Modern Applied Science; Vol. 13, No. 4; 2019
ISSN 1913-1844 E-ISSN 1913-1852
Published by Canadian Center of Science and Education

ANN and DOE Analysis of Corrosion Resistance Inhibitor for Mild Steel Structures in Iraq

Jehad A A Yamin[1], Eiman Ali Eh. Sheet[2] & Ahmed A Al-Amiery[3]

[1] The University of Jordan, School of Engineering, Mechanical Engineering Department, Amman, Jordan

[2] Lecturer, Energy and Renewable Energies Technology Center, University of Technology-Iraq, Baghdad, Industrial Street, Iraq

[3] Professor, Energy and Renewable Energies Technology Center, University of Technology-Iraq, Baghdad, Industrial Street, Iraq

Correspondence: Jehad, A. A. Yamin, The Universiy of Jordan, School of Engineering, Mechanical Engineering Department, Amman 11942, Jordan. Tel: 962-799-609-750. E-mail: yamin@ju.edu.jo

Received: December 27, 2018 Accepted: January 8, 2019 Online Published: March 31, 2019

doi:10.5539/mas.v13n4p80 URL: https://doi.org/10.5539/mas.v13n4p80

Abstract

Modelling of the effect of newly developed mild steel (MS) corrosion inhibitor in Iraq was investigated using artificial neural network (ANN) and Response Surface Methodology Design of Experiment (RSM-DOE) methods. The most significant parameters among the parameters studied and the optimum coating conditions was also investigated. Weight loss method (WLM) as well as Scanning electron microscope (SEM) were used in the experimental work to obtain data for modelling.

The inhibitor used was made in the center's laboratories called N-(3-Nitrobenzylidene)-2-aminobenzothiazole. The MS specimens were tested for different immersion times and corrosive solution temperatures. Different concentrations of the inhibitor from of 0, to 1000 mg/L were used in the study.

The results showed that within the concentrations studied, the corrosion inhibition performance increased with increasing N-(3-Nitrobenzylidene)-2-aminobenzothiazole concentration. The ANN model proposed with the Gaussian activation function was accurate for both testing and validation up to 99%. The RSM method used indicated that comparing time and concentration alone, inhibitor concentration was more significant than the immersion time in the corrosive solution. On the other hand, the effect of temperature and time were opposite to one another.

While increasing time of immersion increased corrosion rate, temperature effect was the opposite.

Keywords: materials science, ANN, DOE-RSM, corrosion inhibitor, mild steel corrosion

1. Introduction

Degradation of materials by corrosion represents a challenging task to chemical and materials science engineers. This phenomenon still needs to be clearly and fully understood and characterized.

This puts heavy pressure on materials scientists, researchers and industries to try to find solution for this disaster causing phenomena that causes the industrial components to be life-limited (Guthrie et al., 2010).

Several studies in the literature tried to shed light on this serious phenomenon (Melchers et al., 2010, Han and Song, 2008, Melchers, 2006, Xue et al., 2001, Melchers, 2009, Jones, 1996, Eiman and Sameh, 2012). Several factors were tried and suggested to counter the effect of this reaction including proper selection of materials, improved service and maintenance operation and provide certain coating materials to prevent or reduce the effect of corrosion, hence, improve its service life.

Seawater is the most aggressive working environment that causes corrosion. However, due to its variability, it is difficult to simulate its effect on corrosion on the laboratory scale (Mo¨ller. Et al., 2006). Researchers reported that the rate of corrosion increased four times when put for 3 weeks in a 3.5 % NaCl solution than in natural seawater (Siddique et al., 2011).

In Iraq, one of the significant source of corrosion (especially to oil industry) is the higher-than-normal levels of sulfur and water in crude oil and its products. This makes the working environment in oil industry more aggressive to piping materials. Moreover, most of the oil reserve and supply in Iraq is in the city of Basra. This city lies at the gulf shore. This makes an additional factor for corrosion to its components. Organic inhibitors offer good performance against corrosion.

They have several advantages including being economical and more practical to be used to reduce/prevent loss of materials as a result of corrosion (Porcayo-Calderón et al., 2015, Finšgar and Milošev, 2010).

Organic compounds have corrosion inhibition characteristics as they contain heteroatoms such as nitrogen, oxygen, sulfur, phosphorous or p bonds, which act as active centers to adsorb on the metal surface (Otmaˇci´c and Stupnišek-Lisac, 2003, Marusic et al., 2011, Gece, 2011, Tüken, 2014, Millán-Ocampo et al., 2018).

