

## Effect of *Terminalia catappa* Leaves Extract on Corrosion of Mild Steel using Response Surface Methodology

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### ABSTRACT

Effect of *Terminalia catappa* leaves (TCL) extract in inhibiting corrosion of mild steel was investigated. In order to obtain the maximum inhibition efficiency, optimization of the process variables affecting corrosion of mild steel was carried out using the Box – Behnken Design plan and desirability function of Response Surface Methodology (RSM). The three parameters - varied include; TCL concentration (inhibitor), immersion time, and temperature and their effects in corrosion inhibition were established. The optimum conditions predicted from the quadratic model were inhibitor's concentration (0.39 g/l), exposure time (8.68 hours), and temperature (36.06 °C) with the inhibition efficiency of 91.95 %. The data fitted well to the quadratic model which was validated. Adsorption of the extract's component on the mild steel was responsible for the inhibitory effect of the TCL extract. The results showed that 97.92% of the total variation in the inhibition efficiency of TCL can be connected to the variables studied.

**Keywords:** Mild steel, acid, *Terminalia catappa*, Corrosion, Response surface methodology (RSM).

### INTRODUCTION

Alloys of metals and pure metals react chemically with aggressive medium (acid) to form a stable compound in which the loss of metal occurs. The formed compound is called corrosion product (rust) and metal surface becomes corroded (Umoren *et al.*, 2016). Corrosion involves the movement of metal ions into the solution at active areas (anode), passage of electrons from the metal to an acceptor at less active areas (cathode), an ionic current in the solution and an electronic current in the metal. The cathodic process requires the presence of an electron acceptor such as oxygen or oxidizing agents or hydrogen ions (Bentiss *et al.*, 2000; Hukovic-Metikos *et al.*, 2002; Raja and Sethuraman, 2008).

Metals corrosion is main problems that need to be tackled for economic, safety, and environmental reasons. This can be reduced by using appropriate approaches which will in turn suppress, impede or totally halt the cathodic or anodic reactions or both (Raja and Sethuraman, 2009). Amongst the numerous techniques of

controlling and preventing corrosion, the application of corrosion inhibitors, is becoming very prevalent in mitigating corrosion in chemical process industries. Majority of the inhibitors that have been proven to be effective are natural complexes that contain mainly atoms of nitrogen, sulphur, or oxygen in their compositions. The use of some of these inorganic inhibitors has been restricted since their production is frequently very expensive. They can be hazardous and poisonous to wildlife, human beings, aquatic, and as well as the environment. This has led to the pursuit of environmentally friendly corrosion inhibitors as a substitute to changing the inorganic and organic inhibitors that will promote ecological green ecosystem. These readily available alternative green corrosion inhibitors are non-hazardous, economical, and can be regenerated. These inhibitors have been found in different parts of plant extract (Okafor *et al.*, 2008; Okafor *et al.*, 2010, Oguzie, 2008, Umoren *et al.*, 2016).

The use of plant products as corrosion inhibitors are justified by the phytochemical compounds

present therein, with molecular and electronic structures bearing close similarity to those of conventional organic inhibitor molecules (Oguzie *et al.*, 2010). Some researchers have reported the inhibitory effect of some plants extracts on mild steel corrosion. These plants are; *Andrographis paniculata* (Singh and Quraishi, 2010), *Spirulina platensis* (Kamal *et al.*, 2012), *Jasminum ndiflorum* L. (Li *et al.*, 2010), *Bridelia retusa* (Patela *et al.*, 2010), *Dacryodis edulis* (Oguzie *et al.*, 2010), aqueous extracts of mango, orange, passion fruit and cashew peels (da Rocha *et al.*, 2010), *Artemisia pallens* (Kalaiselvi *et al.*, 2010), *Gongronema latifolium* (Eddy and Odiongenyi, 2010), *Murraya koenigii* (Quraishi *et al.*, 2010), *Azadirachta indica* (Sharma *et al.*, 2010; Okafor *et al.*, 2010), *Garcinia kola* and *Cola nitida* (Okafor, *et al.*, 2007), rubber seed oil (Okewale and Olaitan, 2017), *Sida acuta* (Umoren *et al.*, 2016), *Kopsia singaporensis* (Raja *et al.*, 2010).

