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OPTIMIZATION OF CORROSION INHIBITION OF ESSENTIAL OILS OF ALPINIA GALANGA ON MILD STEEL USING RESPONSE SURFACE METHODOLOGY

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ABSTRACT

The use of plant extracts as corrosion inhibitors has gained prominence as replacement for synthetic organic compounds. The plant natural products have been found to be effective, cheap and eco-friendly anticorrosion agents. Corrosion inhibitions of essential oils of Alpinia galanga were investigated on mild steel in hydrochloric acid solution using weight loss method. The interactive effects of inhibitor concentration, temperature and time were optimized for maximum response of inhibition efficiency using Response Surface Methodology with Central Composite Design. The optimum inhibition efficiency of 88.5% at 775 ppm of inhibitor concentration, temperature of 320.4 K and reaction time of 3.75 hours was accomplished. The effectiveness of the inhibitor was also supported using scanning electron microscopy. The mechanism of interaction of both the inhibitor on mild steel surface was found to conform to the Langmuir adsorption isotherm.

Keywords: corrosion inhibition, Alpinia galanga, essential oils, RSM, adsorption isotherm.

INTRODUCTION

Corrosion is a risk to both the environments and production processes and as such the deleterious consequences of the corrosion process have become a problem of worldwide significance. Corrosion is detrimental, persistent and insidious in its action. Its effect is threatening to the big as well as small industries. Its total prevention and elimination is practically impossible, hence the only effective antidote lies in controlling it.

The studies of plant products as cost effective and eco-friendly corrosion inhibitors have attracted great interests from investigators. There is a promising prospect for green corrosion inhibitors owing to their renewability and absence of heavy and toxic substances in their compounds in their molecules. Constituents of plants such as alkaloids, tannins, phenolics, amino acids are organic in nature some of which has been exploited as corrosion inhibitors. The inhibition effectiveness of plant extracts is credited to the presence of complex organic compounds containing oxygen, nitrogen and sulphur hetero atoms. The presence of aromatic rings, conjugated double bonds as well as triple bonds have also been identified to provide adsorption centres with which to interact with active sites on metal surface.

Several plants have been studied and found to be effective as corrosion inhibitors. Plants such as *Medicago sative* [4], *Tinospora cripsa* [19], *Sida acuta* [12], Ginko leaves [9], Thyme leaves [20], Apricot juice [30], *Jasminum nudiflorum* Lindl [10] and *Coffee senna* [2] have all been used as green corrosion inhibitors with varying degree of inhibition suitable at the applied concentration. They were all found to be composed of constituents whose interactions lead to improved inhibition on the surface of the applied metal specimens.

Essential oils of plants have also been employed to inhibit corrosion of metals. They function as vapor-phase corrosion inhibitors as a result of their volatility [25]. Several plant oils have been established as effective for inhibiting corrosion of mild steel in acidic solution. The work involving Pennyroyal oil [7]; Jojoba oil [8]; Eucalyptus oil [16, 28]; Artemisia oils [5]; Rosemary oil [6]; Lavender oil [32]; *Salvia aucheri mesatlantica* [33] have all shown varying levels of effectiveness of the plants oils.

Alpinia galanga belonging to the Zingiberaceae family has been chosen based on relating phylogenic and phytochemical considerations whose approach is premised on the existence of similar biochemical properties in closely related plant species. It is pertinent to note that turmeric and ginger which also belong to the Zingiberaceae family as *A. galanga* have been shown to possess corrosion inhibition properties [3, 14].

The plant has been recognized as an antioxidant and a therapeutic agent for several diseases [21, 31]. Its major constituents are phenolics which have resemblance with structures of common organic corrosion inhibitors. In their works, [23, 27], the essential oils of *A. galanga* been shown to be mainly composed of 1, 8 -cineole, α-terpineol, germacrene-D, α-pinene, β-pinene and several other monoterpenoids. However, the essential oils of the plant are yet to be considered as corrosion inhibitors. In addition to this, only few works have been carried out on enhancing the process variables for optimization of corrosion inhibition processes. In this work, the essential oils of *A. galanga* are being optimized as an eco-friendly corrosion inhibitor of mild steel in hydrochloric acid solution.

The Design of Experiment studies the effects of interactions of the process factors leading to establishment of a model correlating the process parameters

(independent variables) with the inhibition efficiency (response variable). The usual one factor at a time (OFAT) method of analysis fails to account for the interaction effects between all factors and therefore a greater number of experimental runs are needed.

