer the effect of pH. According to the mentioned examples, it can be concluded that quantitative analysis is not able to predict and justify corrosion rate change pattern without qualitative analysis.

5. Conclusion

1. Both quantitative and qualitative analyses have been applied to consider the synergistic effect of different parameters on the rate of corrosion. Quantitative analysis calculates the contribution percentage of parameters on changing the average rate of corrosion whiles the qualitative one, represents the variation direction of reference corrosion rate as a result of individual or synergistic action of parameters.

2. According to the results of quantitative analysis, salinity and pH have the least value of contribution percentage on varying the average value of corrosion rate, among all the single acting parameters. In the same way, the simultaneous action of salinity and pH shows the least contribution percentage among the considered dual acting parameters.

3. The simultaneous action of all parameters has the largest contribution percentage.

4. pH is the only single acting parameter that applies a decreasing effect on the rate of corrosion , all the other parameters increases the rate of corrosion individually.

5. The effect of parameters can be moderated, strengthened or weakened by the simultaneous action of other parameters. In most cases, the parameters with the larger value of contribution percentage according to the quantitative analysis play a dominant role in qualitative analysis and control the variation direction of reference corrosion rate.

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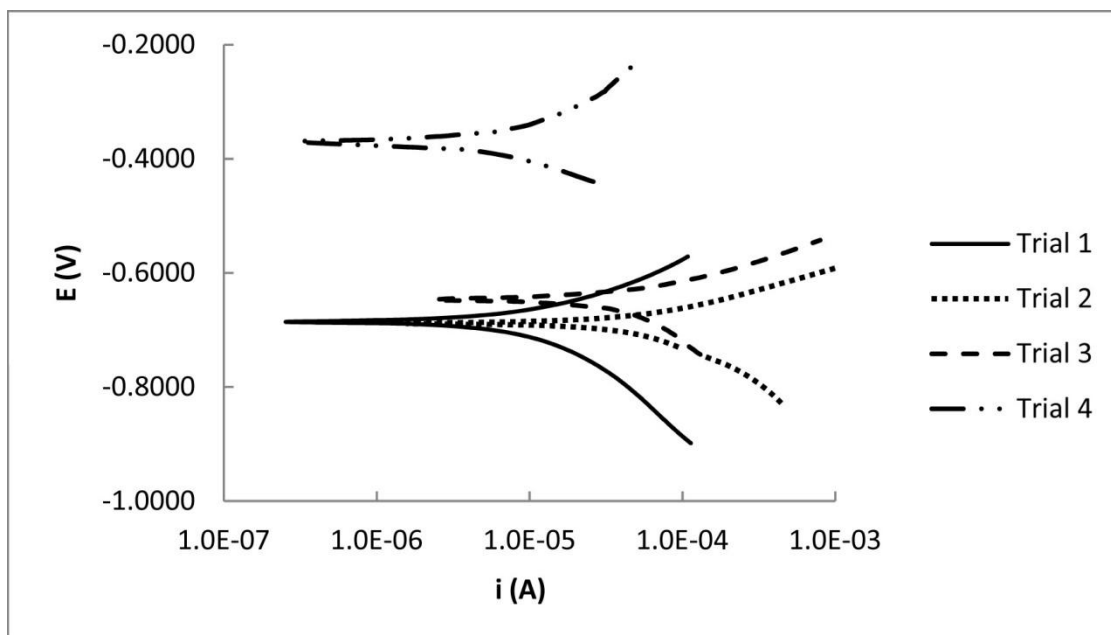[37] J. Zar, *Biostatistical Analysis*, fourth ed., Prentice Hall, New Jersey, USA, 1999.

