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Technical Report

Modelling the Galvanic Behaviour of Cu: Fe: Zn Couple in Acidic Media at Various Temperatures, Speed of Agitation and Area Ratio

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This work is focused on modelling the galvanic behaviour of Cu: Fe: Zn couple in aerated 0.2 N HCl at various operating conditions as independent variables; temperatures (30 and 40 ^oC), speed of agitation (0, 300, 600 & 900 RPM) and area ratio (0.5:1:1) and (1:1:0.5) against the average galvanic current (I_g) as dependent variable using a patented electrochemical system to measure the galvanic currents and coupling potentials. Multiple regression analysis was utilized to generate two models for the given area ratios (0.5:1:1) and (1:1:0.5) based on the highest R^2 and R^2_{adj} with reference to ANOVA statistical analysis based on F and P tests:

$$I_g = 363.863 + 14.866T - 0.489S + 0.017\ T\times S$$
$$R^2 = 97.9\%,\ R^2_{adj.} = 96.3\%$$
$$I_g = 598.757 - 5.764T + 0.399S - 0.0126\ T\times S$$
$$R^2 = 92.7\%,\ R^2_{adj.} = 87.3\%$$

Three dimensional mapping was generated using MATLAB in order to reflect the interactions among these independent variables. It was concluded that varying the area ratio with the other operating conditions (temp. and speed of agitation) have direct influence on changing the average galvanic current generated among the suggested couple based on the results which showed that increasing the temperature and the speed of agitation will increase the average galvanic current for the area ratio 0.5:1:1, while increasing the temperature and the speed of agitation will decrease the average galvanic current for the area ratio 1:1:0.5 because the area of Cu and Fe (cathode) in the Cu:Fe:Zn couple determined the extent of galvanic current. In other word the reduction of oxygen on Cu and Fe samples which is facilitated by large surface area of the exposed metals was the rate determining process. In contrast variations in the area of Zn in the Cu:Fe:Zn couple had negligible effect on the extent of galvanic current. It was also concluded that the patented electrochemical system gave better understanding for the behavior of Cu:Fe:Zn couple in the given corrosive environment under the various operating conditions.

Keywords: Modeling, Galvanic Corrosion, Aerated HCl, different operating conditions

1. INTRODUCTION

A potential difference usually exists between dissimilar metals when they are immersed in a corrosive environment, because this potential difference produces electron flow .Corrosion of the less noble or less corrosion resistant metal is usually increased and the attack of the more resistant metal is decreased [1,2]. On the other hand Brass as an alloy of copper and zinc; has wide applications (i.e. gears, bearings, and valves etc.) [3,4] and when connected to steel under corrosive environment, a galvanic couple will be initiated between the brass alloy and steel.

On the other hand Corrosion attributes to mass transfer which is caused mainly by the transport of reactants and reaction products. The flow dependency of the corrosion system is greater or smaller according to whether it is predominantly mass transport controlled reaction [5,6].

The present work sets to examine the different features of galvanic corrosion due to the coupling of three industrially different feasible metals (i.e. copper, iron and zinc) in aerated 0.2 N HCl solution at various operating conditions as independent variables; temperatures (30 and 40 ^{o}C), speed of agitation (0,300, 600 & 900 RPM) to be applied for the two area ratios (0.5:1:1) and (1:1:0.5) of Cu:Fe:Zn.The results withdrawn from the applied tests in this work have been represented and correlated in two models based on ANOVA statistical method. The significance of each regressor in these models have been estimated and surface responses were drawn using three dimensional mapping facilities in MATLAB to show the behaviour of the average galvanic corrosion current as a function of temperature and speed of agitation.

