ISSN: 2277-3754
ISO 9001:2008 Certified
International Journal of Engineering and Innovative Technology (IJEIT)
Volume 4, Issue 4, October 2014

Parametric Study of Al-Si-Mg Alloy in Simulated Seawater in the Presence of Natural Cyperusesculentus l. Oil as Inhibitor using Response Surface Method

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Abstract- Aluminum is one of the most widely used metals in industry and domestic applications. Thus, the investigation of its corrosion inhibition in different aggressive media has continued to generate lots of interest. One of the best methods to determine corrosion rate of a material is by laboratory experiments which involve experimenting different formulations and selecting the best composition for the specific operation. This exercise is not only time-consuming considering the amount of time required, but also expensive. This paper presents the parametric design and functional optimization of aluminum in simulated sea water environment in the presence of natural Cyperusesculentus L. oil.In order to find out the effect of tool geometry parameters on the surface of the samples, response surface method (RSM) was used and a prediction model was developed using experimental data. The model developed was simple, in agreement with the experimental data used and can be implemented using an ordinary simple calculator.In addition, a good agreement between the predicted and measured surface roughness was observed. Therefore, the developed model can be effectively used to predict the effectiveness of Cyperusesculentus L. oil on Al-Si-Mgsurface.

Keywords: **Process optimization; Statistical experiment design; Response surface methodology; simulated sea water; Corrosion; Al-Si-Mg.**

I. INTRODUCTION

Due to the different industrial applications and economic importance of aluminum and its alloys, its protection against corrosion has attracted much attention [1-6]. Most aluminum alloys have good corrosion resistance, but in the presence of aggressive ions, the protective layer can be locally destroyed and corrosive attack takes place [7]. One of the methods of protecting metals against corrosion is addition of species to the aggressive solution for the purpose of inhibiting the corrosion reaction and reduces the corrosion rate [8]. Corrosion inhibitors are substances which when added in small concentrations to corrosive media decrease or prevent the reaction of the metal with the media. Unfortunately, many of the inhibitors used are inorganic salts or organic compounds with toxic properties. The development of environmentally friendly pre-treatment for metals is now a field of growing interest due to banning of the use of chromates as protective pre-treatments [9].These toxic effects have led to the use of natural products as anti-corrosion agents which are eco-friendly and harmless. In recent days many alternative eco-friendly corrosion inhibitors have been developed, Kliskic et al. [10] reports extract of rosemary leaves as corrosion inhibitor for Al-2.5Mg alloy in a 3% NaCl solution at 25^{O}C. Experimental results show that the additive adsorbs on the alloy surface according to the Freundlich isotherm. The authors concluded that an increase in the catech in fraction concentration increases the resistance and decreases the capacitance of systems considered, while the catechin fraction in the concentrations >10^{-6} M completely prevents localized corrosion attacks. It has-been reported by Ambrish Singh et al.[11] that the corrosion of 7075 aluminum alloy in 3.5 wt.% NaCl solution can be inhibited by 5,6-dihydro-9,10-dimethoxybenzo[g]-1,3-benzodioxolo [5,6-a]quinolizinium (berberine). The experimental results show that inhibition efficiency of the inhibitor increases with an increase in the concentration of berberine. The adsorption of the berberine on the 7075 aluminum alloy surface obeyed the Langmuir adsorption isotherm. The authors concluded from the obtained results that berberine could serve as an effective inhibitor for the corrosion of 7075 aluminum alloy in 3.5% NaCl.The effects of corrosion inhibition on AA 2024-T3 using cerium cation and cinnamateanion were investigated by Hongwei Shi et al. [12]. It was found that cerium ion and cinnamate group have synergistic inhibiting effects. The results show that CeCinis an effective inhibitor pigment for improving the corrosion resistance of epoxy coatings on AA2024-T3,as reflected by the much higher coating resistance than that of the blank epoxy coating. Howida et

