OPTIMIZATION AND DEVELOPMENT OF PREDICTIVE MODELS FOR THE CORROSION INHIBITION OF MILD STEEL IN SULPHURIC ACID BY METHYL-5-BENZOYL-2-BENZIMIDAZOLE CARBAMATE (MEBENDAZOLE)

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Abstract: In this paper, we report the optimization and development of predictive models for the corrosion inhibition of mild steel in sulphuric acid by Methyl-5-benzoyl-2-benzimidazole Carbamate (Mebendazole). Design expert was used to analyze the corrosion inhibition related process parameters, such as corrosion penetration rate, inhibition efficiency, inhibitor concentration, acid concentration, weight loss, and their relationships. An attempt is made to obtain the optimal settings for this corrosion inhibition related process parameters. Design methodology, weight loss measurement, open-circuit potential analysis, Tafel polarization, etc. were used for the evaluation of inhibition efficiency of mebendazole for mild steel in H2SO4. The corrosion inhibition process parameters were optimized and predictive mathematical models developed using the Response Surface Methodology (RSM) using the central composite design (CCD) tool of Design Expert software version 11. Experimental and theoretical corrosion inhibition parameters were used to develop a nonlinear regression model to predict the optimal inhibition efficiency. Also, a quadratic model was generated, with predicted optimum inhibition efficiency of 88.4095% obtained having very near unity desirability of 0.914. The developed model shows that inhibition efficiency is related to the inhibitor concentration.

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PUBLIC INTEREST STATEMENT

The economic impact and problems posed by metallic corrosion has drawn strong attention from researchers and engineers worldwide. Corrosion of mild steel is a major concern in chemical processing and metallurgical plants, oil and gas industry, manufacturing industries, marine operations, boiler plants, and power generation plants. High cost is associated with the replacement of metallic parts in these applications. The consequence often leads to plant shutdowns, breakdown of industrial equipment, reduced efficiency, industrial downtime, high maintenance cost due to replacement of damaged part, wastage of valuable resources, and expensive overdesign. Corrosion inhibition is of great practical importance, being extensively employed in curtailing wastage of engineering materials and minimizing costs of corrosion control.
immersion time, and acid concentration. The regression models generated are successfully used to predict the corrosion inhibition behaviour of mebendazole for low carbon steel in sulphuric acid medium.

Subjects: Materials Science; Mining, Mineral & Petroleum Engineering; Chemical Engineering

Keywords: mebendazole; optimization; corrosion inhibition; predictive modelling; response surface methodology

1. Introduction

Mild steel finds wide application in various industries due to its low cost, availability, and its amenability to various fabrication techniques (Akintola et al., 2019; Odoni, Edoziuno, & Chukwurah, 2017; Ofuyekpone, Akaluzia, & Edibo, 2017; Olawale, Aderin, Talabi, Nwokocha, & Ameh, 2017). Mild steel corrosion in acidic environment occurs due to an electrochemical reaction with the service environment (Alasvand Zarasvand & Rai, 2014; Fiori-Bimbi, Alvarez, Vaca, & Gervasi, 2015; Loto, Popoola, Fayomi, & Loto, 2012; Uhlig & King, 1972). To prolong the design and service life span of mild steel, effective corrosion protection techniques, such as the use of corrosion inhibitors, are required (Arthur, Jonathan, Ameh, & Anya, 2013; Gece, 2011; Patni, Agarwal, & Shah, 2013; Rani & Basu, 2012).

