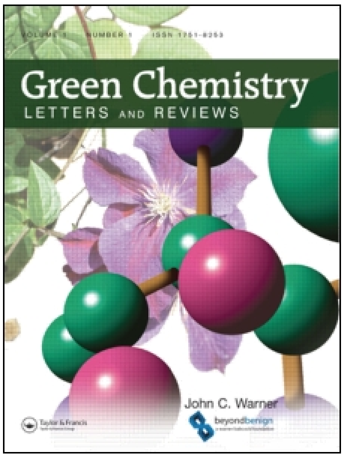

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RESEARCH LETTER

Ethanol extract of *Ocimum gratissimum* as a green corrosion inhibitor for the corrosion of mild steel in H_2SO_4

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The potential of ethanol extract of *Ocimum gratissimum* as a green inhibitor for the corrosion of mild steel in H_2SO_4 was investigated using gasometric, gravimetric, and thermometric methods of monitoring corrosion. The results obtained, indicated that ethanol extract of *O. gratissimum* is an adsorption inhibitor for the corrosion of mild steel. The inhibition efficiency of the inhibitor was found to decrease with increase in the period of contact and with increasing temperature, but increased with increase in the concentration of the inhibitor. From the observed trend for the variation of inhibition efficiency with temperature and the range of values obtained for activation energy (52.24–55.24 kJ mol^{-1}) and free energy of adsorption (-19.15 to -17.06 kJ mol^{-1}), a physical adsorption mechanism is proposed for the adsorption of the inhibitor on the surface of mild steel. In addition, the adsorption is exothermic, spontaneous, and is best described by Langmuir adsorption isotherm.

Keywords: corrosion; mild steel; green inhibitor; *Ocimum gratissimum*

Introduction

Green corrosion inhibitors have the advantage of being inexpensive, non-toxic, and eco-friendly. These advantages have provoked numerous and intensive searches on the use of naturally occurring substances or their extracts for the inhibition of the corrosion of metals (including mild steel). Extracts of *Gnetum africanum*, *Raphia hookeri*, exudate gum, gum arabic, *Datura metel*, *Andrographis paniculata, Carica papaya, Occinum viridis, Telfairia occidentalis, Vernonia amygdalina,* mango juice, *Rosmmarius officinalis, Azadirachta indica, Lasianthera africana, Aloe vera, Musa sapientum, Colocasia esculenta, Phyllanthus amarus, Piper guineense, Opuntia,* and tobacco leaves (*1–30*) have been investigated for their corrosion inhibition potential.

It has been established that the inhibitory actions of plant extracts are due to the presence of some organic compounds such as saponin, tannin, alkaloid, steroids, glycosides, and amino acids (*30*). Most of these compounds have centers for π-electrons and functional groups (such as $-C=C-$, $-OR$, $-OH$, $-COOH$ $-NR_2$, $-NH_2$, and $-SR$), which provide electrons that facilitate the adsorption of the inhibitor on the metal surface. Also, the presence of hetero atoms such as N, P, O, and S enhances the adsorption (which is the initial mechanism of inhibition) of the inhibitor on the metal surface. Recent researches have also indicated that amino acid contents of the plant extracts have a significant role to play in the corrosion inhibition mechanism of plant extracts (*31–40*).

Several researches have been carried out on the inhibition of the corrosion of mild steel by some plant extracts. However, literature is scanty on the use of ethanol extract of *Ocimum gratissimum* as an inhibitor for the corrosion of mild steel in H_2SO_4. Therefore, the present study seeks to investigate the inhibitory potentials of ethanol extract of *O. gratissimum* for the corrosion of mild steel in H_2SO_4 solutions. Effect of temperature on the inhibition potential of the extract shall be considered using Arrhenius equation. The adsorption characteristics of the extract shall be investigated using different adsorption models, while thermodynamics principles shall be used to discuss the data obtained from experiment. Finally, in order to know the active constituents of the plant responsible for the corrosion inhibition process, the phytochemical and amino acid constituents of the extract shall be determined.

The African basil (*O. gratissimum*) is an herbaceous perennial grass, which is woody at its base. It is pantropical and widely naturalized in many regions. *O. gratissimum* prefers wet and fertile conditions, but can tolerate drought after flowering. The leaves of

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this plant (shown in Figure 1) are known for their medicinal purpose and as a spice for the consumption of some tubers.

