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Comparative Studying of Marine Parameters' Effect, via Qualitative Method

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Abstract: In this study the marine corrosion resistivity of the austenitic stainless steels type 304 and 316 has been studied qualitatively. This study is under the synergistic effect of environmental factors. Salinity, velocity, pH and temperature are the factors which induce effects. Corrosion rates are calculated in aerated synthetic seawater, via potentiodynamic polarization scan. Qualitative analysis has been used to predict the variation direction of corrosion rate while applying the most important marine parameters, synergistically. This analysis has an important role in marine applications and predicts the individual effect of considered parameters, as well as their synergistic action effect on the rate of corrosion.

Key words: Environmental parameters, marine corrosion, synergistic effect, qualitative analysis

INTRODUCTION

Seawater is a kind of corrosive environment and its importance has increased during the last few decades because of offshore exploration of oil and gas (Bardal, 2004). It threats all materials to some extent such as bridges, wharfs, platforms pipeline systems and structures immersed in seawater (Duan *et al*., 2008; Shifler, 2005; Melchers, 2005; Möller, 2006). Types 304 and 316 stainless steels are the most widely used of any stainless steels in marine applications and seawater is the most challenging environment for them (Schumacher, 1979; Jones, 2001; Schweitzer, 2007). Some parameters such as seawater temperature, flow velocity, pH and salinity, play the most important role in the corrosivity of seawater (Cristiani *et al*., 2008; Al-Fozan and Malik, 2008; Ozturk and Grubb, 2005). These factors influence on the corrosion rate by effecting the value of dissolved oxygen or stability of the passive layer. Temperature and flow velocity affect the corrosion rate by affecting the oxygen transport to the metal surface (Sasaki and Burstein, 2007; Bardal, 2004). The corrosion behavior of these two alloys in seawater is under the cathodic control and any action which affects the rate of oxygen delivery can have a marked effect on the rate of attack. Oxygen influences the corrosion of stainless steels in seawater by two methods. First by maintaining the passivity of stainless steels and then, by acting as the depolarizer for passive-active cells, created by the breakdown of passivity at a specific point or area. The chloride ion, present in seawater, is singularly efficient in accomplishing this breakdown. (Schumacher, 1979). On the other hand, variation in corrosion rate is also caused by salinity and pH, because they strongly affect the barrier properties of surface layers (Bardal, 2004). These concepts can describe the individual effect of parameters, while corrosion generally, is a statistical effect governed by

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several variables such as environmental ones (Ekuma and Idenyi, 2007) and an alteration in one parameter can affect the relative effect of others (Schumacher, 1979). In recent years some research has been done on the individual effects of environmental parameters (Melchers, 2006; Mor and Beccaria, 2004; Schleich *et al*., 2008; Al-Malahy and Hodgkiess, 2003; Sefaja and Malina, 1985), but the importance of the synergistic effect, has not been evaluated completely. In this research the synergistic effect of marine parameters has been evaluated synergistically, which can result in predicting the variations applied on the corrosion rate under the synergistic action of considered parameters.

MATERIALS AND METHODS

Study Area

Experiments were conducted in the Corrosion Laboratory of Materials Engineering Department of Shiraz University (Iran) from November 2008 to October 2009.

Solution Preparation

Experimental solution of seawater was prepared according to ASTM D1141 (1999). The pH of solution, fixed by 5 molar sodium hydroxide solution (Zamanzade *et al*., 2007). In order to prevent variations in species' concentrations during the formation of deposits, a large volume glass electrochemical cell (1000 mL) was used (Zamanzade *et al*., 2007). The temperature was set by the hot plate HP-840 and the fluid velocity was fixed by the mechanical stirrer, Heidolph RZR 2021. Salinity (the total weight in grams of solid matter dissolved in 1000 g of water) determined by measurement of chlorinity in the seawater. Equation 1 shows the exact relation (Schumacher, 1979; Roberge, 2000):

$$\text{Salinity} = 1.805\ \text{chlorinity} + 0.030 \quad (1)$$

To achieve the desired salinity, the chloride-ion content was adjusted by changing the quantity of sodium chloride as the main part of seawater composition, while the value of other spices was fixed according to standard.