The RSM approach adopted in this work involves the following steps:

a) The use of CCD to obtain the experimental schedule points where investigations are to be executed.

b) Carrying out the weight loss experiments at the designed points.

c) Establishing a mathematical model showing the existence of relationship between the various process factors inhibitor concentration, temperature and time) and the response variable (Inhibition Efficiency).

d) Statistical evaluation of the model using ANOVA.

e) Predicting the optimum values of the process variables for maximum efficiency.

EXPERIMENTAL METHODS

Hydrodistillation of essential oils of *Alpinia galanga*

Freshly cut rhizomes of *A. galanga* were washed, sliced into small bits and placed in a 5-litre round bottomed flask in the Clevenger-type hydrodistillation set up. The hydrodistillation process was carried out for about 12 hours with intermittent collection of the extracted volatile constituents of the rhizome. The essential oil collected was extracted by adopting liquid-liquid extraction process using diethyl ether. The organic layer containing the essential oils was further dehydrated using anhydrous magnesium sulphate. The hydrodistillation process gave a yellowish liquid (0.05 g, 0.005%) characterized by spicy odour and was stored in the refrigerator until used for corrosion experiments.

Metal specimen preparation

Experiments were performed using mild steel with the percentage composition determined by glow discharge spectroscopy. Pre-cut mild steel specimens of 10cm^2 surface area (2 cm x 2 cm x 0.25 cm) with composition C 0.036; Mn 0.172; Cu 0.082; Ni 0.108; Cr 0.053; Zr 0.146 and balance Fe were used. In order to acquire a reproducible clear surface, the specimens were abraded with a series of silicon carbide abrasive paper (grades 180, 400, 800 and 1500). They were then degreased with acetone, rinsed in distilled water, dried and stored in the desiccator in readiness for weight loss experiments and electrochemical measurements according to test method of ASTM G1-03 [15, 29].

Corrosion medium

The corrosion medium of 1 M HCl was used throughout the experiments. Different concentrations of the oils were prepared ranging from of 100 to 1000 ppm in the 1 M HCl solution. The inhibitor solutions were sonicated to obtain homogeneity before the inhibition process. The 1 M HCl prepared also functioned as the blank solution in all the experiments.

Weight loss measurement

The weight loss method is a very useful, effective and reliable method of investigating corrosion of metals. Its use forms the model technique for the laboratory investigation of corrosion inhibition [26]. It is the most extensively and frequently used of all the experimental methods because of its simplicity [11, 18, 24]. The basic quantity determined from corrosion specimens is weight loss which is taken over a certain immersion period and is expressed as ΔW (in mg).

The pre-weighed mild steel specimens were immersed in 100 ml test solutions with and without an inhibitor varying the concentration at different temperatures and time intervals. All the weight loss experiments were performed in unstirred aerated hydrochloric acid solutions containing various concentrations of inhibitors as shown in the experimental set up in Table-1.

At the end of the immersion time, the mild steel specimens were cautiously removed from the test solution with the aid of a tong carefully washed with distilled water, acetone and dried using hot air and allowed to cool down. The masses of the specimens were thereafter recorded and the difference in mass was used to estimate the corrosion rates, surface coverage and the inhibition efficiency using Eqs. 1 to 3 respectively.

$$Rc = \frac{\Delta W}{A.T} \quad (1)$$

$$IE_{WL}\% = \left(1 - \frac{\Delta W_{inh}}{\Delta W_{blank}}\right) \times 100 \quad (2)$$

$$\theta = \frac{\Delta W_{blank} - \Delta W_{inh}}{\Delta W_{blank}} \quad (3)$$

Where ΔW, A, T, R_c, $IE_{WL}\%$ and θ are mass loss (mg), surface area of specimen (cm^2), time (h), corrosion rate (mgcm^{-2}h^{-1}), percentage inhibition efficiency and surface coverage respectively.

Experimental design and optimization procedure using RSM

Preliminary results of weight loss experiments have shown that the inhibitor concentrations, temperatures and immersion times have significant effects on both the weight loss and the efficiencies of the inhibitors. A three-

factor-five-level central composite design (CCD) involving the effects of inhibitor concentration, temperature and exposure time was adopted to examine their combined effects on the inhibition efficiencies and corrosion rates as responses. The experimental design range showing the independent variable levels for the corrosion inhibition is shown in Table-1. The experiments were randomly performed to circumvent systematical errors. A total of twenty runs were generated in the design matrix. The result were analysed using the DX7 Design Expert 7 1 6 software.