Response surface methodology (RSM) is a statistical and experimental design tool in which a selected dependent variable responds to variations in one or more independent or factor variables (Adesina, Fatoba, Farotade, & Aderin, 2017; Kothari & Garg, 2014, Mohammed, Abakr, Yusup, & Kazi, 2016). It is a useful technique for predictive modelling and optimization of the corrosion inhibition properties (Kothari & Garg, 2014). The aim is to optimize the response of the independent variable (output) which is also affected by varying the design settings for one or more independent variables (inputs) (Kothari & Garg, 2014). Many researchers on corrosion inhibition have applied experimental design software packages and techniques like the RSM in optimizing the corrosion inhibition process parameters and predicting their responses (Anadebe, Onukwuli, Omotioma, & Okafor, 2018; Evelin, Rodriguez, Borbolla, Alvarado-Rodriguez, & Pandiyan, 2016; Okafor, Anadebe, & Onukwuli, 2019; Omotioma & Onukwuli, 2016; Onukwuli, Nwanekezie, Anadebe, & Omotioma, 2017; Onukwuli & Omotioma, 2016; Praveen, Manjunatha, Gaurav, & Mohammed, 2016). The results of gravimetric measurement for the corrosion inhibition study of *Sapium ellipticum* leaf extract was modelled and a regression model was generated for predicting its inhibition efficiency (IE) at optimal combination of corrosion inhibition factors (Okafor et al., 2019). Anadebe et al. (2018) carried out an optimization study on the use of ethanol extract of bitter kola leaf as corrosion inhibitor for mild steel in HCl solution. RSM was utilized to predict the optimum range of parameters for controlling corrosion and hence reduced the number of experimental runs. Bitter kola leaf extract was also used to inhibit corrosion of mild steel in H₂SO₄ by Onukwuli et al. (2017), and the results was optimized and modelled using the central composite design (CCD) and RSM of design expert software. A quadratic regression model was generated that adequately explained the relationship between IE and the considered factors of the corrosion inhibition process. Evelin et al. (2016) optimized and developed predictive model for the corrosion inhibition of carbon steel in acidic service by imidazole and benzimidazole derivatives. Multiple linear regression models were obtained using the Moby Digs software. The model shows a quantitative structure–property relationship with descriptors that contribute to predict the corrosion IE of the imidazole and benzimidazole derivatives. The predictive model obtained indicates that IE of the inhibitors is related to the molecule volume charge, electronegativity, and aromaticity. Values of IE agree for both model proposed and experimental results, which indicates that theoretical model can be used to predict experimental results adequately. Omotioma and Onukwuli (2016) modelled and optimized the IE of the extract of pawpaw leaves for mild steel in HCl using RSM. The analysis of the experimental results by RSM shows the simultaneous dependency of weight loss
WL), corrosion rate (CR), and IE on inhibitor concentration, temperature, and time. A quadratic model was generated with optimum IE of 80.29%. Onukwuli and Omotioma (2016) also modelled and optimized the IE of mango extract using RSM and found that IE of the extract was concentration-, temperature-, and time dependent. Odejobi and Akinbulumo (2019) carried out modelling and optimization of the IE of Euphorbia heterophylla extract for mild steel in HCl media using the CCD of RSM. 89.8% IE was obtained at optimum combination of the extract concentration, acid concentration, temperature, and immersion time.

This study is aimed at optimizing the IE of Methyl-5-benzoyl-2-benzimidazole Carbamate (Mebendazole; MBZ) as a suitable corrosion inhibitor for mild steel in H\textsubscript{2}SO\textsubscript{4} and generating mathematical models for predicting the IE in the acid medium. It is important to examine the optimal inhibitive property of mebendazole and develop mathematical regression models for predicting the corrosion inhibition properties.

2. Experimental methods

2.1. Data collection

The materials used for this design optimization and modelling work were collected from experimentally generated data. The experimental corrosion data were obtained by both gravimetric (WL) and electrochemical techniques (potentiodynamic/Tafel polarization) (Edoziuno & Odoni, 2018). These techniques were used to evaluate the effectiveness of various concentrations of Methyl-5-benzoyl-2-benzimidazole Carbamate (0.5, 1.0, 1.5, 2.0, and 2.5 g) in inhibiting the corrosion of mild steel in 0.5, 1.0, and 1.5 M solutions of H\textsubscript{2}SO\textsubscript{4} at room temperature (298 K) and elevated temperature (333 K), respectively.

For the weight-loss measurements, mild steel test coupons with average dimensions of 15 mm thickness, 10 mm length, and a 5 mm hole machined at the centre for suspension were weighed and completely submerged by suspension in each of the prepared test media contained in a 100-ml beaker with cover for different immersion period of 24, 48, 72, 96, and 120 h with and without MBZ inhibitor addition. They were taken out after the required exposure time had elapsed. The corrosion products were removed by washing with distilled water, rinsing in acetone, and drying in ambient air. The coupons were then re-weighed to determine their final weights. The WL are then computed and recorded.

The result data for the potentiodynamic polarization was obtained using an autolab potentiostat/galvanostat, VersaSTAT 4 Electrochemical System, controlled from a suitably equipped PC by a Universal Serial Bus (USB) interface using the Versa Studio electrochemistry software package. A conventional three-electrode Pyrex glass cell was used for each of the experiments. Test coupons encapsulated in a polyester resin with total area of 1 cm\textsuperscript{2} exposed to the corrosive media were used as working electrode and a platinum rod as counter electrode. Ag/AgCl rod was the reference electrode. These electrodes were connected by means of a luggin capillary to the electrolytic cell. All electrochemical tests were performed in stagnant aerated solutions at room temperature and 60°C. The test coupon was submersed in the test solution for one hour to attain an equilibrium open circuit potential. The potentiodynamic polarization test was maintained between a potential range of $-250$ mV and $+250$ mV with respect to the corrosion potential using linear sweep technique at a scan rate of 1 mV/s. To obtain the corrosion current density ($i_{corr}$) values, the linear Tafel segments of the anodic and cathodic curves were extrapolated to equilibrium corrosion potential. The polarization experiment was carried out in the blank sulphuric acid solution and the solutions containing known amount of the inhibitor.

2.2. Optimization of inhibition efficiency and other corrosion properties using RSM

Employing RSM, the IE and other experimental corrosion properties were modelled and optimized based on CCD tool of Design Expert (DoE) software version 11. A quadratic design model was applied during the analysis. Inhibitor concentration (0.5–2.5 g/100 ml), acid concentration
and immersion time (24–120 h) were set as the independent variables (Factors 1, 2, and 3), while WL, CR, and IE were set as the response variables (Responses 1–3). A total of 90 runs of experiments were carried out to obtain the responses of the dependent variables shown in the design layout (Table 1).

