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Experimental Investigation and Machine Learning Prediction of Corrosion Rate and Inhibition Efficiency of ASTM A36 Steel in NaCl Media

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Abstract

This work aims to investigate the effects of corrosion factors including variations in inhibitor type, inhibitor concentration, and immersion duration on the corrosion rate and inhibition efficiency of ASTM A36 steel in a 3.5% NaCl solution. Corrosion rate measurement was performed by adding different inhibitors, which are sodium nitrite and ascorbic acid inhibitors. Each sample was immersed for 7, 14, and 21 days in different inhibitor concentrations of 250, 500, and 750 ppm. Weight loss was obtained to calculate the corrosion rates, then it obtained inhibition efficiency. The experimental data were collected as the dataset to develop corrosion rate and inhibition efficiency models. The dataset was trained using several regression algorithms, and the selected model was evaluated by the highest R^2 value. The best performance of predictive corrosion rate model was 1.00 from XGBR and 0.86 for the inhibition efficiency model obtained from RFR. The Feature effect was analyzed by SHapley Additive exPlanations (SHAP) and Pearson correlation to provide model interpretability. The most significant factor affecting corrosion rate was inhibitor concentration, while immersion duration had the strongest effect on inhibitor efficiency. Therefore, the provides a comprehensive understanding to explain the relationship between corrosion control conditions in ASTM S36 steel.

Keywords: *Corrosion, inhibitor, concentration, machine learning.*

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INTRODUCTION

The electrochemical interaction between a metal and its surrounding environment can lead to irreversible metal degradation, known as corrosion [1]. This corrosion phenomenon is one of the primary causes of component or system failure in industry and has the potential to increase maintenance expenses and occupational safety risks [2]. The likelihood of corrosion increases when a component or system interacts with an environment containing Cl^-, such as in seawater or brackish water. This effect is because the presence of Cl^- ions can damage the passive layer on the metal surface [3]. In structural applications, ASTM A36 carbon steel is one of the most commonly used steels. This phenomenon is because ASTM A36 steel possesses excellent mechanical properties [4] and has low production costs. However, this steel has a chemical composition that does not support its corrosion resistance performance, particularly when interacting with environments containing Cl^- ions. Therefore, corrosion control methods are one of the key measures to extend the service life of ASTM A36 steel [5].

One of the most common methods of corrosion control is the addition of corrosion inhibitors. The mechanism of these corrosion inhibitors involves the formation of a protective layer on the metal surface or the reduction of the corrosion rate [6]. Compared to other methods for protecting metal from corrosion, such as coating or cathodic protection, the application of corrosion inhibitors is considered more economical, particularly when the system is in a liquid medium or solution [7]. Sodium nitrite is a type of inorganic inhibitor that has long been used to protect carbon steel against corrosion [8]. The presence of sodium nitrite promotes the formation of a protective layer on the metal surface, thereby inhibiting metal oxidation [9]. In the use of sodium nitrate as a corrosion inhibitor, one of the important process variables is its concentration. It has been reported that increasing the concentration of sodium nitrate can improve corrosion inhibition performance, and this performance is influenced by chloride ion concentration and time [10].

In addition to inorganic inhibitors, organic inhibitors known as green inhibitors offer a more environmentally friendly alternative [11]. One type of organic inhibitor that can be used to inhibit corrosion is ascorbic acid [12]. Ascorbic acid slows down the rate of corrosion by sticking to the surface of the steel. However, its inhibitory performance is also influenced by the concentration of the inhibitor used and the environmental conditions of application. For example, in a 3.5% NaCl solution, the maximum inhibition efficiency for low-carbon steel is achieved at an inhibitor concentration of 50 ppm and an immersion time of 15 days [13].

To the best of the author's knowledge, research on these corrosion inhibitors has so far focused primarily on experimental studies. Although the results are considered accurate, such experimental studies require a significant amount of time, resources, and a large number of research variables. Furthermore, it is often difficult to determine the relationships between variables in experimental studies. Currently, advancements in machine learning are opening new opportunities in the field of corrosion [14]. The use of the Random Forest machine learning algorithm can predict the corrosion rate profile of carbon steel with a mean squared error ranging from 0.005 to 0.093 [15]. This study aims to analyze the effects of the inhibitor type, added inhibitor concentration, and specimen immersion time on the corrosion rate and inhibitor efficiency of ASTM A36 steel in a 3.5% NaCl solution. This study combines experimental research with a machine learning approach, utilizing several regression algorithms to predict the corrosion rate and inhibition efficiency.

