Communication in Physical Sciences, 2022, 8(4):429-441

Ethanol Extract of *Curcuma longa* as a green corrosion inhibitor for carbon steel in solution of HCl

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Received: 14 June 2022/Accepted 30 August 2022/Published online: 24 September 2022

Abstract: *Investigation of the potential of ethanol extract of Mango leaf as a green corrosion inhibitor for the corrosion of carbon steel was implemented using gravimetric, FTIR and scanning electron microscopy analytical techniques. Results obtained from the gravimetric analysis indicated that the inhibition efficiency of the extract were 57.38, 72.13 and 78.69% at the inhibitor's concentration of 0.1, 0.2 and 0.3 g/L of the extract respectively. The corrosion and the corrosion inhibition process were found to followed pseudo first order kinetics and at various concentrations, the inhibitor demonstrated the potential to extend the half-life of the metal in the acid solution. The adsorption of the inhibitor suited the Langmuir and Frumkin adsorption models. The inference drawn from the isotherms revealed that there exists an interaction between the inhibitor's molecules and that the lateral interaction parameter revealed the attractive behaviour of the inhibitor. The adsorption was spontaneous and agreed with the mechanism of physical adsorption. The scanning electron micrograph of the metal surface after inhibition revealed the formation of a protective layer on the metal surface. Some functional groups that were native to the leaf extract were found to be missing in the FTIR spectrum of corrosion product which suggested that they were used for adsorption while some were shifted, an indication that there was an interaction between the adsorbed species.*

Keywords: *Corrosion, carbon steel, green inhibition, mango leaf extract*

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1.0 Introduction

Researches in green corrosion inhibition is attracting global interest because green corrosion inhibitors are less toxic and biodegradable (Uwah *et al*., 2013). These requirements are hardly met by most synthetic and available chemical compounds but by extract of living organisms (Majd *et al*., 2019; Matos *et al*.,2019; Marzorati *et al*., 2019). In our research group, we have investigated the corrosion inhibition potentials of several plant extracts including, *Vernonia amygdalina* (Odiongenyi *et al*., 2009),*Lasianthera Africana* (Eddy *et al*., 2009c), *Aloe vera* (Eddy and Odoemelam, 2009e), *Azadirachta indica* (Eddy and Mamza, 2009), *Phyllanthus amarus* (Eddy, 2009;Eddy and Awe, 2018), *Gongronema latifolium* (Eddy and Ebenso, 2010), *Gnetum Africana* (Eddy *et al*., 2009), *Heinsia crinata* (Eddy and Odiongenyi, 2010), *Anogessus leocarpus* (Ameh *et al*.,

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2012), *Terminalia catappa* (Eddy *et al*., 2009b), *Hibiscus sabdariffa calyx* (Eddy *et al*., 2011), *Occimium vegratissimum* (Eddy *et al*., 2010), *Solanum melongena (*Eddy *et al*., 2010), *Gloriosa superba* (Eddy *et al*., 2014) and others. We were able to establish that the leaf extracts of these plants are very good corrosion inhibitors. Other researchers have also investigated and confirmed other leaf extracts as good corrosion inhibitors. Examples, *Saraca ashoka extract* (Saxena *et al*., 2018) and *Glycyrrhiza glabra leaf extract* (Alibakhshi *et al*., 2018). Others have used plant seeds, for example, Bahlakeh *et al.* (2019) reported effectiveness of Mustard seed while Eddy (2010) found Garcina *kola and Cola nitida* seeds extracts. Fruits extracts of *Citrullus lanatus* have been reported by Dehghani, *et al*. (2019) while Majd *et al*. (2019) deduced good corrosion inhibition efficiencies from various concentrations of aqueous extract of *Primula Vulgaris* flower. According to Bouknana *et al.* (2015), aqueous extracts of olive roots, stems and leaves are good eco-friendly inhibitors for steel in an acidic medium. Eddy *et al.* (2012) have also reported that *Daniella Oliverri* gum exudate is a good corrosion inhibitor for mild steel in an acidic medium. The use of extracts of plant waste has not been excluded from the list of good corrosion inhibitors (Abdel-Graber *et al*., 2006; Marzorati *et al.,* 2019; Matos *et al*., 2019). The general selection rules for good corrosion inhibitors are the possession of hetero atoms that can act as center for adsorption, and the presence of double or π-bond, conjugated and aromatic systems. Most plant extracts can meet the environmental compliance to corrosion inhibition because they are majorly rich in phytochemicals which are useful to humans. This implies that the adsorption process which is the initial mechanism in corrosion inhibition would be unique when plant extracts are involved. Ameh and Eddy (2014) stated that inhibition by plant extract occurs via cooperative adsorption mostly through the formation of a multimolecular layer and that most plants display good inhibition efficiencies because of a synergistic combination of the various phytochemical constituents in them.