Table-1. Experimental design range and independent variable levels for corrosion inhibition of CHE of *A. galangal.*

Independent variables	Unit	Range				
		Lowest-α	**Low-1**	**Centre 0**	**High+1**	**Highest +α**
A: Concentration	ppm	100	325	550	775	1000
B: Temperature	K	300	308.25	316.5	324.75	333
C: Time	h	1	3.75	6.5	9.25	12

Scanning electron microscopy

The Philip XL40 model of scanning electron microscope was used to evaluate the surface morphological modifications of mild steel immersed in inhibited and uninhibited acid solutions. The mild surface was well polished before immersion into the test solutions for 6 hours at the room temperature after which they were retrieved, rinsed with distilled water, dried and examined by scanning. The imageries were taken from that segment of specimen from where enhanced information was obtained.

RESULTS AND DISCUSSIONS

Optimization of process variables using Response Surface Method

The concentration of inhibitor, exposure time and temperature were considered as the independent variables while the percentage inhibition efficiency was taken as the response. The result showing the experimental schedule and the response is shown in Table-2.

Table-2. Experimental result of corrosion inhibition of essential oils of *A. galangal.*

Standard order	**Factor 1 A: Concentration** (ppm)	**Factor 2 B: Temperature** (K)	**Factor 3 C: Time** (h)	**Actual Inhibition efficiency** (%)	**Predicted inhibition efficiency** (%)
1	325	308.25	3.75	64.3	63.2
2	775	308.25	3.75	85.8	87.27
3	325	324.75	3.75	68.7	69.5
4	775	324.75	3.75	88.4	88.37
5	325	308.25	9.25	66.2	68.37
6	775	308.25	9.25	81.8	83.15
7	325	324.75	9.25	71.9	72.57
8	775	324.75	9.25	78.9	82.15
9	100	316.5	6.5	57.8	57.6
10	1000	316.5	6.5	93.2	91.25
11	550	300	6.5	74.9	74.03
12	550	333	6.5	80.6	79.33
13	550	316.5	1	77.2	77.7
14	550	316.5	12	79.3	76.65
15	550	316.5	6.5	81.9	79.01
16	550	316.5	6.5	77.3	79.01
17	550	316.5	6.5	80.9	79.01
18	550	316.5	6.5	78.9	79.01
19	550	316.5	6.5	77.3	79.01
20	550	316.5	6.5	79.9	79.01

Statistical model of the inhibition process and analysis

The interactions were determined to optimize the process conditions of concentration, temperature and time. A well-defined general second-order model is represented by Eq. 4

$$\hat{y} = \hat{\beta}_0 + \sum_{i=1}^{k} \hat{\beta}_i x_i + \sum_{i=1}^{k} \hat{\beta}_{ii} x_i^2 + \sum_{i=1}^{k} \sum_{j=1}^{i-1} \hat{\beta}_{ij} x_i x_j \quad (4)$$

Where *xi* and *xj* represent the design variables and β the tuning parameters.

The interactive significance of the process variables to the corrosion inhibition process was evaluated using ANOVA. The p-value serves as a measuring yardstick to determine the significance of the model while the model adequacy is tested by the coefficient of determination (R^2) in comparison with the adjusted coefficient of determination (R^2). The model in coded and actual values showing the relative impact of the factors for the inhibition process is given respectively in Eqs. 5 and 6 as follows:

$$\begin{aligned} IE\ (\%) = &+ 79.01 + 8.41*A + 1.33*B - 0.26*C \\ &- 1.30*A*B - 2.33*A*C - 0.53*B*C \\ &- 1.15*A^2 - 0.58*B^2 - 0.46*C^2 \end{aligned} \quad (5)$$

$$\begin{aligned} IE\ (\%) = &- 1042.09352 + 0.30836*Concentration \\ &+ 6.11784*Temperature + 10.08241*Time \\ &- 7.00337E{-}004*Concentration*Temperature \\ &- 3.75758E{-}003*Concentration*Time \\ &- 0.023140*Temperature*Time \\ &- 2.26263E{-}005*Concentration^2 \\ &- 8.56499E{-}003*Temperature^2 \\ &- 0.060556*Time^2 \end{aligned} \quad (6)$$

From the ANOVA, the Model F-value of 26.05 implies the model is significant. There is only a 0.01% chance that a "Model F-Value" this large could ever occur due to noise. Values of "Prob > F" less than 0.0500 designate the model terms to be significant. Therefore, A, B, AC, A2 are the only significant model terms in this work.

The "Lack of Fit F-value" of 2.01 implies that the Lack of Fit is not significant relative to the pure error. Therefore there is only a 23.10% chance that "Lack of Fit F-value" this large could occur due to noise. Non-significant lack of fit is desireable. Values greater than 0.1000 designate that the model terms are insignificant. The "Pred R-Squared" of 0.7649 agrees reasonably with the "Adj R-Squared" of 0.9223. "Adeq Precision" is a measure of signal to noise ratio and a ratio larger than 4 is desirable. The ratio of 20.552 in this work indicates an adequate signal and hence this model can be employed to for optimization of the inhibition process.