METHODS

The specimens are ASTM A36 steel with a dimension of 40 mm × 30 mm × 3 mm. The specimen surfaces were polished using 400 grit SiC paper to remove corrosion and oxides. Each sample was weighed on an analytical balance and the data were recorded as the initial mass. The corrosive media for corrosion measurement is a 3.5 % NaCl solution, representing the artificial seawater. A 3.5% NaCl solution was made by dissolving NaCl in

200 mL distilled water and pH of the solution was 6.59. Two types of inhibitors were used in this study: sodium nitrite ($NaNO_2$, Merck) and ascorbic acid ($C_6H_8O_6$, Smart Lab Indonesia) in inhibitor concentrations of 250, 500 and 750 ppm. The mass of inhibitors was varied in 0.05, 0.1, and 0.15 grams to obtain the desired concentrations and solubilized in 200 mL of a corrosive medium of 3% NaCl. The corrosion resistance was evaluated by the weight loss method, determining the weight of the specimens before and after immersion for 7, 14 and 21 days, which got the corrosion rate. All conditions (various inhibitors, concentration of inhibitor, and immersion time) were replicated three times and 18 specimens were obtained in total. The specimens were removed after corrosion immersion, washed with distilled water and cleaned with a soft-bristled toothbrush. The rinse process lasted 20–30 s on either side of the specimen without considerable mass loss. Then the specimens were rinsed with acetone and dried. The pH of the solution after sample immersion was determined by pH meter as well.

Data collection consists of the weight loss before and after corrosion measurement. The corrosion rate was calculated by equation (1) [16] whereas CR is the corrosion rate (mpy), K is 3.45×10^6 as a constant, W is the weight loss (gram), A is the surface area of specimen (cm^2), T is duration of immersion (hour), and D is density of specimen (g/cm^3). Moreover, inhibition efficiency was also determined by equation (2) [17]. Two values in the inhibition efficiency formula, such as CR_0 and CR_{inh} as the corrosion rate without and using inhibitors.

$$CR = \frac{K\ x\ W}{A\ x\ T\ x\ D} \tag{1}$$

$$Inhibition\ efficiency = \frac{CR_0 - CR_{inh}}{CR_0}\ x\ 100\% \tag{2}$$

Machine Learning Procedure

Data from experiments, including the conditions of various inhibitors, inhibitor concentration, and duration of immersion in corrosion measurement were used as a dataset for machine learning analysis. The schematic procedure of this research was illustrated in Figure 1. The main purpose of machine learning analysis is to provide information about the effect of each variable (features) on targets. In this case, the features are various inhibitors, inhibitor concentration, and duration. However, various inhibitors consist of categorical data and were transformed into binary data. Therefore, there are four features: inhibitor_sodium_nitrite, inhibitor_ascorbic_acid, concentration, and duration. The targets for predictive models are corrosion rate and inhibition efficiency. The dataset is utilized from 18 data points from the experiment. In the step of data preprocessing, the feature and target were determined. Each value was scaled using StandardScaler method. This standardization obtained values in the range of 0 until 1 to avoid the impact of scale differences between features. Data was trained to develop model predictions using various regression algorithms, including linear regression (LR), support vector regression (SVR), random forest regression (RFR), and eXtreme gradient boosting regression (XGBR). The model was validated using the Leave-One-Out Cross Validation (LOOCV) method. Hyperparameter optimizations, except linear regression were performed by Optuna [18] through 30 trials. The final hyperparameters are summarized in Table 1. The performance of models were evaluated using the coefficient of determination (R^2), root mean squared error (RMSE), and mean absolute error (MAE). SHapley Additive exPlanations (SHAP) and Pearson correlation analysis were provided to improve model interpretability. SHAP was only performed on the best-performing model for corrosion rate and inhibition efficiency. Also, Pearson's correlation is calculated to find the linear relationship between features and target. Python programming language