Results and discussion

Effect of concentration of Ocimum gratissimum

Figures 2 and 3 show the variation of volume of hydrogen gas evolved with time for the corrosion of mild steel in 2.5 M H_2SO_4 containing various concentrations of ethanol extract of *O. gratissimum* at 303 and 333 K, respectively. From the figures, it is evident that the volumes of hydrogen gas evolved by the blank solutions are higher than those evolved by solutions containing various concentrations of ethanol extract of *O. gratissimum* indicating that ethanol extract of *O. gratissimum* is an inhibitor for the corrosion of mild steel in H_2SO_4. In addition, the rate of evolution of hydrogen gas decreases with increasing concentration of ethanol extract of *O. gratissimum* but increases with increase in temperature suggesting that ethanol extract of *O. gratissimum* is an adsorption inhibitor for the corrosion of mild steel in H_2SO_4 and that the adsorption of the extract supports the mechanism of physical adsorption.

Figure 4 shows the variation of weight loss with time for the corrosion of mild steel in 0.1 M H_2SO_4 containing various concentrations of ethanol extract of *O. gratissimum* at 303 K. From Figure 4, it is evident that weight loss of mild steel increases with increase in the period of contact. This also indicates that the rate of corrosion of mild steel in H_2SO_4 solutions (containing various concentration of ethanol extract of *O. gratissimum*) increases with increasing period of contact. Weight loss of mild steel was also found to decrease with increase in the concentration of the inhibitor. Therefore, ethanol extract of *O. gratissimum* is an adsorption inhibitor for the corrosion of mild steel in H_2SO_4.

Values of the corrosion rate (CR) of mild steel in H_2SO_4 are presented in Table 1. The results obtained,

Figure 1. Leaves of *Ocimum gratissimum*.

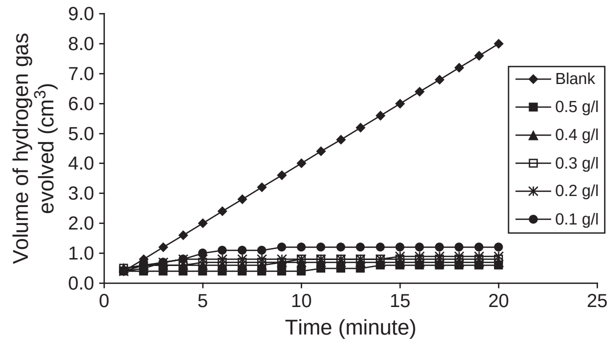

Figure 2. Variation of the volume of hydrogen gas evolved with time for the corrosion of mild steel in 2.5 M (blank) sulfuric acid containing various concentrations of ethanol extract of *Ocimum gratissimum* at 303 K.

indicate that the CRs of mild steel decrease with increase in the concentration of ethanol extract of *O. gratissimum*. Hence, ethanol extract of *O. gratissimum* retarded the rate of corrosion of mild steel in H_2SO_4. On the other hand, from the values of inhibition efficiencies obtained for various concentration of ethanol extract of *O. gratissimum* (Table 1), it is significant to note that the inhibition efficiency increases with increasing concentration of ethanol extract of *O. gratissimum*. This confirms that ethanol extract of *O. gratissimum* is an adsorption inhibitor for the corrosion of mild steel in H_2SO_4 solution. The present results compare favorably with those obtained by Eddy et al. (*30*) for *Gnetum africanum*.

Kinetic study

The kinetic of the corrosion of mild steel in the absence and presence of ethanol extract of *O. gratissimum* was investigated by plotting values of –log(weight loss) versus time. Straight line plots (with R^2 values approaching unity) were obtained (plots not shown). Therefore, the corrosion of mild steel in H_2SO_4 (containing various concentrations of ethanol

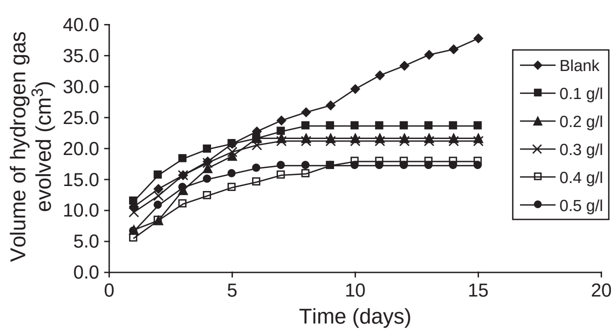

Figure 3. Variation of the volume of hydrogen gas evolved with time for the corrosion of mild steel in 2.5 M tetraoxosulfate (VI) containing various concentrations of ethanol extract of *Ocimum gratissimum* at 333k.