3.0 Results and Discussion

3.1 Weight loss

Fig. 1 shows the variation of weight loss with time for the corrosion of carbon steel in 3 M HCl in the absence and presence of various concentrations of ethanol extract of Mango leaf (inserted is the plot of weight loss versus time for the blank). The figure illustrates that the weight loss of carbon steel increases with an increase in the period of contact but decreased with an increase in the concentration of the extract. This indicates that various concentrations of ethanol extract of Mango leaf inhibited the corrosion of carbon steel and that the inhibition efficiency increased with an increase in concentration. The observed increase in inhibition efficiency with concentration suggests that the inhibitor is an adsorption inhibitor. The decrease in inhibition efficiency with the period of contact is an indication that the rate of corrosion of carbon steel in 3 M HCl increases with an increase in the period of contact.

The corrosion rate of the metal in 3 M HCl in the absence and presence of various concentrations of the inhibitor was calculated and the results are recorded in Table 1 while the plot of corrosion rate versus time is shown in Fig.2. The plots reveal that the corrosion rate is proportional to weight loss and followed a similar trend. The inhibition efficiency of ethanol extract of Mango leaf is shown in Table 2. The results clearly reveal that the inhibition efficiency increased with an increase in the concentration of the extract and ranged from 57.39 to 78.69%. A similar trend was followed for the variation of the degree of surface coverage (Table 2), that is, the surface coverage increases with concentration due to an increase in the number of inhibitor molecules that diffuse to the metal surfaces.

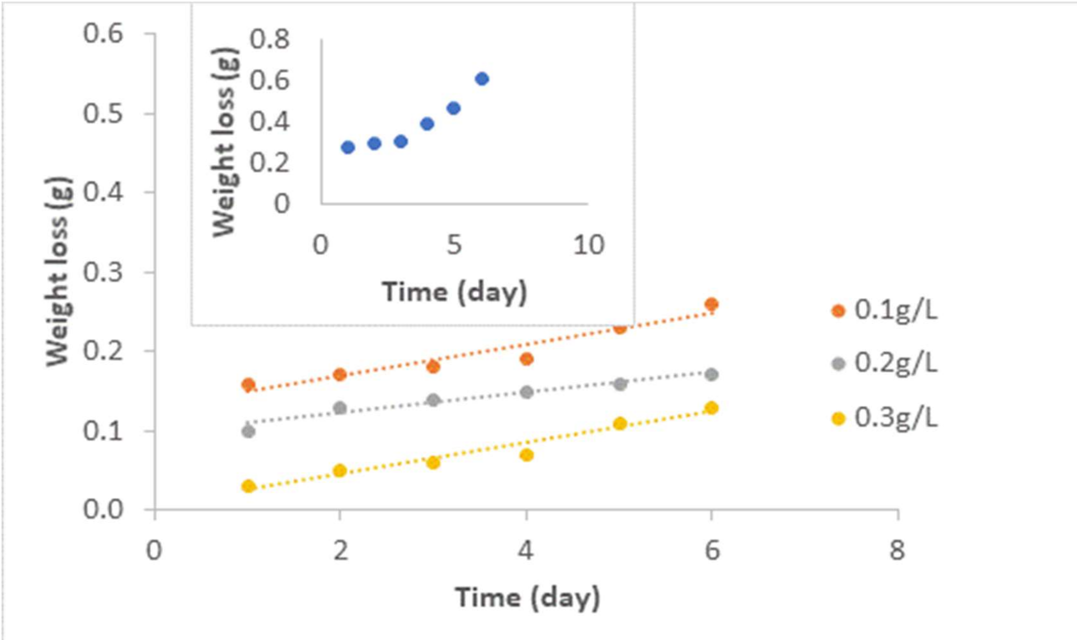


Fig. 1: Variation of the corrosion rate with time for the corrosion of carbon steel in the absence and presence of various concentrations of ethanol extract of Mango leaf

Table 1: Corrosion rate ($gm^{-2}h^{-1}$) of carbon steel in 3 M HCl in the absence and presence of various concentrations of ethanol extract of Mango leaf.

Time (day)	Blank	0.1g/L	0.2g/L	0.3g/L
1	0.5833	0.3333	0.2083	0.0625
2	0.6250	0.3542	0.2708	0.1042
3	0.6458	0.3750	0.2917	0.1250
4	0.8125	0.3958	0.3125	0.1458
5	0.9792	0.4792	0.3333	0.2292
6	1.2708	0.5417	0.3542	0.2708

The inhibition efficiencies recorded in Table 2 are average inhibition efficiency which was obtained after six days of immersion. Instantaneous inhibition calculated after every 24 hours of immersion is shown in Table 3 and depicted graphically in Fig. 3. From the figure, it can be seen that the inhibition efficiency first decreased before it increased with time. Therefore, there is a period required for the passivation or development of the protective film on the surface of the carbon steel.