Fig. 1. Potentiodynamic polarization curves of trials 1 to 4.
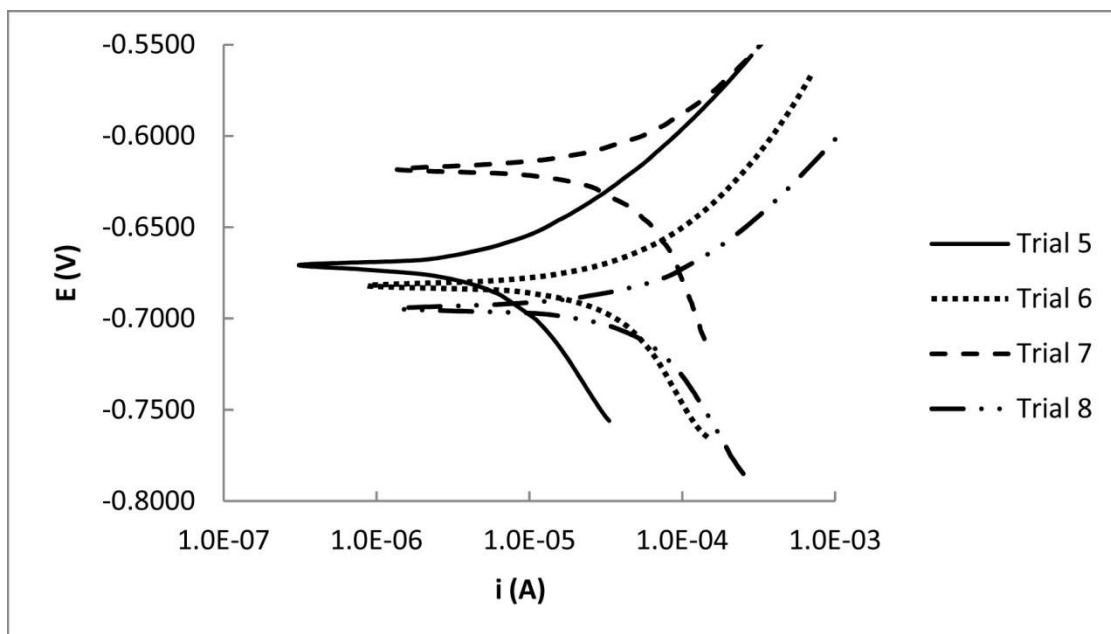

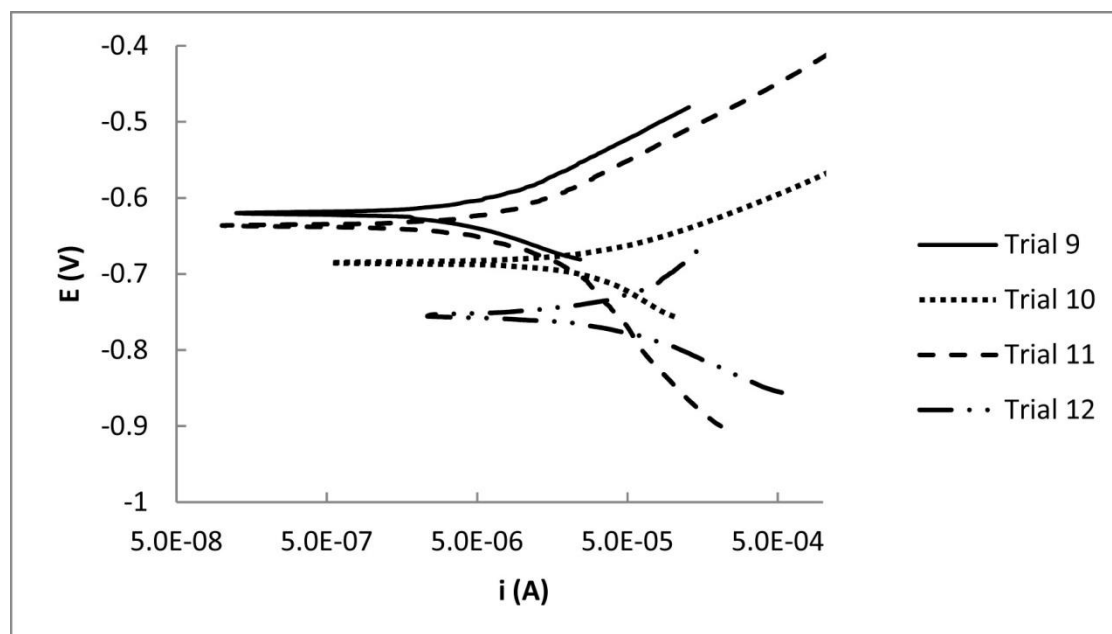**Fig. 2. Potentiodynamic polarization curves of trials 5 to 8.**