Due to experimental limitations, it becomes necessary to develop other alternatives to better understand corrosion phenomena, reduce time, the number of experiments, as well as control the process. One such alternative is the use of mathematical tools. ANN, GA DOE models represent a good option to describe corrosion behavior (Chen et al., 2016, Ndukwe and Anyakwo, 2017).

ANN is based on the biological functions of the brain where connections of neurons form a network. The prediction performance depends on a learning stage and corresponds to the correlation of the inputs and outputs of the model (Khaled and Mobarak, 2012, Khaled, 2010, Millán-Ocampo et al., 2018). Several researchers used ANN in the prediction of corrosion inhibition in pipeline steel (Colorado-Garrido et al., 2009, Efimov et al, 2011, Hernández et al, 2009, Komijani et al., 2017).

Based on the above, this research was undertaken in the University of Technology, Energy and Renewable Energies Technologies Center to try to find solution for this problem. The main aim is to try to find a locally available, cheap, efficient and non-toxic corrosion inhibitor for mild steel. This inhibitor should not react or interfere with the material from which the targeted component is made of.

2. Experimental Work

2.1 Synthesis of the "N-(3-Nitrobenzylidene)-2-aminobenzothiazole"

The inhibitor was synthesized following the steps shown in Figure (1). The raw chemical components were procured from Sigma Aldrich. The reflux of the amino compound namely 2-aminobenzothiazole was done with carbonyl compound namely 3-nitrobenzaldehyde in molar ratio 1:1 in the presence of ethanol as solvent. The final product was then tested for its chemical and physical properties.

The melting point was found to be 226-229 oC; the yield was about 59% by volume, the CHN constituents of the product were, 60.02 (59.35) 3.35 (3.20) 15.11 (14.83) respectively. The NMR signals were; 7.42–7.97 (d, 1H, aromatic ring), 8.15 (d, 1H, HC=N). Preparation of samples: mild steel (MS) with composition C (0.21%), Si (0.38%), Mn (0.05%), P (0.09%), Al (0.01%), and Fe (99.21%) was used in this study. The samples were embedded in epoxy resin leaving a working area of 1 cm2. The samples were cleaned before the experiment by acetone with water.

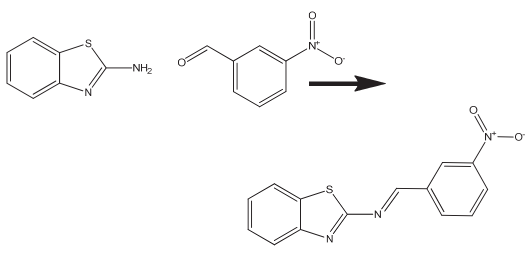


Figure 1. Synthesis steps of N-(3-Nitrobenzylidene)-2-aminobenzothiazole

2.2 Weight Loss Method

In this method, MS samples were immersed in 1 M HCL with/without the inhibitor for comparison. The study was divided into two phases. The first phase aimed at finding the best inhibitor concentration.

The inhibitor concentrations were varied for each test as follows (25, 100, 250, 500, 1000 mg/L) for the period of 1, 5, and 10 hrs and the performance was tested using weight loss technique. This part of the experiment was done at atmospheric conditions of about 303 K. In the second phase, the best performing concentration was chosen, and the temperature effect on its performance was studied.

The temperature was also varied between 303 to 333 K at an increment of 10 K. The samples were then removed, washed, dried and weighed. The inhibition efficiency (IE) was estimated using the following equation:

$$IE\% = \frac{W_o - W_1}{W_o} \times 100 \tag{1}$$

Where Wo is the weight of the MS without inhibitor while $W1$ is the weight of the MS with inhibitor in gm.

3. Artificial Neural Networks

The data consisted of three inputs: Temperature (K), Time (hr) and inhibitor concentration (mg/L). The outputs for each model were represented by the amount of material corroded and inhibition efficiency. SAS Jmp® software was used for the development of the models, evaluating different combinations of activation functions and the number of neurons was increased until the best correlation between input and output variables was achieved.

The training process was proposed to minimize the prediction error of the ANN through the different connections between weights and biases. It was possible using the Gaussian function. The outputs are produced using data from neurons in the input and hidden layers, and the bias, summation and activation functions.

The net output of each layer is calculated by summation function. The summation function used in this study is given in the equation (2) below

$$NET_i = \sum_{j=1}^{n} w_{ij} x_j + b_i \tag{2}$$

The activation function provides a curvilinear match between the input and output layers. Besides, it determines the output of the cell by processing the net input to the cell.