3. Results and discussion

3.1. Results of RSM analysis

Figures 1 and 2 are representative output showing the variations in the analysis of variance (ANOVA), predictive modelling, and optimization results. The results indicated that the WL, corrosion penetration rate, and IE are functions of inhibitor concentration, acid concentration, and immersion time. As observed in Figure 1a–c, a linear trend was noticeable in the diagnostics plots of predicted IE against the actual IE. A similar trend was noticed in the same plots of predicted CR against the actual CR and predicted versus actual WL. These trends show that the design model is adequate to predict the IE of MBZ, the corrosion penetration rate and the WL of mild steel in the inhibited environment. The predicted versus actual values of the IE, corrosion penetration rate, and WL were highly correlated. Surface plots are very useful in understanding the behaviour of chosen output/response variable, when independent variables/factors are varied within their respective ranges or targets considered in the experimental design. Two factors are normally controlled concurrently while automatically keeping the remaining factors at their respective optimal values. The 3-D surface plot (Figure 2a–c) showed the relationship between the independent factors (inhibitor concentration and acid concentration) and IE of mebendazole, corrosion penetration rate of the inhibited solution on mild steel and the WL of mild steel in the inhibited environment. The nature of the 3-D surface plots suggests significant interactions among the concerned factors and response variables during the corrosion protection process by the MBZ inhibitor. The summary of the design parameters are contained in Table 2.

3.2. Mathematical model

The experimental data collected with regard to CCD is utilized in developing nonlinear mathematical models that indicate the responses of the dependent variables at various levels of the factors. Final predictive model equations of IE of MBZ, CR, and WL as a function of the considered factors both in terms of coded and actual factors were generated. The quadratic model with combined significant and insignificant terms is presented in Equations (1)–(3), respectively. The regression models relates the IE, corrosion penetration rate, and WL with acid concentration (A), inhibitor concentration (B), and exposure time (C), respectively. A nonlinear, regression model sufficiently described the relationship between the dependent/response variables; IE, CR, WL, and the independent factors; acid concentration (A), inhibitor concentration (B), and exposure time (C), respectively. The predictive equations can be expressed both in terms of coded factors and actual factors. When the model equations are expressed in terms of coded factors they are useful in making predictions about the responses of the dependent variables for specified levels of each factor. By default, the high levels of the factors are given positive coding as +1, on the other hand the low levels of the factors are negatively coded as −1. The usefulness of the coded equation lies in the identification of the relative impact of the factors through comparison of the coefficients of the factor. Positive and negative factor coefficients respectively indicate maximum positive and negative impact of the factors to the response of the variable. Conversely, the mathematical models expressed in terms of actual factors can be used to make predictions about the response for given levels of each factor. Here, the levels should be specified in the original units for each factor. It is noteworthy that the model equation given in terms of the actual factors should not be used to ascertain the relative impact of each factor because the coefficients in this case are scaled to accommodate the units of each factor and the intercept is not at the centre of the design space. The generated regression models are significant and can be used to navigate the design space.

Final Mathematical Models for Inhibitor Efficiency Expressed Based on the Coded Factors (Equation (1)).
| Std | Run | Factor 1 | Factor 2 | Factor 3 | Response 1 | Response 2 | Response 3 |
|-----|-----|----------|----------|----------|------------|------------|------------|
|     |     | A: Acid Conc (M) | B: Inhibitor Conc (g) | C: Exposure Time (hours) | Weight Loss (g) | Corrosion Rate (mm/yr) | Inhibitor Efficiency (%) |
| 45  | 1   | 0.5      | 0        | 24       | 1.77       | 1.30984    | 0          |
| 72  | 2   | 0.5      | 0        | 48       | 3.18       | 1.17663    | 0          |
| 28  | 3   | 0.5      | 0        | 72       | 1.76       | 0.434145   | 0          |
| 84  | 4   | 0.5      | 0        | 96       | 2.11       | 0.390361   | 0          |
| 37  | 5   | 0.5      | 0        | 120      | 3.74       | 0.553535   | 0          |
| 16  | 6   | 0.5      | 0.5      | 24       | 0.14       | 0.103603   | 92.0904    |
| 85  | 7   | 0.5      | 0.5      | 48       | 0.17       | 0.0629017  | 94.6541    |
| 21  | 8   | 0.5      | 0.5      | 72       | 0.36       | 0.0888024  | 79.5455    |
| 9   | 9   | 0.5      | 0.5      | 96       | 0.5        | 0.0925025  | 76.3033    |
| 68  | 10  | 0.5      | 0.5      | 120      | 0.41       | 0.0606816  | 89.0374    |
| 30  | 11  | 0.5      | 1        | 24       | 0.11       | 0.0814022  | 93.7853    |
| 36  | 12  | 0.5      | 1        | 48       | 0.27       | 0.0814022  | 93.0818    |
| 66  | 13  | 0.5      | 1        | 72       | 0.22       | 0.0542681  | 87.5       |
| 56  | 14  | 0.5      | 1        | 96       | 0.45       | 0.0832522  | 78.673     |
| 24  | 15  | 0.5      | 1        | 120      | 0.35       | 0.0518014  | 90.6417    |
| 61  | 16  | 0.5      | 1.5      | 24       | 0.07       | 0.0518014  | 96.0452    |
| 53  | 17  | 0.5      | 1.5      | 48       | 0.22       | 0.0814022  | 93.0818    |
| 88  | 18  | 0.5      | 1.5      | 72       | 0.1        | 0.024673   | 94.3182    |
| 29  | 19  | 0.5      | 1.5      | 96       | 0.28       | 0.0518014  | 86.7299    |
| 2   | 20  | 0.5      | 1.5      | 120      | 0.32       | 0.0473613  | 91.4439    |
| 7   | 21  | 0.5      | 2        | 24       | 0.06       | 0.0444012  | 96.6102    |
| 50  | 22  | 0.5      | 2        | 48       | 0.13       | 0.0481013  | 95.9119    |

