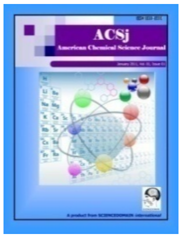

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Corrosion Inhibition of Aluminium in Acidic Medium by Different Extracts of *Ocimum gratissimum*

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Authors' contributions

This work was carried out in collaboration between the authors. Author IJA designed the study, performed the graphics and tables, wrote the first protocol, wrote the first manuscript. Author PME managed the analysis of the study and literature research. All authors read and approved the final manuscript.

Research Article

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ABSTRACT

Aims: This study is aimed at using plant extracts to prevent corrosion of aluminium in acidic medium. The inhibition efficiency of different extracts of 1M HCl, ethanol and distilled water were determined.
Study Design: Gravimetric method was used for the analysis.
Place and Duration of Study: Department of Chemistry, Federal University of Technology, Owerri, Nigeria, between March and November, 2011.
Methodology: Aluminium sheets of AA1060 and purity 98.98% was used. Each sheet was mechanically press-cut into coupons of dimension 3 cm x 3 cm. The solvents used for extraction of *ocimum gratissimum* leaf were 1M HCl, ethanol and distilled water, respectively. A hole with diameter 0.5 cm was drilled in each aluminium coupon and suspended in beakers containing test solution using glass hook. The weight loss was determined by retrieving coupons from test solutions at intervals. The coupons were weighed after retrieving. The different in weight was taken as weight loss of aluminium.

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Results: The percentage of inhibition efficiency (% I.E) was calculated at 303 K and 333 K, respectively. The result indicates that % I.E increases as concentration of inhibitor increases. The trend of inhibition efficiency was in order: Distilled H_2O > C_2H_5OH > 1M HCl. The result indicates that increase in temperature decreases inhibition efficiency and degree of surface coverage. The result shows that apparent activation energy E_a increases as inhibitor concentration increases. The activation energy of distilled water extract range between 45.02 to 79.90 kJ/mol. While the activation energies of ethanol and 1 M HCl extracts of the inhibitor ranges between 35.12 to 72.93 kJ/mol and 26.60 to 65.25 kJ/mol, respectively. The experimental data obtained corroborated with Langmuir and Flory Huggins adsorption isotherms.

Conclusion: This study indicates that different extracts of *Ocimum gratissimum* inhibits aluminium surface in presence of 1M HCl. The result of the analysis shows that inhibition efficiency and degree of surface coverage decreases as temperature increases. The activation energy of different extracts increases as concentration of inhibitor increases. The negative values of ΔG_{ads} shows that adsorption of inhibitor on surface of aluminium is spontaneous.

Keywords: Acid corrosion; weight loss; aluminium; adsorption; Ocimum gratissimum.

1. INTRODUCTION

Aluminium is a hard, strong, white metal. It is highly electropositive and resistant to corrosion because a hard, tough film of oxide is formed on the surface (Cotton and Wilkinson, 1972). The surface film is amphoteric, hence the metal could dissolve readily in both strong acid and alkaline media. Hydrochloric acid solutions are normally used for pickling of aluminium and electrochemical etching processes that normally lead to substantial loss of metal to corrosion. Aluminium is extensively used in industry as well as domestic applications.

Attempts have been made to reduce aluminium surface film dissolution and hence protect the metal in aggressive acid and alkaline media. Some of the methods employed to reduce corrosion of aluminium is application of sulphur, oxygen or nitrogen containing organic compounds as corrosion inhibitors to hinder corrosion reaction and thus reduce corrosion rate (Moussa et al., 1998; Madkour et al., 1999; Ebenso et al., 2001; Aytac et al., 2005). These organic compounds function by forming a protective adsorption layer on aluminium surface which isolates the corroding metal from action of corrodent.

Corrosion behaviour of aluminium has been studied in acid media. It has been reported that addition of halide salt to sulphuric acid solution containing organic inhibitor, effectively inhibits iron corrosion (Oguzie et al., 2004; Elewady et al., 2008). Halides have been reported to inhibit corrosion of some metals in strong acids and this effect depends on ionic size and charge. The use of chemical inhibitors has been a concern about its toxicity which affects the living organism as well as poisoning of environment. The use of plant extracts to prevent corrosion has become important because they are environmentally acceptable, readily available and renewable source for a wide range of inhibitors. Plant extracts are rich source of naturally synthesized chemical compounds that can be extracted by simple methods with low cost (Abdel-Gaber et al., 2008). These extracts are biodegradable and do not contain heavy metals or other toxic compounds. The use of plant extracts as corrosion inhibitor of metals in acidic and alkaline media have been reported (Abiola et al., 2007; Kliskic et al., 2000; Umoren and Ebenso, 2008; Oguzie, 2005; Eddy and Ebenso, 2008).