Fig. 3. Potentiodynamic polarization curves of trials 9 to 12.
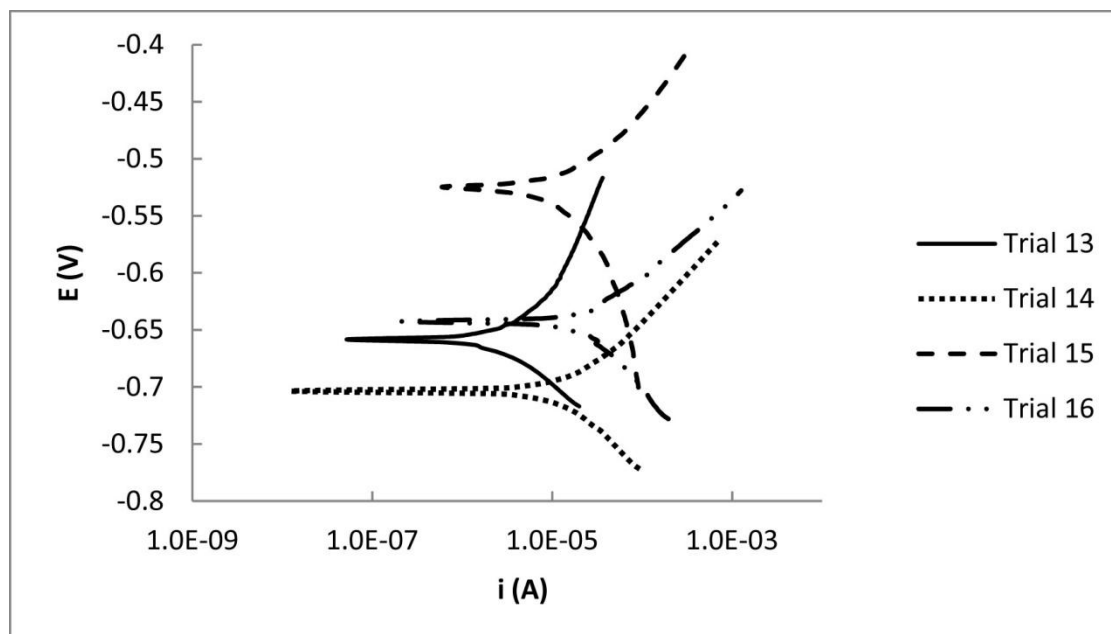

Fig. 4. Potentiodynamic polarization curves of trials 13 to 16.

Table 1. Experimental conditions and corresponding corrosion rates for all the considered trials

Test number	Temperature	Velocity	Salinity	pH	Corrosion rate
1	1	1	1	1	C_1
2	2	1	1	1	C_2
3	1	2	1	1	C_3
4	2	2	1	1	C_4
5	1	1	2	1	C_5
6	2	1	2	1	C_6
7	1	2	2	1	C_7
8	2	2	2	1	C_8
9	1	1	1	2	C_9
10	2	1	1	2	C_{10}
11	1	2	1	2	C_{11}
12	2	2	1	2	C_{12}
13	1	1	2	2	C_{13}
14	2	1	2	2	C_{14}
15	1	2	2	2	C_{15}
16	2	2	2	2	C_{16}

Table 2. Corrosion rate in all 16 trials.