(Continued)
| Std | Run | Factor 1 A: Acid Conc (M) | Factor 2 B: Inhibitor Conc (g) | Factor 3 C: Exposure Time (hours) | Response 1 Weight Loss (g) | Response 2 Corrosion Rate (mm/yr) | Response 3 Inhibitor Efficiency (%) |
|-----|-----|--------------------------|-------------------------------|----------------------------------|---------------------------|-----------------------------------|----------------------------------|
| 71  | 23  | 0.5                      | 2                             | 72                               | 0.32                      | 0.789355                          | 81.8182                          |
| 69  | 24  | 0.5                      | 2                             | 96                               | 0.38                      | 0.703019                          | 81.9905                          |
| 4   | 25  | 0.5                      | 2                             | 120                              | 0.48                      | 0.710419                          | 87.1658                          |
| 10  | 26  | 0.5                      | 2.5                           | 24                               | 0.08                      | 0.592016                          | 95.4802                          |
| 80  | 27  | 0.5                      | 2.5                           | 48                               | 0.15                      | 0.555015                          | 95.283                           |
| 32  | 28  | 0.5                      | 2.5                           | 72                               | 0.13                      | 0.320675                          | 92.6136                          |
| 62  | 29  | 0.5                      | 2.5                           | 96                               | 0.31                      | 0.573515                          | 85.3081                          |
| 13  | 30  | 0.5                      | 2.5                           | 120                              | 0.35                      | 0.518014                          | 90.6417                          |
| 64  | 31  | 1                        | 2.5                           | 24                               | 2.06                      | 1.52444                           | 0                                |
| 90  | 32  | 1                        | 0                             | 48                               | 2.23                      | 0.825122                          | 0                                |
| 39  | 33  | 1                        | 0                             | 72                               | 2.91                      | 0.717819                          | 0                                |
| 20  | 34  | 1                        | 0                             | 96                               | 4.42                      | 0.817722                          | 0                                |
| 82  | 35  | 1                        | 0                             | 120                              | 2.82                      | 0.417371                          | 0                                |
| 27  | 36  | 1                        | 0.5                           | 24                               | 0.19                      | 0.140604                          | 90.7767                          |
| 79  | 37  | 1                        | 0.5                           | 48                               | 0.16                      | 0.592016                          | 92.8251                          |
| 25  | 38  | 1                        | 0.5                           | 72                               | 0.37                      | 0.912691                          | 87.2852                          |
| 57  | 39  | 1                        | 0.5                           | 96                               | 0.54                      | 0.999027                          | 87.7828                          |
| 11  | 40  | 1                        | 0.5                           | 120                              | 0.72                      | 0.106563                          | 74.4681                          |
| 12  | 41  | 1                        | 1                             | 24                               | 0.14                      | 0.103603                          | 93.2039                          |
| 3   | 42  | 1                        | 1                             | 48                               | 0.2                       | 0.074002                          | 91.0314                          |
| 78  | 43  | 1                        | 1                             | 72                               | 0.46                      | 0.11347                           | 84.1924                          |
| 49  | 44  | 1                        | 1                             | 96                               | 0.41                      | 0.075852                          | 90.724                           |