There are several activation functions that can be used for training. In this study, Gaussian function showed the best accuracy with least hidden neurons. The Gaussian transfer function of the ANN model in this study is given in the equation (3) below:

$$f(NET_i) = e^{-x^2} \tag{3}$$

Where "x" is a linear combination of the x variables.

In the output layer, the output of the network is generated by the data being processed by the hidden layer and transferred to the external world. The significant advantages of ANN are their learning ability and the freedom to use different learning algorithms.

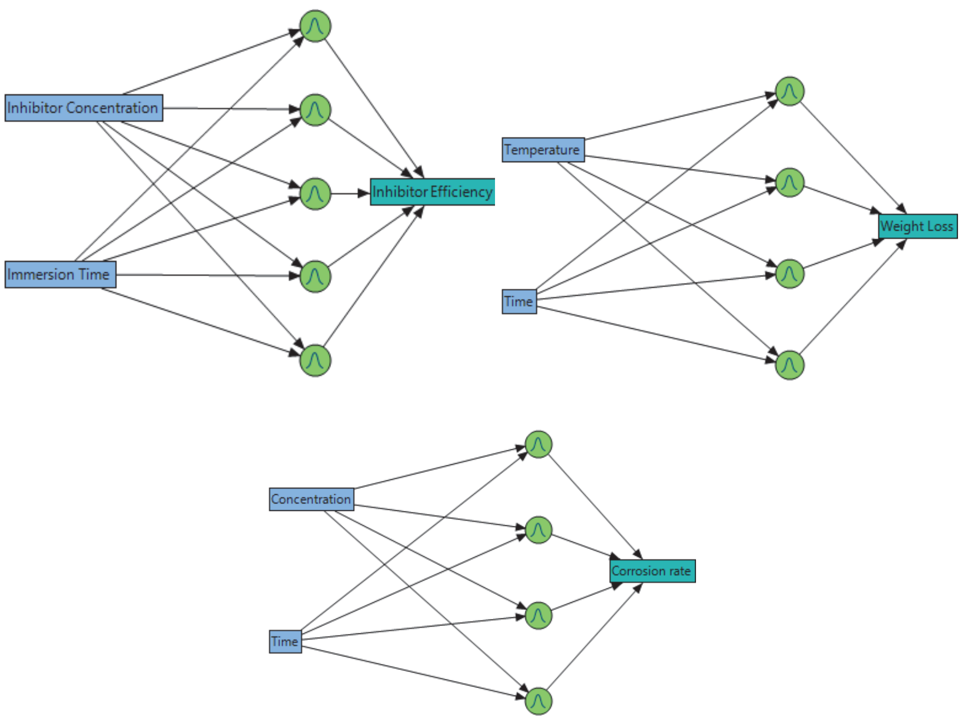
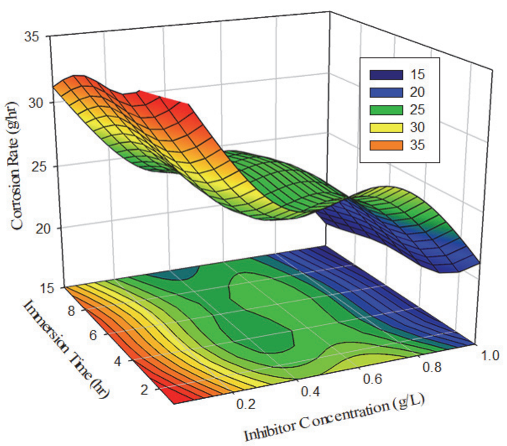
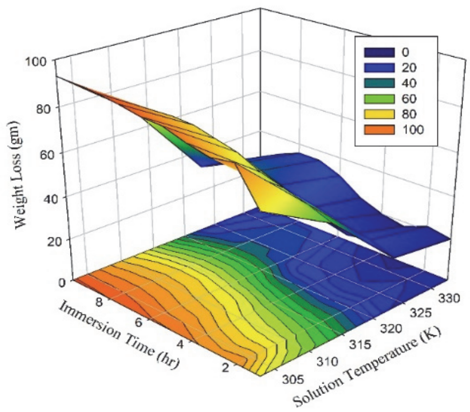


Figure 2. Sample of Artificial Neural Network Design

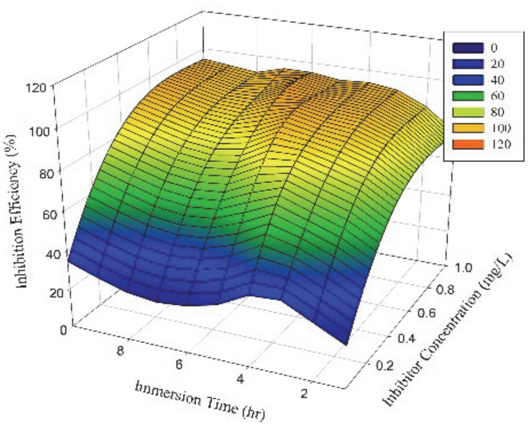
4. Results and Discussion

Results of the experimental work is shown below in Figure (3-a and 3-b). Both figures show the effect of concentration, time and immersion temperature on corrosion rate.