Parameter	C_1	C_2	C_3	C_4
Corrosion rate (mm/year)	2.263E-2	3.434E-1	1.237E-1	3.325E-2

Parameter	C_5	C_6	C_7	C_8
Corrosion rate (mm/year)	4.061E-2	2.392E-1	1.236E-1	3.268E-1

Parameter	C_9	C_{10}	C_{11}	C_{12}
Corrosion rate (mm/year)	1.592E-2	8.815E-2	1.088E-1	9.240E-2

Parameter	C_{13}	C_{14}	C_{15}	C_{16}
Corrosion rate (mm/year)	6.800E-3	5.655E-2	3.174E-1	1.683E-1

Table 3. Average effect of parameters in low and high levels.

Parameter	Calculation formula	Value
(C_{IV})	$1/8(C_1+C_2+C_5+C_6+C_9+C_{10}+C_{13}+C_{14})$	1.017E-1
(C_{hV})	$1/8(C_3+C_4+C_7+C_8+C_{11}+C_{12}+C_{15}+C_{16})$	1.618E-1
(C_{IS})	$1/8(C_1+C_2+C_3+C_4+C_9+C_{10}+C_{11}+C_{12})$	1.035E-1
(C_{hS})	$1/8(C_5+C_6+C_7+C_8+C_{13}+C_{14}+C_{15}+C_{16})$	1.599E-1
(C_{IP})	$1/8(C_1+C_2+C_3+C_4+C_5+C_6+C_7+C_8)$	1.566E-1
(C_{hP})	$1/8(C_9+C_{10}+C_{11}+C_{12}+C_{13}+C_{14}+C_{15}+C_{16})$	1.068E-1
(C_{IT})	$1/8(C_1+C_3+C_5+C_7+C_9+C_{11}+C_{13}+C_{15})$	9.493E-2
(C_{hT})	$1/8(C_2+C_4+C_6+C_8+C_{10}+C_{12}+C_{14}+C_{16})$	1.685E-1

Table 4. Sums of squares for all the possible interactions, values and calculation formula.

| Parameter	Calculation formula	Value
SS_V	$2(C_{lv}-C_g)^2+2(C_{hv}-C_g)^2$	3.615E-3
SS_S	$2(C_{ls}-C_g)^2+2(C_{hs}-C_g)^2$	3.178 E-3
SS_p	$2(C_{lp}-C_g)^2+2(C_{hp}-C_g)^2$	2.486 E-3
SS_T	$2(C_{lT}-C_g)^2+2(C_{hT}-C_g)^2$	5.413 E-3
SS_{VS}	$2(C_{lv}-C_g)^2+2(C_{hv}-C_g)^2+2(C_{ls}-C_g)^2+2(C_{hs}-C_g)^2$	6.793 E-3
SS_{Vp}	$2(C_{lv}-C_g)^2+2(C_{hv}-C_g)^2+2(C_{lp}-C_g)^2+2(C_{hp}-C_g)^2$	6.101 E-3
SS_{pS}	$2(C_{lp}-C_g)^2+2(C_{hp}-C_g)^2+2(C_{lS}-C_g)^2+2(C_{hS}-C_g)^2$	5.664 E-3
SS_{ST}	$2(C_{lS}-C_g)^2+2(C_{hS}-C_g)^2+2(C_{lT}-C_g)^2+2(C_{hT}-C_g)^2$	8.591 E-3
SS_{pT}	$2(C_{lp}-C_g)^2+2(C_{hp}-C_g)^2+2(C_{lT}-C_g)^2+2(C_{hT}-C_g)^2$	7.899 E-3
SS_{VT}	$2(C_{lv}-C_g)^2+2(C_{hv}-C_g)^2+2(C_{lT}-C_g)^2+2(C_{hT}-C_g)^2$	9.028 E-3
SS_{SVT}	$2(C_{ls}-C_g)^2+2(C_{hs}-C_g)^2+2(C_{lv}-C_g)^2+2(C_{hv}-C_g)^2$ $+2(C_{lT}-C_g)^2+2(C_{hT}-C_g)^2$	1.221 E-2
SS_{VpT}	$2(C_{lv}-C_g)^2+2(C_{hv}-C_g)^2+2(C_{lp}-C_g)^2+2(C_{hp}-C_g)^2$ $+2(C_{lT}-C_g)^2+2(C_{hT}-C_g)^2$	1.151 E-2
SS_{pST}	$2(C_{lp}-C_g)^2+2(C_{hp}-C_g)^2+2(C_{lS}-C_g)^2+2(C_{hS}-C_g)^2$ $+ 2(C_{lT}-C_g)^2+2(C_{hT}-C_g)^2$	1.108 E-2
SS_{VSp}	$2(C_{lv}-C_g)^2+2(C_{hv}-C_g)^2+2(C_{lS}-C_g)^2+2(C_{hS}-C_g)^2$ $+ 2(C_{lp}-C_g)^2+2(C_{hp}-C_g)^2$	9.279 E-3
SS_{VpST}	$2(C_{lv}-C_g)^2+2(C_{hv}-C_g)^2+2(C_{lp}-C_g)^2+2(C_{hp}-C_g)^2$ $+2(C_{lS}-C_g)^2+2(C_{hS}-C_g)^2+2(C_{lT}-C_g)^2+2(C_{hT}-C_g)^2$	1.469 E-2