Sample Preparation

This study examined the corrosion rate of commercially supplied austenitic stainless steels type 304 and 316 specimens. The nominal bulk composition of used materials is shown in Table 1. These alloys were obtained in rod form (12 mm diameter). The samples were first embedded in epoxy resin then they polished successively with metallographic emery paper for increasing fineness of up to 1200 grits. Then they were washed by distilled water, digressed by ethanol and dried by cool air.

Electrochemical Method

A Standard Calomel Electrode (SCE) was used as the reference electrode and Pt gauze as the auxiliary. The samples were tested exactly after being immersed for 5 h in the solution of controlled cell. Electrochemical experiments were performed using a μ Autolab type III/FRA2 electrochemical interface potentiostat-galvanostats. For potentiodynamic

Table 1: Nominal compositions of SS 304 and 316 (wt.%, Fe margin)

Elements	Cr	Ni	Mn	N	S	C	Si	P	Mo
SS 304	17	8	2	0.10	0.03	0.15	0.75	0.045	---
SS 316	18	14	2	0.10	0.03	0.08	0.75	0.045	3.0

Table 2: Design matrix for a four-parameter, sixteen-run experiment

Trial	pH	Salinity	Velocity	Temperature	Corresponding corrosion rate
1	1[a]	1	1	1	C_1
2	1	1	1	2	C_2
3	1	1	2	1	C_3
4	1	1	2	2	C_4
5	1	2	1	1	C_5
6	1	2	1	2	C_6
7	1	2	2	1	C_7
8	1	2	2	2	C_8
9	2[a]	1	1	1	C_9
10	2	1	1	2	C_{10}
11	2	1	2	1	C_{11}
12	2	1	2	2	C_{12}
13	2	2	1	1	C_{13}
14	2	2	1	2	C_{14}
15	2	2	2	1	C_{15}
16	2	2	2	2	C_{16}

[a]: The number 2 in this matrix represents high level of all parameters, while 1 represents low level

polarization experiments the potential was scanned from about -0.3 to +0.3 V across the corrosion potential to achieve the linear range and the scan rate was 0.005 mV sec^{-1} to maintain a stable corrosion potential and a steady-state behavior (Perez, 2004). The instruments were controlled by the GPES version 4.9 software program. The corrosion rates, in millimeters per year were determined using Tafel extarapolarization method (Yang, 2008).

Analysis Method

Salinity, pH, temperature and velocity were the parameters considered in this research. Each parameter was set at two levels, so there were $2\times2\times2\times2 = 16$ trials (Meng *et al*., 2007). The salinity was set at 30 and 40; pH trials were at 7.5 and 10.5, velocities were adjusted at 0 and 1000 rpm and temperature was fixed at 23 and 65°C. These ranges were defined according to the variation range of the environmental parameters in the most natural seawater. For each condition three tests were carried at least, to make sure about the reproducibility of the results. Each parameter applied in both high and low intense conditions. All the possible trials, which satisfy this phenomenon, are shown in Table 2. Table 2, each parameter is considered in both low and high levels shown by 1 or 2 signs and all the possible trials resulted from the combined action of parameters in each of the low or high levels are also gathered. For example, trial 3 represents the condition in which pH, temperature and salinity are applied in low levels and velocity is in its high level.

RESULTS AND DISCUSSION

Corrosion Rate Results

The corrosion rates in millimeters per year for different conditions were determined by Tafel Extarapolarization Method and can be seen in the Table 3 and 4. C_n relates to the corrosion rate of trial condition n. The condition, applied to trial condition n, is shown in Table 2.