Statistical modeling and optimization are very vital process in design of experiment this is because it enables improvement in the system and as well as increase in the process efficiency without increase in cost on the process. The classical method of experimentation where by one factor is considered at a time is time consuming, very tedious, and this does not give complete effects of the variables/parameters on the process. It does not give the combined interactions between the physiochemical parameters and can also lead to misinterpretation of the results (Bas and Boyaci, 2007).

Response Surface Methodology (RSM) is a collection of statistical and mathematical techniques that are useful for analysis and modeling of engineering problems in which a response of interest is influenced by several parameters (Montgomery, 2001). RSM is used for designing experiments, building numerical models, evaluating the effects of variables, and searching for optimum combinations of factors (Okewale *et al.*, 2015). This method enable

researcher to know the interactive effect of the parameters on the process. *Terminalia catappa* is a large tropical tree in the leadwood tree family, *Combretaceae*. The tree has been spread widely by humans and the native range is uncertain. It has long been naturalized in a broad belt extending from Northern Australia and New Guinea through Southeast Asia and Micronesia into the Indian Subcontinent. Common names include Indian almond, Bengal almond, Singapore almond, Malabar almond, Tropical almond, Sea almond, Talisay tree, and Umbrella tree. The phytochemical screening carried out on *Terminalia catappa* extract revealed that it is a complex a mixture of many components like tannin, flavonoids, saponins, alkaloids, terpenoid, anthraquinone, cardiac glycosides, and phlobatanin. This work evaluate the effect of *Terminalia catappa* leaf extract as an inhibitor on corrosion of mild steel in acidic medium at 30°C using gravimetric method while RSM was employed to investigate the effects of the corrosion parameters like temperature, time of immersion, and concentration of inhibitor on the inhibition efficiency of the inhibitor on mild steel.

## **MATERIALS AND METHODS**

### **Materials**

*Terminalia catappa* leaves (TCL) were obtained from Ugbomro Community in Effurun, Delta State, Nigeria. Mild steel was procured from accredited iron sheet dealer in Effurun and machined in Mechanical Workshop at Federal University of Petroleum Resources, Delta State. HANNA model pH – 211 (pH meter), Genlab oven model Mino/75/f (oven), weighing balance of model (BH – 600), and beakers, were employed for the corrosion study. Hydrochloric acid, acetone, and ethanol solutions used were of analytical grades and were Sigma Aldrich. Distilled water was procured from the Department of Chemical Engineering Laboratory, Federal University of Petroleum Resources, Effurun, Delta State, Nigeria for sample preparation and solutions.

## Methods

### Pre-treatment of Sample and Sample Characterization

The samples were thoroughly washed thereafter, sun dried and pulverized into powdery form with the aid of laboratory blender. It was then sieved with a sieve of 420 $\mu$ m mesh. The sample was later stored in a desiccator prior to use.

### Analysis of the Phytochemical Constituents in *Terminalia catappa* Leaf Extracts

Screening of the phytochemical constituents of *Terminalia catappa* leaf extract were carried out to identify the chemical active constituents present in the *Terminalia catappa* leaf extract. The qualitative and quantitative methods were used as previously described (Okwu, 2001; Rahilla *et al.*, 1994; Sofowora, 1993; and Harborne, 1973).

### X – ray Fluorescence Analysis (X – RF)

The elemental compositions of the mild steel used were observed with X – supreme 800, Oxford instrument. The metal was machined to flat shape of 20 – 50mm diameter. Sample cup was cleaned and filter fixed in it shape. Thereafter, the metal sample was placed in the cup to cover the filter and ensure a thickness of 3mm at the bottom. The instrument was switched on, the elemental composition method was selected, and the sample identity was then level in the position selected with reading accepted. The instrument was rotated to the position of the sample in the tray at X – ray position after the start button was pressed while the intensity of radiation was now converted to the weight percentage.

