

Response of Current Density to the Concentration and Inhibition Efficiency of Hydroxypropyl Cellulose on Aluminium in Hydrochloric Acid

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Abstract: This paper presents the response of corrosion current density to the concentration and inhibition efficiency of hydroxypropyl cellulose (inhibitor) on aluminum in hydrochloric acid. The response analysis was carried out within a range of process parameter; 18.35-30.11(μAcm^{-2}), 1-5 (g/l) and 74.22-84.29 (%) for current densities, inhibitor concentrations and inhibition efficiencies respectively. A derived empirical model; $i_{\text{corr}} = -0.58\eta - 1.439 + 74.35$ predicts the response of the corrosion current density as sum of two linear parts involving inhibition efficiency and inhibitor concentration. Results predicted by the Derived Response Model (DRM) show that the current density decreases with increase in both inhibitor concentration and inhibition efficiency, in line with previous work. The decrease in the corrosion density basically implies reduction in corrosion attack on the aluminium. The validity of the model was rooted on the core model expression $i_{\text{corr}} - K = -\eta - N\eta$ where both sides of the expression are correspondingly almost equal. The standard error incurred in predicting the model-based current density relative to the actual results was 0.48%. Evaluations from generated results indicate that the corrosion current density per unit inhibitor concentration as obtained from the actual and model-predicted results are 2.94 and 2.89 (μAcm^{-2})/ (g/l) respectively. Maximum deviation of model-predicted results (from actual results) was <3.2%. This translates into over 96% operational confidence levels for the derived model and 0.96 dependency coefficient of the current density on inhibition efficiency and inhibitor concentration. The correlation coefficients between values of current density and inhibition efficiency & inhibitor concentration from model-predicted results were all >98%.

Key words: Current density - Corrosion inhibition - Hydroxypropyl cellulose - Aluminium - Hydrochloric acid

INTRODUCTION

Globally, huge sums of money have been lost to structural failures due to corrosion attack on industrial facilities; an amount which would have paid for series of operational needs. In practice corrosion is unavoidable and can never be completely overcome but can be hindered to a reasonable level.

Aluminum and its alloys has been subjected to numerous studies due to their high technological value and wide range of industrial and domestic applications especially in aerospace, house-hold items, industries, transportation, packaging, construction, electronics, electrical transmission, machinery and chemical batteries.

The non-ferrous metal and its alloys are reactive, remarkable for its density and prone to corrosion. Reports [1-3] have shown that the metal relies on the formation of a compact, strongly adherent and continuous passive oxide film developed on it upon exposure to the atmosphere or aqueous solutions for its corrosion resistance in most environments. This surface film is amphoteric and dissolves substantially when the metal is exposed to high concentrations of acids or bases [4]. In addition, aluminum may be used in neutral solutions containing pitting agents such as chloride ions. These solutions cause pitting corrosion. Under these circumstances, corrosion inhibitors are expected to be used because the solubility of the oxide film increases

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above and below pH4-9 range [5, 6] resulting to uniform attack on aluminum. Hydrochloric acid (HCl) solutions are used for pickling, chemical and electrochemical etching of aluminum, producing hydrogen gas. Chloride ions of the HCl cause a substantial loss of the metal as a result of corrosion through localized attack [7]. It is very important to add corrosion inhibitors to prevent metal dissolution and minimize acid consumption [8]. Introduction of an oxidizing agent like KIO_3 into a corrosive acidic medium can lead to self-passivation of steel. KIO_3 has been identified as an effective inhibitor for corrosion of copper in acidic environment. Several studies [9-17] have appraised various organic compounds to ascertain their viability as corrosion inhibitor. The result of the investigation indicates that they are good corrosion inhibitors for aluminium in hydrochloric acid media.

Results of investigation [18] on the inhibitive effect of potassium iodate on the corrosion of aluminum in 2M HCl using weight loss, polarization and electrochemical impedance spectroscopy (EIS) measurements indicate that KIO_3 acts as an excellent inhibitor. Inhibition efficiency with 100 ppm inhibitor was very high. The results revealed that the inhibitor used is a mixed type following critical analysis of the polarization curves. The investigation also indicates that surface adsorption of KIO_3 led to a decrease in double layer capacitance as well as an increase in polarization resistance. Furthermore, the adsorption of the inhibitor on the aluminum surface was observed to be in agreement with Temkin adsorption isotherm.