(a) Corrosion Efficiency (b) Weight Loss



(C) Inhibition Efficiency

Figure 3. Results of experimental data

In this simulation code, four or five hidden layers using Gaussian activation function were used. This number of hidden layers were the least needed for best accuracy and processing time.

The equation (transfer function) predicted by the software for the corrosion rate is given below in equation (4) :

$$\textit{Corrosion Rate} = 45.2076864774515 + (-8.97520766396585 * H1) + (-12.0081123893159 * H2) + (-21.0926909145727 * H3) + (-4.61094511566863 * H4) \quad (4)$$

Where :

$$H1 = Exp\ (-(0.5 * (2.45461344898404 + -0.470838982914504 * Concentration + -0.18590423834216 * Time)\ \char`^\ 2))$$

$$H2 = Exp\ (-(0.5 * ((-1.54255610403526) + 4.06032251508128 * Concentration + 0.0244779634342122 * Time)\ \char`^\ 2))$$

$$H3 = Exp\ (-(0.5 * (3.4138809823697 + -2.8303837009034 * Concentration + -0.0111200194515011 * Time)\ \char`^\ 2))$$

H4 = Exp (-(0.5 * (1.26403711603913 + -0.188474760040823 * Concentration + -0.402908064011555 * Time) ^ 2))

And that for the inhibition efficiency is given by equation (5) :

*Inhibition Efficiency (%) = (-336.403883795337) -1272.56684882916 * H1 + 6.97300787360302 *H2 -51.9271483590782 * H3 + 1520.03348903796 *H4 + 157.173511293188 * H5* (5)

Where :

H1 = Exp(-(0.5 * ((-2.09493947845829) + 0.0616913939689554 * Concentration +0.0953982424029027 * Time) ^ 2))

H2 = Exp(-(0.5 * (5.2253991758461 + 4.8621884967946 * Concentration + -1.52958939354803 * Time) ^ 2))

H3 = Exp(-(0.5 * (2.40221356488797 + -0.0300892174223173 * Concentration + -0.337216743287327 * Time) ^ 2))

H4 = Exp(-(0.5 * ((-1.73001358693346) + 0.197121885855249 * Concentration +0.071824279566637 * Time) ^ 2))

H5 = Exp(-(0.5 * ((-0.351344184959364) + 1.41475182114702 * Concentration +0.00746193183415178 * Time) ^ 2))

As for the effect of temperature and time on weight loss (gm), the model predicted is shown below in equation (6):

*Weight Loss = 94.1130944324328 + (14.0023294159015 * H1) + (-8.19740814091276 * H2) +(-71.7395728792427 * H3) + (-11.2296111840034 * H4)* (6)

Where :

H1 = Exp (-(0.5 * ((-10.8373839167341) + 0.0255007466696847 *Temperature+0.352748363738454 * Time) ^ 2))

H2 = Exp (-(0.5 * ((-11.5138524959298) + 0.040481643303725 * Temperature +-0.103172855097328 * Time) ^ 2))

H3 = Exp (-(0.5 * ((-27.8290001869095) + 0.0842734476364645 * Temperature+0.00955024148366429 * Time) ^ 2))

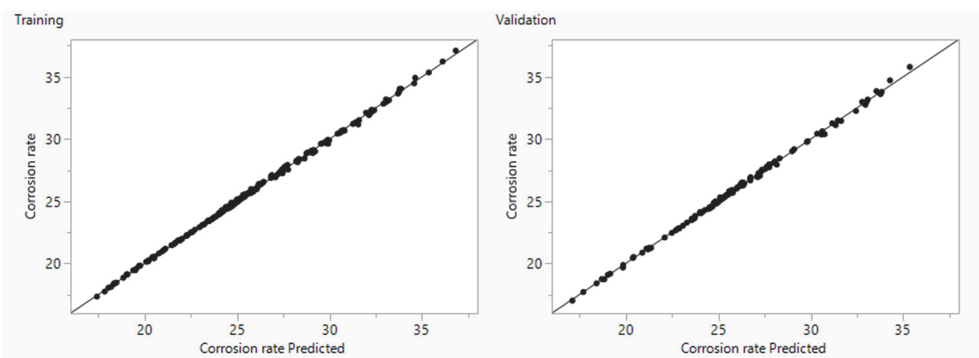
H4 = Exp (-(0.5 * ((-23.5932348468283) + 0.0781045471361819 * Temperature +-0.60396786866101 * Time) ^ 2))