(Continued)
| Std | Run | Factor 1 | Factor 2 | Factor 3 | Response 1 | Response 2 | Response 3 |
|-----|-----|----------|----------|----------|------------|------------|------------|
|     |     | A: Acid Conc (M) | B: Inhibitor Conc (g) | C: Exposure Time (hours) | Weight Loss (g) | Corrosion Rate (mm/yr) | Inhibitor Efficiency (%) |
| 74  | 45  | 1         | 1        | 120       | 0,61       | 0,0902824  | 78,3688    |
| 31  | 46  | 1         | 1,5      | 24        | 0,11       | 0,0814022  | 94,6602    |
| 46  | 47  | 1         | 1,5      | 48        | 0,19       | 0,0703019  | 91,4798    |
| 51  | 48  | 1         | 1,5      | 72        | 0,41       | 0,0246673  | 96,5636    |
| 33  | 49  | 1         | 1,5      | 96        | 0,12       | 0,075852   | 90,724     |
| 35  | 50  | 1         | 1,5      | 120       | 0,12       | 0,0444012  | 89,3617    |
| 81  | 51  | 1         | 2        | 24        | 0,11       | 0,066018   | 95,6311    |
| 34  | 52  | 1         | 2        | 48        | 0,12       | 0,0296008  | 96,4126    |
| 63  | 53  | 1         | 2        | 72        | 0,19       | 0,0468679  | 93,4708    |
| 83  | 54  | 1         | 2        | 96        | 0,39       | 0,0407011  | 95,0226    |
| 19  | 55  | 1         | 2        | 120       | 0,11       | 0,0577216  | 86,1702    |
| 15  | 56  | 1         | 2,5      | 24        | 0,11       | 0,0814022  | 94,6602    |
| 22  | 57  | 1         | 2,5      | 48        | 0,22       | 0,0814022  | 90,1345    |
| 65  | 58  | 1         | 2,5      | 72        | 0,34       | 0,0592016  | 91,7526    |
| 5   | 59  | 1         | 2,5      | 96        | 0,22       | 0,0629017  | 92,3077    |
| 54  | 60  | 1         | 2,5      | 120       | 0,28       | 0,0414411  | 90,0709    |
| 42  | 61  | 1,5       | 0        | 24        | 2,96       | 2,19046    | 0          |
| 55  | 62  | 1,5       | 0        | 48        | 5,64       | 2,0868     | 0          |
| 48  | 63  | 1,5       | 0        | 72        | 3,37       | 0,831289   | 0          |
| 18  | 64  | 1,5       | 0        | 96        | 6,07       | 1,12298    | 0          |
| 86  | 65  | 1,5       | 0        | 120       | 9,31       | 1,37792    | 0          |
| 14  | 66  | 1,5       | 0,5      | 24        | 0,15       | 0,111003   | 94,9324    |

(Continued)
| Std | Run | Factor 1 | Factor 2 | Factor 3 | Response 1 | Response 2 | Response 3 |
|-----|-----|----------|----------|----------|------------|------------|------------|
|     |     | A: Acid Conc (M) | B: Inhibitor Conc (g) | C: Exposure Time (hours) | Weight Loss (g) | Corrosion Rate (mm/yr) | Inhibitor Efficiency (%) |
| 8   | 67  | 1.5      | 0.5      | 48       | 0.22       | 0.0814022  | 96.0993    |
| 89  | 68  | 1.5      | 0.5      | 72       | 0.35       | 0.0863357  | 89.6142    |
| 40  | 69  | 1.5      | 0.5      | 96       | 0.54       | 0.0999027  | 91.1038    |
| 43  | 70  | 1.5      | 0.5      | 120      | 0.46       | 0.0680818  | 95.0591    |
| 76  | 71  | 1.5      | 1        | 24       | 0.1        | 0.074002   | 96.6216    |
| 6   | 72  | 1.5      | 1        | 48       | 0.27       | 0.0999027  | 95.2128    |
| 60  | 73  | 1.5      | 1        | 72       | 0.19       | 0.0468679  | 94.362     |
| 73  | 74  | 1.5      | 1        | 96       | 0.36       | 0.0666018  | 94.0692    |
| 38  | 75  | 1.5      | 1        | 120      | 0.68       | 0.100643   | 92.696     |
| 17  | 76  | 1.5      | 1.5     | 24       | 0.13       | 0.0962026  | 95.6081    |
| 47  | 77  | 1.5      | 1.5     | 48       | 0.3        | 0.111003   | 94.6809    |
| 67  | 78  | 1.5      | 1.5     | 72       | 0.2        | 0.049347   | 94.0653    |
| 26  | 79  | 1.5      | 1.5     | 96       | 0.36       | 0.0666018  | 94.0692    |
| 1   | 80  | 1.5      | 1.5     | 120      | 0.78       | 0.115443   | 91.6219    |
| 58  | 81  | 1.5      | 2       | 24       | 0.12       | 0.0888024  | 95.9459    |
| 44  | 82  | 1.5      | 2       | 48       | 0.22       | 0.0814022  | 96.0993    |
| 77  | 83  | 1.5      | 2       | 72       | 0.13       | 0.0320675  | 96.1424    |
| 59  | 84  | 1.5      | 2       | 96       | 0.19       | 0.0351509  | 96.8699    |
| 52  | 85  | 1.5      | 2       | 120      | 0.42       | 0.0621617  | 95.4887    |
| 41  | 86  | 1.5      | 2.5     | 24       | 0.12       | 0.0888024  | 95.9459    |
| 75  | 87  | 1.5      | 2.5     | 48       | 0.17       | 0.0629017  | 96.9858    |
| 87  | 88  | 1.5      | 2.5     | 72       | 0.08       | 0.0197339  | 97.6261    |
| 70  | 89  | 1.5      | 2.5     | 96       | 0.45       | 0.0832522  | 92.5865    |
| 23  | 90  | 1.5      | 2.5     | 120      | 0.6        | 0.0888024  | 93.5553    |
Figure 1. (a) Diagnostic plot of predicted versus actual inhibition efficiency. (b) Diagnostic plot of predicted versus actual corrosion rate. (c) Diagnostic plot of predicted versus actual weight loss.
Figure 2. (a) 3-D surface plot of inhibition efficiency. (b) 3-D surface plot of corrosion rate. (c) 3-D surface plot of weight loss.