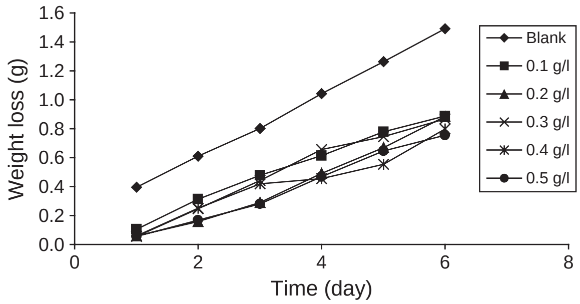

Figure 4. Variation of weight loss with time for the corrosion of mild steel in 0.1 M sulfuric acid by ethanol extract of *Ocimum gratissimum* at 303 K.

extract of *O. gratissimum*) is first order. Values of rates constants obtained from the slopes of the plots are recorded in Table 2. These values were seen to be constant for all concentrations of the extract. For a first order reaction, the half life ($t_{1/2}$) and the rate constant (k_1) are related according to Equation (1):

$$t_{1/2} = 0.963/k_1. \quad (1)$$

Calculated values of $t_{1/2}$ are also presented in Table 2. The half lives for various concentrations of ethanol extract of *O. gratissimum* are similar, indicating that the assumption of the rate models (i.e. $t_{1/2}$ is independent on the concentration of the inhibitor) is valid for the adsorption of ethanol extract of *O. gratissimum* on mild steel surface.

Effect of temperature

The effect of temperature on the inhibition of the corrosion of mild steel by ethanol extract of *O. gratissimum* was investigated using the logarithm form of Arrhenius equation, which can be written as follows (*41,42*):

$$\log\frac{CR_2}{CR_1} = \frac{E_a}{2.303R}\left(\frac{1}{T_1} - \frac{1}{T_2}\right), \quad (2)$$

where CR_1 and CR_2 are the corrosion rates of mild steel at the temperatures T_1 (303 K) and T_2 (333 K), respectively. E_a is the activation energy and R is the gas constant. Values of E_a calculated from Equation (*2*) are presented in Table 2. From the results obtained, the activation energies are within the range of values expected for a physical adsorption mechanism, hence the adsorption of ethanol extract of *O. gratissimum* is consistent with the mechanism of physical adsorption.

Thermodynamic and adsorption considerations

The heat of adsorption (Q_{ads}) of ethanol extract of *O. gratissimum* on the surface of mild steel was calculated using Equation (3) (*43*):

$$Q_{ads} = 2.303R\left[\log\left(\frac{\theta_2}{1-\theta_2}\right) - \log\left(\frac{\theta_1}{1-\theta_1}\right)\right] \times \left(\frac{T_1 \times T_2}{T_2 - T_1}\right)\text{kJ mol}^{-1}, \quad (3)$$

where Q_{ads} is the heat of adsorption of the inhibitor, R is the gas constant, θ_1 and θ_2 are the degrees of surface coverage of ethanol extract of *O. gratissimum* at 303 K (T_1) and 333 K (T_2), respectively. Calculated values of Q_{ads} are also presented in Table 2. These values are negative and tend to increase with increase in the concentration of ethanol extract of *O. gratissimum*, indicating that the adsorption of ethanol extract of *O. gratissimum* on mild steel surface is exothermic.

The adsorption behavior of ethanol extract of *O. gratissimum* was also studied by fitting data obtained for degree of surface coverage to different adsorption isotherms including Langmuir, Temkin, Freundlich, Frumkin, and Flory–Huggins adsorption isotherms. The tests reveal that the adsorption of ethanol extract of *O. gratissimum* on mild steel surface is best described by Langmuir and Temkin adsorption isotherms.

The expression for the Langmuir adsorption model can be written as follows:

$$\log(C/\theta) = \log C - \log K_{ads}, \quad (4)$$

where C is the concentration of the inhibitor in the electrolyte, θ is the degree of surface coverage of the

Table 1. Corrosion rate (CR), reaction number (RN) of mild steel in H_2SO_4 solutions and inhibition efficiencies (%I) of various concentrations of ethanol extract of *Ocimum gratissimum*.