Ocimum gratissimum (scent leaf) is widely distributed in tropical Africa especially Nigeria. It belongs to lamiacea family and most abundant of genus *Ocimum*. The Igbos of Eastern part of Nigeria calls it Nchonwu. The Yorubas of Western part of Nigeria calls it Efinrin ajase, whereas Hausas of Northern part of Nigeria call it Daidoya (USDA, 2008). It is a perennial plant and woody at base. It has an average height of 1-3 m high and leaves are broad, narrowly ovate, usually 5-13 cm long and 3-9 cm wide. It is scented shrub with lime-green fuzzy leaves. It is used in Nigeria for nutritional and therapeutic purposes. In coastal area of Nigeria it is used for treatment of epilepsy, high fever and diarrhea, while in savannah areas, decoctions of its leaves are used to treat mental illness. The Igbos of Eastern Nigeria use *Ocimum gratissimum* for management of baby's cord by keeping the wound sterile. It is used as antifungal, antibacterial and for seasoning of foods.

2. MATERIALS AND METHODS

2.1 Material Preparation

Aluminium sheets of AAI060 and purity 98.98% were obtained from Material and Metallurgical Engineering Workshop, Federal University of Technology, Owerri, Nigeria. Each sheet was 0.1 cm in thickness and mechanically press-cut into coupons of dimension 3 cm x 3 cm. The coupons were descaled using wire brush and degreased in absolute ethanol, dried in acetone, weighed and stored in moisture- free desicator prior to use. The solvents used for extraction of *Ocimum gratissimum* leaf were 1 M HCl, ethanol and distilled water, respectively. All the solvents used in this study were of analytical reagent grade.

2.2 Extraction of Plant

Sample leaves of *Ocimum gratissimum* were obtained from Ihiagwa market in Owerri, Imo State, Nigeria. The leaf samples were washed and dried under the sunlight and ground to fine powder. 10 g of leaf extract were extracted with Soxhlet extractor for 3 hours using 1 M HCl, ethanol and distilled water, respectively. After extraction the samples were cooled and filtered. The filtrates obtained were used to prepare inhibitor concentrations between 20 mg/l and 100 mg/l in HCl corrodents.

2.3 Gravimetric Method

In weight loss experiment, a hole with diameter 0.5 cm was drilled in each aluminium coupon so that it could hang freely in solution. The aluminium coupons were suspended in beakers containing 200 ml of test solution maintained at 303--333 K with glass hooks and rods in a thermo-stated water bath. The weight loss was determined by retrieving coupons from test solutions at 3 hours intervals. The coupons after retrieving from test solution were scrubbed with bristle brush under running water, dried in acetone and weighed (Ebenso et al., 2001). The difference in weight was taken as weight loss of aluminium. From weight loss, the inhibition efficiency (%I.E) of extract and corrosion rate (CR) of aluminium were calculated using equations 1 and 2, respectively.

$$\%\ I.E = (1 - w_2/w_1) \times 100 \quad (1)$$

$$CR\ (gh^{-1}\ cm^{-2}) = \Delta w/AT \quad (2)$$

Where w_1, and w_2 are weight loss of aluminium in absence and presence of inhibitor, respectively. A is area of coupon in cm^2, T is period of immersion in hours and $\Delta w = w_1 - w_2$. The degree of surface coverage was calculated using:

$$\Theta = 1 - [C_{Rinh}/C_{RBC}] \quad (3)$$

Where θ is surface coverage; C_{Rinh} is corrosion rate for aluminium in presence of inhibitor, C_{RBC} is corrosion rate for aluminium in the absence of inhibitor.

3. RESULTS AND DISCUSSION

3.1 Weight Loss Consideration

Figs. 1 and 2 represented data of weight loss against concentration for aluminium corrosion in 1M HCl in absence and presence of different concentrations of inhibitor, *Ocimum gratissimum* at 303 k and 333 k, respectively. As shown in Figs. 1 and 2, increase in concentration of inhibitor, decreases weight loss of aluminium samples. The result shows that weight loss of aluminium increases as temperature increases as depicted in Fig. 2. The decrease in weight loss as inhibitor concentration increases shows that components of inhibitor retards corrosion of Al samples. The reduction in weight loss of Al samples as inhibitor concentration increases may be attributed to adsorption of inhibitor on surface of Al and thereafter impede corrosion either by merely blocking reaction sites (anodic and cathodic) or by altering mechanism of anodic and cathodic processes. The result indicates that weight loss of Al sample using different extracts of inhibitor were in order: 1M HCl > C_2H_5OH>distilled H_2O.

