Analyses Corrosion Prediction Software for CO₂ Corrosion of Carbon Steel Using Statistical Formulas

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Abstract—The statistical formulas are capable tools to find a regression of corrosion rate effectively among combining factors. One type of statistical model which is response surface methodology (RSM) has shown a proven method in minimizing number of running. Through this technique, this research study predicting corrosion rate of carbon steel as effects of pH, CO_2 pressure and temperature. It can be used to run 3 dependent factors, 3 level experiment with only 16 number of running. The result reveals that NORSOK corrosion prediction software with second order model regression has 98 % of coefficient determination. Model prediction of Cassandra has 99.3% of coefficient determination. Second order model also has been verified with experimental data which shows a good correlation.

Index Terms— CO_2 Corrosion, carbon steel, corrosion models

I. INTRODUCTION

Predicting of corrosion behavior is an important issue in project of corrosion design to decrease a failure risk. Degree of corrosiveness is influenced by several factors that potentially complicate to predict. Corrosion reaction behaves in many mechanisms [1-4]. Multiple factors variables create difficulty to select an interest factors. Therefore, the building experiment has to able to reduce a robust data and simplify factors involved besides providingareasonably agreement between the experimentally observed and the real condition. Appropriate design experiment is also useful tool to minimize cost, time and energy.

Response surface methodology offers a simple design to process dependent factorseconomically [5-17]. Compare with full randomized design, response surface methodology is far simple [18]. This experiment design suggests that it is not necessary to randomized all factors during an experiment. Response surface is powerful technique, especially in determining stationer point which is the maximum or minimum curve located. Multi-variant regression prediction is presented in the three dimension (3D) graphic contour. So, the focus of response interest or predicted value will be easily identified.

A. Response Surface Methodology

Response surface methodology (RSM) deals the correlation between several response variables[19]. The main idea of RSM is to set an optimal design experiment. First step is determining suspected variables that only significantly influenced response. Once a dominant variables interest found the others are left, then starting to design a central composite design. The second-degree model can be used to optimize (maximize, minimize, or attain a specific target for) a response[20].

A contour plot can make an easier way to find the responses result of two variables. To find regression coefficients is calculated by least sum square methods and using matrices manipulation to predict a statistically parameters (residual, deviation, variant analyze, coefficient determination).

In this work, two types of models regression are used to facilitate interpretation of experiment data. The model regressions are exponential and second order model that will predict corrosion rate data calculated by two predicting model programs- "NORSOK[21] and Cassandra[22]".

NORSOK [7] standard is owned by Norwegian Oil Industry Association and Federation of Norwegian Manufacturing Industries. The program covers only calculation of corrosion rates where CO_2 is the corrosive agent. It does not include the corrosivity, e.g. contamination's of O_2 , H_2S etc. The model is an empirical containing CO_2 at different temperatures, pH, CO_2 fugacities, wall shear stresses, and temperatures from 20 to 160 °C.

Cassandra [8] is a model as a implementation of de Waard and BP's experiences. The input includes pH, CO_2 concentration, temperature, and water contaminant. This model does not consider a scaling temperature. The user must set an assumption of scaling temperature.

II. EXPERIMENTAL SETUP

A. Building an Experiment Design on CO₂ Corrosion

Relationship between corrosion rate and factors involved in CO_2 environment have been published intensively. Majority result stated that the corrosion rate

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haslinearand polynomial functions. The first one-linear function- is for corrosion rate under scaling temperature. While, the later-polynomial functions-is occurred if oxide film is formed.

Therefore, a suitable design to estimate simple curvature model regression, in the widely range of temperature, ison an assumption that corrosion rate (response) will behaves a second-degree polynomial model.So, the first order model is definitely not suitable. Then with the aim to optimally research, design experiment response surface try to be applied. The minimum number of levels required for each factor to quantify that behavior is three. Further,by adding center points with some repetition variantswould satisfy the requirement for pure error analysis.