Table 2: Inhibition efficiencies of various concentrations of ethanol extract of Mango leaf for carbon steel in a solution of HCl

C (g/L)	%IE	(θ)
0.1	57.38	0.5738
0.2	72.13	0.7213
0.3	78.69	0.7869

Table 3: Instantaneous inhibition efficiency of ethanol extract of Mango leaf for carbon steel corrosion

Time (hours)	0.1g/L	0.2g/L	0.3g/L
24	42.85	64.28	89.29
48	43.33	56.67	83.33
72	41.93	54.84	80.64
96	51.28	61.54	82.05
120	51.07	65.96	76.60
144	57.38	72.13	78.69

3.2 Kinetic study

The kinetic of corrosion and corrosion inhibition is known to be consistent with a pseudo first order kinetics, which implies that equation 1 is applicable

$$-\log(weight\ loss) = k_1 t + constant \quad (4)$$

Based on equation 4, a plot of -log (weight loss) versus time was found to be linear as shown in Fig. 4.

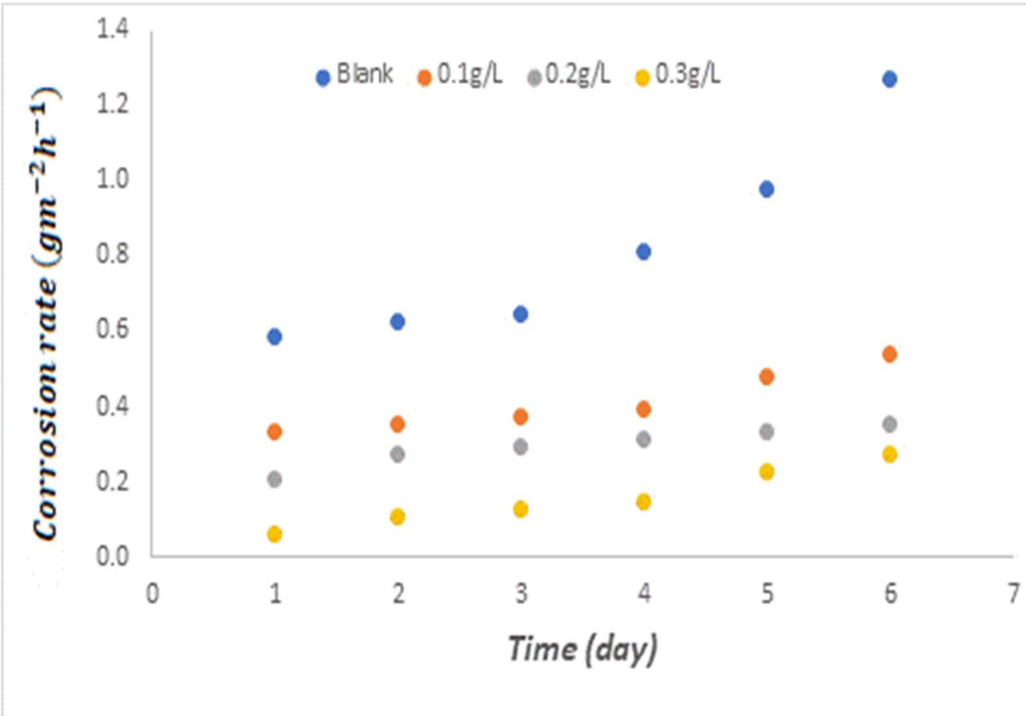


Fig. 2: Variation of the corrosion rate with time for the corrosion of carbon steel in 3 M HCl in the absence and presence of various concentrations of ethanol extract of Mango leaf