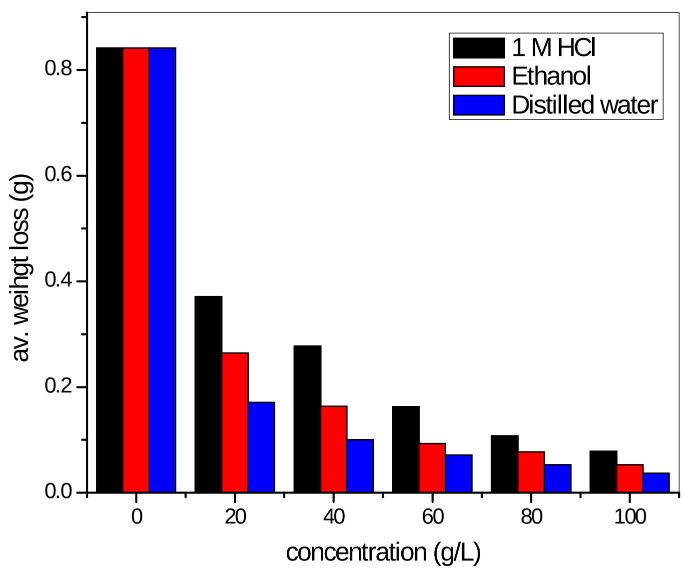

Fig. 1. Variation of weight loss of aluminium in HCl at various concentrations of 1 M HCl, ethanol and distilled water extracts of *Ocimum gratissimum* at 303 k

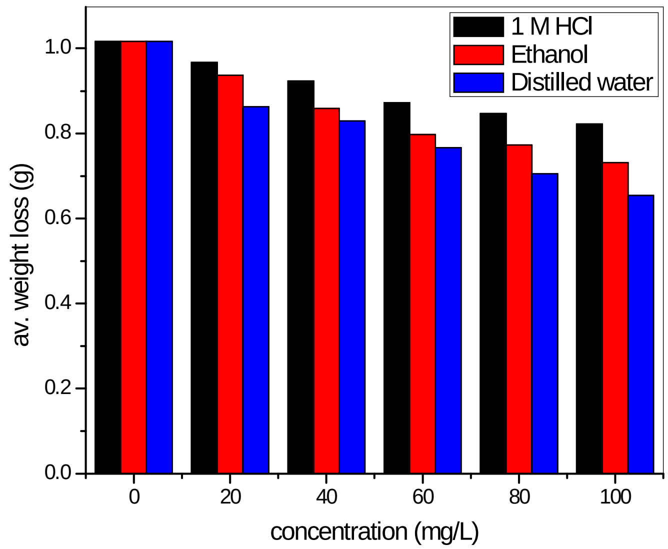

Fig. 2. Variation of weight loss of aluminium in HCl at various concentrations of 1 M HCl, ethanol and distilled water extracts of *Ocimum gratissimum* at 333 K

3.2 Inhibition Efficiency and Surface Coverage

The percentage of inhibition efficiency (% I.E) was calculated and represented in tables 1 and 2 at 303k and 333k, respectively. The result revealed that different extracts of inhibitor inhibits corrosion of Al sample in hydrochloric acid solution. The result indicates that % I.E increases as concentration of inhibitor increases. A parameter (θ), which represents part of metal surface covered by inhibitor molecules were calculated for different inhibitor concentrations and represented in tables 1 and 2. The result shows that surface coverage (θ) increases as inhibitor concentration increases. The increase in inhibition efficiency and surface coverage as concentration of extracts increases may be due to the presence of complex chemical composition of extracts. The increase in inhibition efficiency with increase in inhibitor concentration suggests that inhibitor molecule were adsorbed at aluminium sample /solution interface where adsorbed species mechanically screen the coated part of aluminium surface from action of corrosive medium.

Table 1. Effect of various inhibitor extracts on % I.E and θ for Al corrosion in 1M HCl at 303 k

Concentration mg/l	1m HCl extract of *Ocimum gratissimum*		Ethanol extract of *Ocimum gratissimum*		Distilled water extract of *Ocimum gratissimum*	
	% I.E.	**θ**	**% I.E**	**θ**	**% I.E.**	**θ**
20.00	55.96	0.56	68.62	0.69	79.79	0.80
40.00	67.04	0.67	80.60	0.81	88.11	0.88
60.00	80.70	0.81	88.96	0.89	91.53	0.92
80.00	87.31	0.87	90.87	0.91	93.80	0.94
100.00	90.72	0.91	93.74	0.94	95.65	0.96

Table 2. Effect of various inhibitor extracts on % I.E and θ for Al corrosion in 1M HCl at 333k

Concentration mg/l	1M HCl extract of *Ocimum gratissimum*		Ethanol extract of *Ocimum gratissimum*		Distilled water extract of *Ocimum gratissimum*	
	% I.E.	**θ**	**% I.E**	**θ**	**% I.E.**	**θ**
20.00	4.83	0.05	7.81	0.08	15.11	0.15
40.00	9.15	0.09	15.50	0.16	18.37	0.18
60.00	14.16	0.14	21.55	0.22	24.60	0.25
80.00	16.67	0.17	23.94	0.24	30.64	0.31
100.00	19.10	0.19	28.05	0.28	35.69	0.36

The result shows that distilled water extract exhibited highest inhibition efficiency and surface coverage when compared to values obtained for ethanolic and 1M HCl extracts, respectively. The highest inhibition efficiency and surface coverage of distilled water extract may probably be attributed to the presence of more complex chemical composition of distilled water extract. It has been reported (Alinnor and Aneke, 2009) that ethanolic extract of *Ocimum gratissimum* leaf contains alkaloid, saponins, flavonoids and tannins. The adsorption of these species from extracts on aluminium surface reduces surface area available for corrosion, thereby increasing inhibition efficiency and surface coverage.