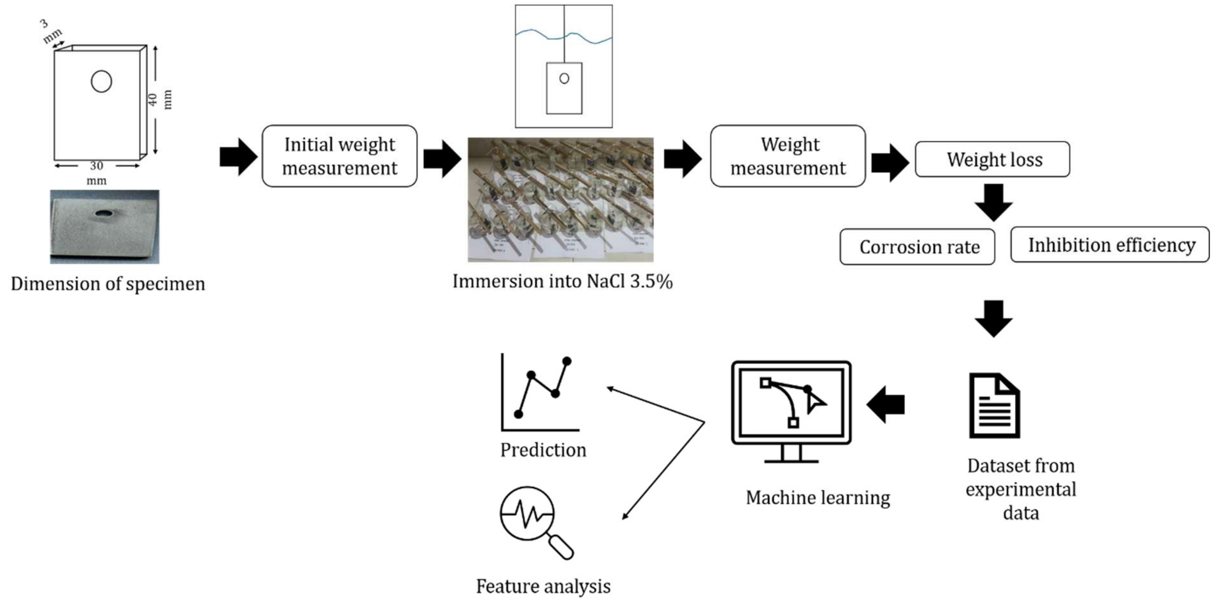


Figure 1. Schematic of experimental steps and machine learning framework

Table 1. The optimized hyperparameters for various regression

Models	Optimized hyperparameters for corrosion rate models	Optimized hyperparameters for inhibition efficiency models
LR	-	-
SVR	'kernel': 'rbf', 'C': 5.17, 'epsilon': 0.69	'kernel': 'linear', 'C': 7.81, 'epsilon': 0.93
RFR	'n_estimators': 162, 'max_depth': 18	'n_estimators': 148, 'max_depth': 9
XGBR	‘n_estimators': 84, learning_rate': 0.20, 'max_depth': 8	'n_estimators': 69, 'learning_rate': 0.012, 'max_depth': 9

is used for the development of machine learning prediction models, feature analysis and calculation of Pearson’s correlation.