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ISO 9001:2008 Certified
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al [13] investigated the effect of aqueous extracts of Damsissa, Lupine and Half bar on the corrosion of 7075-T6 aluminum alloy in an aqueous solution of 0.5 M sodium chloride. The polarization curves showed that the three extracts act as cathodic inhibitors. Inhibitive mechanism was discussed assuming the adsorption of the three extracts on the electrode surface. Loto et al. [14] studied the effect of *Camellia Sinensis*(green tea) extract as a green inhibitor on the corrosion of aluminum alloy in 0.8M sulphuric acid at room temperature. The results obtained showed effective corrosion inhibition of the extract on the aluminum alloy test specimens in the different concentrations of sulphuric acid used. The value of Gibb's free energy of adsorption obtained show that the mechanism of adsorption of plant extract molecules on the metal surface was by physiosorption. Recently, response surface methodology (RSM) has been employed to optimize and understand the performance of complex systems [15–20]. By application of RSM, it is possible to evaluate the interactions of factors influencing treatment efficiency with a limited number of planned experiments. The analysis of the data during manufacturing by using suitable statistical designs is of high importance for precise evaluation to be obtained from the process. Design and methods such as factorial design, response surface methodology (RSM) and Taguchi methods are now widely in use in place of one factor at a time experimental approaches which is time consuming and very expensive [21]. Response surface methodology (RSM) experimental design is one of the emerging technique which helps in carrying out the analysis of experiments with the least experimental effort [22]. Yang and Tarng [23] studied optimal cutting parameters using Taguchi method in turning.ChoudhuryandEl-Baradie [24] used RSM and 23 factorial designs to estimate the surface roughness during the turning process of high strength steel. On the other hand, Salam et al. [25] investigated the influence of operating parameters on weight of wax deposit in oil-pipeline using response surface methodology (RSM). The RSM was used to develop polynomial regression model and investigate the effect changes in the level of wax temperature differential, flow rate and residence time on weight of wax deposit using Box Behnken design. The authors concluded that the agreement between the predicted and experimental values describe the accuracy of the model developed and can be used to navigate within the design space. In this paper, the application of RSM on the corrosion ofAl-Si-Mg alloy was carried out to develop the mathematical model so as to investigate the influences of the different operating parameters. For finding optimum value of geometry parameters, the quadratic model of response surface methodology was used. This research will be beneficial for future applications. Especially, as introduced in a real-world application, such as in manufacturing plant. Also, just as is found in a production environment, this provides an efficient project in an academic environment as well. In the present study, RSM, trial version of Design Expert 6.0.8was employed for the optimization of natural *Cyperusesculentus* L. oil used in the experimental investigation reported in the work of Mohammed*et al.* [26]. The main objective was to optimize the process from the model obtained via experimental data.

II. MATERIALS AND METHOD

The solution of natural tiger nut oil was used as the inhibitor. The detail of the experimental procedure and results were reported in the work of Mohammed*et al.* [26]. The work is targeted at determining the influence of identified process variables; immersion time, inhibitor concentration and temperature on corrosion of aluminum alloy in simulated seawater.

MODEL DEVELOPMENT

Response surface methodology is a statistical technique based on simple multiple regressions. The effect of two or more factors on quality criteria can be investigated and optimum values are obtained using this method [27]. In response surface methodology, there should be at least three levels for each factor. Thus, the factor values that are not actually tested using less experimental combinations and the combinations themselves can be estimated [28]. It was reported that exposure time, temperature and inhibitor concentration significantly affect the corrosion rate of aluminum alloy in 3.5% Nacl solution using natural *CyperusesculentusL.*oil as the inhibitor[26]. After the identification of the important parameters, the experimental range was tabulated in Table 1. A Box-Behnken statistical design with three factors and three levels was employed to solve second order polynomial model. A total number of seventeen experiments were required for the successful model development of influence of identified variables on corrosion rate. Design-Expert software (version 6.0.3, Stat-Ease Inc., Minneapolis, USA) was used to generate design of experiment shown in Table 2, which will be used for both model analysis and optimization. In most RSM problems, there is a functional relation between responses and independent variables and this relation can be explained using the model by Salam et al. [25]. The main and interactive values between the independent variables and response were evaluated by generating a quadratic model equation in the form represented in equation 1.