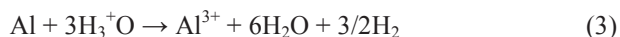
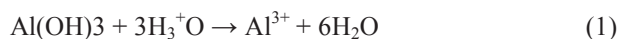
Research [19] has shown the possibility of inhibiting inhibition of aluminium corrosion in HCl solution by ortho substituted aniline-N-salicylidene using the mass loss method. Results culled from the research revealed that the value of inhibition efficiency obtained through the highlighted method is in good agreement and depends upon the inhibitor, the acid, period of exposure and temperature.

Researchers [20] have successfully evaluated the effectiveness of 3-nitrobenzoic acid as inhibitor against corrosion of aluminium in HCl solution using theoretical and experimental methods (weight loss, thermometric, polarization, FTIR and SEM techniques). Results of the research revealed that inhibition efficiency put forth by the inhibitor (3-nitrobenzoic acid) as evaluated from weight loss technique ranged from 71% to 82%. The research further revealed calculated kinetic, thermodynamic and adsorption parameters which expounded the adsorption of the inhibitor on the surface of the respective metal as being clearly accompanied by molecular association. The adsorption was also observed to be endothermic, spontaneous and favoured the mechanism of physical adsorption. It was discovered that

best-fitted adsorption isotherms were Langmuir and Frumkin models, which gave evidences for the existence of interaction, characterized by attractive behaviour of the inhibitor on aluminium surfaces. Comparative analysis of the scanning electron micrographs emanating from the research [20] showed the metal before and after inhibition. The micrograph clearly revealed that the inhibitor prevented crevice and pitting corrosion by forming adsorbed protective layer on the respective metal surface. In addition, FTIR spectra of the inhibitor and corrosion products clearly revealed the formation of new bond, existence of interaction between the inhibitor molecules and the involvement of some functional groups in the adsorption and inhibition processes.

Mechanism of Corrosion of Aluminium in Hydrochloric Acid:

In the presence of hydrochloric acid, aluminium can react to liberate hydrogen gas. However, the reactions maybe somewhat slowed down by the formation of hydroxide or oxide. The formation of aluminium oxide or hydroxide retards corrosion attack on the metal as shown in the following equations [21]:



Following the above mechanism, it is evident that before the corrosion of aluminium in hydrochloric acid solution can proceed, the oxide or hydroxide protective layer must be dissolved. Based on the foregoing, hydrochloric acid seems to catalyse corrosion in aluminium. This explains why the rate of corrosion of aluminium in HCl is higher than the rate of corrosion of mild steel in similar medium.

Hydroxypropyl cellulose has been appraised [22] and recognized as a desirable metal corrosion inhibitor considering its sterling properties and characteristics. These characteristics include solubility in water, appreciably safe in handling, and applicability in drilling operations.

Investigation [23] has been carried out on the adsorption and inhibition performance of hydroxypropyl cellulose (HPC) on aluminium corrosion in 0.5 M HCl and 2M H_2SO_4 at 30–65°C using techniques such as potentiodynamic polarization, gravimetric measurement and quantum chemical computation. Results of the analysis on potentiodynamic polarization confirmed that HPC acted as a mixed-type inhibitor in both aggressive solutions with a more dominant anodic effect. The results of the investigation revealed clearly that aluminium dissolved with ease in hydrochloric acid

compared to sulphuric acid. The results indicated that inhibition efficiency of HPC increased with increase in inhibitor concentration within a range of increased temperature. It was observed that adsorption of HPC molecules onto aluminium surface took to the Langmuir adsorption isotherm. In addition, the quantum chemical calculations, through the aid density functional theory revealed the adsorption strength attractiveness of HPC molecules towards aluminium surface.

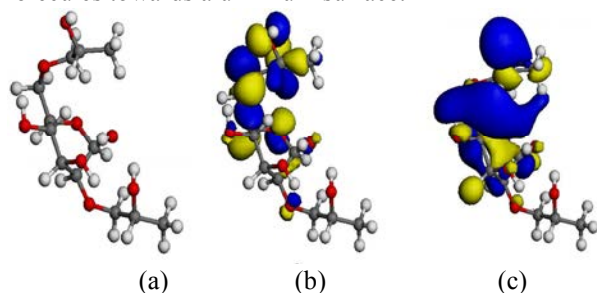


Fig. 1: Quantum chemical parameters for a single Hydroxypropyl Cellulose molecule a optimized Structure, b HOMO Orbital, c LUMO Orbital [23] [Active legend: C = grey; H = white; O = oxygen]