### Extraction of *Terminalia catappa* Leaf Extracts (TCL)

A 500ml soxhlet apparatus and ethanol as solvent was used for the extraction of *Terminalia catappa* leaf extract. A quantity (100g) of the powdery sample was poured onto a Mushin cloth and placed inside the thimble of the apparatus. A round bottom flask containing 300ml of ethanol is fixed to the end of the apparatus and the

condenser was tightly fixed at the bottom end of the extractor.

The whole set up is heated on a heating mantle at a temperature of 78 $^{\circ}$ C. The solvent was allowed to remain in contact with the powdery *Terminalia catappa* leaf extract sample for 3 hours and the excess solvent in the extract was recovered by heating.

### Experimental Procedure (Gravimetric Measurements)

The effect of exposure time, inhibitor concentrations and temperatures was quantified using weight loss measurement. The corrosion study was carried out using the method described by Nwigbo *et al.* (2012). The mild steel coupons having dimensions of 2cm x 3cm x 0.12cm were polished with abrasive paper, greased, degreased with ether of petroleum and thereafter rinsed with distilled water and dried. The mild steel coupon was suspended with the aid of a thread in 100 ml beaker that contains 100 ml of 1.5M HCl. Each coupon after corrosion study was inserted in distilled water and dipped in methanol solution, which was scrubbed to remove remaining acids, and residual inhibitor and thereafter dried in acetone before it was re-weighed. The inhibition efficiency was calculate from Equation 1.

$$E(\%) = \frac{W_b - W_c}{W_b} \times 100 \quad (1)$$

Where,  $W_b$  is the weight loss in uninhibited medium (blank), and  $W_c$  is the weight loss in inhibited medium.

### Statistical Analysis

#### Experimental Design using

The design expert software (Design – Expert 7.00) was used for the experimental design and modeling of experimental data. Seventeen (17) runs of experiments were generated with Box - Behnken Design (BBD) as depicted in Table 2. The process variables studied were concentration of inhibitor ( $X_1$ ), time of exposure ( $X_2$ ), and temperature ( $X_3$ ) as shown in Table 1.

**Table 1:** Box – Behnken design independent variables levels

Independent variable	Low level	Mid-point	High level
Exposure time (hours)	3	6	9
Inhibitor concentration, (g/L)	0.1	0.3	0.5
Temperature, (oC)	35	45	55

**Table 2:** Box-Behnken Design of Experiment plan (Actual Value)

Time (hours)	Inhibitor concentration (g/L)	Temperature (°C)
6	0.5	55
6	0.3	45
6	0.1	55
3	0.3	35
9	0.1	45
6	0.3	45
6	0.5	35
3	0.5	45
6	0.1	35
6	0.3	45
6	0.3	45
9	0.5	45
3	0.1	45
6	0.3	45
9	0.3	55
3	0.3	55
9	0.3	35

**Optimization of the RSM Regression Model**

The quadratic model generated from the Response Surface Methodology was optimized using the global response surface equation below;

$$Y = \beta_0 + \sum_{i=1}^k \beta_i X_i + \sum_{i=1}^k \beta_{ii} X_i^2 + \sum_{i<j} \beta_{ij} X_i X_j + e(2)$$

For three factor inputs of  $x_1$ ,  $x_2$  and  $x_3$ , the equation of the quadratic response is given as;

$$Y = b_0 + b_1 X_1 + b_2 X_2 + b_3 X_3 + b_{12} X_1 X_2 + b_{13} X_1 X_3 + b_{23} X_2 X_3 + b_{11} X_1^2 + b_{22} X_2^2 + b_{33} X_3^2 (3)$$

Y is the response predicted by the Response Surface Methodology, the linear coefficient is i and j is the quadratic coefficients, regression

coefficient is  $\beta$ , while parameters studied is k, and optimized in the experiment, and the random error is e (Ghorbana *et al.*, 2008).

**RESULTS AND DISCUSSION**

**Characterization of Mild Steel and *Terminalia catappaleaf* extract**

Table 3 shows the result of the X – ray fluorescence of the mild steel used. It can be seen that iron has the highest percentage abundance.