An equation missing

$$Y = \beta_o + \beta_1 A + \beta_2 B + \beta_3 C + \beta_{11} A^2 + \beta_{22} B^2 + \beta_{33} C^2 + \beta_{12} AB + \beta_{13} AC + \beta_{23} BC \quad 1$$

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Where: Y is the predicted value, β0 is the constant coefficient, βi, βij and βii the coefficients of linear, interaction and quadratic term respectively. The above coefficient parameters were estimated using a multiple linear regression analysis employing the Design-Expert software. Factor interaction, ANOVA and 3-D surface plots are used for analyzing the developed model.

Table 1 Range of independent variables used for model development

Factor	Unit	Low (-1)	Mid (0)	High (+1)
Temperature (A)	Degree (^{O}C)	30	50	70
Immersion time (B)	Hour	1	3	5
Inhibitor concentration(C)	g/v	0.2	0.5	0.8

Table 2: Experimental sequence of the three factors

Std	Run	A	B	C	Std	Run	A	B	C
6	1	1	0	-1	10	10	0	1	-1
11	2	0	-1	1	7	11	-1	0	1
2	3	1	-1	0	1	12	-1	-1	0
9	4	0	-1	-1	4	13	1	1	0
8	5	1	0	1	12	14	0	1	1
17	6	0	0	0	3	15	-1	1	0
14	7	0	0	0	16	16	0	0	0
13	8	0	0	0	5	17	-1	0	-1
15	9	0	0	0					

III. RESULTS AND DISCUSSION

This section will evaluate the response from the mathematical model developed from box behnken design using response surface method by study of effect of the three variables selected on corrosion rate, effect of interactions between the three variables on corrosion rate, the influence of each of the model parameters of the developed model, validation of model using performance plot and ANOVA.

IV. QUANTIFICATION OF VARIABLES ON MODEL DEVELOPED

The effect of each input parameter on the response was shown in Figure 1.At fixed immersion time of 3minutes and inhibitor concentration of 0.5mg/L, corrosion rate increased as temperature increased from 0.845 at 30^{o}C to 6.745 at 70^{o}C. Also, the effect of immersion time on the output performance at constant temperature 50^{o}C and 0.5mg/L inhibitor concentration was shown in the figure 1ii. At 1minute, the corrosion rate was 10.695 and as time increases to 5minutes, the corrosion rate was decreased to 4.595, thereby generalizing it to be increment in time leads to decrease in corrosion rate. The effect of inhibitor concentration on corrosion rate was also shown at fixed temperature 50^{o}C and immersion time 3 minutes. The corrosion rate was found to be 7.795 at 0.2mg/L and decreases to 5.398 at increased inhibitor concentration 0.8mg/L. The effect of input combinations on the corrosion rate was shown in Figure 2. The effect of temperature (A) and immersion time (B) on the response at fixed inhibitor concentration (C) 0.5mg/L was shown in the Figure 2(i). At low immersion time 1 minute, corrosion rate increased from 4.0 to 14.3 as temperature increased from 30 to 70oC. At high immersion time 5 minutes, the corrosion rate was found to be 2.3 at 30oC and increased to 3.8 at 70oC. Figure 2ii show the relationship between temperature and inhibitor concentration at fixed immersion time. At fixed immersion time of 3 minutes, the effect of temperature (A) and inhibitor concentration (C) on the corrosion rate show that corrosion rate increased as temperature and inhibitor concentration increased. At low inhibitor

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ISO 9001:2008 Certified
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concentration of 0.2mg/L, the corrosion rate was found to be 2.8 at 30oC and 9.7 at 70oC.