Design-Expert® Software
Factor Coding: Actual

Inhibitor Efficiency (%)
- Design points above predicted value
- Design points below predicted value

Actual Factor
C: Exposure Time = 72

Corrosion Rate (mm/yr)
- Design points above predicted value
- Design points below predicted value

Actual Factor
C: Exposure Time = 72

Weight Loss (g)
- Design points above predicted value
- Design points below predicted value

Actual Factor
C: Exposure Time = 72
Table 2. (a & b) The summary of the design parameters for both the factors and response variables

(a) Factors

| Factor | Name             | Units | Type  | Minimum | Maximum | Coded Low | Coded High | Mean   | Std. Dev. |
|--------|------------------|-------|-------|---------|---------|-----------|-----------|--------|-----------|
|        | Acid Conc        | M     | Numeric | 0.5000  | 1.50    | -1 ↔ 0.50 | +1 ↔ 1.50 | 1.0000 | 0.4105    |
| B      | Inhibitor Conc   | g     | Numeric | 0.0000  | 2.50    | -1 ↔ 0.50 | +1 ↔ 2.50 | 1.25   | 0.8587    |
| C      | Exposure Time    | hours | Numeric | 24.00   | 120.00  | -1 ↔ 24.00 | +1 ↔ 120.00 | 72.00  | 34.13     |

(b) Responses

| Response | Name             | Units | Obs | Analysis  | Minimum | Maximum | Mean   | Std. Dev. | Ratio | Transform | Model  |
|----------|------------------|-------|-----|-----------|---------|---------|--------|-----------|-------|-----------|--------|
| R1       | Weight Loss      | g     | 90  | Polynomial | 0.06    | 9.31    | 0.8369 | 1.50      | 155.17 | None      | Quadratic |
| R2       | Corrosion Rate   | mm/yr | 90  | Polynomial | 0.0197339 | 2.19,046 | 0.2344 | 0.4318 | 111,00  | None      | Quadratic |
| R3       | Inhibitor Efficiency | % | 90 | Polynomial | 0 | 97,6261 | 76.29 | 34.63 | N/A | None | Quadratic |

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https://doi.org/10.1080/23311916.2020.1714100
| Source               | Sum of Squares | df | Mean Square | F-value | p-value | p-value | p-value |
|---------------------|----------------|----|-------------|---------|---------|---------|---------|
| Model Terms         | 86,086,67      | 9  | 9565,19     | 37.03   | < 0.0001| Significant |
| A—Acid Conc.       | 273,37         | 1  | 273,37      | 1.06    | 0.3067  |          |
| B—Inhibitor Conc.  | 5753,66        | 1  | 5753,66     | 22.28   | < 0.0001|          |
| C—Exposure Time     | 348,34         | 1  | 348,34      | 1.35    | 0.2490  |          |
| AB                  | 7,33           | 1  | 7,33        | 0.0284  | 0.8667  |          |
| AC                  | 55,38          | 1  | 55,38       | 0.2144  | 0.6446  |          |
| BC                  | 6,21           | 1  | 6,21        | 0.0241  | 0.8771  |          |
| A²                  | 43,66          | 1  | 43,66       | 0.1691  | 0.6820  |          |
| B²                  | 36,075,43      | 1  | 36,075,43   | 139.67  | < 0.0001|          |
| C²                  | 8.62           | 1  | 8.62        | 0.0334  | 0.8555  |          |
| Residual            | 20,662.57      | 80 | 258.28      |         |         |          |
| Cor Total           | 1.067E+05      | 89 |             |         |         |          |
**Table 4. Fit statistics for the response parameter considered (inhibitor efficiency)**

| Std. Dev. | 16.07 | $R^2$ | 0.8064 |
|-----------|-------|-------|--------|
| Mean      | 76.29 | Adjusted $R^2$ | 0.7847 |
| C.V. %    | 21.07 | Predicted $R^2$ | 0.7535 |
|           |       | Adeq Precision | 18.3380 |