Experiments carried out at these optimum conditions relate closely with those predicted. The plot showing the correlation between the actual and predicted inhibition efficiency is shown in Figure-1 while the plot of the residual and predicted values as well as the internal normal plots of residuals are shown as Figures 3 and 4 respectively.

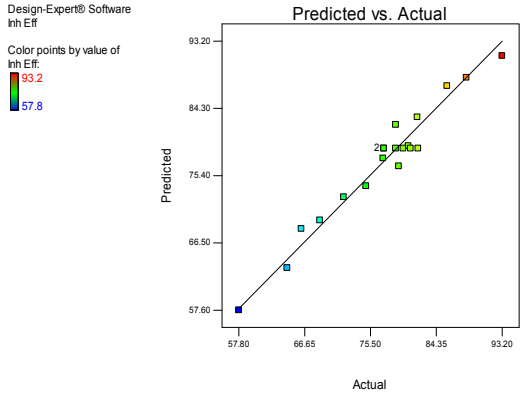

Figure-1. Plot of the predicted and actual experimental values of inhibition efficiency.

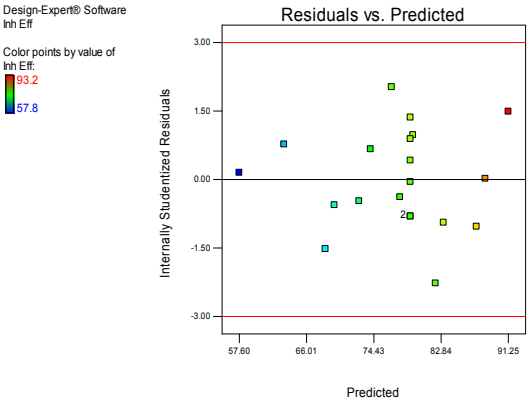

Figure-2. Plot of the residual and predicted values of inhibition efficiency.

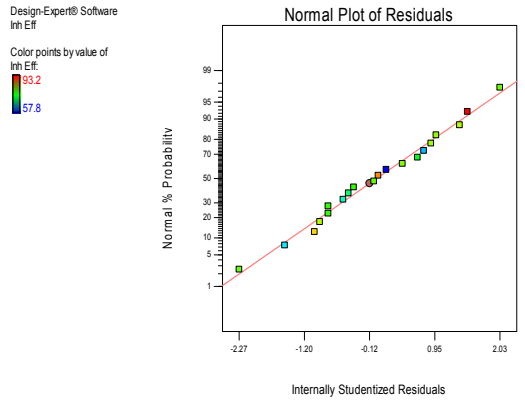

Figure-3. Internally normal plot of residuals for the inhibition efficiency.

The contour plots representation showing the interactions between the variables on mild steel are shown in Figures 4 to 6.

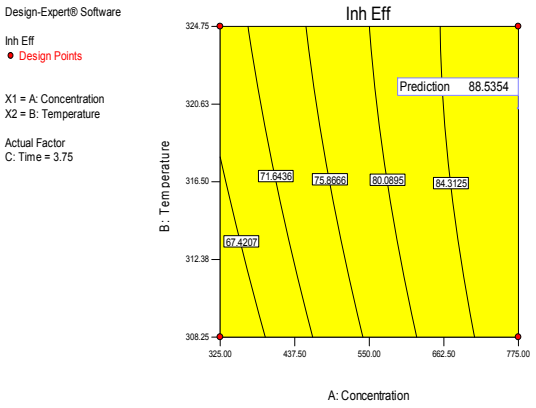

Figure-4. Effect of inhibitor concentration and temperature on inhibition efficiency.

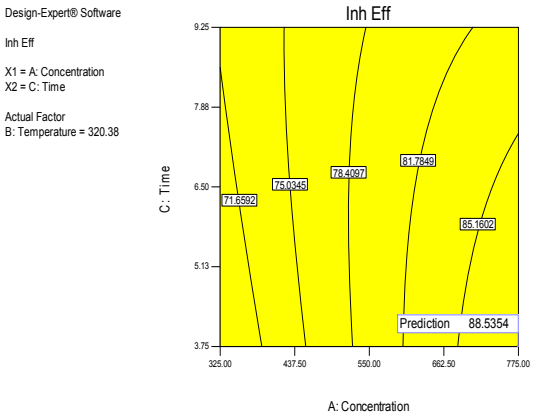

Figure-5. Effect of inhibitor concentration and time on inhibition efficiency.