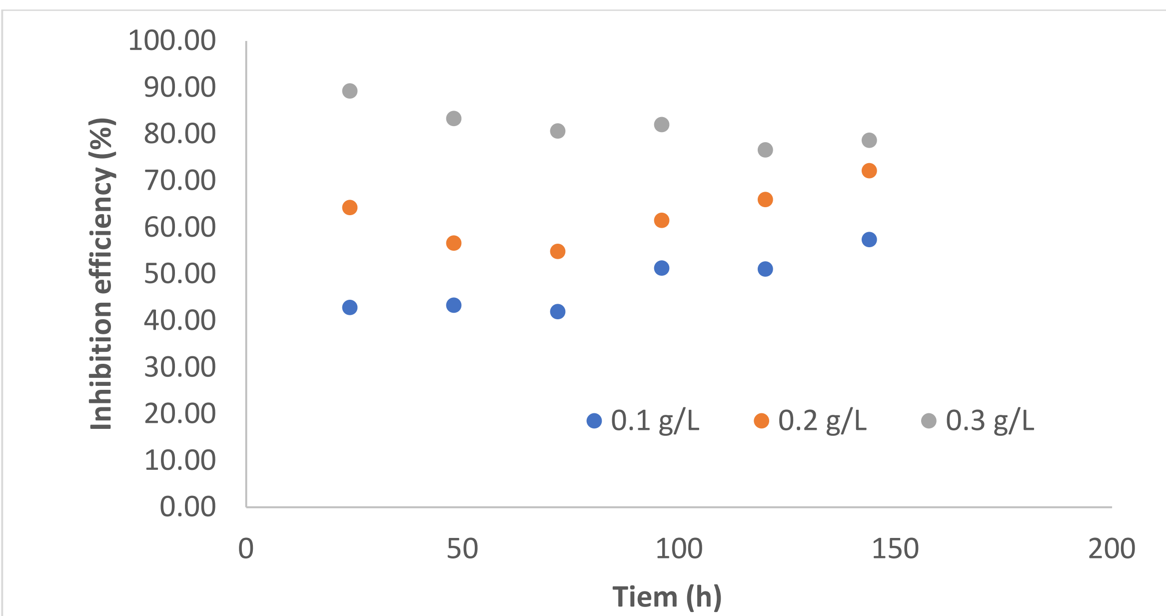


Fig. 3: Variation of instantaneous inhibition efficiency of ethanol extract of Mango leaf with time

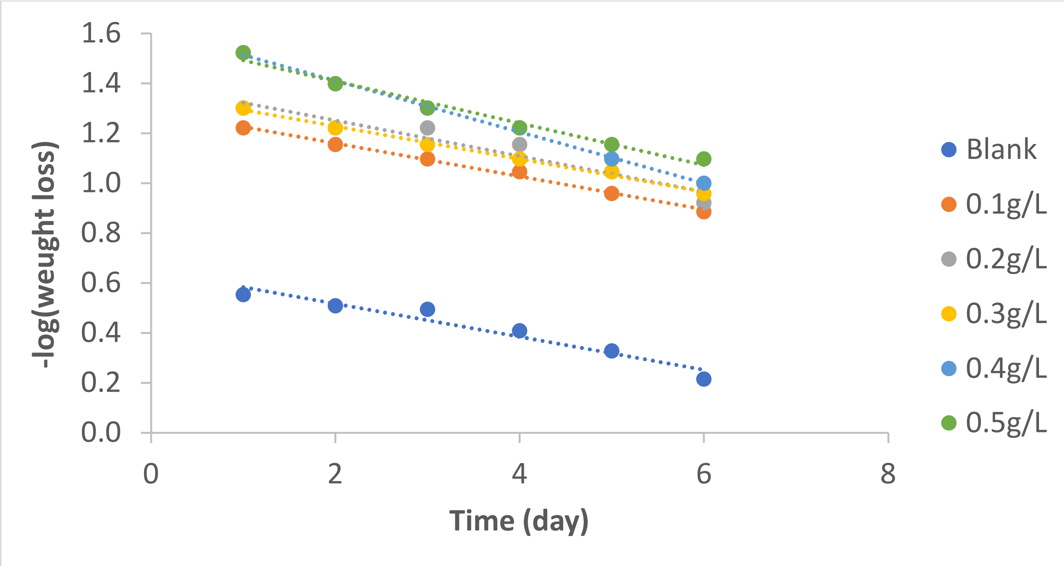


Fig. 4: Variation of -log (weight loss) with time for the corrosion of carbon steel in 3 M HCl containing various concentrations of ethanol extract of Mango leaf.

The plots have an excellent degree of linearity (R^2 ranged from 0.9238 to 0.9967). The calculated rate constants are recorded in Table 4. The rate constant (k_1) is related to the half-life according to equation 5

$$t_{1/2} = \frac{0.693}{k_1} \quad (5)$$

The half-life of the metal in the presence of various concentrations of the inhibitor was higher compared to the half-life of the metal in the blank solution (3 M HCl) which was 9 days. Therefore, ethanol extract of Mango leaf extended the half-life of carbon steel in 3 M HCl.

3.3 Adsorption isotherm

Adsorption isotherm is useful in explaining the adsorption characteristics of a corrosion inhibitor, the mechanism of inhibition and other processes occurring in the corrosion inhibition process (Eddy *et al*., 2022).