**Table 5. ANOVA results for the response parameter (corrosion rate)**

| Source          | Sum of Squares | df | Mean Square | $F$-value | $p$-value |
|-----------------|----------------|----|-------------|-----------|-----------|
| Model Terms     | 11.49          | 9  | 1.28        | 20.02     | < 0.0001  |
| A—Acid Conc.   | 0.0965         | 1  | 0.0965      | 1.51      | 0.2223    |
| B—Inhibitor Conc. | 0.6077      | 1  | 0.6077      | 9.53      | 0.0028    |
| C—Exposure Time | 0.1047         | 1  | 0.1047      | 1.64      | 0.2039    |
| AB              | 0.4782         | 1  | 0.4782      | 7.50      | 0.0076    |
| AC              | 0.0009         | 1  | 0.0009      | 0.0145    | 0.9045    |
| BC              | 0.5968         | 1  | 0.5968      | 9.36      | 0.0030    |
| $A^2$           | 0.0415         | 1  | 0.0415      | 0.6509    | 0.4222    |
| $B^2$           | 4.18           | 1  | 4.18        | 65.50     | < 0.0001  |
| $C^2$           | 0.1066         | 1  | 0.1066      | 1.67      | 0.1997    |
| Residual        | 5.10           | 80 | 0.0638      |           |           |
| Cor Total       | 16.59          | 89 |             |           |           |

**Table 6. Fit statistics for the response parameter considered (corrosion rate)**

| Std. Dev. | 0.2525 | $R^2$ | 0.6925 |
|-----------|--------|-------|--------|
| Mean      | 0.2344 | Adjusted $R^2$ | 0.6579 |
| C.V. %    | 107.72 | Predicted $R^2$ | 0.5786 |
|           |        | Adeq Precision | 18.3107 |

Inhibitor Efficiency = $+103.19 + 2.22A + 11.35B - 2.90C + 0.4093AB + 1.36AC - 0.4352BC + 1.48A^2 - 32.10B^2 + 0.7397C^2$  \( (1) \)

Final Mathematical Models for Corrosion Rate Expressed Based on the Coded Factors (Equation (2)).

Corrosion Rate = $-0.1397 + 0.0418A - 0.1166B - 0.0503C - 0.1045AB - 0.0056AC + 0.1349BC + 0.0456A^2 + 0.3455B^2 + 0.0823C^2$  \( (2) \)

Final Mathematical Models for Weight Loss Expressed Based on the Coded Factors (Equation (3)).
Table 7. ANOVA results for the response parameter (weight loss)

| Source            | Sum of Squares | df | Mean Square | F-value | p-value |
|-------------------|----------------|----|-------------|---------|---------|
| Model Terms       | 136.61         | 9  | 15.18       | 19.06   | < 0.0001| Significant |
| A—Acid Conc.     | 1.49           | 1  | 1.49        | 1.87    | 0.1758  |
| B—Inhibitor Conc.| 7.28           | 1  | 7.28        | 9.14    | 0.0034  |
| C—Exposure Time   | 3.38           | 1  | 3.38        | 4.25    | 0.0426  |
| AB                | 7.55           | 1  | 7.55        | 9.49    | 0.0028  |
| AC                | 3.83           | 1  | 3.83        | 4.81    | 0.0312  |
| BC                | 0.6468         | 1  | 0.6468      | 0.8124  | 0.3701  |
| A<sup>2</sup>     | 48.28          | 1  | 48.28       | 60.64   | < 0.0001|
| B<sup>2</sup>     | 0.2408         | 1  | 0.2408      | 0.3024  | 0.5839  |
| Residual          | 63.70          | 80 | 0.7962      |         |         |
| Cor Total         | 200.30         | 89 |             |         |         |
3.3. Statistical test and analysis of models

The CCD generated nonlinear mathematical models showing the levels of relationship between the response variables and various factors were tested to ascertain their statistical adequacy, practical usefulness and significance with the aid of ANOVA test. The statistical test (significance, ANOVA, coefficient of determination, and prediction accuracy) and model analysis (surface plots) were carried out in order to provide complete insight into the corrosion inhibition process (Chate, Manjunath, Deshpande, & Parappagoudar, 2017). The statistical analysis of the data is presented in Tables 3–8. Factor coding used for the ANOVA test for all the response parameters is coded, while the Sum of squares is Type III—Partial.

The Model F-value of 37.03 obtained as shown in Table 3 is a significant model as there is only a 0.01% likelihood that an F-value that is this large could occur as a result of noise. The source of noise may be attributed to non-controllable factors of the inhibition process. On the other hand, P-values below 0.0500 is an indication of significant model terms. From the ANOVA results in Table 3, B, and B² are significant model terms. P-values above 0.1000 is indicative of insignificant model terms. When there are many model terms that are not significant in a predictive model equation (excluding those model terms required to support hierarchy), model reduction in the analysis settings is recommended for the improving the model.

The F-value of this model presented in Table 5 (20.02) is a significant model, because there is only a 0.01% possibility that an F-value of this magnitude could be obtained from the model owing to noise. Significant model terms are usually associated with P-values that are less than 0.0500. In the case of this response model, the significant model terms are B, AB, BC, and B². Conversely, P-values higher than 0.1000 are used to indicate the insignificant model terms.