Table 2 revealed that % I.E and θ decreases at 333 k when compared to values obtained at 303 k. The decrease in % I.E and θ at 333 k may be explained as a result of increase in average kinetic energy of components of extracts, thus making adsorption between components of extracts and Al surface insufficient to retain the species at binding site. This could lead to desorption or cause species to bounce off surface of aluminium instead of colliding and combining with it. Therefore, increase in temperature may be associated with decrease in stability of components – aluminium surface complex. Report (Odiongenyi et al., 2009) have shown that increase in temperature decreases inhibition efficiency of corrosion of mild steel in H_2SO_4 using ethanolic extract of *Vernonia amygdalina*. Ebenso et al., (2008) working on corrosion of mild steel in H_2SO_4 using ethanolic extract of *Piper guinensis* as green corrosion inhibitor reported decrease in inhibition efficiency at higher temperature. Noor (2007) working on corrosion inhibition of mild steel in acidic solutions with aqueous extract of Fenugreek leaves reported decrease in inhibition efficiency at higher temperature.

Figs. 3 and 4 show plot of inhibition efficiency versus different concentrations of inhibitor at 303 k and 333 k, respectively. The result indicates that inhibition efficiency was high at low temperature of 303 k when compared to values obtained at 333 k. This observation has been explained earlier in this report. The trend of inhibition efficiency was in order: Distilled $H_2O>C_2H_5OH>1M\ HCl$.

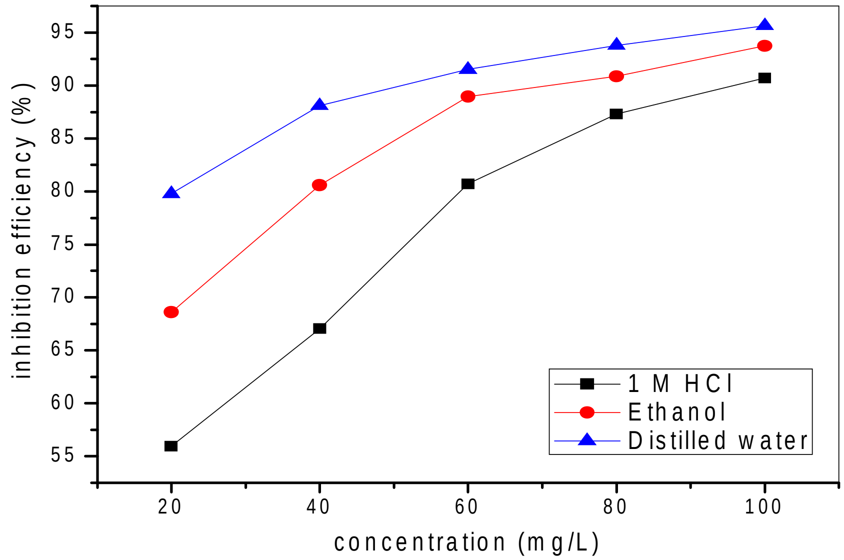

Fig. 3. Variation of inhibition efficiency with concentrations of 1 M HCl, ethanol and distilled water extract of *Ocimum gratissimum* at 303 K

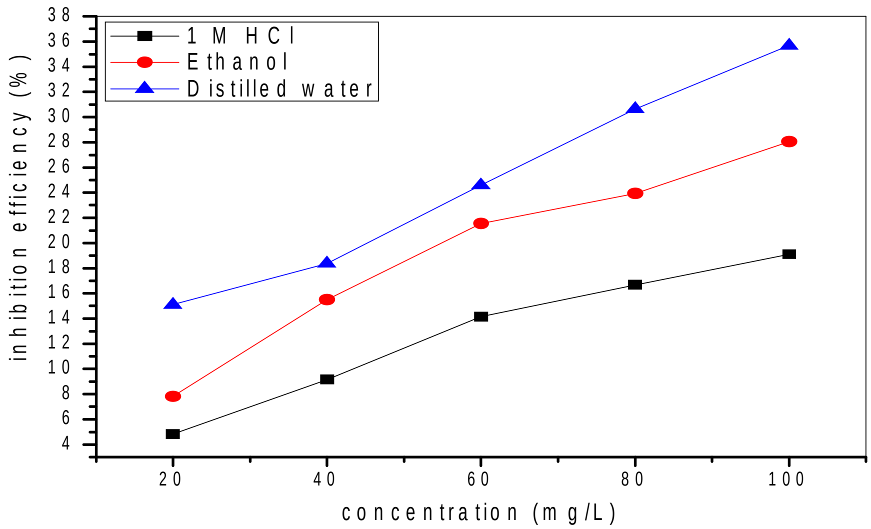

Fig. 4. Variation of inhibition efficiency with concentration of 1 M HCl, ethanol and distilled water extract of *Ocimum gratissimum* at 333 K

3.3 Adsorption Consideration

The adsorption of inhibitor on surface of corroding aluminium sample may be regarded as a substitution process between inhibitor compound in aqueous phase and water molecules adsorbed on aluminium surface:

$$Inh_{aq} + xH_2O_{ads} \rightleftharpoons Inh_{ads} + xH_2O_{aq} \quad (4)$$

Where x is size ratio, the number of water molecules displaced by one molecule of organic inhibitor. When the equilibrium as described in Eq (4) is reached, it is possible to obtain different forms of adsorption isotherm.