**Table 3:** Result of the X – RF Analysis on mild steel

Elements	Abundance (%)
Magnesium	0.341
Aluminium	0.304
Silicon	0.190
Sulphur	0.065
Chlorine	0.031
Calcium	0.221
Titanium	0.125
Chromium	0.067
Manganese	0.203
Iron	98.362
Cobalt	0.061
Bromine	0.03

Table 4 shows the presence of tannin, alkaloids, terpenoid, and saponins while the anthraquinone presence in the TCL leaf extract also suggest that the sample inhibit corrosion process of mild steel in acidic medium studied. The inhibitory action of this extract was attributed to the presence of organic compounds in their chemical constituents and the blocking of the metal surface via adsorption of these organic compounds onto the surface of metal in acidic solutions. The presence of these compounds has been reported to promote the corrosion inhibition of mild steel in aggressive acid media and also corroborating the works of other researchers (Prithiba *et al.*, 2014; Owate *et al.*, 2014; Umoren *et al.*, 2006; Nwigbo *et al.*, 2012).

**Table 4:** Phytochemical Composition of TCL Extract

Phytochemical constituents	Indicator of constituent present
Tannin	+++
Saponin	+++
Flavonoid	++
Alkaloid	+++
Terpenoid	+++
Phlobatannin	++
Anthraquinone	++
Cardiac glycosides	+++
Reducing sugar	-

+++ : More abundance, ++: Abundance, -: Absent

**Statistical Analysis of the Response Surface Methodology Quadratic Model of BBD**

Statistical testing of the quadratic model was done using analysis of variance (ANOVA) as depicted in Table 5. The significance of the model was adjudged by the F – value. The high F – value of 36.57 and p – value < 0.0001 showed the model is significant. Thus, the data obtained experimentally is represented well with the quadratic model. The standard deviation value of 2.87 is realized. Coefficient of correlation ( $R^2$ ) value of 0.9792 which is closer to 1 was achieved for the model signifying that the predicted values are close to the actual values, suggesting that the model is accurate (Mohd and Rasyidah, 2010). The results from this investigation showed that the selected factors were sufficiently represented by the obtained model. This also described that there is an actual correlation among the factors selected in the experiment. It showed that 97.92% of the total variation in the inhibitor efficiency of TCL can be connected to the variables studied experimentally. The p – values were used to check the significance of each coefficient in the model. An adequate precision ratio of 23.517 is achieved in this work

which indicates an adequate signal. It can also be suggested that the model can be used to navigate the design space. Adequate signal for the model obtained was confirmed with the precision ratio value gotten. The goodness – of – fit of the regression equation was measured using the adjusted determination of coefficient ( $R^2$ ). The predicted  $R^2$  value of 0.888, which indicates the model power in response prediction. The coefficient of variation (CV) for the standard deviation of the mean and experimental data is 3.91%. It showed a better precision and reliability of the experiments carried out (Li *et al.*, 2011; Rodrigues *et al.*, 2012). This is less than 10% which is an indication that the data from the experiment is reproducible. The significance of each coefficient is also ascertained using the P – value and F – value at 5% confidence level. It can be seen that corrosion of mild using TCL as inhibitor was significantly affected by ( $X_2$ , and  $X_3$ ,  $p < 0.0001$ ) linear terms. It is obvious that the interaction terms ( $X_1X_3$ , and  $X_2X_3$ ) were also significant at the level of  $p < 0.05$  and the quadratic terms ( $X_2^2$ , and  $X_3^2$ ) were also significant at the level of  $p < 0.05$ . This allude an interaction between the process variables studied which were noted to be the core factors that affect the rate of corrosion, and weight loss of the mild which in turn affect the efficiency of the inhibitor. The Response Surface Methodology model for the corrosion inhibition of mild steel using TCL as an inhibitor was optimized using the desirability function of the Response Surface Methodology. The optimum conditions predicted from the quadratic model were inhibitor’s concentration (0.39 g/l), exposure time (8.68 hours), and temperature (36.06°C) corresponding to the inhibitor’s efficiency of 91.95%. These values were validated with an average inhibitor’s efficiency of 92.15% from three replicates and this was in agreement with the obtained optimum result predicted by the model.