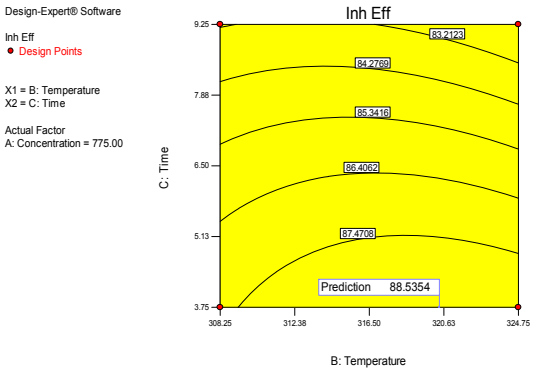

Figure-6. Effect of temperature and time on inhibition efficiency.

In Figure-4, showing the plot of interactions between concentration and temperature, it shows that the inhibition efficiency increases with increase in concentration of the inhibitor but there is a decrease in the efficiency as the temperature is increased. As a result of increased concentration, more molecules of the extracts are more readily available to block the active sites on the mild steel surface. Figure-5 depicts the effect of time on the efficiency of the inhibitor in which the efficiency is reduced over time at constant temperature. At constant concentration, increase in temperature and time is seen to reduce the efficiency. This observation is depicted in Figure-6. At higher temperatures and longer contact of mild steel with acid solution, corrosion rate increases because the rate of dissolution of the metal is higher than the rate of surface coverage.

The optimum inhibition efficiency of the essential oils of *A. galanga* 88.5% at the optimal inhibitor concentration of 775 ppm, temperature of 320.4 K and time of 3.75 h. The inhibition process is therefore shown to be concentration, time and temperature dependent.

Adsorption isotherm considerations

Surface coverage information is valuable in evaluating the inhibitor behaviour. This was adopted for fitting experimental data into adsorption isotherms which gives an insight into the mechanism of inhibition process. Establishing appropriate adsorption isotherms that suitably describes the adsorption behaviours of the inhibitors is an essential part of this study. The different kinds of interaction between the inhibitors and the metal surface are defined by the adsorption isotherms [17, 22]. These models offer the association between the coverage of an interface by the adsorbed molecules and the concentrations in solution.

The process of adsorption is dependent on the electronic characteristics, the nature of the metal surface, the process temperature and pressure, steric effects, multilayer adsorption as well as changing degree of surface-site action. The role of an inhibitor in a corrosion measurement is of both kinetic and thermodynamic importance and therefore treatment of corrosion adsorption phenomena is considered incomplete without their consideration [13].

Principally, all existing isotherms are obtainable in the form of Eq. 7 as follows;

$$f(\theta, x)\exp(-a\theta) = KC \tag{7}$$

Where a represents the molecular interaction parameter which depends on the molecular interactions of the adsorbed layer and also on the heterogeneity level of the surface. The $f(\theta, x)$ represents the configurational factor and this depends basically on the model of physical interactions and assumptions fundamental to the isotherm derivation [13]. The adsorption type was tested by theoretically appropriating the surface coverage (θ) data into adsorption different isotherms. The adsorption data fitted well into the Langmuir adsorption isotherm and is articulated in Eq. 4 as:

$$\frac{C_{inh}}{\theta} = \frac{1}{K_{ads}} + C_{inh} \quad (8)$$

Where θ = surface coverage, C_{inh} = inhibitor concentration, K_{ads} = equilibrium constant for the adsorption process and the isotherm plot is shown in Figure-7.

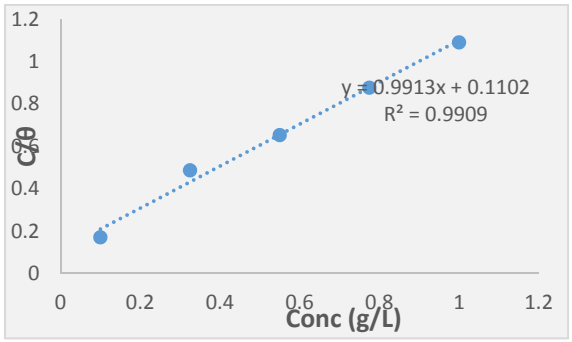

Figure-7. Adsorption isotherm model for the inhibition of mild steel using essential oils of *A. galangal.*

The standard free energy of adsorption ΔG_{ads}, is calculated using expression in Eq. 9. After substituting the concentration of water in solution into the equation the following equation is obtained:

$$K_{ads} = \frac{1}{55.5} exp\left(\frac{-\Delta G^0_{ads}}{RT}\right) \quad (9)$$

On reordering the equation, the following Eq. 10 is obtained;

$$\Delta G_{ads} = -RTln\left(55.5K_{ads}\right) \quad (10)$$

Eq. 10 is engaged to calculate the adsorption parameters at temperatures 300 – 333 K for ESS OIL at the optimal conditions. The adsorption parameter values deduced from the isotherm (Figures 6) for the inhibitor are enlisted in Tables 3.