2. EXPERIMENTAL WORK

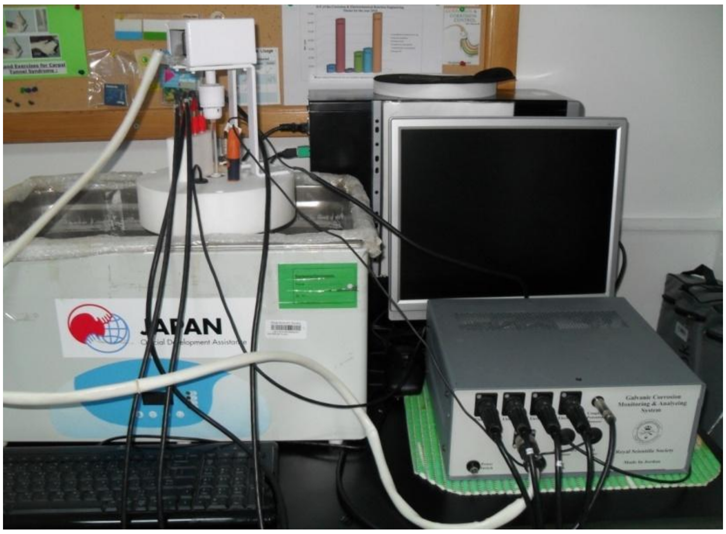

Figure 1. Galvanic Corrosion monitoring and analyzing system

Figure 2. Galvanic Corrosion Cell

The experiments were conducted using electrochemical system (Software, Hardware and Galvanic Corrosion Cell) shown in Figures 1,2 , designed and fabricated to measure the Galvanic Currents (I_g) for two, three and four specimens at a time as well as measuring the coupling potential ($E_{coupling}$) with respect to SCE.

Materials:

Rectangular copper, iron and zinc coupons of 20x10 mm were used in the experiments.

Electrolyte:

The electrolyte used in this work was aerated 0.2N HCl solution.

Experimental procedure:

The specimens were connected to the galvanic corrosion cell shown in Figure.2, then kept inside the electrolyte at the needed temperature and connected to the hardware and the data acquisition card shown in Figure.1.The system is turned on after setting the experimental time, speed of agitation in order to record and map the galvanic current Vs. time for each specimen (i.e. Cu, Fe and Zn). The experiments were repeated twice to assure reproducibility of data.

3. RESULTS AND DISCUSSION

3.1 Model generation for the area ratio

A model was generated in Eqn (1) using the data given in Table 1 based on T test with 95% confidence limit as shown in Table 2 with highest R^2 and $R^2_{adj.}$ to show the effect of different variables on the average galvanic current of the couple (Cu: Fe: Zn) at the given area ratio (0.5:1:1):

$$I_g = 363.863 + 14.866T - 0.489S + 0.017\, T \times S \qquad (1)$$
$$R^2 = 97.9\%,\ R^2_{adj.} = 96.3\%$$

Where T is temperature (^oC), S is the speed (RPM), I_g is the galvanic current (μA)

Table 1. Average galvanic current of Cu-Fe-Zn at various temperatures and speed of agitation

Area ratio	Temperature (^oC)	Speed (RPM)	Average galvanic current (μA)
0.5: 1 :1	30	0	808
0.5 : 1 :1	30	300	818
0.5 : 1 : 1	30	600	824
0.5 : 1 : 1	30	900	827
0.5 : 1 : 1	40	0	938
0.5 : 1 : 1	40	300	1029
0.5 : 1 : 1	40	600	1107
0.5 :1 : 1	40	900	1103

Table 2. Estimation of regression coefficient for the suggested model based on T-test with 95% confidence limit

Regression Statistics	
Multiple R	0.99
R square	0.98
Adjusted R square	0.96
Standard error	25.04
Observations	8

Table 3. ANOVA table of the adopted model

	Degree of freedom	Sum Square	Mean Square	F	Significance F
Regression	3	117843.7	39280.9	62.6	8×10^{-4}
Residual	4	2508.5	627.1		
Total	7	120351.2			

Table 3 shows the analysis of variance of the model given in Eqn(1) which emphasize that the model is significant based on F test .The numerical estimates of the regression coefficients given in Eqn(1) are shown in Table 4.