Concentration of *Ocimum gratissimum* (g l^{-1})	0.1	0.2	0.3	0.4	0.5
CR ($cm^3 min^{-1}$) $\times 10^{-1}$ at 303 K (gasometric)	1.40	1.25	1.15	1.10	1.10
CR ($cm^3 min^{-1}$) $\times 10^{-1}$ at 333 K (gasometric)	9.87	8.13	8.27	7.73	7.13
CR ($gh^{-1}cm^2$) $\times 10^{-4}$ at 303 K (gravimetric)	2.98	2.90	2.68	2.57	2.29
RN (°C min^{-1}) $\times 10^{-2}$ at 303 K (thermometric)	1.86	1.72	1.70	1.59	1.44
%I at 303 K (gasometric)	89.47	93.42	96.05	97.37	97.37
%I at 333 K (gasometric)	36.50	39.00	66.43	72.34	73.70
%I at 303 K (gravimetric)	32.89	34.56	39.60	42.15	48.39
%I at 303 K (thermometric)	74.24	76.44	78.39	80.35	82.67

Table 2. Kinetic parameters and some thermodynamics parameters for the adsorption of ethanol extract of *Ocimum gratissimum* on the surface of mild steel.

Concentration of *Ocimum gratissimum* ($g\ l^{-1}$)	k_1	$t_{1/2}$ (day)	R^2	E_a ($kJ\ mol^{-1}$)	Q_{ads} ($kJ\ mol^{-1}$)
0.1	0.5	2	0.9876	54.68	−45.47
0.2	0.5	2	0.8976	52.42	−55.93
0.3	0.5	2	0.9231	55.24	−66.68
0.4	0.5	2	0.8976	54.59	−75.52
0.5	0.5	2	0.8976	52.33	−75.50

Note: k_1 = rate constant; $t_{1/2}$ = half life; R^2 = degree of linearity of the kinetic plots; E_a = activation energy; Q_{ads} = heat of adsorption.

inhibitor, and K_{ads} is the equilibrium constant of adsorption. Using Equation (4), the plot of log(C/θ) versus logC (Figure 5) was found to be linear, indicating the application of the Langmuir model to the adsorption of ethanol extract of *O. gratissimum* on mild steel surface. Values of Langmuir adsorption parameters deduced from the slopes and intercepts of the plots are presented in Table 3. From the results obtained, the slopes and values of R^2 (obtained for lines on the plots) are very close to unity indicating that there is a strong adherence of the inhibitor's adsorption of to the assumptions establishing Langmuir isotherm.

The adsorption of ethanol extract of *O. gratissimum* was also found to occurred according to Temkin adsorption isotherm which can be expressed as follows:

$$\exp(-2a\theta) = KC, \tag{5}$$

where a is the Temkin interaction parameter, θ is the degree of surface coverage of the inhibitor, K is the equilibrium constant of adsorption, and C is the concentration of the inhibitor in the bulk electrolyte. Rearranging and taking logarithm of Equation (5), Equation (6) is obtained:

$$\theta = \frac{-2.303\log K}{2a} - \frac{2.303\log C}{2a}. \tag{6}$$

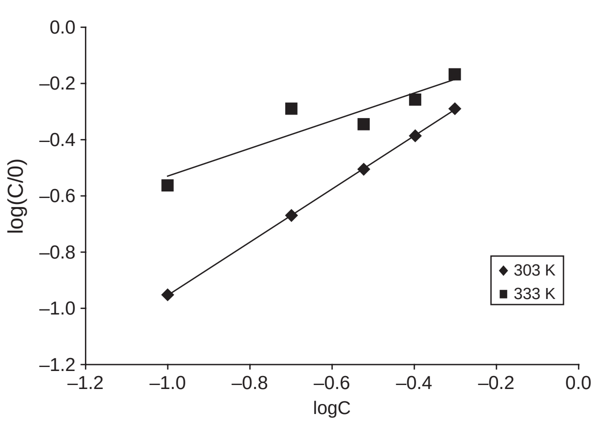

Figure 5. Langmuir isotherm for the adsorption of ethanol extract of *Ocimum gratissimum* on the surface of mild steel.

Using Equation (6), the plots of θ versus logC were also linear (Figure 6), which also indicate the application of the Temkin isotherm to the adsorption of ethanol extract of *O. gratissimum* on mild steel surface. Values of adsorption parameters deduced from the Temkin isotherms are also presented in Table 3. From the results the interaction parameters were found to be negative indicating the non-attractive behavior of the inhibitor.