The F-value of the model in Table 7 (19.06) is a significant model since there is only a 0.01% likelihood that such large F-value could be obtained due to noise. P-values below 0.0500 indicate significant model terms. Thus, B, C, AB, BC, and B² are significant model terms in the WL mathematical model. P-values that are higher than 0.1000 show insignificant model terms. When there are many insignificant model terms in a response model (excluding those model terms required to support hierarchy), applying model reduction technique may enhance the developed model.

The $R^2$ values predicted (Tables 4, 6 and 8) reasonably agreed with the Adjusted $R^2$ values, since their differences are less than 0.2, respectively. The $R^2$ are close to unity, thus the fitted models are considered to be good ones (Kothari & Garg, 2014).

Adeq Precision is a useful measure of the signal to noise ratio. A ratio that is above 4 is adequate and desirable. The ratio of 18.338, 18.311, and 18.469 obtained in Tables 4, 6 and 8, respectively, indicates an adequate signal. Hence these models can be satisfactorily used to navigate the design space (Saha & Banerjee, 2013).

\[
\text{Weight Loss} = -0.3755 + 0.1640A - 0.4035B + 0.2856C - 0.4155AB + 0.1948AC \\
- 0.3417BC + 0.1798A^2 + 1.17B^2 + 0.1237C^2
\]  

(3)
| Number | Acid Conc | Inhibitor Conc | Exposure Time | Weight Loss | Corrosion Rate | Inhibitor Efficiency | Desirability |
|--------|-----------|----------------|---------------|-------------|----------------|----------------------|--------------|
| 1      | 0.500     | 0.957          | 56,363        | -0.096      | 0.020          | 88,411               | 0.914        |
| 2      | 0.500     | 0.956          | 56,424        | -0.096      | 0.020          | 88,381               | 0.914        |
| 3      | 0.500     | 0.961          | 55,873        | -0.103      | 0.020          | 88,642               | 0.914        |
| 4      | 0.500     | 0.953          | 56,805        | -0.090      | 0.020          | 88,199               | 0.914        |
| 5      | 0.500     | 0.952          | 56,934        | -0.088      | 0.020          | 88,136               | 0.914        |
| 6      | 0.500     | 0.964          | 55,456        | -0.109      | 0.020          | 88,837               | 0.914        |
| 7      | 0.500     | 0.946          | 57,632        | -0.079      | 0.020          | 87,806               | 0.914        |
| 8      | 0.500     | 0.969          | 54,859        | -0.117      | 0.020          | 89,130               | 0.914        |
| 9      | 0.504     | 0.956          | 56,528        | -0.095      | 0.020          | 88,328               | 0.914        |
| 10     | 0.500     | 0.943          | 58,091        | -0.073      | 0.020          | 87,593               | 0.914        |
| 11     | 0.505     | 0.947          | 57,513        | -0.081      | 0.020          | 87,861               | 0.914        |
| 12     | 0.500     | 0.936          | 58,871        | -0.062      | 0.020          | 87,231               | 0.914        |
| 13     | 0.512     | 0.949          | 57,409        | -0.083      | 0.020          | 87,912               | 0.914        |
| 14     | 0.514     | 0.958          | 56,315        | -0.099      | 0.020          | 88,427               | 0.914        |
| 15     | 0.500     | 0.927          | 60,059        | -0.045      | 0.020          | 86,680               | 0.914        |
| 16     | 0.500     | 0.947          | 56,104        | -0.087      | 0.025          | 88,001               | 0.914        |
| 17     | 0.530     | 0.970          | 55,000        | -0.118      | 0.020          | 89,047               | 0.914        |
| 18     | 0.500     | 0.955          | 54,445        | -0.102      | 0.028          | 88,492               | 0.914        |
| 19     | 0.500     | 0.923          | 61,410        | -0.032      | 0.017          | 86,339               | 0.914        |
| 20     | 0.500     | 0.901          | 63,590        | 0.007       | 0.020          | 85,091               | 0.914        |
| 21     | 0.846     | 0.960          | 64,306        | 0.020       | 0.020          | 87,724               | 0.912        |
3.4. Optimization results

Twenty one optimized solutions (Table 9) for IE (%), CR, and WL respectively were obtained for various predicted optimal levels of immersion time (h) acid concentration and inhibitor concentration (g/100 ml), respectively. One automatically selected optimized solution having a near unity desirability (0.914) (with optimum IE of 88.41% was achieved at optimal acid concentration of 0.5 M H₂SO₄, inhibitor concentration of 0.957 g/100 ml and immersion duration of 56.363 h) is depicted in the overlay plot (Figure 3).

4. Conclusions

A quadratic model was generated, with high IE of 88.4095%. The inhibitor was effective on the surface treatment of mild steel in acidic environment. It is therefore recommended that me bendazole be utilized as corrosion inhibitor in oil well acidizing. Additionally, it can serve in an industrial acid pickling and cleaning of other related surface treatment of mild steel in acidic environment. The quadratic predictive model equations adequately explains the relationship among: WL, CR, IE of MBZ and the considered independent factors of the inhibition process in the operating environment.

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