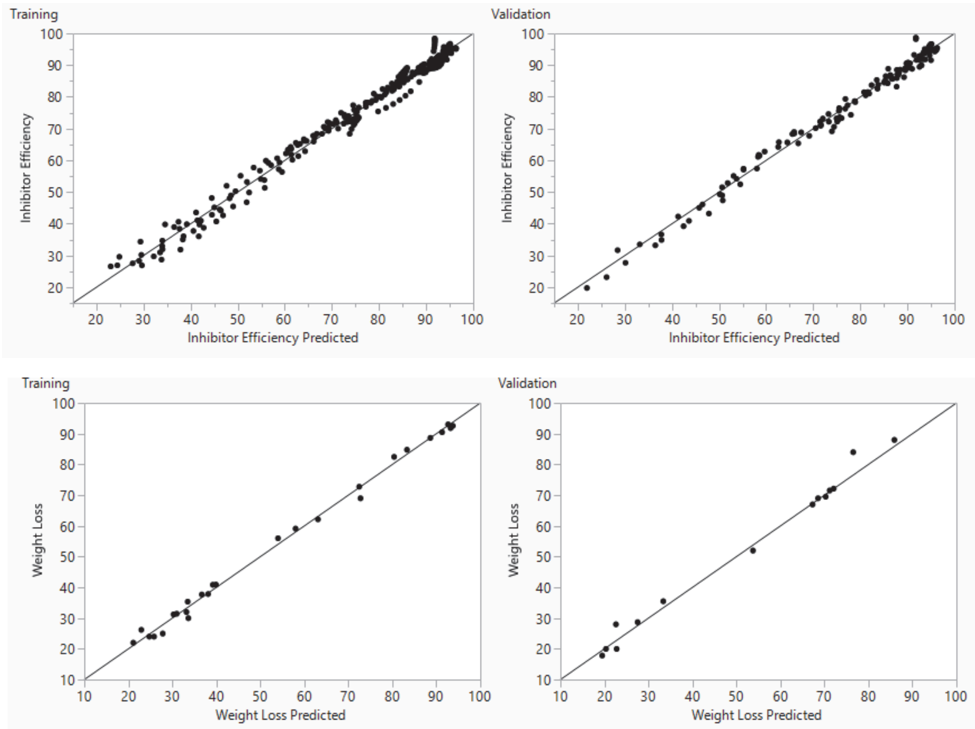
The mathematical model predicted by the algorithm has the following accuracy features:

Table 1. Model specifications

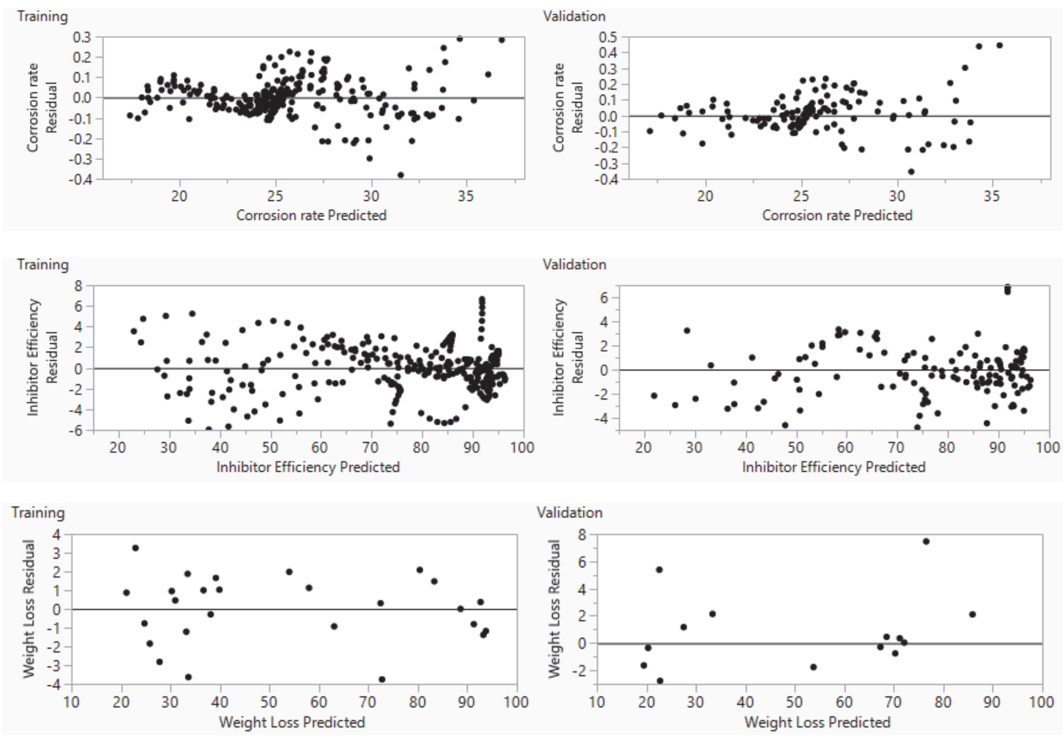
	Corrosion rate (mg/hr)		Inhibition Efficiency		Weight Loss (mg)	
Measures	Training	Validation	Training	Validation	Training	Validation
R^2	0.995	0.995	0.986	0.987	0.99	0.98
RMSE	0.0906	0.117	2.275	2.152	1.73	2.8
Mean Abs Error	0.0668	0.084	1.74	1.63	1.42	1.9
SSE	2.184	1.85	1377.48	620.905	78.45	109.6