The equilibrium constant of adsorption of ethanol extract of *O. gratissimum* is related to the free energy of adsorption (ΔG_{ads}) according to the following equation (*41,42*):

$$\Delta G_{ads} = -2.303RT\log(55.5K_{ads}), \tag{7}$$

where R is the gas constant, T is the temperature, K_{ads} is the equilibrium constant of adsorption, and 55.5 is the molar concentration of H_2SO_4. Values of K_{ads} obtained from the intercepts of Langmuir and Temkin isotherms were used to compute the free energies for the adsorption of ethanol extract of *O. gratissimum* on mild steel surface. From the calculated values (Table 3), the free energies are negatively less than the threshold value of −40 kJ mol^{-1} required for chemical adsorption, therefore, the adsorption of ethanol extract of *O. gratissimum* on mild steel surface is spontaneous and supports the mechanism of physical adsorption.

Phytochemical constituents/inhibition mechanism

Phytochemical screening of ethanol and aqueous extract of *O. gratissimum* reveals that ethanol extract of *O. gratissimum* contains alkaloid, saponin, tannin, phlobatanins, anthraquinone, steroids, terpenoid, flavanoids, and cardiac glycoside (with steroid ring and deoxy-sugar), while the aqueous extract contains all the above listed phytochemicals except saponin and anthraquinone. These results are presented in Table 4. The concentration of some amino acids on ethanol extracts of *O. gratissimum* is also presented in Table 4. From the results of the present study, it can also be proposed that the inhibitory potentials of ethanol extract of *O. gratissimum* are due to its

Table 3. Langmuir and Temkin adsorption parameters for the adsorption of ethanol extract of *Ocimum gratissimum* mild steel surface.

Langmuir	Temperature (K)	log*K*	Slope	ΔG_{ads} (kJ mol^{-1})	R^2
	303	0.0089	0.9443	−10.15	0.9999
	333	0.0365	0.4934	−11.33	0.8524
Temkin	**Temperature (K)**	**log*K***	***a***	**ΔG_{ads} (kJ mol^{-1})**	**R^2**
	303	1.0175	1.1300	−16.00	0.9743
	333	0.9355	1.2300	−17.06	0.8578

phytochemical constituents (*4*). In addition, from the results of research carried out by Assashi-Sorkhabi et al. (*42*), it is evident that amino acids are good corrosion inhibitors. Therefore, the inhibition of the corrosion of mild steel by ethanol extract of *O. gratissimum* may be due to synergistic interactions of its phytochemicals and amino acid constituents.

Experimental

Materials

Materials used for the study were mild steel sheet of composition (wt%) Mn (0.6), P (0.36), C (0.15), Si (0.03), and the rest Fe. The sheet was mechanically pressed cut into different coupons, each of dimension $5 \times 4 \times 0.11$ cm. Each coupon was degreased by washing with ethanol, rinsed with acetone, and allowed to dry in air before it was preserved in a desiccator. All reagents used for the study were analar grade and double distilled water was used for their preparation.

Extraction of plants

Samples of *O. gratissimum* leaves were obtained from the Akwa Ibom State Polytechnic botanical garden in Ikot Ekpene, Nigeria. The leaves were dried, ground, and soaked in a solution of ethanol for 48 hours. After 48 hours, the samples were cooled and filtered. The filtrates were further subjected to evaporation at 352 K in order to leave the sample free of the ethanol. The sample so obtained was used in preparing different concentrations of the extract by dissolving 0.1, 0.2, 0.3, 0.4, and 0.5 g of the extract in 1 l of 2.5 M for use in gasometric and thermometric experiments. However, for gravimetric (weight loss) experiments, similar concentrations of the extract were, respectively, dissolved in 0.1 M H_2SO_4.

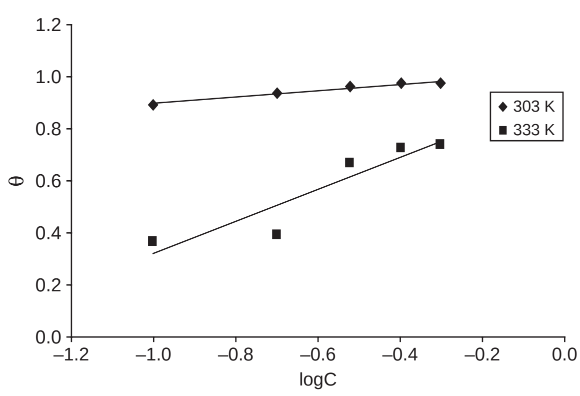

Figure 6. Temkin isotherm for the adsorption of ethanol extract of *Ocimum gratissimum* on mild steel surface.