Table 4. Numerical estimates of the regression coefficients

	Coefficient	Standard error	T-tset	P-value	Lower 95%	Upper 95%
Intercept	363.9	104.8	3.5	0.03	73.0	654.7
Temperature(T)	14.9	3.0	5.0	0.007	6.6	23.1
Speed (S)	-0.5	0.2	-2.6	0.06	-1.0	0.03
T.S	0.02	0.005	3.2	0.03	0.002	0.03

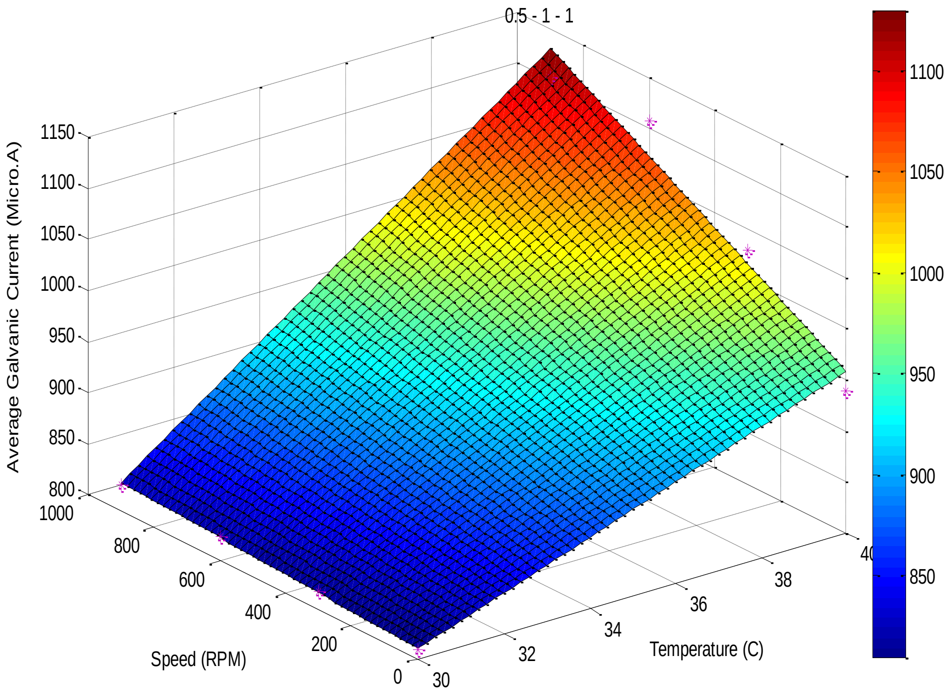

Figure 3. Surface response of Eqn (1) showing the average galvanic (I_g) Vs. temperature (^oC) and speed of agitation (RPM)

It is clear from Eqn (1) that increasing the temperature and the speed of agitation will increase the average galvanic current for the area ratio 0.5:1:1 and this is obvious from the three dimensional mapping given in Figure 3. These results agree with the opinion given by Kim *et. al* [5], Saeed [6,7] Belasco-Tamarit *et.al* [8] and Sanchez-Tovar *et.al* [9].

3.2 Model generation for the area ratio (1:1:0.5)

A model was generated in Eqn (2) using the data given in Table 5 based on T test with 95% confidence limit as shown in Table 6 with highest R^2 and $R^2_{adj.}$ to show the effect of different variables on the average galvanic current of the couple (Cu: Fe: Zn) at the given area ratio (1:1:0.5):

$$I_g = 598.757 - 5.764T + 0.399S - 0.0126\, T \times S \qquad (2)$$
$$R^2 = 92.7\%,\ R^2_{adj.} = 87.3\%$$

Where T is temperature (°C), S is the speed (RPM), I_g is the galvanic current (μA)

Table 5. Average galvanic current of Cu-Fe-Zn at various temperatures and speed of agitation

Area ratio	Temperature (°C)	Speed (RPM)	Average galvanic current (μA)
1 : 1 : 0.5	30	0	410
1 : 1: 0.5	30	300	443
1 : 1 : 0.5	30	600	464
1 : 1 : 0.5	30	900	425
1: 1 : 0.5	40	0	385
1 : 1 : 0.5	40	300	318
1 :1 : 0.5	40	600	293
1 : 1 : 0.5	40	900	289

Table 6. Estimation of regression coefficient for the suggested model based on T-test with 95% confidence limit

Regression Statistics	
Multiple R	0.96
R square	0.93
Adjusted R square	0.87
Standard error	24.77
Observations	8

Table 7 shows the analysis of variance of the model given in Eqn(2) which emphasize that the model is significant based on F test .The numerical estimates of the regression coefficients given in Eqn(2) are shown in Table 8.