Experimental data were tested for the fitness of various adsorption isotherms using values of surface coverage at various concentrations of the inhibitor. The test indicated that the adsorption of the inhibitor obeyed the Langmuir and the Frumkin adsorption isotherms. The assumptions establishing the Langmuir adsorption isotherm can be expressed according to equation 6 (Yurt *et al*., 2014),

$$\ln\left(\frac{C}{\theta}\right) = \ln C - \ln k_{ad} \quad (6)$$

where C is the concentration of the inhibitor in the bulk solution, θ is the degree of surface coverage and k_{ads} is the Langmuir equilibrium constant of adsorption which is related to the standard free energy of adsorption as follows (Odiongenyi and Afangide, 2019):

$$\Delta G^0_{ads} = -RT\ln(55.5k_{ads}) \quad (7)$$

where R is the gas constant, T is the temperature and 55.5 is the molar heat of adsorption of water. The Langmuir isotherm for the adsorption of ethanol extract of Mango leaf on the surface of carbon steel is shown in Fig. 5 while parameters calculated from the plot are shown in Table 5. This included the equilibrium constant of adsorption, the free energy change, values of R^2, slope and intercept of the plot. The application of the Langmuir isotherm is confirmed by an excellent degree of linearity of the plot (R^2 = 0.9982). An ideal Langmuir isotherm should have a slope equal to unity. Values of slope greater or less than unity indicate that there is an interaction between the adsorbed specie (Eddy and Ita, 2011a,b). Consequently, there

is an interaction between the adsorbed species. Therefore, there is some level of interaction between the adsorbed specie. Estimated value of the standard free energy of adsorption was -9.81 kJ/mol which is negatively less than the threshold value (- 40 kJ/mol) required for the mechanism of chemical adsorption.
Therefore, the adsorption of ethanol extract of Mango leaf is spontaneous and occurred through the mechanism of physical adsorption (Odiongenyi *et al*., 2015).

Table 4: Kinetic data for the corrosion of carbon steel in 3 M HCl containing various concentrations of ethanol extract of Mango leaf at 303 K

C (g/L)	Slope	$t_{1/2}$ (min)	R^2
Blank	0.0028	10	0.9286
0.1	0.0018	16	0.9371
0.2	0.0017	17	0.8932
0.3	0.0015	19	0.9691

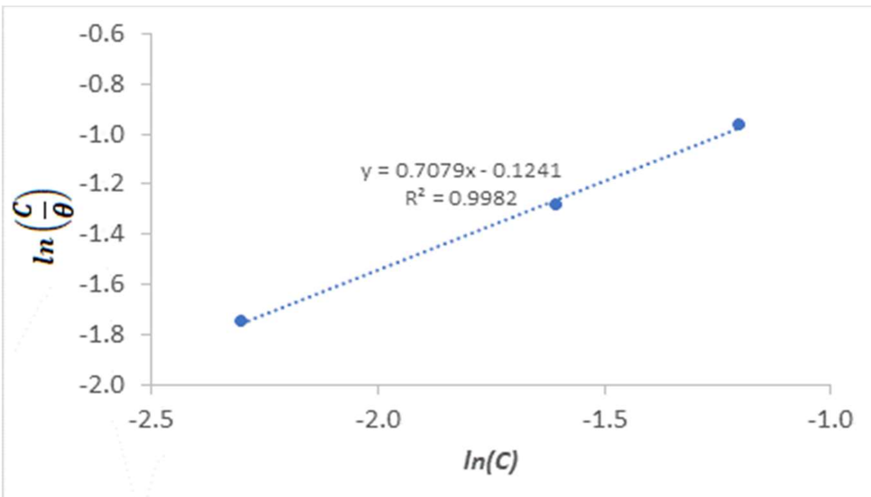


Fig. 5: Langmuir isotherm for the adsorption of ethanol extract of Mango leaf on the surface of carbon steel

According to Eddy and Ita (2011b), when the calculated free energy is negatively less than – 20 kJ/mol, the mechanism of physical adsorption is sustained but free energy values between -20 to -40 kJ/mol point toward both mechanisms while those above -40 kJ/mol is consistent with the mechanism of chemical adsorption.
The model of the Frumkin adsorption isotherm is expressed by equation 8 (Almzarzie *et al*., 2020),

$$ln\left[C.\left(\frac{\theta}{1-\theta}\right)\right] = lnk_{ads} + 2\alpha\theta \qquad (8)$$

α is the lateral interaction parameter and describes the interaction in the adsorbed layer. Frumkin isotherm for the adsorption of ethanol extract of Mango leaf on the surface of the carbon steel is shown in Fig. 6. The degree of linearity of the plot was very high (R^2 = 0.9965) and implies the application of the Frumkin isotherm to the adsorption of ethanol extract of Mango leaf on the surface of the carbon steel. The lateral interaction parameter (4.8852) was positive and indicate the attractive behaviour of the inhibitor's molecules.

3.4 Scanning electron microscopy

The scanning electron micrograph of the carbon steel in 3 M solution of HCl is shown in Fig. 7. The micrograph reveals an uneven and unsmooth surface of the carbon steel due to corrosion (Fig. 7a) but in the presence of the inhibitor the surface of the metal is seen to be covered by a protective film.

3.5 Fourier transformed infrared absorption study

The FTIR spectrum of the Mango leaf is shown in Fig. 8. After adsorption on the surface of the carbon steel, the spectrum of the scratch corrosion product is shown in Fig. 9. Functional groups, frequencies of IR absorption and associated assignment are presented in Table 5.