The results of the ANN model for both testing and validation is shown below in Figures (4 a and b).





(a) Plot of predicted with experimental for both testing and validation



(b) Plot of residual error for both prediction and validation

Figure 4. Model performance

The last stage is to use this data obtained in the design of experiment (Response Surface Methodology) to find the most significant factor. This was done using Minitab R18.

This is shown by what is called Pareto chart. These charts are used to identify the most frequent factors affecting certain output, the most common causes of this type of output. Pareto charts can help to focus improvement efforts on areas where the largest gains can be made.

Main effects plots are used to the differences between level means for one or more factors. There is a main effect when different levels of a factor affect the response differently. A main effects plot graphs the response mean for each factor level connected by a line.

The Pareto and main effect charts for all factors of this study are shown below in Figures (5).

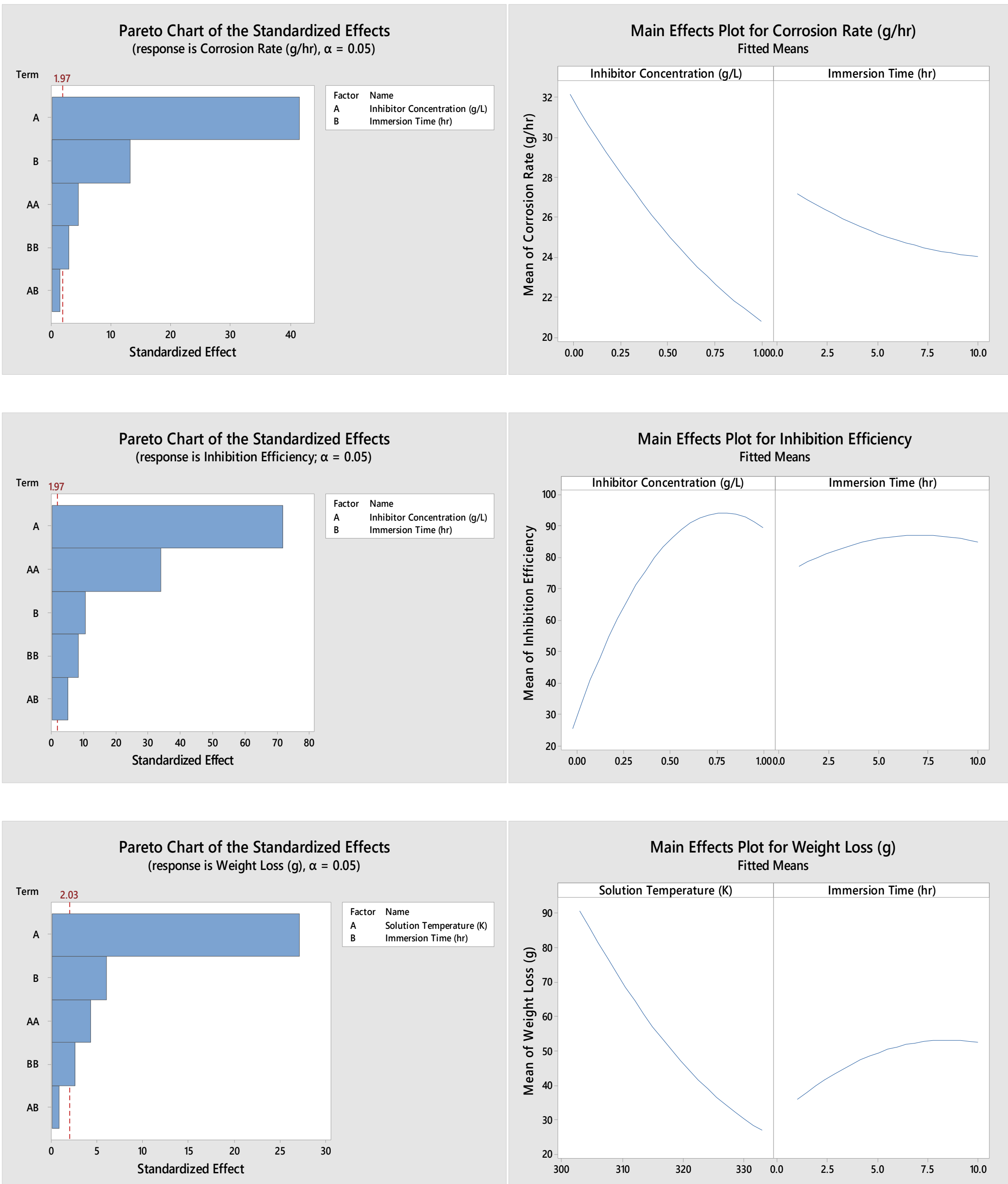


Figure 5. Response Surface Methodology

Based on these figures, inhibitor concentration showed most significant effect on corrosion (inhibition of corrosion) and inhibition efficiency compared with immersion time.

Figures also showed no significant interaction between those two variables (AB), and that concentration has some sort of squared effect on preventing inhibitor.

The main effect clearly shows that both factors affect the rate of corrosion, but to different levels. There is clear significant effect for inhibitor concentration over the immersion time.

Regarding inhibition efficiency, main effect figures clearly show that there is an optimum value for both factors that gives best efficiency. This at around 0.75 g/L concentration with 7.5 hrs immersion rate.

Based on the figures, it is clear that corrosive solution temperature showed most significant effect on corrosion (indicated by weight loss) compared with immersion time.

The figure also showed no significant interaction between those two variables (AB), and that both variables have some sort of second order effect on preventing corrosion.