In the present study Langmuir adsorption isotherm was found to be suitable for the experimental findings and had been used to describe the adsorption characteristic of inhibitor. Langmuir adsorption isotherm is expressed in Eq (5) (Schockry et al., 1998).

$$\frac{C_{inh}}{\Theta} = \frac{1}{k} + C_{inh} \qquad (5)$$

where C_{inh} is inhibitor concentration, K is equilibrium constant of adsorption, θ is degree of surface coverage.

Figs. 5 and 6 show plot of C_{inh}/θ versus C_{inh} at 303 k and 333 k, respectively for 1M HCl, ethanol and distilled water extracts of inhibitor. Figs. 5 and 6 were linear indicating that adsorption of inhibitor is consistent with assumptions of Langmuir adsorption isotherm meaning there is no interaction between adsorbed species.

Adsorption of inhibitor extracts on Al surface was found to follow Flory – Huggins isotherm expressed in Eq (6) (Oguzie et al., 2004):

$$\log (\theta/C_{inh}) = \log K_c + x \log (1- \theta) \qquad (6)$$

Where θ is degree of surface coverage, C_{inh} is inhibitor concentration, x is number of water molecules replaced by one inhibitor molecule and K_c is equilibrium constant for adsorption process. Figs. 7, 8 and 9 show Flory – Huggins adsorption isotherm for different extracts of inhibitor using 1M HCl, ethanol and distilled water at 303 k and 333 k, respectively. The calculated values for x and k_c for 1 M HCl extract of inhibitor were 1.00 and 0.007 for 303 k and 0.40 and 0.74 for 333 k, respectively. The values for x and K_c obtained with ethanol extract of inhibitor were 1.0 and 0.006 for 303 k, and 0.30 and 0.63 for 333 k, respectively. Also calculated values for x and k_c with distilled water extract of inhibitor were 1.51 and 0.005 for 303 k, and 0.10 and 0.58 for 333 k, respectively. The result indicates that values of x are unity for both 1 M HCl and ethanol extracts, while distilled water extract of inhibitor is more than unity, indicating that each molecule of inhibitor is attached to one active site on Al surface.

The values of free energy of adsorption, ΔG_{ads} of 1 M HCl, ethanol and distilled water extracts of inhibitor on Al surface were calculated using Eq (7) (Noor, 2007):

$$\Delta G_{ads} = - 2.303 RT \log (55.5K) \qquad (7)$$

Where R is gas constant, T is temperature and K is equilibrium constant of adsorption, given as $K = \theta/1\text{-}\theta$ and 55.5 is concentration of water in solution. The values of ΔG_{ads} were negative in 303k and 333k indicating spontaneous adsorption of inhibitor on surface of aluminium, and the propose mechanism is physical adsorption (ΔG_{ads} < 40 kJ/mol) (Ebenso, 2004; Bilgic and Sahin, 2001).

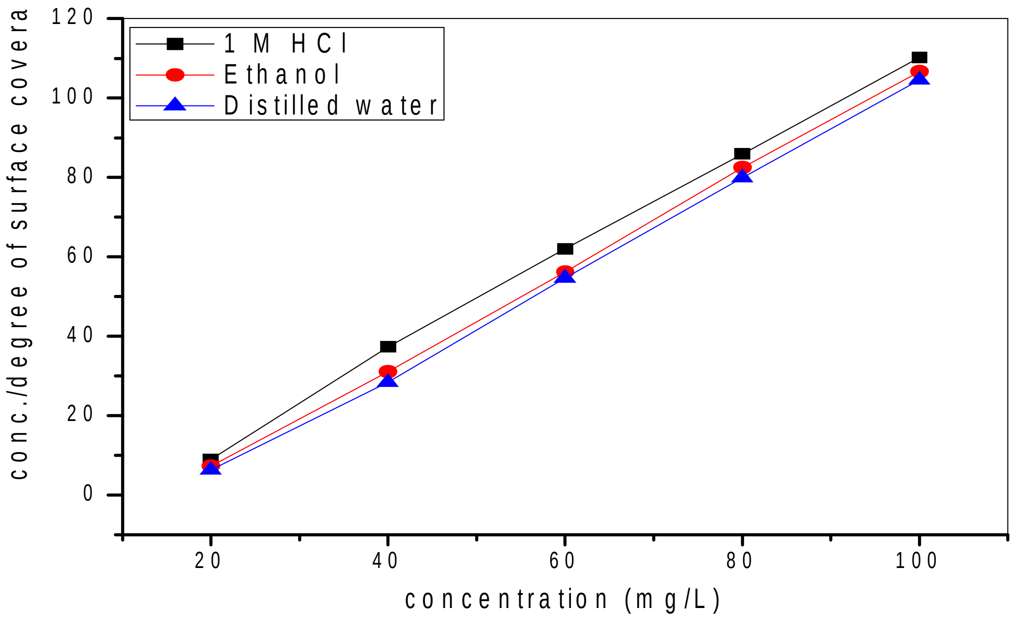

Fig. 5. Langmuir adsorption isotherm at 303 K for 1 M HCl, ethanol and distilled water *Ocimum gratissimum* extracts