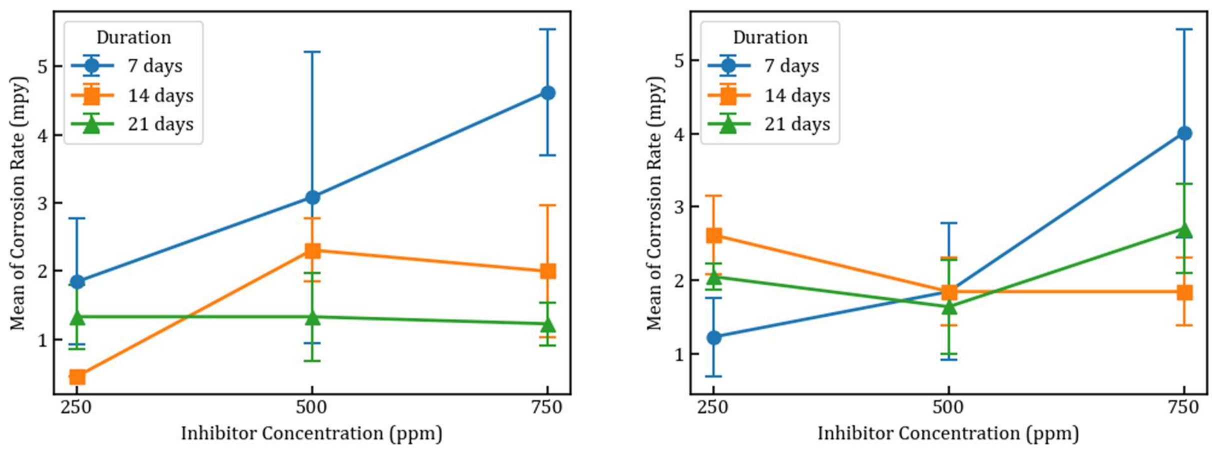
RESULTS AND DISCUSSIONS

Corrosion Rate and Inhibition Efficiency

The addition of sodium nitrite and ascorbic acid determines the effect of various inhibitors to the corrosion resistance of ASTM A36 steel. The average corrosion rates for each sample with different inhibitor concentrations and immersion durations in a 3.5% NaCl solution, are shown in Figures 2(a) and 2(b) for sodium nitrite and ascorbic acid inhibitors, respectively. The corrosion measurement was conducted three times for each experimental condition. The averages for each condition regarding corrosion rate and inhibition efficiency are summarized in Table 2. The highest corrosion rate using sodium nitrite inhibitors occurred at the shortest immersion duration, while the longest duration produced a lower corrosion rate. This result indicates that the effectiveness of the inhibitor in slowing the corrosion rate had not yet reached its optimum at 7 days, thus requiring more time to slow the corrosion rate. The trend of the highest corrosion rate during the shortest duration also occurred at higher inhibitor concentrations at 7 days, but this trend did not perform at durations of 14 and 21 days. Conversely, the use of ascorbic acid inhibitors at the shortest duration showed the lowest corrosion rate at an inhibitor concentration of 250 ppm. At concentrations more than 500 ppm, the corrosion rate tended to decrease, except for the 7-day duration sample. The decrease of corrosion rate is attributed to the increasing amount of sodium nitrite inhibitors, which form a thin oxide layer as a protective layer. Therefore, the increases in resistance at the higher inhibitor concentrations leads to decrease in ion transfer [19].

Table 2. Experimental data of corrosion rate and inhibition efficiency using various inhibitors, inhibitor concentration, and duration of immersion

No	Inhibitor	Inhibitor concentration (ppm)	Duration of immersion (days)	Mean of corrosion rate (mpy)	Mean of inhibition efficiency (%)
1	None	0	7	9.25	-
			14	3.70	-
			21	3.08	-
2	Sodium Nitrite	250	7	1.85	80.04
			14	0.46	87.56
			21	1.33	56.81
		500	7	3.08	66.67
			14	2.31	37.56
			21	1.33	56.81
		750	7	4.62	50.02
			14	2.00	45.95
			21	1.23	60.06
3	Ascorbic Acid	250	7	1.23	86.7
			14	2.62	29.18
			21	2.05	33.44
		500	7	1.85	80.04
			14	1.85	50
			21	1.64	46.75
		750	7	4.01	56.64
			14	1.85	50
			21	2.77	10.06



(a) (b)

Figure 2. (a) Mean of corrosion rate in various inhibitor concentration and duration for corrosion measurement using (a) sodium nitrite and (b) ascorbic acid inhibitor