Qualitative Analysis of Data

Qualitative analysis can describe the way that these factors influence the rate of corrosion or change the direction of its variations. In this research, the quality of mentioned environmental effects was survived individually and synergistically using data in Table 2-4, then they gathered in Table 5 and 6. The symmetric 16×16 matrixes were designed. Each array represents the result that was obtained by comparing the corrosion rates corresponding to

Table 3: Corrosion rate results gathered in trial conditions for SS304

Parameter	C_1	C_2	C_3	C_4	C_5	C_6	C_7	C_8
Corrosion rate (mm $year^{-1}$)	1.048E-3	3.762E-3	5.121E-4	4.303E-3	4.676E-4	3.311E-3	1.029E-3	1.480E-3
Parameter	C_9	C_{10}	C_{11}	C_{12}	C_{13}	C_{14}	C_{15}	C_{16}
Corrosion rate (mm $year^{-1}$)	2.371E-4	7.060E-2	2.511E-2	4.736E-3	1.325E-5	1.761E-3	1.944E-3	6.119E-2

Table 4: Corrosion rate results gathered in trial conditions for SS316

Parameter	C_1	C_2	C_3	C_4	C_5	C_6	C_7	C_8
Corrosion rate (mm $year^{-1}$)	6.472E-4	1.122E-2	3.392E-4	3.112E-3	8.474E-4	2.650E-3	7.862E-4	5.568E-2
Parameter	C_9	C_{10}	C_{11}	C_{12}	C_{13}	C_{14}	C_{15}	C_{16}
Corrosion rate (mm $year^{-1}$)	2.969E-4	2.424E-2	1.027E-3	7.908E-2	3.805E-4	1.184E-3	3.638E-3	1.637E-2

Table 5: A symmetric matrix of qualitative effect for SS304

No.	1	2	3	4	5	6	7	8
1								
2	T>0							
3	V<0							
4	VT>0	T≈VT■	[a]V<VT●					
5	S<0	[a]T>S	V≈S	[a]VT>S				
6	ST>0	T>ST▲	[a]V<ST	VT>ST	[a]S<ST●			
7	SV≈0	T>SV	V>VS▲	VT>SV	S>SV▲	ST>SV		
8	SVT≈0	T>SVT▲	V>SVT▲	VT>SVT▲	S>SVT▲	ST>SVT▲	SV≈SVT■	
9	P<0	[a]T>p	V≈P	[a]VT>p	S≈p	[a]ST>p	SV>p	SVT>p
10	pT>0	T<pT●	[a]V<pT	VT<pT	[a]S<pT	ST<pT	SV<pT	SVT<pT
11	pV>0	T<pV	[a]V<pV●	VT<pV	[a]S<pV	ST<pV	SV<pV	SVT<pV
12	VpT>0	T<VpT●	[a]V<VpT●	VT<VpT●	[a]S<VpT	ST<VpT	SV<VpT	SVT<VpT
13	pS<0	[a]T>pS	V≈pS	[*]VT>pS	S≈pS■	[a]ST>pS	SV>pS	SVT>pS
14	pST>0	T>pST▲	[b]V≈pST	VT>pST	[b]S≈pST■	ST>pST▲	SV<pST	SVT<pST
15	VpS>0	T>VpS	[b]V≈VpS■	VT>VpS	[b]S≈VpS■	ST>VpS	SV<VpS●	SVT<VpS
16	VpST>0	T<VpST●	[a]V<VpST●	VT<VpST●	[a]S<VpST■	ST<VpST●	SV<VpST●	SVT<VpST●
No.	9	10	11	12	13	14	15	16
1								
2								
3								
4								
5								
6								
7								
8								
9								
10	[a]p<pT●							
11	[a]p<pV●	pT>pV						
12	[a]p<VpT●	pT>VpT▲	pV>VpT▲					
13	p≈pS■	[a]pT>pS	[a]pV>pS	[a]VpT>pS				
14	[b]p≈pST■	pT>pST▲	pV>pST	VpT>pST	[b]pS≈pST■			
15	[b]P≈VpS■	pT>VpS	pV>VpS▲	VpT>VpS	[b]pS≈VpS■	pST≈VpS		
16	[a]P<VpST●	pT>VpST▲	pV<VpST▲	VpT<VpST●	[a]pS<VpST●	pST<VpST●	VpS<VpST●	