The main effect clearly shows that both factors affect the rate of corrosion, but to different levels. There is clear significant effect for solution temperature over the immersion time.

5. Conclusion

An experimental and numerical analysis of the corrosion inhibition performance for local materials made in Iraq.

The effect of concentration of inhibitor coating as well as the corrosive solution temperature and time of exposure to such environment was studied.

Further, mathematical modelling was made with the aim of finding the most significant parameter on corrosion rate.

Results showed that corrosive solution temperature as well as the inhibitor concentration were most influential to corrosion phenomena. Exposure time was effective to lesser extent.

References

Chen, F. F., Breedon, M., White, P., Chu, C., Dwaipayan, M., Thomas, S., Sapper, E., & Cole, I. (2016). Correlation between molecular features and electrochemical properties using an artificial neural network. *Mater. Des., 112,* 410–418. https://doi.org/10.1016/j.matdes.2016.09.084

Colorado-Garrido, D., Ortega-Toledo, D. M., Hernández, J. A., González-Rodríguez, J. G., & Uruchurtu, J. (2009). Neural networks for Nyquist plots prediction during corrosion inhibition of a pipeline steel. *J. Solid State Electrochem, 13*, 1715–1722. https://doi.org/10.1007/s10008-008-0728-7

Efimov, A. A., Moskvin, L. N., Pykhteev, O. Y., & Epimakhov, T. V. (2011). Simulation of corrosion processes in the closed system steel-water coolant. *Radiochemistry, 53,* 19–25. https://doi.org/10.1134/S1066362211010036

Eiman, A. E. S., & Sameh, S. A. (2012). Evaluation of Corrosion Inhibitor Blend Efficiency in Recirculation Cooling Water of Al-Doura Refinery. *Journal of Petroleum Research & Studies, 212,* 138-157.

Finšgar, M., & Milošev, I. (2010). Inhibition of copper corrosion by 1, 2, 3-benzotriazole: A review. *Corros. Sci., 52,* 2737–2749. https://doi.org/10.1016/j.corsci.2010.05.002

Gece, G. (2011) Drugs: A review of promising novel corrosion inhibitors. *Corros. Sci., 53*, 3873–3898. https://doi.org/10.1016/j.corsci.2011.08.006

Guthrie, J., Battat, B., & Grethlein, C. (2010). Accelerated corrosion testing, Jeffrey Guthrie. *The AMPTIAC Quarterly, 6*(3), 11–15.