Fukui function has been successfully used to analyze [23] the chemical reactivity of HPC molecule. The analysis indicates the sites for a nucleophilic and electrophilic attack on the molecule. Results of the analysis show that the HOMO orbital (b) carries the sites for the electrophilic attack which represent the regions where the inhibitor molecule and metal surface exhibit highest bonding ability. The LUMO orbital (c) was found to contain the sites for nucleophilic attack. The research identified this region as area of interaction between the inhibitor molecule and metal surface to exhibit anti-bonding orbital to form feedback bond which strengthens the interaction between the inhibitor and Al surface. In HPC molecule, the HOMO and LUMO orbital respectively cut across the hydroxy group, propyl group and phenyl group within the molecule.

The present research aims at evaluating the response of corrosion current density to the concentration and inhibition efficiency of hydroxypropyl cellulose on aluminium in hydrochloric acid. Hydroxypropyl cellulose has been successful used [23] as an effective corrosion inhibitor for aluminium in acidic environments. However, no work has been published to empirically predict very important process parameters relative to some others.

MATERIALS AND METHODS

Materials Preparation: The aluminium sheet used for the study has the following chemical composition (wt%): Si (0.842%), Fe (0.898%), Cu (0.028%), Mn (0.081%),

Mg (0.026%), Zn (0.010%), Ti (0.0155), Cr (0.0065), Ni (0.003%), V (0.009%) and balance Al.

The sheet was press cut mechanically into $4 \times 3 \times 0.2$ cm coupons. The test coupons were polished with fine emery papers 1000 grades to ascertain smooth surface, degreased with acetone, rinsed in distilled water and dried with warm air. The prepared coupons were stored in a desiccator before use for the corrosion test. The acid solutions were prepared with analytical grade (BDH) of HCl and double distilled water. The blank acid solution used for the research was 0.5M HCl. For the inhibited solution, powdered HPC (product of Sigma Aldrich chemical company) was added to blank acid solutions to obtain concentrations of 1 g/L, 2 g/L, 3 g/L, 4 g/L and 5 g/L respectively. The HPC was used as obtained without any purification [23].

Potentiodynamic Polarization Measurements:

Advanced electrochemical corrosion equipment (PARC-263 model) was used to conduct the experimental test. The system consists of cylindrical computer glass electrolytic cell display system and power suite software. The electrolytic cell contains three conventional electrodes (counter electrode (graphite rod), reference electrode (saturated calomel electrode) and the working electrode [aluminium coupons coated with epoxy resin and exposing a surface area of 1 cm^2 to the test solution]) and test solution. The electrodes were connected to the electrolytic cell via the lugging capillary of the electrochemical workstation before the equipment was switched on. Open potential circuit steady-state values were allowed for 30 min of immersion before each potentiodynamic polarization measurement was made in unstirred solution and aerated condition maintained at $30 \pm 1^\circ\text{C}$ room temperature and in the potential range ± 250 mV versus corrosion potential at a scan rate of 0.333 mV/s. Power suite software was used to extrapolate the polarization data. Each test was repeated in triplicates to verify the reproducibility of the system [23].

Gravimetric Loss Measurement: The weight loss measurement was done by complete immersion of aluminium coupons in 200 mL of blank and inhibited solution respectively contained in 300 mL glass beaker kept at $30 \pm ^\circ\text{C}$ with the aid of a nylon thread, glass rod, and hooks and thermostatic water bath. The aluminium coupons were retrieved after 24 h interval progressively for 144 h. Also, the temperature variation effect was monitored by equilibrating the beakers at $35\text{--}65^\circ\text{C}$ in a thermostatic water bath and the coupons were retrieved at an interval of 8 h. The experiments were conducted in triplicates to confirm reproducibility of results and the average value of the weight [23].