B. Corrosion Rate Models

The exponential model of corrosion rate equation is confirmed with Henry's reaction constant's temperature dependence as[23],

$$log (V_{cor}) = 5.8 - 1710/T + 0.67 log (pCO_2)$$
$$CR_t = K_t x fCO_2^{0.62} x (S/19)^{0.146 + 0.0324 log (fCO2)} x f(pH)_t$$

where

 V_{cor} = corrosion rate in mm/yr T = operating temperature in K pCO_2 = partial pressure of CO₂ in bar

Y = response

 $X_k = k^{th} predictor$

 $\beta_k = k^{th}$ population regression coefficient

The exponential first order model is also argued by Kapusta which build a corrosion rate prediction involving the role of several factors as following:

$$CR = (CR_{NAP} + C_{RS} + CR_{HSR}) x f_f x f_{sc} x f_{al}$$

where C_{RNAP} , C_{RS} and C_{RHSR} represent the corrosion induced by naphthenic acids, sulfur and mercaptans, respectively. Then f_f is the flow enhancement factor, f_{sc} is a scale factor that reflects the protectiveness of iron sulfide, and f_{al} is the alloy factor, i.e., the ratio of the corrosion rate of an alloy to that of carbon steel.

Considering the effect of temperature dependence with an activation energy of general corrosion rate was reported by Jon Lee[24]. According to the Arrhenius relation below, this model corrosion rate follows an exponential behavior as:

$$\operatorname{Ln}\left(\mathrm{C}_{\mathrm{RT}}\right) = \mathrm{C}_{\mathrm{o}} + \mathrm{C}_{\mathrm{l}}/\mathrm{T}$$

The entire variance of that uncertainty is around ± 3 standard deviations. He compared with analysis of the literature data. The result has shown that the temperature dependence of general corrosion of an alloy behave a

similar to a prediction model. Corrosion rate model presented by Arrhenius seems the same as what Song written. Song related the current density with the respect to the temperature as:

$$i^{o} = I_{reff} Exp (E/R(1/T_{reff} - 1/T))$$

C. Second- order Model Regression

There is a curvature of general second order model which expressed as:

$$Y = \beta_0 + \sum_{i=1}^k \beta_i X_i + \sum_{i=1}^k \beta_{ii} X_i^2 + \sum_{i< j} \sum \beta_{ij} X_i X_j + \varepsilon$$

where Y = response that can fit the following linear, quadratic, or cubic regression models:

$$y_i$$
 = fitted response

 \wedge

Another way of modeling curvature is to generate additional models by using the log10 of x and/or y for linear, quadratic, and cubic models.

Model regression for second order for three dependence variable is as follows:

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_{11} X_1^2 + \beta_{22} X_2^2 + \beta_{33} X_3^2 + \beta_{12} X_1 X_2 + \beta_{13} X_1 X_3 + \beta_{23} X_2 X_3 + \varepsilon$$

Analogically, exponential model regression can be written as:

 $\hat{LnY} = \beta_0 + \beta_1 \ln X_1 + \beta_2 \ln X_2 + \beta_3 \ln X_3 + \beta_{11} \ln X_1^2 + \beta_{22} \ln X_2^2 + \beta_{33} \ln X_3^2 + \beta_{12} \ln X_1 \ln X_2 + \beta_{13} \ln X_1 \ln X_3 + \beta_{23} \ln X_2 \ln X_3 + \varepsilon$

Which is developed from simple exponential equation regression model of first degree:

$$\hat{Y} = a * X_1^a * b * X_2^b * c * X_3^c$$

 $Ln\hat{Y} = aLnX_1 + bLnX_2 + LnX_3$

Y = predicted value (independence factors)

 β = regression constant

X = main effect of dependence factors

XX= interaction effects between dependence factors

Where constant prediction is calculated through equation

$$b = [X^{T}.X]^{-1}.X^{T}Y$$

Then,

$$\hat{Y} = Xb$$

And residual is defined by

$$\mathbf{r} = \mathbf{Y} - \mathbf{\hat{Y}}$$

Coefficient determination (\mathbb{R}^2) is defined as,

$$r^{2} = 1 - \frac{SSerror}{SStotal} = 1 - \frac{\sum \left(y_{i} - \bar{y}_{i}\right)^{2}}{\sum \left(y_{i} - \bar{y}_{i}\right)}$$