[a]: They have reverse effects and the absolute values of effects are compared, [b]: These two factors have equal effects but in reverse directions. ●: Increasing effect of combining action, ▲: Decreasing effect of combining action, ■: Insignificant effect of combining action

the trials' numbers coming in the index of the mentioned array. The trial condition number 1 was selected as a reference and the other trial conditions were compared two by two for the variation that they caused on the corrosion rate of the reference trial. In detail, to obtain the logical result of the first column of the matrixes, all trials had come in analogy with trial one. For example, to obtain the array $A_{2,1}$ the corrosion rates in trial 2 and 1 were compared. Since, these two trials differed only in the temperature factor, this comparison resulted in surveying the effect of temperature. For example in Table 6, Corrosion rate in trial 2, which represents the higher temperature, is higher in the amount. It shows the accelerating effect of temperature on the rate of corrosion. All other arrays in the first column

Table 6: A symmetric matrix of qualitative effect for SS316

No.	1	2	3	4	5	6	7	8
1								
2	T>0							
3	V<0	[a]T>V						
4	VT>0	T>VT▲	[a]V<VT■					
5	S>0	T>S	[b]V≈S	VT>S				
6	ST>0	T>ST	[a]V<ST	VT ST	S<ST■			
7	SV>0	T>SV	[b]V SV■	VT>SV	S≈SV	ST>SV		
8	SVT>0	T<SVT●	[a]V<SVT●	VT<SVT●	S<SVT●	ST<SVT●	SV<SVT●	
9	P<0	[a]T>p	V≈P	[a]VT>p	[b]S≈p	[a]ST>p	[b]SV≈p	[a]SVT>p
10	pT>0	T<pT●	[a]V<pT	VT<pT	S<pT	ST<pT	SV<pT	SVT>pT
11	pV>0	T>pV	[b]V≈pV■	VT>pV	S≈pV	ST>pV	SV≈pV	SVT>pV
12	VpT>0	●T<VpT	●[a]V<VpT	VT<VpT●	S<VpT	ST<VpT	SV<VpT	SVT<VpT
13	pS<0	[a]T>pS	V≈pS	[a]VT>pS	[b]S≈pS■	[a]ST>pS	[b]SV pS	[a]SVT>pS
14	pST>0	T>pST▲	[b]V≈pST	VT>pST	S≈pST■	ST>pST	SV≈pST	SVT>pST
15	VpS>0	T>VpS	[b]V VpS■	VT<VpS	S<VpS	ST<VpS	SV<VpS	SVT>VpS
16	VpST>0	T<VpST●	[a]V<VpST●	VT<VpST●	S<VpST●	ST<VpST●	SV<VpST●	SVT>VpST▲

No.	9	10	11	12	13	14	15	16
1								
2								
3								
4								
5								
6								
7								
8								
9								
10	[a]P<pT●							
11	[b]P pV■	pT>pV						
12	[a]P<VpT●	pT<VpT●	pV<VpT●					
13	p≈pS■	[a]pT>pS	bpV≈pS	[a]VpT>pS				
14	[b]p≈pST■	pT>pST▲	pV≈pST	VpT>pST	[b]pS≈pST■			
15	[a]P<VpS■	pT>VpS	pV<VpS■	VpT>VpS	[a]pS<VpS■	pST<VpS		
16	[a]P<VpST●	pT>VpST▲	pV<VpST	VpT>VpST▲	[a]pS<VpST●	pST<VpST●	VpS<VpST●	