RESULTS AND DISCUSSION

Table 1: Variation of current density i_{corr} with inhibition efficiency η and concentration of inhibitor θ [23]

(η)	(i_{corr})	(θ)
74.22	30.11	1
78.39	25.24	2
80.43	22.85	3
82.69	20.22	4
84.29	18.35	5

Computational analysis of the actual results shown in Table 1, gave rise to Table 2 which indicate that;

$$i_{corr} - K = -h\eta - N\theta \quad (4)$$

Introducing the values of K, h and N into equation (4) reduces it to;

$$i_{corr} - 74.35 = -0.58\eta - 1.43\theta \quad (5)$$

$$i_{corr} = -0.58\eta - 1.43\theta + 74.35 \quad (6)$$

where,

K = 74.35, h = 0.58 and N = 1.43; equalizing constant (determined using C-NIKBRAN [24])

(η) = Corrosion inhibition efficiency (%)

(θ) = Concentration of inhibitor (g/l)

(i_{corr}) = Current density ($\mu\text{A cm}^{-2}$)

Boundary and Initial Conditions: Consider short cylindrically shaped aluminium coupon submerged in hydrochloric acid, interacting with some corrosion-induced agents. The solution is assumed to be affected by undesirable dissolved gases. The considered range of the current densities, inhibitor concentrations and inhibition efficiencies are 18.35-30.11 ($\mu\text{A cm}^{-2}$), 1 - 5 (g/l) and 74.22- 84.29 (%) respectively.

Table 2: Variation of $i_{corr} - K$ with $-h\eta - N\theta$

$i_{corr} - K$	$-h\eta - N\theta$
- 44.24	- 44.4776
- 49.11	- 48.3262
- 51.50	- 50.9394
- 54.13	- 53.6802
- 56.00	- 56.0382

Model Validity: The validity of the model is strongly rooted on the core model equation (4) where both sides of the equation are correspondingly almost equal. Table 2 also agrees with equation (4) following the values of $i_{corr} - K$ and $-h\eta - N\theta$ evaluated from the actual results in Table 1. Furthermore, the derived model was validated by comparing the current density predicted by the model and that obtained from the experiment. This was done using various analytical techniques which includes

computational, statistical, graphical and deviational analyses.

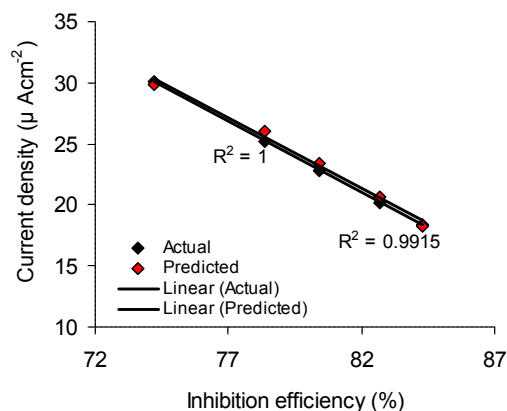


Fig. 2: Coefficient of determination between current density and inhibition efficiency as obtained from actual and model-predicted results

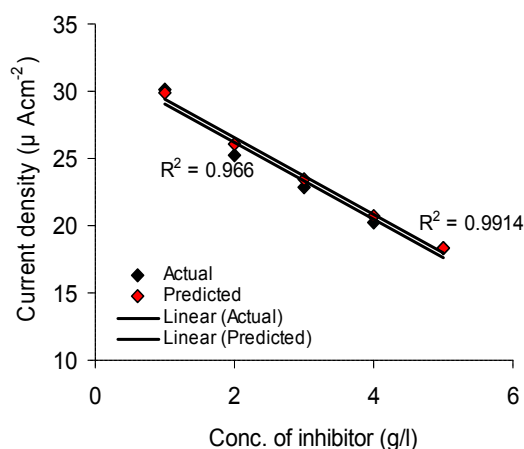


Fig. 3: Coefficient of determination between current density and concentration of inhibitor as obtained from actual and model-predicted results

Computational Analysis: Corrosion current density per unit inhibitor concentration.

Corrosion current density per unit inhibitor concentration $i_{corr-\theta}$ ($\mu\text{A cm}^{-2}$) / (g/l), was calculated from the equation;

$$i_{corr-\theta} = i_{corr} / \theta \quad (7)$$

Re-written as

$$i_{corr-\theta} = \Delta i_{corr} / \Delta \theta \quad (8)$$

Equation (8) is detailed as

$$i_{\text{corr}-9} = \frac{i_{\text{corr}2} - i_{\text{corr}1}}{\eta_2 - \eta_1} \quad (9)$$

where,

$i_{\text{corr}-9}$ = Change in the current densities $i_{\text{corr}2}$, $i_{\text{corr}1}$ at inhibitor concentrations η_2 , η_1 .