Table 7. ANOVA table of the adopted model

	Degree of freedom	Sum Square	Mean Square	F	Significance F
Regression	3	31285.9	10428.6	17.0	1×10^{-2}
Residual	4	2455.8	614.0		
Total	7	33741.7			

Table 8. Numerical estimates of the regression coefficients

	Coefficient	Standard error	T-test	P-value	Lower 95%	Upper 95%
Intercept	598.8	103.7	5.8	0.004	311.0	886.5
Temperature (T)	-5.8	2.9	-2.0	0.1	-13.9	2.4
Speed (S)	0.4	0.2	2.2	0.1	-0.1	0.9
T.S	-0.01	0.005	-2.4	0.07	-0.03	0.002

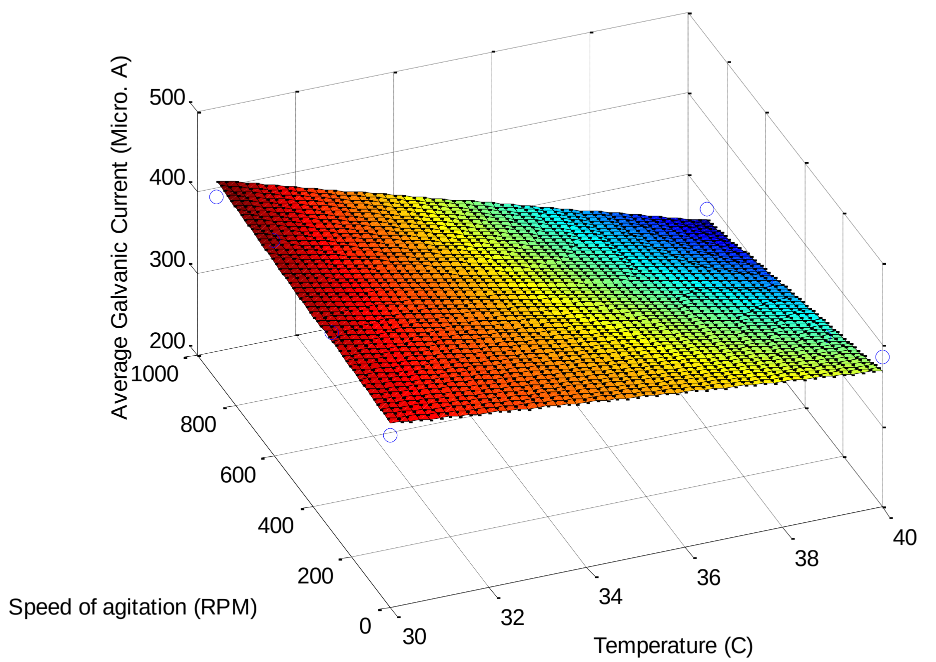

Figure 4. Surface response of Eqn (2) showing the average galvanic (I_g) Vs. temperature (^oC) and speed of agitation (RPM)

It is clear from Eqn (2) that increasing the temperature and the speed of agitation will decrease the average galvanic current for the area ratio 1:1:0.5 and this is obvious from the three dimensional mapping given in Figure 4. These results agree with the opinion given by Kim *et. al* [5], Saeed [6,7] Belasco-Tamarit *et.al* [8] and Sanchez-Tovar *et.al* [9]

3.3 Combining the surface response of the both models given in eqns (1&2):

The surface responses of the both models given in Eqns (1&2) were combined in Figure 5 in order to give an explanation for their behavior. It is obvious from the two models that Zn sacrificially corroded to protect Cu and Fe. In other word Zn played as an anode against Cu and Fe at the given aggressive environment. The given regressors (temperature and speed) played an important role as shown in Table 1 in which increasing the temperature and speed of agitation increased the galvanic current of the couple. On the other hand the given regressors (temperature and speed) reduced the average galvanic current in an obvious way as shown in Table 5. It is worthy to mention that the effect of changing the area ratio of on the galvanic currents was studied also by Sout *et. al.*[10]. In fact Sout *et. al.* showed that the area of Fe in Zn:Fe couple determines the extent of the galvanic process.