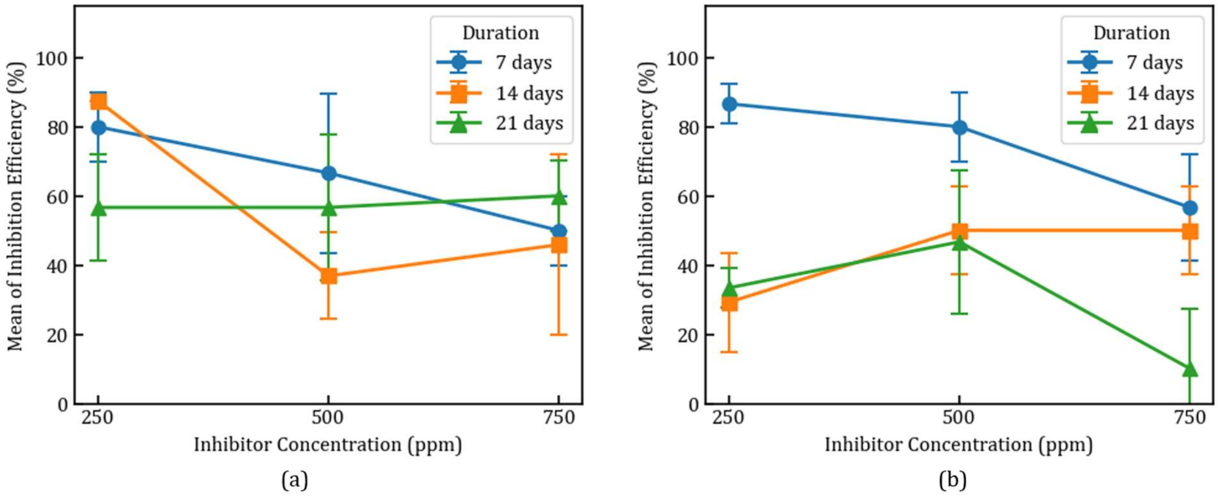


Figure 3. (a) Mean of inhibition efficiency in various inhibitor concentration and duration for corrosion measurement using (a) sodium nitrite and (b) ascorbic acid inhibitor

Figure 3 presents the effect of various experimental variables on inhibition efficiency. The results indicated that the efficiency of inhibitors was reduced, especially for the duration of 7 days for both sodium nitrite and ascorbic acid inhibitors. Moreover, the inhibitor concentration at 250 ppm to 500 ppm dramatically decreased for sodium nitrate inhibitors. Conversely, ascorbic acid inhibitor causes a significant increment over 14 days of immersion contributing to the fact that the effectiveness of inhibitor to reduce the corrosion rate of ASTM A36 in NaCl 3.5% affected by inhibitor type, concentration, and duration of immersion. Among different inhibitors, sodium nitrite demonstrated the most effective inhibition efficiency because it consistently showed higher efficiency across various concentrations and durations of immersion. The performance of sodium nitrite inhibitor is attributed to its oxidizing properties and promotes the formation of protective passive oxide for improving corrosion resistance [20].

Machine Learning Prediction and Feature Analysis

The effect of features on corrosion rate and inhibition efficiency is too complex for drawing conclusions only from experimental data. Therefore, a machine learning approach assists in developing a predictive model and identifying the influence of features on the target variables. The data obtained from the experiments was used as a dataset to train various algorithms, such as SVR, RFR, XGBR, and LR. Four desired algorithms were used for compairing the ability of predictions. SVR is a linear regression which solves an optimization using the regularization parameter and error sensitivity [21]. RFR belongs to the ensemble learning group which start of bootstrapping and end with agrregating. XGBR is able to show outstanding performance and versality [22]. The model with the best performance was selected to further explain information related to feature analysis. Model evaluation metrics used R^2, MAE, and RMSE values as shown in Table 2. The R^2, MAE, and RMSE values represent model performance on the training data, while the R^2-LOOCV value represents the validation of model performance using leave-one-out cross validation. The best model for predicting corrosion rate was obtained from the XGBR algorithm and RFR for inhibition efficiency. The low validation performance occurs due to the limited number of data points. Therefore, model generalization can be improved by increasing data points. Even though machine learning models in this study are able to make predictions, they also explain the feature effect on corrosion resistance.