Considering the points (1, 30.11) & (5, 18.35) and (1, 29.8724) & (5, 18.3118) as shown in Fig. 3, designating them as $(i_{\text{corr}1}, \eta_1)$ & $(i_{\text{corr}2}, \eta_2)$ for actual and model-predicted results, and then substituting them into equation (9), gives the slopes: - 2.94 and - 2.89 (μAcm^{-2})/(g/l) respectively as corrosion current density per unit inhibitor concentration. The negative sign preceding the values is an indication that the corrosion current density-inhibitor concentration slopes of (as shown in Fig.3) are all negative. Therefore the real values of the corrosion current density per unit inhibitor are 2.94 and 2.89 (μAcm^{-2})/(g/l) for the actual and model-predicted results respectively.

Results predicted by the Derived Response Model (DRM) show that the current density decreases with increase in both inhibitor concentration and efficiency, line with previous work [24]. The decrease in the corrosion density basically implies reduction in corrosion attack on the aluminium.

Statistical Analysis

Correlation: The correlation coefficient between current density and inhibition efficiency & inhibitor concentration were evaluated (using Microsoft Excel Version 2003) from the coefficient of determinants on the actual and model-predicted results of Figs. 2 and 3, using equation (10). These results are 1.0000 and 0.9957 & 0.9829 and 0.9957 respectively.

$$R = \sqrt{R^2} \quad (10)$$

Standard Error (STEYX): The standard error incurred in predicting the model-based current density relative to values of the actual results is 0.48%. The standard error was evaluated using Microsoft Excel version 2003.

Graphical Analysis: The validity of the derived model was further verified by plotting values of the actual, besides the model-predicted results using Microsoft Excel (version 2003) to evaluate the trend of both results. Comparative analysis of Figs. 4 and 5 indicate very close alignment of curves which depicted significantly similar trend of data point's distribution for the actual and derived model-predicted current density. This shows proximate agreement between both results.

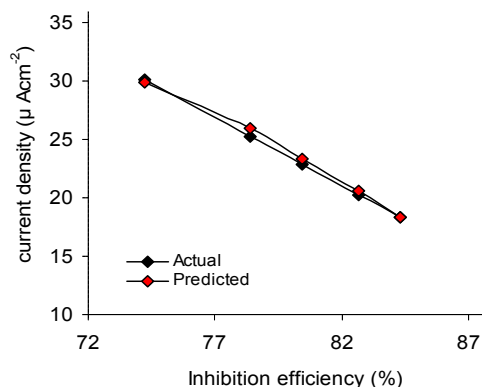


Fig. 4: Variation of current densities with inhibition efficiency as obtained from actual and model-predicted results

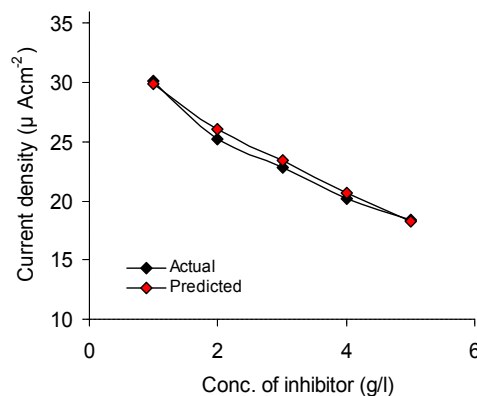


Fig. 5: Variation of current densities with inhibitor concentration as obtained from actual and model-predicted results

Deviational Analysis: Analysis of the current density obtained from the actual and model-predicted results show very low deviation between actual and the model-predicted results. This was attributed to the fact that the effects of the surface properties of the aluminium which played vital roles during corrosion in hydrochloric acid were not considered during the model formulation. This necessitated the introduction of correction factor, to bring the model-predicted current density to those of the corresponding experimental values.

The deviation D_v , of model-predicted current density from the corresponding actual result was given by;

$$D_v = \left(\frac{i_{\text{corr}-P} - i_{\text{corr}-E}}{i_{\text{corr}-E}} \right) \times 100 \quad (11)$$

where,

$i_{\text{corr}-E}$ and $i_{\text{corr}-P}$ are inhibition efficiencies evaluated from actual and model-predicted respectively.

Figure 6 shows that maximum deviation of model-predicted current density from the actual results was less than 3.2 %. This translates into over 96% model operational confidence. The figure shows that the least and highest deviations of model-predicted results (from actual results) are 0.21 and -3.11 %.