[a]: They have reverse effects and the absolute values of effects are compared, [b]: These two factors have equal effects but in reverse directions. ●: Increasing effect of combining action, ▲: Decreasing effect of combining action, ■: Insignificant effect of combining action

were computed in the same manner. The effects were shown >0 while having the accelerating effect and <0 while having the inhibitory effect on corrosion rate. For example, in Table 6, the array $A_{10,1}$ represents pT>0, which shows the accelerating effect of pH and temperature while acting synergistically. To continue the procedure of finding the other arrays, the value of $|C_x-C_1|$ and $|C_y-C_1|$ were calculated and compared and the logical results of this comparison were shown as the array $A_{x,y}$. C_x, C_y and C_1 were the corrosion rates corresponding the trial condition x, y and 1, as were defined in Table 2. For example in Table 6, to calculate $A_{3,2}$, $|C_3-C_1|$ and $|C_2-C_1|$ were calculated. The value $|C_3-C_1|$ was related to $A_{3,1}$ which shows the effect of velocity, because trials 3 and 1 differ only in parameter velocity. $|C_2-C_1|$ was related to $A_{2,1}$, respectively which defines temperature effect. Since $|C_2-C_1| > |C_3-C_1|$, The conclusion T>V was obtained, which corresponds to the higher effect of temperature, compared with velocity. It should be admitted that the absolute values, equal or more than 0.001 was considered in comparisons. To analyze the other arrays of Table 5 and 6 and survey the synergistic effect of factors and methods of influencing on one other, three groups of arrays were considered. The first group of arrays defines the increasing effect of factors on the corrosion rate while acting together. We can take example $A_{8,2}$ in Table 6 as a sample. This array presents the result T<SVT. This conclusion has obtained according to the method described in previous

part. Trials 1 and 8 differ in salinity, velocity and temperature parameters and $|C_8\text{-}C_1|$ illustrates the SVT effect. Trials 2 and 1 differ in temperature parameter and $|C_2\text{-}C_1|$ illustrates the temperature effect. $|C_8\text{-}C_1| > |C_2\text{-}C_1|$, So, the result SVT>T is obtained. This corresponds to the higher effect of salinity, velocity and temperature cooperation in comparison to temperature alone on heightening the rate of corrosion. Arrays that belong to this group were shown by a circle sign in the cell. The second group of arrays corresponds to the decreasing effect of factors' cooperation on the corrosion rate. For example, $A_{14,2}$ in Table 6 shows the lower effect of combining action of temperature and salinity and pH in comparison to temperature alone. The arrays belong to this group were shown by a triangle sign in the cell. The third group of arrays shows the insignificant effect of combining action in comparison to individual ones. For example, $A_{11,3}$ in Table 6 shows the effect of velocity nearly as much as combining action of velocity and pH. It shows nearly the worthless effect of pH while acting with velocity. The components of this group were shown by a square sign in the cell. These three groups can show the sharp difference in the combining action of parameters considered above. At last, it should be noted that all these results were obtained over the range of conditions considered. Although this range is the most common one in worldwide seawater, any variation of this range must be considered carefully. Since the variation range of factors is not comparable, we can't calculate the effectiveness of the factor by the variation value that they cause on corrosion rate; and this can only offer the variation direction and overwhelming parameter to some extent as described in this research.

QUALITATIVE RESULTS AND DISCUSSION

Stainless Steel 304

Temperature is the only factor among the other single acted factors considered (salinity, pH and velocity) that increases the rate of degradation, as shown in the first column of Table 5. This result is expected according to previous researches (Sasaki and Burstein, 2007; Bardal, 2004; Möller, 2006; Melchers, 2005; Jones, 2001; Al-Malahy and Hodgkiess, 2003). The obtained results support the previous outcomes. In higher temperature the amount of dissolved oxygen is lower and it prevents the formation of passive layer and increases the corrosion rate.

The arrays in this column, which correspond to the combining action of temperature with other factors, have increasing effect on the corrosion rate. It shows that temperature is always dominant while acting with other factors and neutralizes their reducing effect, to some extent. This result has not been considered in previous researches. In the previous researches the effect of parameters are considered individually and the synergistic action and the effect of dominant parameters are not considered. This analysis has a kind of nobility to predict the dominant parameter, when acting under the real situation, in synergistic manner.