	L 27	I ONEITIAL REGRESSION				
Code	pН	Total Pressure (bar) Temperature				
			(°C)			
√3	6.6	3.7	107			
1	5.8	3	80			
0	4.9	2.2	53			
-1	4.1	1.6	35			
-√3	3.6	1.3	26			

TABEL I. CORRELATION BETWEEN CODE AND UN-CODED FACTORS ON EXPONENTIAL REGRESSION

TABEL II. CORRELATION BETWEEN CODE AND UN-CODED FACTORS ON SECOND ORDER MODEL REGRESSION

Code	рН	Total Pressure (bar)	Temperature (⁰ C)
√3	6.4	3.5	95.8
1	5.8	3	80
0	4.95	2.3	57
-1	4.1	1.6	35
-√3	3.5	1.1	19.3

 $\bar{y}_{i} = i^{th}$ observed response value

$y_i =$ mean response

Another formula is presented on regression model as,

$$r^{2} = \frac{ss_{regression}}{ss_{total}} = \frac{\sum \left(\hat{y} - \bar{y_{i}} \right)}{\sum \left(y_{i} - \bar{y_{i}} \right)}$$

Coefficient of determination; indicates how much variation in the response is explained by the model.

Calculation of code from the un-code factors

$$X_{code} = \frac{2(X - X_{max})}{(X_{max} - X_{min})} + 1$$

For exponential model, code is calculated from equation:

$$X_1 = \frac{2(\ln v - \ln v_{atas})}{(\ln v_{atas} - \ln v_{bawab})} + 1$$

TABELIII. EXPERIMENT VARIANTS CODED AND RESULT

Ν	Tempera	Total	р	Corr.	Corr.	Corr.	Corr
0	ture	Press	Η	rate	rate	rate	
	(^{0}C)	ure		(Casdra/	(Casd	(NRSK/	Rate
		(bar)		Ln)	ra)	Ln)	(Nrs
							k)
1	-1	-1	-1	9.02	9.02	7.5	7.5
2	1	-1	-1	12.78	12.78	10	10
3	-1	1	-1	14.23	14.23	11	11
4	1	1	-1	22.17	22.17	14	14
5	-1	-1	1	4.16	4.16	2.4	2.4
6	1	-1	1	8.53	8.53	1.6	1.6
7	-1	1	1	5.87	5.87	3.5	3.5
8	1	1	1	13.29	13.29	2.4	2.4
9	0	0	0	10.83	11.56	8	7.9
1	0	0	0	_			
0							
1	0	0	0	P	ire erro)r	
1				> 10		Л	
1	0	0	0				
2				J			
1	$\sqrt{3}$	0	0	14.88		2.7	
3					13.77		2.7
1	-√3	0	0	6.77		5.1	
4					5.78		3.6
1	0	√3	0	15.4	15.48	11	10

5 1	0	-1/3	0	7 37	6.6	59	
6	0	- 15	Ū	1.51	0.0	5.7	5.2
1	0	0	\checkmark	4.8	5.72	1	1.2
7			3				
1	0	0	-,	15.82	17.2	12	
8							
			3				13

III. RESULT AND DISCUSSION

TABLE IV. FIRST ORDER MODEL REGRESSION ANALYSIS OF VARIANCE CALCULATED BY CASSANDRA AND NORSOKCORROSION RATE PROGRAM.

Source DF Seq SS Adj SS Adj MS F P
Regression 3 3.91843 3.91843 1.30614 36.45 0.000
Linear 3 3.91843 3.91843 1.30614 36.45 0.000
Residual Error 11 0.39422 0.39422 0.03584
Total 14 4.31264
R-Sq = 90.86%
Analysis of Variance calculated by NORSOK corrosion rate program
Source DF Seq SS Adj SS Adj MS F P
Regression 3 7.95662 7.95662 2.65221 20.32 0.000
Linear 3 7.95662 7.95662 2.65221 20.32 0.000
Residual Error 11 1.43584 1.43584 0.13053
Total 14 9.39246
R-Sq = 84.71%

At the first order model regression, both of Cassandra and NORSOK corrosion calculation program shows a quite unsatisfied of coefficient determination. It has 84% for NORSOK and 90.86% for Cassandra. It is because of those model regressions are only influenced by main effects factors. Linear dependence of the corrosion rate on independent factors (temperature, CO₂pressure, and pH) indicates that along the temperature applied there is no scaling effect considered. But, theoretically, at the high temperature and pH, corrosion rate must decrease. Therefore, the coefficient determination of the regression may be able to be increased by involved the quadratic effects of each factors. This conclusion suggests that the model must be developed to become a second order model regression.