Table-3. Adsorption isotherm parameters for the inhibition process on mild steel.

Adsorption isotherm	Slope	Intcpt (g/L)	R^2	K (L/g)	ΔG_{ads} ($KJmol^{-1}$)
Langmuir	0.9913	0.1102	0.9909	9.0744	-16.80

Application of adsorption isotherm models

An adsorption isotherm model provides the relation that exists between an interface covered by the adsorbed species of the inhibitor and its concentration in solution. The performance clarification of the inhibitor as a type of adsorbent is enhanced by fitting the data into a suitable adsorption isotherm.

Different adsorption isotherms were tested at the optimum condition of temperature and time but the Langmuir Adsorption isotherms were the more fitted for the inhibition of the molecules of the essential oils on the mild steel surface. The Langmuir isotherm given by Eq. 8 assumes a direct relationship between inhibition efficiency and the degree of surface coverage (θ) for different inhibitor concentrations. The inhibitor obeys the Langmuir adsorption isotherms by producing a straight line for the plot of C/θ versus concentrations of the inhibitor.

This straight line obtained reveals that the main process of inhibition is adsorption. It also shows that increase in inhibition efficiency with increase in inhibitor concentration is an indication of an increase in the number of components of the inhibitors adsorbed over the mild steel surface blocking the active sites thereby leading to the protection of the metal from corrosion. The closeness of the slope to unity and the non-zero intercept is due to interactions between inhibitor molecules on the metal surface as well as changes in the adsorption heat with increasing surface coverage. This also denotes that the inhibitor molecules occupy more than one active site on the metal surface. The K_{ads} value gives a pointer to the force of adherence that exists between an adsorbate and an adsorbent. Large values of K_{ads} surmise enhanced adsorption and therefore improved inhibition efficiency (Li *et al*., 2009).

From the value of ΔG_{ads} calculated, negative value connotes the spontaneity of the adsorption process and stability of the adsorbed layer on the mild steel surface. The ΔG_{ads} value is lower than - 20 $kJmol^{-1}$ which is consistent with physiosorption.

SEM examinations

The SEM images were taken to study the surface morphology of mild steel under the influence of hydrochloric acid and the corrosion inhibitor. This helps to establish the interaction between the inhibitor molecules and the metal surface. These analyses are carried on both the corroded and protected metal surfaces. The SEM micrographs of mild steel surfaces for both the uninhibited and inhibited systems for 6 hours of exposure at room temperature are shown in Figure-8.

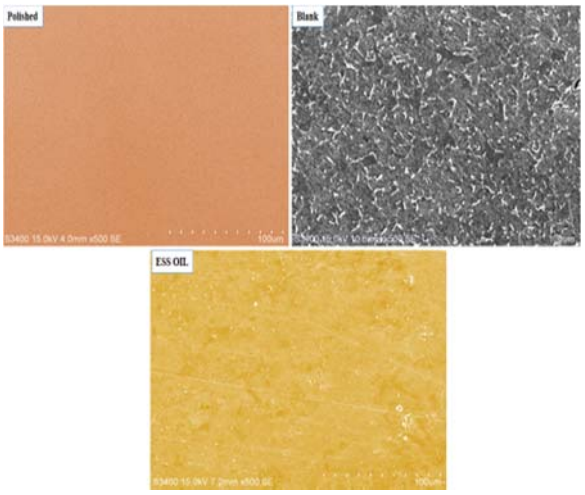

Figure-8. SEM micrographs of polished mild steel surface and uninhibited and inhibited surfaces on immersion for 6 hours at room temperature in essential oils of *A. galangal.*

Figure-8 shows the uniform smoothness of polished mild steel surface before immersion. Aggressive attack is seen on blank mild steel sample after 6 hours immersion in uninhibited 1 M HCl solution. In the sample inhibited, the mild steel surface is in a better condition having smooth surface compared with the metal immersed in uninhibited HCl solution. In the incidence of the inhibitor tested, the images show that a good protective coverage film produced on the metal surface, which subdues the corrosion rate, is accountable for the inhibition. There is a clear evidence that the inhibition is as result of formation of insoluble stable protective coverage by adsorption process [1]. The SEM results show that the ESS OIL possess molecules that inhibit mild steel from corrosion.