Tabel 2. Metric evaluation of corrosion rate and inhibition efficiency models in various algorithms

Algorithm	Model : Corrosion rate (mpy)				Model : Inhibition efficiency (%)			
	R^2	R^2-LOOCV	MAE	RMSE	R^2	R^2-LOOCV	MAE	RMSE
SVR	0.70	0.16	0.49	0.54	0.47	0.24	0.62	0.73
RFR	0.90	0.18	0.27	0.31	0.86	-0.03	0.30	0.37
XGBR	1.00	0.28	0.00	0.00	0.52	0.24	0.52	0.69
LR	0.42	-0.00	0.66	0.76	0.48	0..14	0.60	0.72

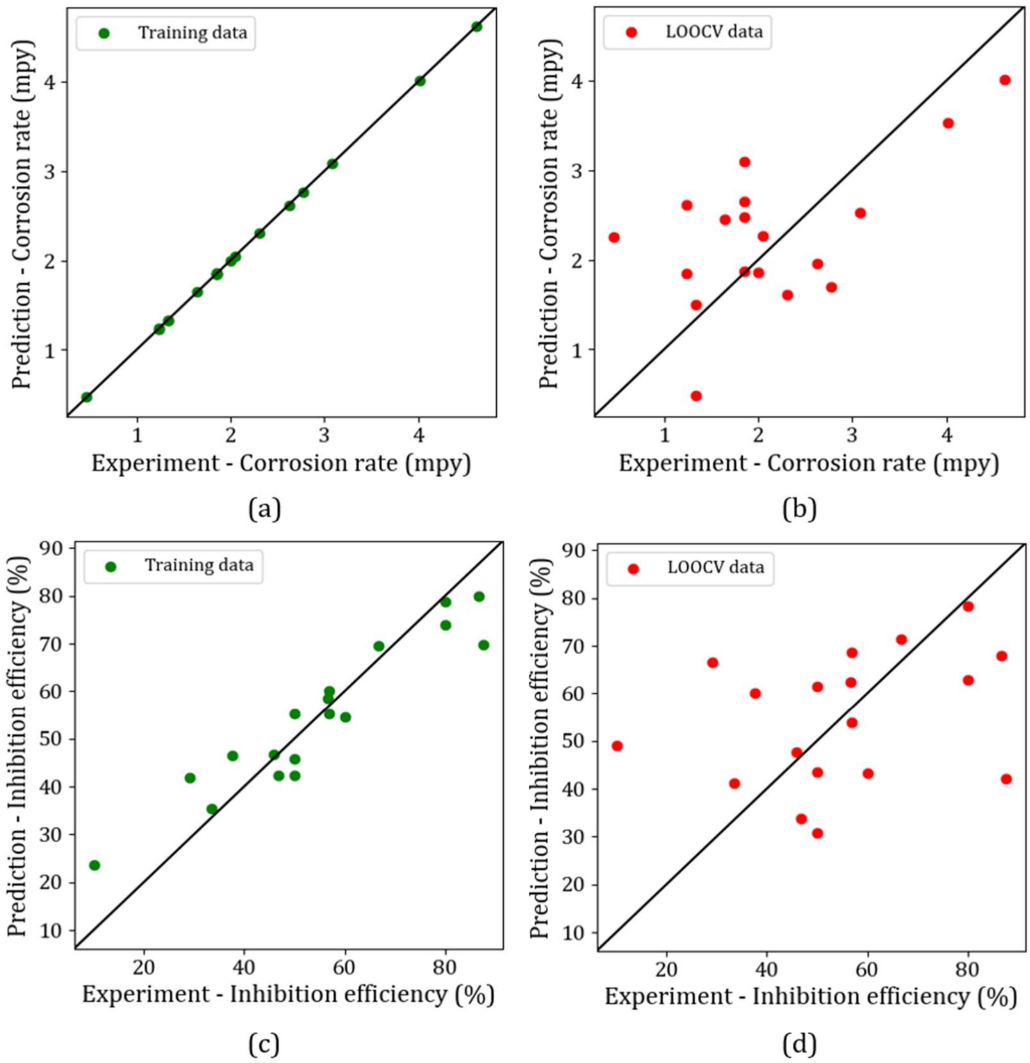


Figure 4. The comparison of experimental data and predicted corrsosion rate by XGBR algorithm on (a) training data and (b) a leave-one-out cross-validation (LOOCV), and comparison for inhibitor models developed by RFR algorithm on (c) training data and (d) a leave-one-out cross-validation (LOOCV)