A. Second Order Model Regression

TABLE V. CORROSION RATE CALCULATED BY" NORSOK" WITH SECOND ORDER MODEL REGRESSION

Source	DF Seq SS Adj SS Adj MS F P	
Regression	9 260.627 260.627 28.9585 30.85 0.001	
Linear	3 223.927 223.927 74.6423 79.52 0.000	
Square	3 25.930 25.930 8.6432 9.21 0.018	
Interaction	n 3 10.770 10.770 3.5900 3.82 0.091	
Residual Er	rror 5 4.693 4.693 0.9387	
Total	14 265.320	
ObsStdOrde	er C4 Fit SE Fit Residual St Resid	
15 15	7.900 7.816 0.967 0.084 1.26 X	

Estimated second order model Regression by NORSOK

R-Sq = 98.23%



Figure 1. Surface plot calculated by NORSOK program showsdependence of corrosion rate on Temperature and CO₂ pressure at pH of 4.9



Figure 2. Contour plot calculated by NORSOK program

TABLE VI. ANALYSIS OF VARIANCE CALCULATED BY NORSOK CORROSION RATE PROGRAM FOR EXPONENTIAL MODEL REGRESSION

Source	DF Seq SS Adj SS Adj MS F P	
Regression	9 248.472 248.472 27.6080 18.06 0.003	
Linear	3 214.691 214.691 71.5637 46.81 0.000	
Square	3 23.011 23.011 7.6703 5.02 0.057	
Interaction	3 10.770 10.770 3.5900 2.35 0.189	
Residual Er	ror 5 7.644 7.644 1.5288	
Total	14 256.116	
R-Sq = 97.0)2%	
		_

Figs. 1 and 2 illustrate possible behaviors of responses (corrosion rate) as functions of factor settings (temperature, CO₂ pressure, pH) which calculated by NORSOK program software. The polynomial coefficient regression analyzed based on design experiment response surface shows a 98% confidence level. This regression is better than calculation by exponential regression model (97%). In this case, selecting point of variant factors assume the value of the response increases from the low temperature exceed to scaling temperature. The setting range of temperature is appropriate to proof that scaling temperature is located between that ranges which is indicated by point of transition corrosion rate. From the contour plot on Fig. (2) is shown clearly that scaling temperature occurred at 75°C. This scaling temperature will decrease with the increase of pH.

TABLE VII. CORROSION RATE CALCULATED BY" CASSANDRA" WITH SECOND ORDER MODEL REGRESSION

Source	DF	Seq SS	Adj SS	Adj MS	F	Р	
Regression	9	365.382	2 365.382	40.598	90.22	0.000	
Linear	33	47.321	347.321	115.774 2	257.29	0.000	
Square	3	3.264	3.264 1	.088 2.4	2 0.18	2	
Interaction	3	14.797	14.797	4.932 10	0.96 0.	012	
Residual Err	or :	5 2.250	2.250	0.450			
Total	14	4 367.	632				
ObsStdOrde	r	C4 Fit	SE Fit	Residual 3	St Resid	ł	
9 9 13	3.770) 14.647	0.525	-0.877	-2.10 F	ł	
15 15	11.56	50 11.49	4 0.669	0.066	1.42	Х	
R-Sq = 99.3	9%						

Estimated regression second order model regression for corrosion rate by Cassandra

Y= 11.4943+2.69035*x*1+2.62453*x*2-3.32845*x*3-0.491541*X*1.*x*1-0.0538244*x*2*x*2+0.0915044*x*3.*x*3+ 0.903750*x*1.*x*2+0.0112500*x*1.*x*3-1.01625*x*2.*x*3



Figure 3-Surface plot calculated by CASSANDRA program at pCO₂ of 1.1 bar



Figure 4. Contour plot calculated by CASSANDRA program at $p\mathrm{CO}_2$ of 1.1 bar

TABLE VIII. ANALYSIS OF VARIANCE CALCULATED BY CASSANDRA
CORROSION RATE PROGRAM FOR EXPONENTIAL MODEL REGRESSION.