**Table 5:** ANOVA of regression equation for quadratic model

Sources	Sum of squares	DF	Mean square	F – Value	P - value
Model	2708.51	9	300.95	36.57	<0.0001
Exposure time	22.31	1	22.31	2.71	0.1437
Inhibitor concentration	562.80	1	562.80	68.38	<0.0001
Temperature	1566.88	1	1566.88	190.39	<0.0001
Exposure time and Conc	0.67	1	0.67	0.082	0.7833
Exposure time and Temperature	73.44	1	73.44	8.92	0.0203
Concentration and Temperature	3.10	1	73.10	8.88	0.0205
Exposure time <sup>2</sup>	3.74	1	3.74	0.45	0.5219
Concentration <sup>2</sup>	155.52	1	155.52	18.90	0.0034
Temperature <sup>2</sup>	221.16	1	221.16	26.87	0.0013
Residual error	57.61	7	8.23		
Lack of fit	57.61	7	19.20		
Pure error	0.00	4	0.00		
Corelation Total	2766.12	16			

The experimental testing of full quadratic model is expressed by the second order polynomial equation in term of actual values is as follows:

$$Y = - 45.52875 + 8.03583x_1 + 32.81250x_2 + 5.33900x_3 + 0.6833x_1x_2 - 0.14283x_1x_3 + 2.1375x_2x_3 - 0.10472x_1^2 - 151.93750x_2^2 - 0.07247x_3^2 \quad (5)$$

The final model equation after eliminating the insignificant terms is as follows;

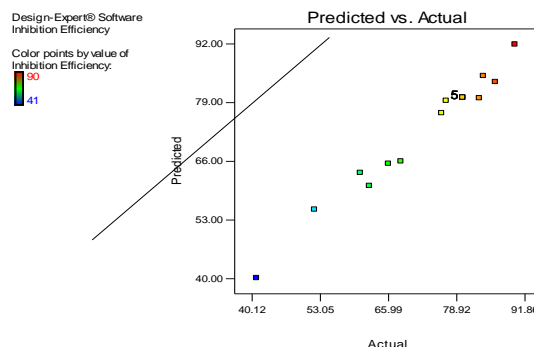
$$Y = - 45.52875 + 2.81250x_2 + 5.33900x_3 - 0.14283x_1x_3 + 2.1375x_2x_3 - 151.93750x_2^2 - 0.07247x_3^2 \quad (6)$$

where, Y is the inhibition efficiency, x<sub>1</sub> is exposure time (hr), x<sub>2</sub> is inhibitor concentration (g/L), and x<sub>3</sub> is the temperature (°C). The statistical estimate of the BBD obtained is shown in Table 6.

**Table 6:** Statistical estimates of BBD

Parameters	Value
Standard deviation	2.87
Mean	73.39
Coefficient of variation (%)	3.91
Coefficient of determination (R <sup>2</sup> )	0.9792
Predicted coefficient of determination (R <sup>2</sup> )	0.888
Adjusted coefficient of determination (R <sup>2</sup> )	0.9524
Adequate precision	23.517

The predicted and experimental data of the inhibition efficiency (%) were also investigated to determine their correlation as depicted in Figure 1. The data points on the plot were moderately spread and very close to the straight line point. This suggests a sound connection between the predicted values and values obtained from the experiment for the response, and therefore the strong predictive power of the model. This further confirmed the assumptions made that the data is appropriate for the design space.



**Figure 1:** Predicted values of the model versus the experimental data values.

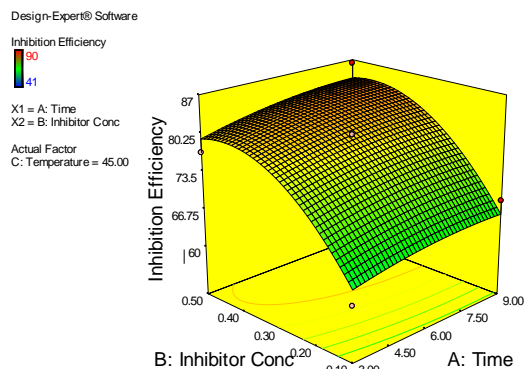
### Surface Response Plots

3-D surface response plots were presented in Figures (2 – 4) based on the analysis of the full quadratic model. It was used to give an important evidence on the system behaviour within the experimental design on the corrosion of mild steel using TCL as an inhibitor. The different shapes of the contour plots indicated different interactions between the variables. An elliptical contour plot indicated interactions between the variables were significant while a circular contour plot means otherwise (Li-Chun Zhao *et al.*, 2012).