At high inhibitor concentration 0.8mg/L, the corrosion rate increased from 1.4 to 6.3 when temperature was increased from 30 to 70oC. Similarly, effect of immersion time and inhibitor on the response at fixed temperature 50^oC was as well shown in the Figure 2iii. At low inhibitor concentration 0.2mg/L, the corrosion rate was found to be 15.9 at 1minute and decreased to 4.3 at 5minutes. A slight decrease in corrosion rate was observed at high inhibitor concentration 0.8mg/L with increase in time.

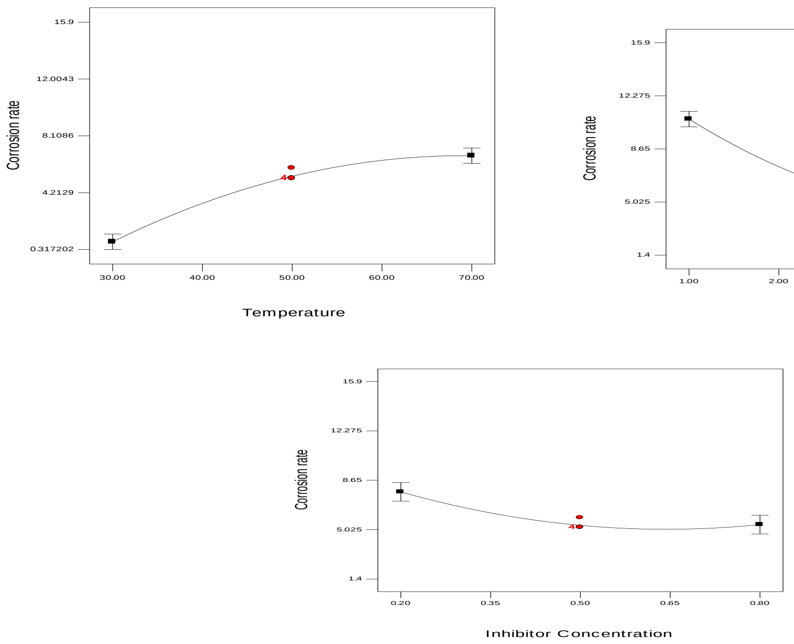

Fig 1: One factor plots of input parameters on corrosion rate.

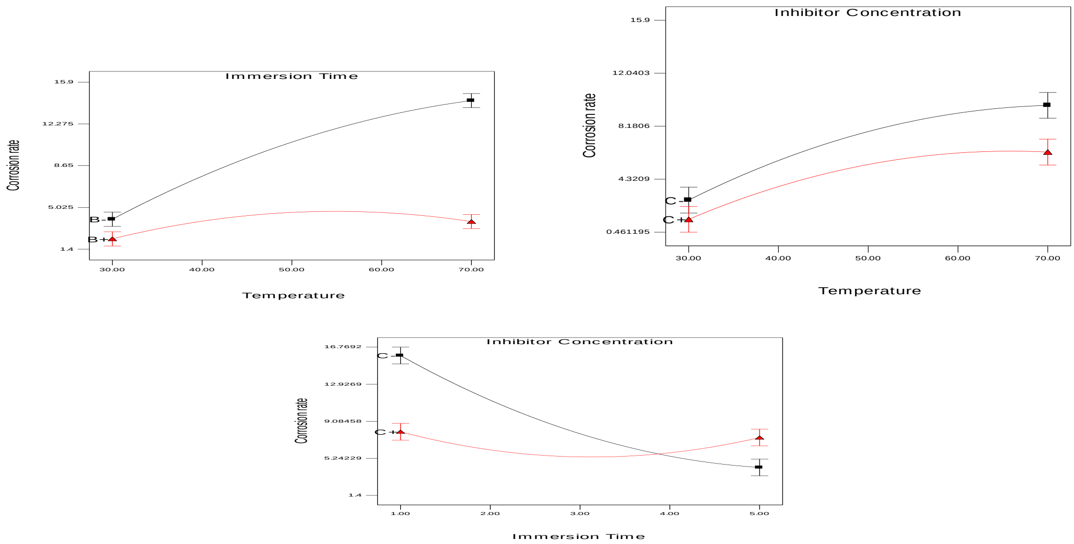

Fig 2: Interaction plots of input parameter on the corrosion rate

V. MODEL EQUATION

ISSN: 2277-3754
ISO 9001:2008 Certified
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The experimental value generated using design in Table 2 was used to generate a quadratic model. Multiple regression analysis was used to correlate the corrosion rate with the three identified variables that affects corrosion rate using a second order polynomial as shown in Equation (2) below:

$$Y = 5.34 + 2.95A - 3.05B - 1.02C - 1.54A^2 + 2.3B^2 + 1.25C^2 - 2.2AB - 0.5AC + 2.75BC \quad (2)$$

A, B and C represents the coded values for temperature, immersion time and inhibitor concentration. The variables with a positive sign denote synergistic effect, while those with negative sign denote antagonistic effect.

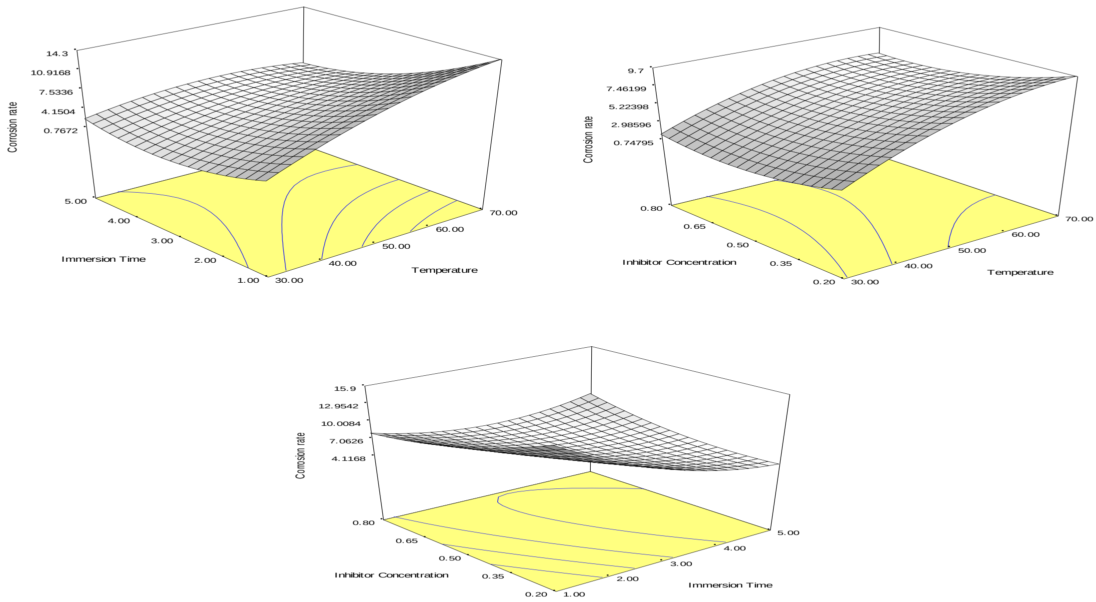

Fig 3: Surface interaction plot of the variables

Figure 3 described the surface behavior of A and B and C under different combinations. Figure 3i show the interaction between varying values of A and B at constant value of C. Minimum corrosion rate of 2.3 was observed when B was 5minutes at low A at 30°C. There was a slight increase in corrosion rate when A increased to 70°C whereas a high corrosion rate was experienced at low A and increased with change in B. Figure 3ii described the relationship between different values of A and C at fixed B. Low corrosion rate of 1.4 was recorded at high C and low A, a corresponding low corrosion rate was observed too at low C at low A but was increased as A was increased. Though corrosion rate was lower in high C but it didn't prevent progressive increase in corrosion rate as A was increased. Surface behavior of different values of B and C at constant A in Figure 3iii.Low corrosion rate of 8 mm/yr was experienced at high C with low B. This value decreased with increased in value of B to 3minutes, beyond 3minutes there was increased in corrosion rate to 7.4 at high B.