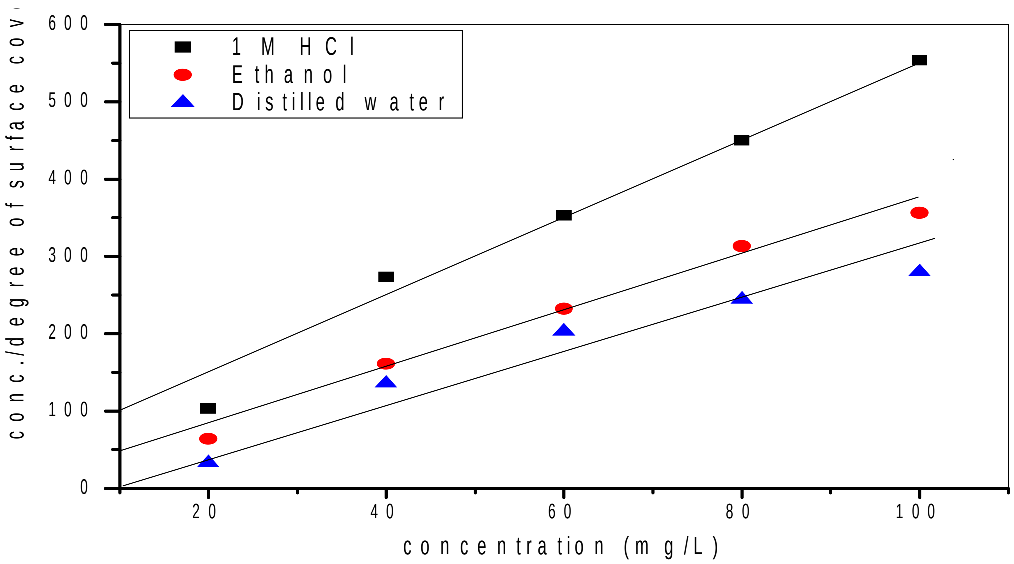

Fig. 6. Langmuir adsorption isotherm at 333 K for 1 M HCl, ethanol and distilled water *Ocimum gratissimum* extracts

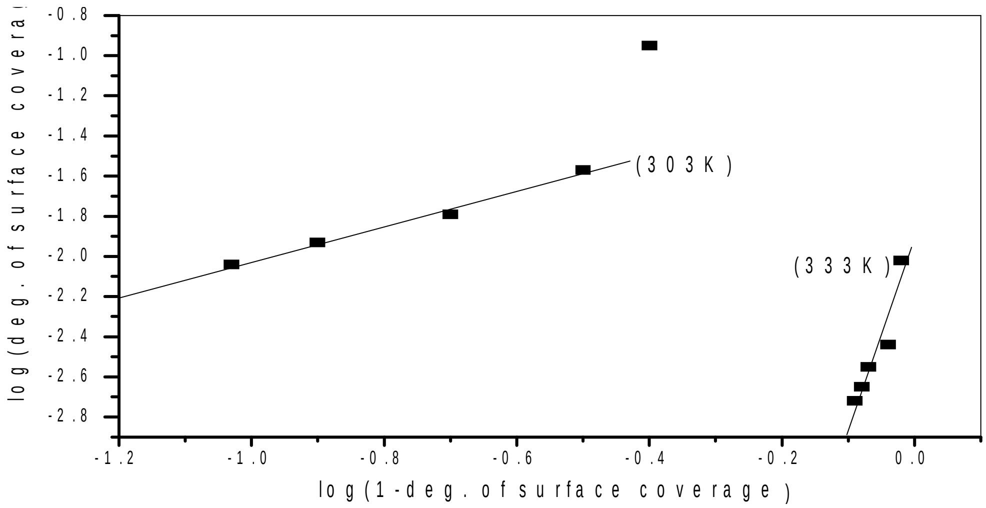

Fig. 7. Flory-Huggins adsorption isotherm at 303 K and 333 K for 1M HCl *Ocimum gratissimum* extract.