The surprising result that can be obtained from the array $A_{11,1}$ is the increasing effect of the combined action of pH and velocity, on the corrosion rate (pV>0), while pH and velocity, both have decreasing function when alone. The similar surprising result can be observed in the array $A_{15,1}$ too. This array shows the increasing effect of combining action of salinity, velocity and pH (SVp>0) while these three factors have a decreasing effect individually. As another example of the combining action effect, we can consider the simultaneous action of salinity and velocity ($A_{7,1}$), which shows no significant effect on corrosion rate, while both have reducing effect when alone. All these results illustrate the surprising effect of combining action, which is strongly in contrarily to what is reported in researches which consider the individual role of parameters (Sasaki and Burstein, 2007; Bardal, 2004; Möller, 2006; Melchers, 2005; Jones, 2001; Al-Malahy and Hodgkiess, 2003). These new results, in comparison with the previous outcomes are the obvious witnesses that show the importance of synergistic action. This novel result has a valuable role in engineering designs.

Stainless Steel 316

By considering some arrays in first column of Table 6 ($A_{2,1}$, $A_{3,1}$, $A_{5,1}$ and $A_{9,1}$), it is obvious that temperature and salinity apply the accelerating effects, while pH and velocity have reducing function. High temperature reduces the amount of dissolved oxygen and prevents the formation of passive layer. The other parameter, salinity, increases the corrosion rate by influencing the passive layer's property. These results are expected and similar to which is reported in previous researches (Sasaki and Burstein, 2007; Bardal, 2004; Möller, 2006; Melchers, 2005; Jones, 2001; Al-Malahy and Hodgkiess, 2003).

The arrays in the first column, which correspond to the combining action of temperature with other factors, have increasing effect on the corrosion rate. It shows that temperature is always dominant while acting with other factors and neutralizes their reducing effect, to some extent. It is a surprising result which has not been considered before. In previous studying the parameters' effect had been evaluated individually, so this kind of results has not been reported.

The surprising result that can be obtained from the array $A_{11,1}$ is the accelerating effect of combined action of pH and velocity, on the corrosion rate (pV>0), while pH and velocity, both have decreasing function when alone. As another example of combining action effect, the simultaneous action of pH and salinity ($A_{13,1}$) can be considered, which shows the dominant effect of pH while acting synergistically with salinity. This array illustrates that although the salinity has an increasing effect on the corrosion rate, the combining action of this parameter with pH has the reducing function and surprisingly, while velocity adds to these two factors ($A_{15,1}$), the synergistic action of parameters has an increasing function, although the added parameter is velocity, which has a reducing function by alone. All these results which show the unexpected role of combining action has not been considered before and is not predictable by mere individual studying of parameters' effect (Sasaki and Burstein, 2007; Bardal, 2004; Möller, 2006; Melchers, 2005; Jones, 2001; Al-Malahy and Hodgkiess, 2003). Comparison of the obtained results with the previous ones shows the sharp difference which comes from the synergistic action' ignorance.

CONCLUSIONS

The following conclusions can be drawn from the present investigation:

- Temperature has a marked effect among the other single-acted factors (p, V and S), on corrosion rate of both 304 and 316 samples
- Temperature and salinity are the factors that increase the corrosion rate of type 316 steel, while only temperature increase the corrosion rate of type 304 steel, among the other single-acted factors considered
- Salinity increases the corrosion rate of type 316 steel and decreases the corrosion rate of type 304.
- The environmental factors not always support one other; sometimes weaken or neutralize the others

- The effect of one parameter can be influenced by the others and its variation direction may be reversed. As a case, increasing the temperature in high velocity synthetic seawater can overcome the reducing effect of velocity on the corrosion rate of type 316 and increase the rate of degradation and in the type 304, increasing the temperature in high salinity synthetic seawater can overcome the reducing effect of salinity on corrosion rate and increase the rate of degradation

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