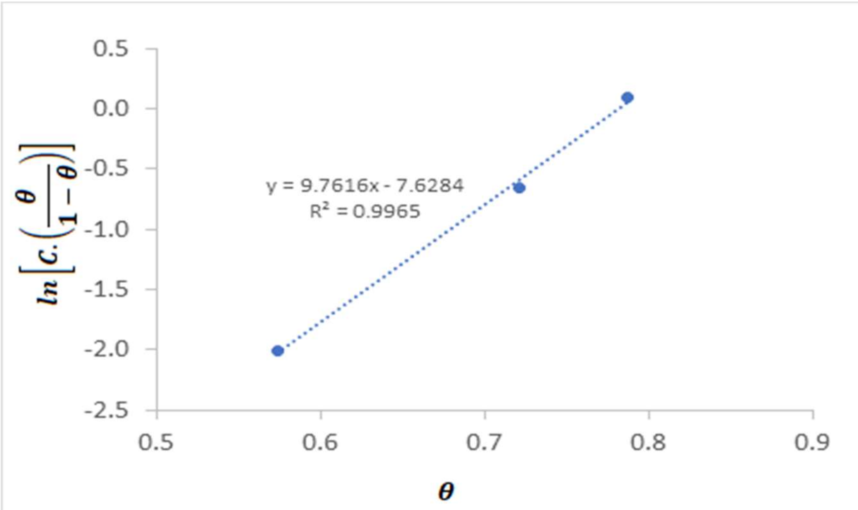
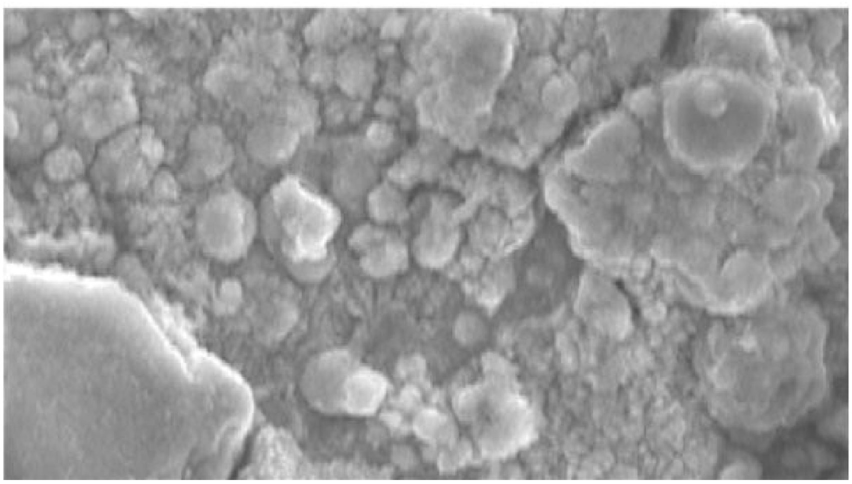


Fig. 6: Frumkin isotherm for the adsorption of ethanol extract of Mango leaf on the surface of carbon steel



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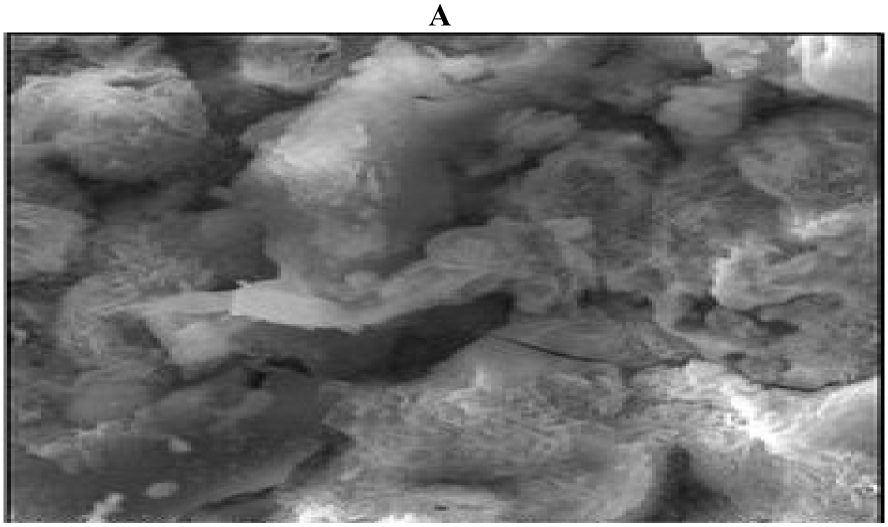

B

Fig. 7: Scanning electron micrograph of (a) carbon steel immersed in 3 M HCl (b) carbon steel immersed in 3 M HCl in the presence of 5 g/L of the inhibitor. (image was taken at 30 kV at x750)