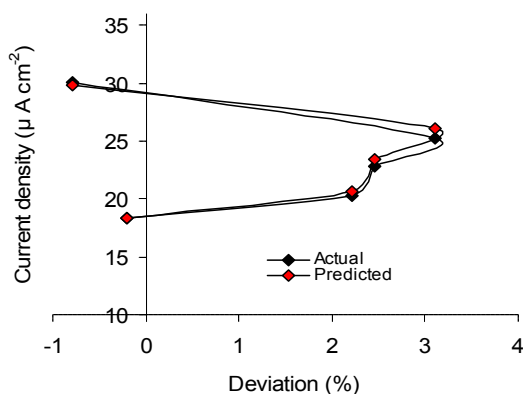


Fig. 6: Deviation of model-predicted results from actual values

These deviations correspond to model-predicted current densities: 18.3118 and 26.0238 (μAcm^{-2}) inhibition efficiencies: 84.29 and 78.39 (%); inhibitor concentrations: 5 and 2(g/l) respectively.

Correction factor, Cf to the model-predicted results was given by;

$$Cf = - \left(\frac{i_{\text{corr-P}} - i_{\text{corr-E}}}{i_{\text{corr-E}}} \right) \times 100 \quad (12)$$

Critical analysis of Fig. 6 and Fig. 7 show that the evaluated correction factors are negative of the deviation as shown in equations (11) and (12).

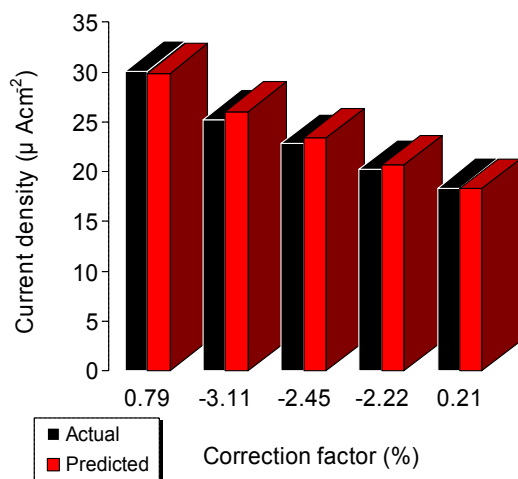


Fig. 7: Correction factor to model-predicted results

The correction factor took care of the negligence of operational contributions of the effects of surface properties of the aluminium which actually affected the corrosion process. Introduction of the corresponding values of Cf from equation (12) into the model gives exactly the corresponding actual current density. Fig. 7 indicates that the maximum correction factor to the model-predicted current density was less than 3.2 %. The figure shows that the least and highest correction factors to the model-predicted results (from actual results) are -0.21 and 3.11%. These correction factors also correspond to model-predicted current densities: 18.3118 and 26.0238 (μAcm^{-2}) inhibition efficiencies: 84.29 and 78.39 (%); inhibitor concentrations: 5 and 2 (g/l) respectively.

The negative and positive signs preceding numerals in reported deviation and correction factors merely indicate deficit and surplus respectively. The actual deviation or correction factor is just the numeral.

CONCLUSION

Response of corrosion current density to the concentration and inhibition efficiency of hydroxypropyl cellulose (inhibitor) on aluminum in hydrochloric acid was evaluated. A derived empirical model; $i_{\text{corr}} = -0.58\eta - 1.439 + 74.35$ predicts the response of the corrosion current density as sum of two linear parts involving inhibition efficiency and inhibitor concentration. Results predicted by the Derived Response Model (DRM) show decrease in current density as both inhibitor concentration and inhibition efficiency increases, in line with previous work. The decrease in the corrosion density basically implies reduction in corrosion attack on the aluminium. The validity of the model was rooted on the core model expression $i_{\text{corr}} - K = -\eta\eta - N\eta$ where both sides of the expression are correspondingly almost equal. The standard error incurred in predicting the model-based current density relative to the actual results was 0.48%. Evaluations from generated results indicate that the corrosion current density per unit inhibitor concentration as obtained from the actual and model-predicted results are 2.94 and 2.89 (μAcm^{-2})/(g/l) respectively. Maximum deviation of model-predicted results (from actual results) was < 3.2%. This translates into over 96% operational confidence levels for the derived model and 0.96 dependency coefficient of the current density on inhibition efficiency and inhibitor concentration. The correlation coefficients between values of current density and inhibition efficiency & inhibitor concentration from model-predicted results were all > 98%.

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