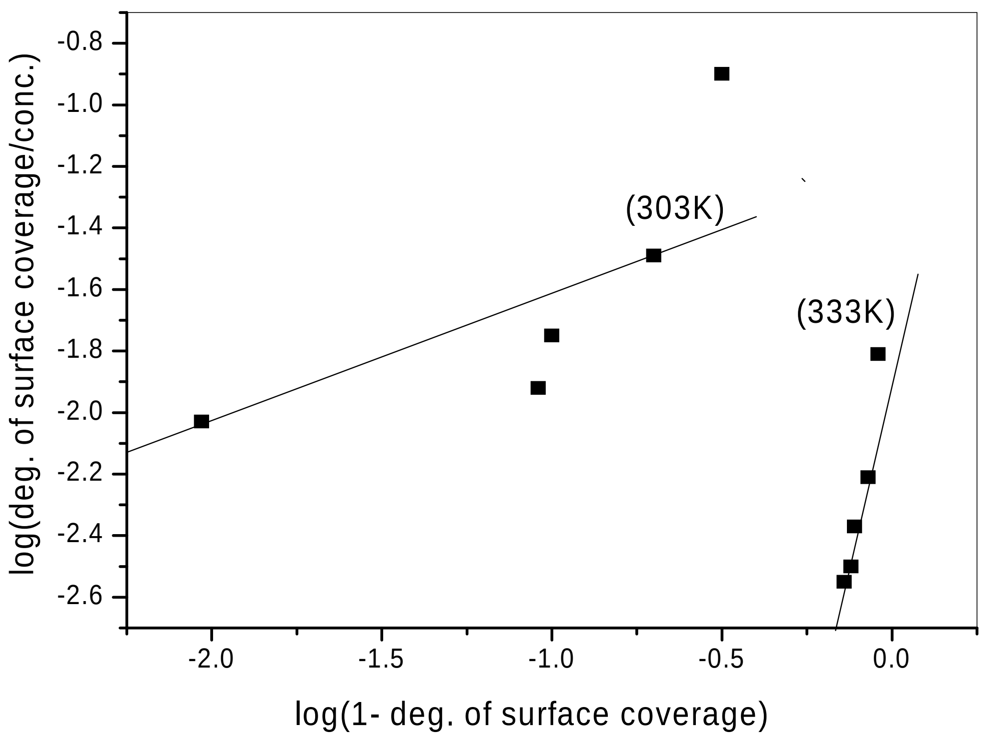

Fig. 8. Flory-Huggins adsorption isotherm at 303 K and 333 K for ethanol *Ocimum gratissimum* extract.

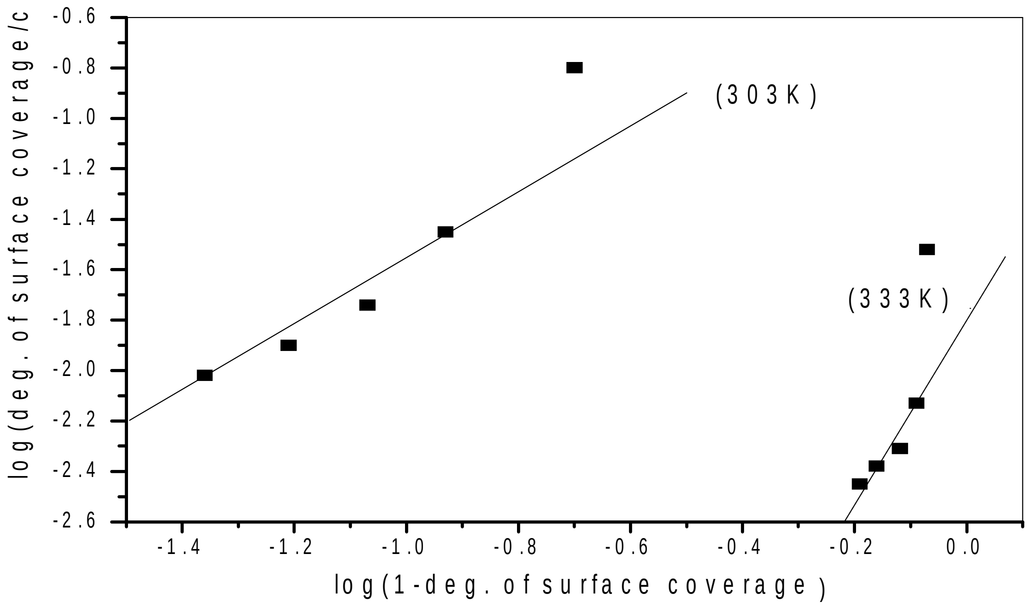

Fig. 9. Flory-Huggins adsorption isotherm at 303 K and 333 K for distilled water *Ocimum gratissimum* extract.

3.4 Effect of Temperature

The influence of temperature on weight loss of Al sample in 1 M HCl in presence and absence of different concentrations of inhibitor has been depicted in figs. 1 and 2. The increase in temperature decreases inhibition efficiency and degree of surface coverage. The reasons for this observation have been explained earlier in this study. In order to calculate apparent activation energy, E_a, for corrosion reaction of Al in absence and presence of various concentrations of inhibitor, the Arrhenius equation was used:

$$\log (\ell_2/\ell_1) = E_a / 2.303R \, [1/T_1 - 1/T_2] \quad (8)$$

Where ℓ_1 and ℓ_2 are corrosion rates at temperatures T_1 and T_2, respectively, and R is gas constant.