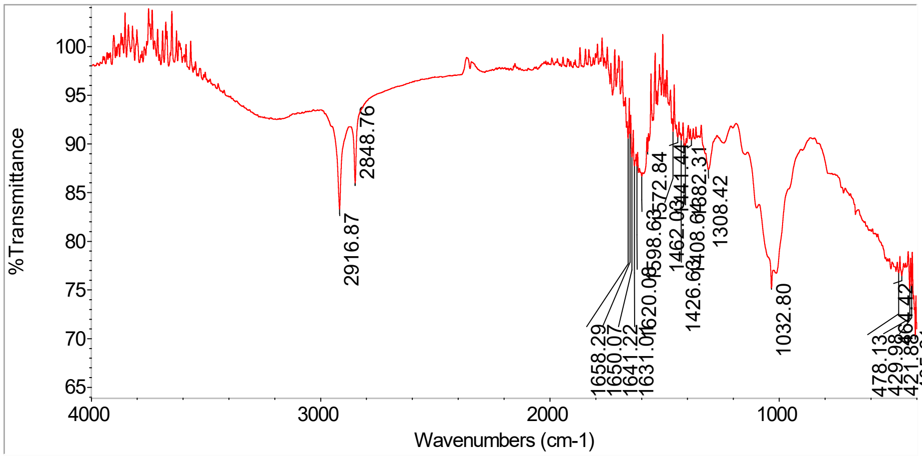

Fig. 8: FTIR of Mango leaf

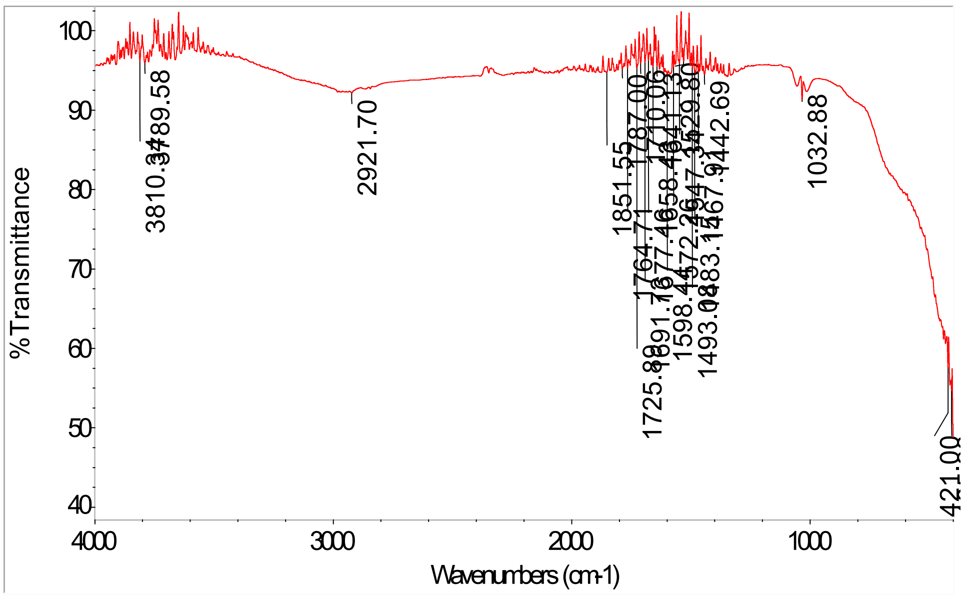

Fig. 9: FTIR of the corrosion product of carbon steel after inhibition by ethanol extract of Mango leaf

Functional groups identified in the IR spectrum of Mango leaf extract included OH stretch at 2917 cm^{-1}, N-H stretch at 2849 cm^{-1}, C=C stretches at 1658, 1631, 1573, 1599 and 1626 cm^{-1}, C-N stretch at 1641 cm^{-1}. Aromatic C=C stretch was found at 1462 cm^{-1}. CH_2 wagging vibration was found at 1382 cm^{-1}, S=O stretch occurred at 1032 and 1308 cm^{-1} while S-S stretches were found at 406, 422, 430 and 478 cm^{-1}. After adsorption, the OH and N-H stretches were shifted from 2917 to 2921 cm^{-1} and from 2848.76 to 2849.94 cm^{-1} respectively. The C=C stretch at 1658.29 was shifted to 1658.43 cm^{-1}. The aromatic C=C stretch was shifted from 1462 to 1464 cm^{-1}, the OH bend was shifted from 1441 to 1443 cm^{-1}, the S=O stretch at 1032.80 was shifted into 1032.88 and the S-S stretch was shifted from 405.61 to 405.93 cm^{-1}. Several new bonds were formed including C=O stretches (at 1716, 1726, 1725, 1764 and 1787 cm^{-1}), C=C stretches (at 1658 and 1677 cm^{-1}), C-O stretch at 1691 cm^{-1}, OH stretches at 3789.88 and 3810.34 cm^{-1} and N-O stretch at 1547 cm^{-1}. Functional groups that were missing in the spectrum of the corrosion product after inhibition were C=C stretches at 1620 and 1650 cm^{-1}, OH bend at 1427 and 1408 cm^{-1}, S=O stretch at 1308 cm^{-1} and S-S stretches at 430, 404 and 438 cm^{-1}. The functional groups that are present in the spectrum of the inhibitor but missing in that of the corrosion product might have been used in the adsorption of the inhibitor onto the surface of the carbon steel (Ameh and Eddy 2018).