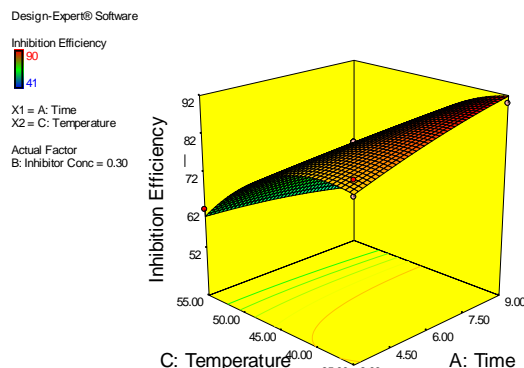
Figure 2 contour plot showed an interaction between the concentration of an inhibitor and time of exposure on the efficiency of the corrosion inhibition. An increase in exposure time from 3 to 6 hours enhanced the inhibition efficiency of the corrosion. Though, when time of exposure was increased more than 6 hours, there was a steady decrease in the response but the inhibitor concentration over 0.4g/L did not indicate any effect that is evident on the efficiency of the corrosion inhibition. This can be explained that, increasing time of exposure may quicken the degradation of the bioactive chemical compound in the inhibitor, which lead to lower inhibition efficiency.

Figure 3 described the effect of temperature ( $x_3$ ) and time of exposure ( $x_1$ ) on the corrosion inhibition efficiency, it can be seen that the inhibition efficiency is reduced as time of exposure is increased with temperature. This also affirmed the insignificant of the time of exposure in the model obtained from this study.

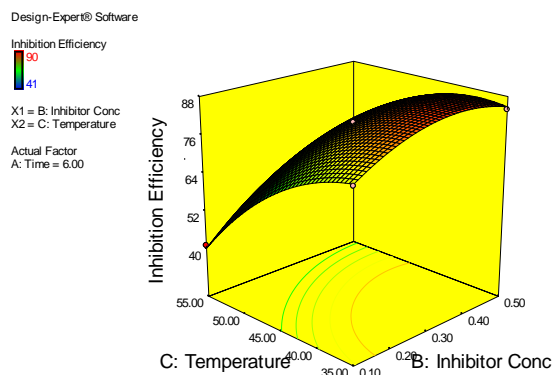
Figure 4 depicted the effect of temperature ( $x_3$ ), and inhibitor concentration ( $x_2$ ), on the inhibition efficiency, it was observed that at lower temperature and inhibitor concentration favoured an increased in inhibition efficiency of the inhibitor on mild steel. This also confirmed the synergetic effect of these two terms on the inhibition efficiency.



**Figure 2:** Surface Plot for the influence of inhibitor's concentration, time, and interface with efficiency of inhibitor.



**Figure 3:** Surface plot for the influence of temperature, time, and interface with efficiency of inhibitor.



**Figure 4:** Surface plot for the influence of temperature, inhibitor's concentration, and interface with efficiency of inhibitor.

## CONCLUSION

The corrosion inhibition of mild steel in acidic medium using TCL inhibitor can be linked to adsorption mechanism. The results showed that 97.92% of the total variation in the inhibition efficiency of TCL can be connected to the variables studied. The quadratic model developed showed that temperature and concentration of the TCL extract significantly affected the inhibition efficiency of the TCL extract on mild steel surface. Correlation coefficient of determination ( $R^2$ ) obtained was 0.9792. The optimum process parameters obtained from the quadratic model developed were 0.39 g/L TCL concentration, 9 hours immersion time, and temperature of 36.06°C with a predicted optimum inhibition efficiency value of 91.95 %. The results from the validated optimum value were in agreement with that predicted by the quadratic model.

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