The comparative value between experimental data and machine learning prediction was visualized by a parity plot. Model accuracy was graphically demonstrated by how data are distributed around the ideal diagonal line. Data points near ideal line indicate better predicted accuracy and show less bias than far-scattered data. Figure 4 (a) and (b) are parity plots for the predictive model of corrosion rate from overall data training and using leave-one-out cross-validation (LOOCV), respectively. Similarly, Figure 4 (c) and (d) present the comparison of inhibition efficiency. Although the highest model performance of corrosion rate shows perfect R^2 ($R^2 = 1$), R^2 from validation is relatively low as evidenced by the long distance data points from ideal line. The significant reduction of validation performance also occurs for inhibitor performance which contributes to low generalization ability to apply unseen data. This behavior is referred to as overfitting due to a small dataset [23].

The model with the highest was selected to provide more accurate interpretation for feature analysis on corrosion rate and inhibition efficiency through SHAP (SHapley Additive exPlanations). SHAP values determine feature analysis by assessing which each feature contributes to decreasing or increasing the target [24], [25]. Figure 5 (a) presents that inhibitor concentration has the most impact on the corrosion rate. Lower inhibitor concentrations, longer duration of immersion, and using sodium nitrite inhibitor can reduce the corrosion rate. Ascorbic acid inhibitor are not significant enough to contribute to the decrease in corrosion rate. Figure 5(b) demonstrates that immersion duration is the most important feature for inhibitor efficiency, while inhibitor concentration is the

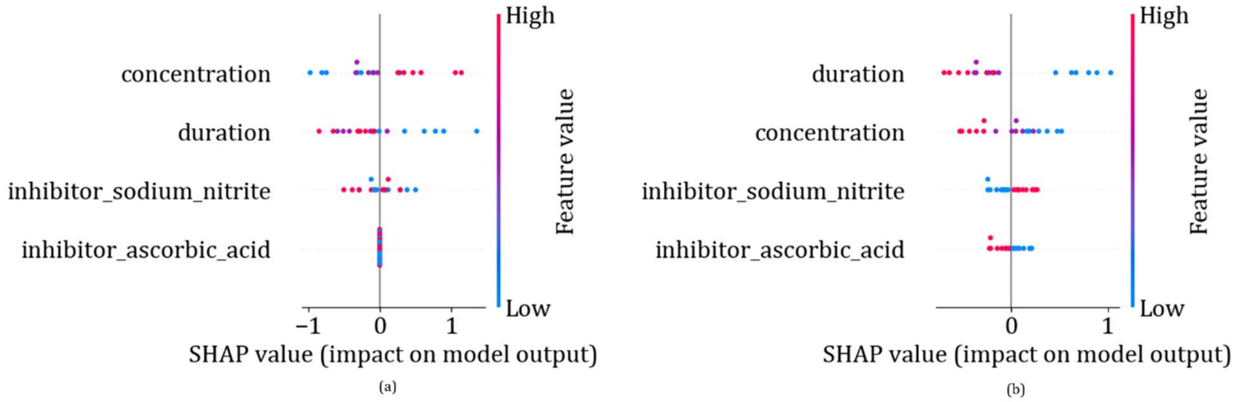


Figure 5. SHAP plots of the best-performing machine learning models (a) corrosion rate developed by XGBR algorithm and (b) inhibition efficiency obtained from RFR algorithm