The heat of adsorption (Q_{ads}) of inhibitor on surface of Al was evaluated using equation (9) (Bhajiwala and Vashi, 2001).

$$Q_{ads} = 2.303R \, [\log (\theta_2/1-\theta_2) - \log (\theta_1/1-\theta_1)] \times T_1T_2/T_2-T_1 \quad (9)$$

Where θ_1 and θ_2 are degree of surface coverage at temperatures T_1 and T_2. The calculated values of apparent activation energy E_a, and heat of adsorption, Q_{ads} of different inhibitor extracts are shown in Tables 3, 4 and 5, respectively.

Table 3. Values of thermodynamic parameters for adsorption of distilled water extract of *Ocimum gratissimum* on surface of aluminium in 1 M HCl

Concentration of *Ocimum gratissimum* (mg /l)	Activation energy, E_a (KJ/mol)	Heat of adsorption Q_{ads} KJ/mol)	Free energy change ΔG_{ads} (KJ/mol) of adsorption at 303k	Free energy change, ΔG_{ads} (KJ/mol) of adsorption at 333k
Blank	5.24	-	-	-
20.00	45.02	-86.68	-13.58	-5.77
40.00	58.64	-97.72	-15.17	-6.36
60.00	65.82	-97.89	-16.12	-7.30
80.00	72.18	-98.82	-16.96	-8.06
100.00	79.90	-102.90	-17.91	-8.64

Table 4. Values of thermodynamic parameters for adsorption of ethanol extract of *Ocimum gratissimum* on surface of aluminium in 1 M HCl

Concentration of *Ocimum gratissimum* (mg /l)	Activation energy, E_a (KJ/mol)	Heat of adsorption Q_{ads} (KJ/mol)	Free energy Change ΔG_{ads} (KJ/mol) of adsorption at 303k	Free energy change, ΔG_{ads} (KJ/mol) of adsorption at 333k
Blank	5.24	-	-	-
20.00	35.12	-71.22	-12.09	-3.90
40.00	46.02	-87.25	-13.71	-5.85
60.00	59.58	-94.50	-15.38	-6.86
80.00	63.99	-96.60	-15.91	-7.21
100.00	72.93	-102.04	-16.94	-7.75

Table 5. Values of thermodynamic parameters for adsorption of 1 M HCl extract of *Ocimum gratissimum* on surface of aluminium in 1 M HCl

Concentration of *Ocimum gratissimum* (mg /l)	Activation energy, E_a (KJ/mol)	Heat of adsorption Q_{ads} (KJ/mol)	Free energy Change ΔG_{ads} (KJ/mol) of adsorption at 303k	Free energy change, ΔG_{ads} (KJ/mol) OF adsorption at 333k
Black	5.24	-	-	-
20.00	26.60	-84.04	-10.72	-2.61
40.00	33.34	-84.06	-11.89	-4.34
60.00	46.60	-90.07	-13.72	-5.58
80.00	57.40	-90.41	-14.98	-6.07
100.00	65.25	-98.95	-15.86	-6.48

The results shown in tables 3, 4 and 5 of different inhibitor extract indicates that apparent activation energy E_a, increases as inhibitor concentration increases. Activation energy of distilled water extract ranged between 45.02 to 79.90 KJ/mol. While the activation energies of ethanol and 1 M HCl extracts of inhibitor ranged between 35.12 to 72.93 KJ/mol and 26.60 to 65.25 KJ/mol, respectively. Since corrosion primarily occurs at surface sites free of adsorbed inhibitor, the higher E_a values in inhibited solutions imply that extracts mechanically screen the active sites of Al surface thereby decreasing the surface area available for corrosion. The order of apparent activation energy, E_a of different extracts was in order: distilled H_2O > C_2H_5OH> 1 M HCl.

The heat of adsorption, Q_{ads} were all negative and ranged from – 86.68 to - 102.90 KJ/mol for distilled water extract, while ethanol and 1 M HCl extracts ranged from – 71.22 to -102.04 KJ/mol and -- 84.04 to -- 98.95 KJ/mol, respectively. The negative Q_{ads} values indicate that adsorption and hence inhibition efficiency decreases as temperature increases, supporting the earlier proposed physisorption mechanism (Bhajiwala and Vashi, 2001).

4. CONCLUSION

This study indicates that extract of *Ocimum gratissimum* inhibits Al surface in presence of 1 M HCl. The corrosion process is inhibited by adsorption of extracts on aluminium surface. Inhibition efficiency increase with increase in inhibitor concentrations. The result of the analysis shows that inhibition efficiency and degree of surface coverage decreases as temperature increases. Activation energies of different extracts increase as concentration of inhibitor increases. The negative values of ΔG_{ads} shows that adsorption of inhibitor on surface of aluminium is spontaneous. The adsorption behaviour was approximated by Langmuir and Flory – Huggins isotherms. The trend of inhibition efficiency of different extracts was in order: Distilled H_2O > C_2H_5OH>1M HCl.

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COMPETING INTERESTS

Authors have declared that no competing interest exist

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