VI. MODEL VALIDATION

The accuracy of the model developed can be further established with the aid of statistical parameter like coefficient of correlation R^2 value as well as Adjusted R^2 value. R^2 indicates the ratio between sum of the squares (SSR) with total sum of the square (SST) and it describes up to what extent perfectly the model estimated experimental data points. The model gave a predicted R-square value of 0.998 which show that the experimental data fitted better and is in reasonable agreement with the Adjusted R-Squared of 0.9945. The accuracy of the predicted and experimental was demonstrated in Figure 4 which show the agreement between the predicted and experimental values.Adequate prediction of 58.805 indicated an adequate signal for the model. The model can be use for prediction within the set experimental range. The Model F-value of 260.64 implies that the model is significant. There is only a 0.01% chance that a "Model F-Value" this large could occur due to noise. Values of "Prob> F" less than 0.0500 indicate model terms are significant. In this case A, B, C, A2, B2, C2, AB, BC are significant model terms. Values greater than 0.1000 indicate the model terms are not significant. If there are many insignificant model terms (not counting those required to support hierarchy), model reduction may improve the model.

Also, the input parameter which has optimum effect on the response was input parameter B (immersion time) because it has the highest F-value of 379.69 and corresponding lowest prob>F value, followed by temperature and the input parameter with least effect is inhibitor concentration having the lowest F-value of 15.69. In terms of two factors interaction, input combination A and B gave the highest F-value of 197.55 which is most significant on the response.

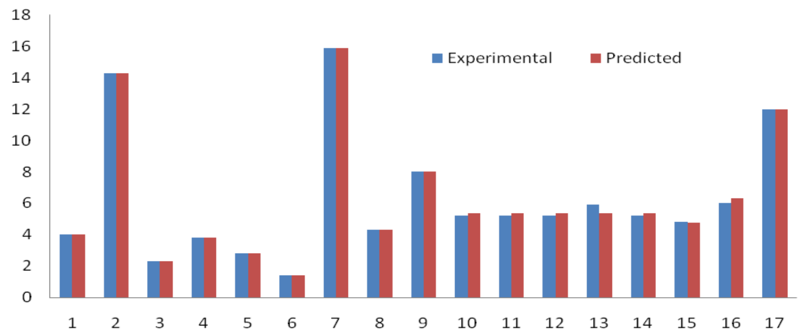

Figure 4: The performance plot of predicted and experimental values

Table 2: ANOVA for the response

Source	Sum of Squares	Degree of Freedom	Mean Square	F-Value	Prob> F	
Model	229.8801	9	25.54224	260.6351	< 0.0001	Significant
A	34.81	1	34.81	355.2041	< 0.0001	
B	37.21	1	37.21	379.6939	< 0.0001	
C	1.92	1	1.92	19.59184	0.0115	
A^2	4.896462	1	4.896462	49.96389	0.0021	
B^2	10.89851	1	10.89851	111.2093	0.0005	
C2	3.230821	1	3.230821	32.96756	0.0046	
AB	19.36	1	19.36	197.551	0.0001	
AC	0.2	1	0.2	2.040816	0.2263	
BC	10.08333	1	10.08333	102.8912	0.0005	

VII. CONCLUSION

The modeling of effect of inhibitor concentration, temperature and exposure time on the corrosion of aluminum alloy was successfully studied with minimum number of experimental runs using response surface method. The behavior of each of the independent variable was observed on dependent variables show that the inhibitor efficiency of inhibitor decreases as temperature increases. The Model F-value implies that the model is significant. The terms A, B, C, A^2, B^2, C^2, AB and BC are significant model terms. There is a close agreement between the experimental and predicted result for all the experimental result reported in the literature used for this study which describes the accuracy of the model developed and can be used within the set experimental condition.

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ISSN: 2277-3754
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