Table 5: Absorption frequencies and peaks of IR absorption by Mango leaf and corrosion product after inhibition by Mango leaf

Mango leaf		**Corrosion product**		**Assignment**
Wave number (cm^{-1})	**Transmittance (%)**	**Wave number (cm^{-1})**	**Transmittance (%)**	
405.61	70.319	405.93	54.611	
421.88	75.435			
429.98	75.125			S-S stretch
464.42	76.441			S-S stretch
478.13	76.756			S-S stretch
1308.42	87.362			S=O stretch
1032.80	75.153	1032.88	91.209	S=O stretch
1382.31	90.383			CH_2 wagging
1426.63	90.353			OH bend
1408.64	89.834			OH bending
1441.44	90.672	1442.69	94.496	OH bend
1462.03	1.245	1467.91	94.705	Aromatic C=C stretch n
		1483.15	94.701	
		1493.08	95.273	
		1529.80	96.078	
		1547.34	95.819	N-O stretch
1572.84	89.172	1572.26	95.434	C=C stretch
1598.63	86.645	1598.44	94.478	C=C stretch
1620.08	87.700			C=C stretch
1631.01	88.638			C=C stretch
1641.22	84.794	1641.13	95.512	C-N stretch
1650.07	91.033			C=C stretch
1658.29	90.530	1658.43	95.453	C=C stretch
		1677.46	96.220	C=C stretch
		1691.73	96.021	C-O stretch
		1710.06	95.922	C=O stretch
		1725.89	95.282	C=O stretch
		1764.71	95.297	C=O stretch
		1787.00	95.144	C=O stretch
		1851.55	94.716	C-H bend
2848.76	85.940	2849.94		NH stretch
2916.87	83.295	2921.70	92.132	OH stretch
		3789.58	95.969	OH strech
		3810.34	96.187	OH strech

However, those that experience shifts in frequency were involved in interactions between the inhibitor's molecule and the metal surface. From the Frumkin adsorption model, it was found that there is an attraction of the inhibitor's molecules while the non-unity value of the Langmuir isotherm also suggests that there is an interaction between the adsorbate and the adsorbent as confirmed by the FTIR spectra of the inhibitor and that of the corrosion product.

4.0 Conclusion

The present study led to the findings that support the following conclusions

(i) Ethanol extract of Mango leaf is a good adsorption inhibitor for the corrosion of carbon steel in a solution of HCl.

(ii) The inhibition efficiency of the extract increases with an increase in the extract concentration but decreased with increasing period of contact

(iii) The average inhibition efficiency mechanism of the extract supports physical adsorption

(iv) The adsorption of the extract on the metal surface is spontaneous and is best described by the Langmuir and Frumkin adsorption isotherms

(v) The corrosion of carbon steel in 3 M HCl in the absence and presence of various concentrations of ethanol extract of Mango leaf followed a pseudo first order kinetics

(vi) The extract has the potential of extending the half life of carbon steel in solution of HCl.

(vii) Functional groups in the Mango extract that were involved in adsorption onto the metal surface are C-N stretch (1641 cm^{-1}), C=C stretch (1591 cm^{-1}), N-O stretch (1552 cm^{-1}), aromatic C=C stretch (1462 cm^{-1}), OH bend (1413 cm^{-1}), CH_2 wagging (1378 cm^{-1}), S=O stretch (1033 cm^{-1}), C-X stretch (523 cm^{-1}) and some S-S stretches (498, 456, 431 and 409 cm^{-1}).

In view of the above conclusions, the following recommendations are made:

(i) There is a need to investigate the effect of temperature, pH and other environmental factors on the inhibition of the corrosion of carbon steel in 3 M HCl by ethanol extract of Mango leaf

(ii) The application of electrochemical techniques to evaluate the inhibition efficiency will provide more information on the kinetic and mechanism of inhibition.

5.0 References

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Consent for publication

Not Applicable

Availability of data and materials

The publisher has the right to make the data public

Competing interests

The authors declared no conflict of interest. This work was carried out in collaboration among all authors.

Funding

There is no source of external funding

Authors' contributions

All the authors contributed equally to the work