CONCLUSIONS

The empirical relationship between the inhibition efficiency and the process variables of inhibitor concentration, temperature and time was accomplished and expressed by the Quadratic model of the CCD. The interactions were determined to optimize the process conditions of concentration, temperature and time and their significance to the corrosion inhibition process was evaluated. The optimum inhibition efficiency of 88.5% at the optimal inhibitor concentration of 775 ppm, temperature of 320.4 K and time of 3.75 h was accomplished. The inhibition process is therefore shown to be concentration, time and temperature dependent. Adsorption of essential oils of *Alpinia galanga* on mild steel follows the linearity of the Langmuir adsorption isotherm model. The low negative values of ΔG_{ads} is an indication of physical interaction between the inhibitor and mild steel. This also signifies the spontaneity of the adsorption process and the stability on mild steel surface. The surface examination tests using SEM on the mild steel also confirms the effectiveness of the inhibitors.

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REFERENCES

[1] Achary G., Naik Y. A., Kumar S. V., Venkatesha T., and Sherigara B. 2008. An electroactive co-polymer as corrosion inhibitor for steel in sulphuric acid medium, Applied surface science. 254: 5569-5573.

[2] Akalezi C. O., Enenebaku C. K. and Oguzie E. E. 2012. Application of aqueous extracts of coffee senna for control of mild steel corrosion in acidic environments. International Journal of Industrial Chemistry. p. 3.

[3] Al-Fakih A., Aziz M. and Sirat H. 2015. Turmeric and ginger as green inhibitors of mild steel corrosion in acidic medium. Journal of Materials and Environmental Science. 6: 1480-1487.

[4] Al-Turkustani A. M., Arab S. T. and Al-Qarni L. S. S. 2011. Medicago Sative plant as safe inhibitor on the corrosion of steel in 2.0M H_2SO_4 solution. Journal of Saudi Chemical Society. 15: 73-82.

[5] Bammou L., Mihit M., Salghi R., Bouyanzer A., Al-Deyab S., Bazzi L., *et al*. 2011 Inhibition effect of natural Artemisia oils towards tinplate corrosion in HCL solution: chemical characterization and electrochemical study. Int. J Electrochem Sci. 6: 1454-1467.

[6] Bendahou M., Benabdellah M., and Hammouti, B. 2006. A study of rosemary oil as a green corrosion inhibitor for steel in 2 MH_3PO_4, Pigment & Resin Technology. 35: 95-100.

[7] Bouyanzer A., Hammouti B. and Majidi L. 2006. Pennyroyal oil from Mentha pulegium as corrosion inhibitor for steel in 1M HCl, materials letters. 60: 2840-2843.

[8] Chetouani A., Hammouti B. and Benkaddour M. 2004. Corrosion inhibition of iron in hydrochloric acid solution by jojoba oil. Pigment & Resin Technology. 33: 26-31.

[9] Deng S. and Li X. 2012. Inhibition by Ginkgo leaves extract of the corrosion of steel in HCl and H_2SO_4 solutions, Corrosion Science. 55: 407-415.

[10] Deng S. and Li X. 2012. Inhibition by Jasminum nudiflorum Lindl. Leaves extract of the corrosion of aluminium in HCl solution, Corrosion Science. 64: 253-262.

[11] Eddy N. O. and Odiongenyi A. O. 2010. Corrosion inhibition and adsorption properties of ethanol extract of Heinsia crinata on mild steel in H_2SO_4, Pigment & Resin Technology. 39: 288-295.

[12] Eduok U. M., Umoren S. A. and Udoh A. P. 2012. Synergistic inhibition effects between leaves and stem extracts of Sida acuta and iodide ion for mild steel corrosion in 1M H_2SO_4 solutions. Arabian Journal of Chemistry. 5: 325-337.

[13] El-Awady A., Abd-El-Nabey B. and Aziz S. 1992. Kinetic-thermodynamic and adsorption isotherms analyses for the inhibition of the acid corrosion of steel by cyclic and open-chain amines. Journal of the Electrochemical Society. 139: 2149-2154.

[14] Fouda A. E.-A. S., Nazeer, A. A., Ibrahim M. and Fakih, M. 2013. Ginger Extract as Green Corrosion Inhibitor for Steel in Sulfide Polluted Salt Water. Journal of the Korean Chemical Society. 57: 272-278.

[15] G1-03 A. 2003. Standard practice for preparing, cleaning, and evaluating corrosion test specimens.