In our case the area of Cu in the Cu:Fe:Zn couple played an important role in altering the galvanic current. In other word the reduction of oxygen on Cu and Fe samples which is facilitated by large surface area of the exposed metals was the rate determining process. In contrast variations in the area of Zn in the Cu:Fe:Zn couple had negligible effect on the extent of galvanic current.

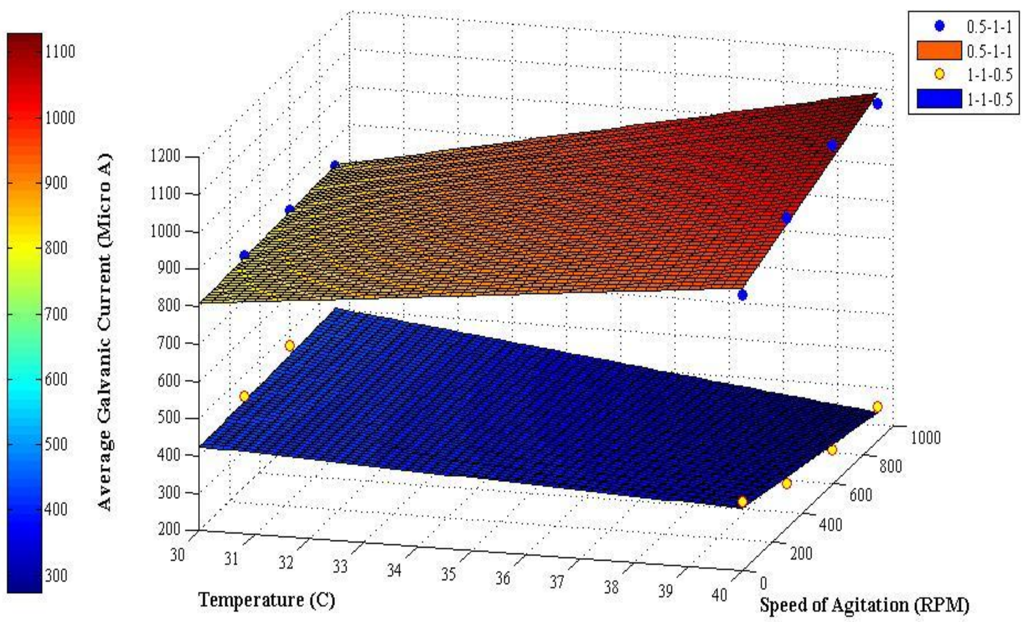

Figure 5. Surface response of Eqn (1&2) showing the average galvanic (I_g) Vs. temperature (^oC) and speed of agitation (RPM) for the area ratios (0.5:1:1) and (1:1:0.5)

4. CONCLUSIONS

1. The area of Cu and Fe in the Cu:Fe:Zn couple determined the extent of galvanic current
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2. Variations in the area of Zn in the Cu:Fe:Zn couple had negligible effect on the extent of galvanic current.
3. The surface response of the given models showed that the given regressors (temperature and speed) played a significant role in the generated models.
4. The generated models were significant in terms of the regression coefficients based on T-test with 95% confidence limit and ANOVA statistical analysis based on F and P tests.
5. The Galvanic Corrosion Monitoring and Analysing System provided an excellent explanation of the behaviour of Cu: Fe: Zn.
6. It was also concluded that the patented electrochemical system gave better understanding for the behavior of Cu:Fe:Zn couple in the given corrosive environment under the various operating conditions.

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