Source DF Seq SS Adj SS Adj MS F P
Regression 9 3.31590 3.31590 0.36843 105.92 0.000
Linear 3 3.18664 3.18664 1.06221 305.37 0.000
Square 3 0.04947 0.04947 0.01649 4.74 0.063
Interaction 3 0.07978 0.07978 0.02659 7.65 0.026
Residual Error 5 0.01739 0.01739 0.00348
Total 14 3.33329
ObsStdOrder C4 Fit SE Fit Residual St Resid
9 9 2.700 2.775 0.046 -0.074 -2.03 R
15 15 2.382 2.379 0.059 0.004 0.87 X
R-Sq = 99.48%

Fig. 3 and Fig. 4 is model corrosion rate behavior calculated by Cassandra software program. This regression has a coefficient determination about of 99%. Only have an uncertainty of 0.6%. And very satisfied confidence level is shown by an exponential regression coefficient (99.48%). A polynomial curve of corrosion rate in CO_2 environment at the range of temperature, pH, and CO_2 pressure reveal that corrosion rate has a transition temperature at the temperature of 80 °C.

The corrosion rate determined by software program calculation of Cassandra confirm with NORSOK program. A reduction of corrosion rate occurred on the surface due to corrosion product at a certain temperature range 70^{0} C– 80^{0} C. According to the figure 1 and 3, this scaling temperature is influenced by factors set.

Parabolic polynomial dependence between the corrosion rate and temperature has been found by researchers. This is attributed to carbonate scale formation. Schmitt[4], recorded that carbonate scale will be observed grow during 48 h of CO2 corrosion of steel grade J 55 in brine at 80° C under 5 bar of CO₂. He stated that oriented texture and morphology carbonate scales grown on steels depend on steel microstructure. Considering of the growth of carbide scale also was studied by by Brown [25]. He observed that scale on steel surface repair at temperature of 80°C and low pH.Critical scaling tendency decrease with increasing of carbon content is explained by Hunnik[26]. He argued that at the temperature interested, degree of carbon saturated have been exceeded. The other researcher in acociated with this is Nesic[27]. Nesic proposed a model of iron carbonate (FeCO) film growth which film growth occurs by precipitation of iron carbonate saturated. Then he recorded that very protective films and low corrosion rates are predicted at high pH, temperature, CO_2 partial pressure and Fe^{2+} ion concentration.

B. Verification with Experimental Data

In the case of pH effect, response surface, Figure 1-3,calculate a linear correlation of pH to corrosion rate. The increase of pH will decrease corrosion rate. This trend result is the same as both NORSOK and Cassandra program calculation. The decrease of corrosion rate, it may relate to barrier properties of thin layer forming layer of corrosion products. This result is supported by Hoffmeister[28]observation. Even more, he found that not only uniform corrosion was retarded but also reduce initiation times of crevice corrosion.





The effect of pH on corrosion rate was studied in solutions saturated with CO_2 with the addition of HAc in the pH range from 4 to 5.5 using RSM. The results are shown in Figure 5. In Figure 5, the model prediction is compared to Nesic's experimental data [27], which had shown a good fit. As the pH was increased from 4 to 6, the corrosion rate decreased from 0.8 mm/y to 0.5 mm/y.

IV. CONCLUSION

Response surface methodology is capable to analyze combine effects and reducenumber of runs requiredin the experiment rather than fully randomizeddesign. This method presents a satisfied coefficient determination of regression (more than 97%). Corrosion rate calculated by NORSOK software has a best fit on second order model regression (98%).Cassandra software model regressionfor corrosion rate shows an exponential pattern, with fitting regression of 99.4%. But, the difference with second order model which is not significant. Its differences is 0.09%.In condition elevated of temperature, determination of temperature scalingwillbe easier when using second order model corrosion regression.

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