Chemical analysis

The analysis of the amino acid composition of the extract was carried out using the amino acid analyzer as reported by Speckman et al. (*44*), while photochemical analysis of the extract was carried out according to the method reported by Eddy et al. (*30*).

Gasometric method

Gasometric methods were carried out at 303 and 333 K as described in the literature (*45*). Triplicate analysis was adopted for gasometric methods and the mean values were used for computation. From the average volume of hydrogen evolved per minute, inhibition efficiency (%*I*), degree of surface coverage (θ), and corrosion rate (CR) in °C min^{-1} were calculated using Equations (8)–(10), respectively:

$$\%I = \left(1 - \frac{V^{I}_{Ht}}{V^{0}_{Ht}}\right) \times 100 \quad (8)$$

$$\theta = \%I/100 \quad (9)$$

$$\mathrm{CR} = V^{I}_{Ht}/t \quad (10)$$

where V^{I}_{Ht} is the volume of hydrogen gas evolved (in cm^3) at time "*t*" (in minute) for inhibited solution and V^{0}_{Ht} for uninhibited solution.

Table 4. Concentrations of amino acid and phytochemicals in ethanol extract of *Ocimum gratissimum*.

Amino acid	Concentration (mg $100g^{-1}$)	Phytochemicals	Concentration (%)
Ihiamine	0.20	Saponin	0.61
Pyrdanine	0.41	Tannin	0.63
Ascorbic acid	15.35	Oxalate	1.05
Glycine	3.84	Phytate	0.65
Cysteine	1.62	Flavanoid	9.98
Hydrolysate	90.01	Phenols	0.33
Nicotinamide	0.88	Alkaloids	10.61

Thermometric method

Thermometric experiment was also carried out as reported elsewhere (*46–48*). From the average (mean of three samples) values obtained for the rise in temperature of the system per minute, the reaction number (RN) was calculated using Equation (11):

$$\mathrm{RN}(^{\circ}\mathrm{C\ min}^{-1}) = \frac{T_m - T_i}{t}, \qquad (11)$$

where T_m and T_i are the maximum and initial temperatures, respectively, and t is the time (min) taken to reach the maximum temperature. The inhibition efficiency (%*I*) was evaluated from percentage reduction in the RN, namely:

$$\%I = \frac{\mathrm{RN_{aq}} - \mathrm{RN_{wi}}}{\mathrm{RN_{aq}}} \times 100, \qquad (12)$$

where $\mathrm{RN_{aq}}$ is the reaction number in the absence of inhibitors (blank solution) and $\mathrm{RN_{wi}}$ is the reaction number for 2.5 M H_2SO_4 containing studied inhibitors.

Gravimetric analysis

In gravimetric experiment, a previously weighed metal (mild steel) coupon was completely immersed in 250 ml of the test solution in an open beaker. The beaker was inserted into a water bath maintained at a temperature of 30°C. After every 24 hours, each sample was withdrawn from the test solution, washed in a solution containing 50% NaOH and 100 gl^{-1} of zinc dust. The washed steel coupon was rinsed in acetone and dried in air before re-weighing. The difference in weight for a period of 168 h was taken as the total weight loss. From the mean values (mean of triplicate analysis) of the weight loss, the inhibition efficiency (%*I*) of the inhibitor and the CR of mild steel in various media were calculated using Equations (13) and (14), respectively:

$$\%I = (1 - W_1/W_2) \times 100 \qquad (13)$$

$$\mathrm{CR} = (W_2 - W_1)/At \qquad (14)$$

where W_1 and W_2 are the weight losses (g dm^{-3}) for mild steel in the presence and absence of the inhibitor, respectively, CR is the corrosion rate of mild steel in $gh^{-1}cm^{-2}$, A is the area of the specimen in cm^2, and t is the period of immersion in hours.

Conclusion

From the study, we conclude that ethanol extract of *O. gratissimum* is an adsorption inhibitor for the corrosion of mild steel in H_2SO_4 solutions. The adsorption of the inhibitor is spontaneous and supports the mechanism of physical adsorption. Inhibitory action of the extract is due to synergistic effect of its photochemical and amino acid constituents. Optimum inhibition efficiency can be obtained from ethanol extract of *O. gratissimum* by taking advantage of period of contact, concentration of the inhibitor, and temperature.

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