Han, L., & Song, S. (2008). A measurement system based on electro-chemical frequency modulation technique for monitoring the early corrosion of mild steel in seawater. *Corros Sci., 50*(6), 1551–1557. https://doi.org/10.1016/j.corsci.2008.02.009

Hernández, M., Genescá, J., Uruchurtu, J., & Barba, A. (2009). Correlation between electrochemical impedance and noise measurements of waterborne coatings. *Corros. Sci., 51,* 499–510. https://doi.org/10.1016/j.corsci.2008.12.011

Jones, D. A. (1996). *Principles and prevention of corrosion*. Prentice-Hall, New Jersey.

Khaled, K. F. (2010). Studies of iron corrosion inhibition using chemical, electrochemical and computer simulation techniques. *Electrochim. Acta., 55,* 6523–6532. https://doi.org/10.1016/j.electacta.2010.06.027

Khaled, K. F., & Mobarak, N. A. (2012). A Predictive Model for Corrosion Inhibition of Mild Steel by Thiophene and Its Derivatives Using Artificial Neural Network. *Int. J. Electrochem. Sci., 7,* 1045–1059.

Komijani, H., Rezaeihassanabadi, S., Parsaei, M. R., & Maleki, S. (2017). Radial basis function neural network for electrochemical impedance prediction at presence of corrosion inhibitor. *Period. Polytech. Chem. Eng., 61*, 128–132. https://doi.org/10.3311/PPch.9295

Marusic, K., Otmaˇci´c-Curkovic, H., & Takenout, H. (2011). Inhibiting effect of 4-methyl-1-p-tolylimidazole to the corrosion of bronze patinated in sulphate medium. *Electrochim. Acta, 56*, 7491–7502.

Melchers, R. E. (1999). Corrosion uncertainty modeling for steel structures. *J Constr Steel Res., 52*, 3–19. https://doi.org/10.1016/S0143-974X(99)00010-3

Melchers, R. E. (2006). Modelling immersion corrosion of structural steels in natural fresh and brackish waters. *Corros Sci., 48*(12), 4174–4201. https://doi.org/10.1016/j.corsci.2006.04.012

Melchers, R. E., & Chernov, B. B. (2010). Corrosion loss of mild steel in high temperature hard freshwater. *Corros Sci., 52,* 449–454. https://doi.org/10.1016/j.corsci.2009.10.002

Millán-Ocampo, D. E., Parrales-Bahena, A., González-Rodríguez, J. G., Silva-Martínez, S., Porcayo-Calderón, J., & Hernández-Pérez J. A. (2018). Modelling of Behavior for Inhibition Corrosion of Bronze Using Artificial Neural Network (ANN). *Entropy, 20*(6), 409. https://doi.org/10.3390/e20060409

Mo¨ller, H., Boshoff, E. T., & Froneman, H. (2006). The corrosion behaviour of a low carbon steel in natural and synthetic seawaters. *J S Afr I Min Metall, 106*, 585–592.

Ndukwe, A. I., & Anyakwo, C. N. (2017). Modelling of Corrosion Inhibition of Mild Steel in Hydrochloric Acid by Crushed Leaves of Sida Acuta (Malvaceae). *Int. J. Eng. Sci., 6*, 22–33. https://doi.org/10.9790/1813-0601032233

Otmaˇci´c, H., & Stupnišek-Lisac, E. (2003). Copper corrosion inhibitors in near neutral media. *Electrochim. Acta, 48,* 985–991. https://doi.org/10.1016/S0013-4686(02)00811-3

Porcayo-Calderón, J., Martínez de la Escalera, L. M., Canto, J., Casales-Díaz, M., & Imidazoline (2015). Derivatives Based on Coffee Oil as CO2 Corrosion Inhibitor. *Int. J. Electrochem. Sci., 10,* 3160–3176.

Siddique, R., Aggarwal, P., & Aggarwal, Y. (2011). Prediction of compressive strength of self-compacting concrete containing bottom ash using artificial neural networks. *Adv Softw Eng., 42*, 780–786. https://doi.org/10.1016/j.advengsoft.2011.05.016

Tüken, T., Giray, E. S., Fındıkkıran, G., & Sıg, G. (2014). A new corrosion inhibitor for copper protection. *Corros. Sci., 84*, 21–29. https://doi.org/10.1016/j.corsci.2014.03.004

Xue, J., Michelle, S., & Fatt, H. (2001). Buckle propagation in pipelines with non-uniform thickness. *Ocean Eng., 28*, 1383–1392. https://doi.org/10.1016/S0029-8018(00)00056-1

Copyrights