[16] Gualdrón A. F., Becerra E. N., Peña D. Y., Gutiérrez J. C. and Becerra H. Q. 2013. Inhibitory effect of Eucalyptus and Lippia Alba essential oils on the corrosion of mild steel in hydrochloric acid, J Mater Environ Sci. 4: 143-158.

[17] Hosseini M., Mertens S. F. and Arshadi M. R. 2003. Synergism and antagonism in mild steel corrosion inhibition by sodium dodecylbenzenesulphonate and hexamethylenetetramine, Corrosion Science. 45: 1473-1489.

[18] Hussein Alwan H. 2014. Adsorption Mechanism for Corrosion Inhibition of Carbon Steel on HCl Solution by Ampicillin Sodium Salt, Global Journal of Research In Engineering. p. 13.

[19] Hussin M. H., Jain Kassim M., Razali N. N., Dahon N. H. and Nasshorudin D. 2011. The effect of Tinospora crispa extracts as a natural mild steel corrosion inhibitor in 1M HCl solution, Arabian Journal of Chemistry.

[20] Ibrahim T., Alayan H. and Mowaqet Y. A. 2012. The effect of Thyme leaves extract on corrosion of mild steel in HCl, Progress in Organic Coatings. 75: 456-462.

[21] Jaju, S., Indurwade, N., Sakarkar, D., Fuloria, N., and Ali, M. 2009 Isolation of galangogalloside from rhizomes ofAlpinia galanga, International Journal of Green Pharmacy. 3: 144.

[22] Leelavathi S. and Rajalakshmi R. 2013. Dodonaea viscosa (L.) Leaves extract as acid Corrosion inhibitor for mild Steel–A Green approach, J. Mater. Environ. Sci. 4: 625-638.

[23] Nampoothiri S. V., Menon A. N., Esakkidurai T. and Pitchumani K. 2016. Essential Oil Composition ofAlpinia calcarata and Alpinia galangaRhizomes-A Comparative Study, Journal of Essential Oil Bearing Plants. 19: 82-87.

[24] Obot I., Obi-Egbedi N., Umoren S. and Ebenso E. 2010. Synergistic and antagonistic effects of anions and Ipomoea invulcrata as green corrosion inhibitor for aluminium dissolution in acidic medium, Int. J. Electrochem. Sci. 5: 994-1007.

[25] Putilova I. N. 1960. Metallic corrosion inhibitors: Pergamon Press.

[26] Rahim A. A., Rocca E., Steinmetz J., Kassim M. J., Adnan R. and Sani Ibrahim M. 2007. Mangrove tannins and their flavanoid monomers as alternative steel corrosion inhibitors in acidic medium. Corrosion Science. 49: 402-417.

[27] Raina A. P., Verma S. and Abraham Z. 2014 Volatile constituents of essential oils isolated from *Alpinia galanga* Willd. (L.) and A. officinarum Hance rhizomes from North East India. Journal of Essential Oil Research. 26: 24-28.

[28] Rekkab S., Zarrok H., Salghi R., Zarrouk A., Bazzi L., Hammouti B., *et al*. 2012. Green corrosion inhibitor from essential oil of Eucalyptus globulus (Myrtaceae) for C38 steel in sulfuric acid solution. Journal of Materials and Environmental Science. 3: 613-627.

[29] Revie R. W. and Uhlig H. H.011. Uhlig's corrosion handbook vol. 51: John Wiley and Sons.

[30] Yaro A. S., Khadom A. A., and Wael R. K. 2013 Apricot juice as green corrosion inhibitor of mild steel in phosphoric acid, Alexandria Engineering Journal. 52: 129-135.

[31] Yasuhara T., Manse Y., Morimoto T., Qilong W., Matsuda H., Yoshikawa M., *et al*. Jun 1 2009 Acetoxybenzhydrols as highly active and stable analogues of 1'S-1'-acetoxychavicol, a potent antiallergic principal from Alpinia galanga, Bioorg Med Chem Lett. 19: 2944-2946.

[32] Zerga B., Sfaira M., Rais Z., Touhami M. E., Taleb M., Hammouti B., *et al*. 2009 Lavender oil as an ecofriendly inhibitor for mild steel in 1 M HCl, Matériaux & techniques. 97: 297-305.

[33] Znini M., Majidi L., Bouyanzer A., Paolini J., Desjobert J.-M., Costa J., *et al*. 2012 Essential oil of Salvia aucheri mesatlantica as a green inhibitor for the corrosion of steel in 0.5 MH_2 SO_4. Arabian Journal of Chemistry. 5: 467-474.