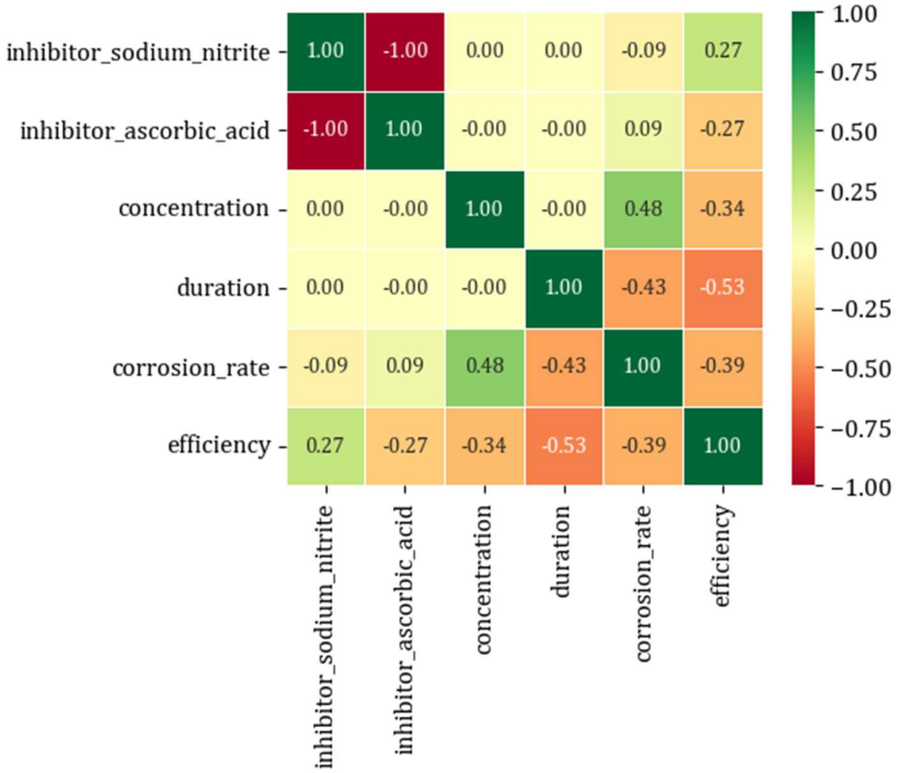


Figure 6. Pearson correlation coefficients between features and targets (corrosion rate and inhibition efficiency)

second-impactful feature. In fact, inhibitor concentration has opposing effects on corrosion rate and inhibition efficiency. Higher inhibitor concentrations increase the corrosion rate but decrease inhibition efficiency. Although these effects are opposite, the feature still contributes significantly.

Another quantitative analysis for feature analysis on corrosion rate and inhibition efficiency is the Pearson Correlation coefficient. The maximum positive and negative value of Pearson correlations indicates the strongest positive and negative correlation between features and targets. Figure 6 shows that inhibitor concentration has the highest Pearson correlation coefficient ($r = 0.48$) with corrosion rate. It indicates that inhibitor concentration shows the highest correlation with corrosion rate trends. A Positive correlation of 0.48 indicates that higher inhibitor concentration increases corrosion rate and vice versa. The negative correlation of duration determines a longer duration of immersion, affecting low corrosion rate. In another target, duration of immersion for corrosion measurement shows the highest correlation value with inhibition efficiency. It describes the highest correlation among other features and shows negative correlations. Inhibitor concentration has the second highest Pearson correlation as similar as the second most impactful feature in SHAP. Therefore, feature analysis based on SHAP is consistent with Pearson correlation. Compared to corrosion rate, various inhibitors have a higher correlation than inhibition efficiency.

CONCLUSIONS

The addition of inhibitors causes a significant effect on corrosion rate and inhibition efficiency. Corrosion resistance of ASTM A36 steel in a 3.5% NaCl medium without inhibitors is quite lower compared to inhibitor additions. The corrosion rate of samples immersed in NaCl without inhibitors for 7 days immersion is 9.25 mpy and decreases to 1.85 and 1.25 mpy with the addition of sodium nitrite and ascorbic Acid, respectively. The lowest corrosion rate for ASTM A36 in NaCl-added sodium nitrite inhibitor is 0.46 mpy in 250 ppm of inhibitor concentration and immersed for 14 days. In the case of ascorbic acid inhibitors, the lowest corrosion rate is 250 ppm for 7 days. SHAP and Pearson correlation reveal feature analysis and demonstrate consistent trends. Inhibitor concentration causes the most effect for corrosion rate and duration of immersion is the most impactful feature on inhibition efficiency. According to experimental and machine learning analysis, the current study suggest that the corrosion rate can be reduced by low inhibitor concentration combined with longer duration of immersion. Therefore, future studies should provide broader feature ranges and validated using other electrochemical techniques to explain corrosion resistance.

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DECLARATION OF CONFLICTING INTERESTS

The authors declare that they have no potential conflicts of interest regarding the research, authorship, and/or publication of this article.

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