Table 5. Contribution percentage of parameters

Parameter	V	S	p
Contribution %	3.076	2.704	2.115
Parameter	T	VS	Vp
Contribution %	4.605	5.780	5.190
Parameter	pS	ST	pT
Contribution %	4.819	7.309	6.720
Parameter	pST	VSp	VpST
Contribution %	9.424	7.895	12.500

Table 6. A symmetric matrix of qualitative effect

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
1																
2	T>0															
3	V>0	T>V														
4	VT>0	▲T>VT	▲V>VT													
5	S>0	T>S	V>S	VT≈S												
6	ST>0	▲T>ST	V<ST	VT<ST	•S<ST											
7	SV>0	T>SV	▪V≈SV	VT<SV	•S<SV	ST>SV										
8	SVT>0	▪T≈SVT	•V<SVT	•VT<SVT	•S<SVT	•ST<SVT	•SV<SVT									
9	P<0	*T>p	*V>P	**VT≈p	**S≈p	*ST>p	*SV>p	*SVT>p								
10	pT>0	▲T>pT	V≈pT	VT<pT	S≈pT	ST>pT	SV≈pT	SVT>pT	••*P<pT							
11	pV>0	T>pV	▪V≈pV	VT<pV	S<pV	ST>pV	SV≈pV	SVT>pV	••*P<pV	pT≈pV						
12	VpT>0	▲T>VpT	▪V≈VpT	•VT<VpT	S<VpT	ST>VpT	SV≈VpT	SVT>VpT	••*P<VpT	▪pT≈VpT	▪pV≈VpT					
13	pS<0	*T>pS	*V>pS	**VT≈pS	▪**S≈pS	*ST>pS	*SV>pS	*SVT>pS	▪p≈pS	**pT≈pS	*pV>pS	*VpT>pS				
14	pST>0	▲T>pST	V>pST	VT≈pST	▪S≈pST	▲ST>pST	SV>pST	SVT>pST	▪**p≈pST	▪pT≈pST	pV>pST	VpT≈pST	▪**pS≈pST			
15	VpS>0	T≈VpS	•V<VpS	VT<VpS	•S<VpS	ST<VpS	•SV<VpS	SVT≈VpS	••*P<VpS	pT<VpS	•pV<VpS	VpT<VpS	••*pS<VpS	pST<VpS		
16	VpST>0	▲T>VpST	●V≈VpST	•VT<VpST	•S<VpST	▲ST>VpST	▪SV≈VpST	▲SVT>VpST	••*P<VpST	•pT<VpST	•pV<VpST	•VpT<VpST	••*pS<VpST	•pST<VpST	▲VpS>VpST	

Legend:
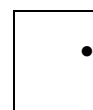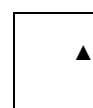
- • Increasing effect of combined action
- ▲ Decreasing effect of combined action
- ▪ Venial effect of combined action
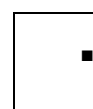
*Absolute effects are compared, although they cause reverse impact.   **The parameters cause reverse effect, with equal magnitude.