ISSN: 2578-7365

Research Article

Journal of Chemistry: Education Research and Practice

Naturally Occurring Exudates Gums as Ecofriendly Inhibitors for Mild Steel Corrosion in Acidic Medium

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Submitted: 05 Nov 2021; **Accepted:** 12 Nov 2021; **Published:** 14 Dec 2021

Citation: *Onukwube ND, Odoemelam SA, Eddy NO (2021) Naturally Occurring Exudates Gums as Ecofriendly Inhibitors for Mild Steel Corrosion in Acidic Medium. J Chem Edu Res Prac 5(2): 149-155.*

Abstract

The corrosion inhibition potentials of gum exudates from Daniella oliveri (DO) and Commiphora Africana (CA) for the corrosion of mild steel in H_2SO_4 have been studied using weight loss and thermometric methods at 303 and 333K. Results show that the exudates gum actually reduced the rates of corrosion of mild steel. Increase in the concentrations of the exudate's gums increased their percentage inhibition efficiencies. Corrosion rate was found to increase with increase in temperature in the presence and absence of the gum exudates, though the corrosion rate was slower in the presence of the exudates gums. Both DO and CA exudates gums were found to obey Temkin and Langmuir adsorption models at all concentrations and temperatures studied. Physical adsorption mechanism was proposed from the adsorption parameters. Kinetic and thermodynamic parameters revealed that the adsorption process is spontaneous, exothermic and no significant difference was found between the inhibition efficiencies of DO and CA.

Keywords: Corrosion Inhibition, Adsorption Model, Mild Steel, Daniella Oliveri, Commiphora Africana

Introduction

Corrosion, a naturally occurring phenomenon is commonly defined as deterioration of metal surfaces caused by its reaction with the surrounding environmental conditions. Corrosion as a major destructive process affects the performance of metallic materials in applications in many construction sectors. The consequences of corrosion have become quite catastrophic and are considered a serious problem in industries, constructions and civil services. The cost of restoring damages due to corrosion has become so alarming that preventive approaches have become undoubtedly the best. For example, the cost of restoring damages due to corrosion in the US was estimated at $170 billion per year in 1999. In the UK, the estimate of annual cost of losses due to corrosion was put at four million dollars ($4 million) as at 1971. The cost due to corrosion was also estimated at 137.9 billion per year in 1998 [1].

With the increase in the advancement of technology and industrialization, these costs are expected to also increase. However, since prevention is always better and cheaper than cure, inhibitors which are substances which when added in small concentration to an environment, effectively reduce the corrosion rate of a metal exposed to them are applied to prevent or minimize corrosion. Most of the organic compounds with corrosion inhibition potentials are not only expensive but also poses health and environmental hazards prompting the search for their replacement [2].

Plants have been recognized as sources of naturally occurring compounds that are generally referred to as 'green' compounds, some with rather complex molecular structures and having a variety of physical, chemical and biological properties [3]. There has been a growing trend in the use of natural products as corrosion inhibitors for metals in various corrosive media. These substances are otherwise tagged 'green inhibitors' and their wide application is attributed to the fact that they are cheap, ecologically friendly and pose no threat to the environment. In addition, they are readily available and renewable source of materials. Literature abound on the adsorption and corrosion inhibition potentials of many exudates' gums eg. Gum Arabic, Acacia seyal var seyal, Acacia drepanolobium and Acacia Senegal, Raphia hookeri, Guar gum, Anogessus leocarpus gum [3-9].

More plants materials have continued to be tested for their corrosion inhibition potentials due to the encouraging results obtained from the researches above. In this paper, gravimetric and thermometric techniques are applied to study and compare the abilities of exudates gums from *Daniella oliveri* (DO) and *Commiphora*

Africana (CA) to inhibit the corrosion of Mild Steel in H_2SO_4 in furtherance of our quest to explore naturally occurring substances (green inhibitors) as corrosion inhibitors for metals in different aqueous media.

Materials and Methods

Materials

The study was carried out using mild steel sheets of composition (wt%) Mn (0.6), P (0.36), C (0.15), Si (0.03) and the rest iron. The sheet was mechanically cut into different coupons, each of dimensions 4 x 3cm. Each coupon was degreased by washing with ethanol, rinsed with acetone and allowed to dry in air before they were preserved in desiccators. All reagents used for the study were Analar grade and double distilled water was used for the preparations.

Sampling

Samples of the gums (*Daniella oliveri* (DO) and *Commiphora Africana* (CA)) were collected from mature stems of the plants during dry season, by breaking the outer bark of the tree trunk, a process known as tapping. The bark was carefully pealed in each case and a cut made. The cut was extended to form a wound of about 3cm wide and 0.5- 1cm deep. The gum samples were slowly collected along the length of the wounded trunk.

Purification of the Gum Samples

The purification process involved the dissolution of the gum in cold distilled water. This was followed by filtration and centrifugation which left a small quantity of dense gel. The supernatant liquor was then separated and acidified with dilute tetraoxosulphate (VI) acid. The gum was allowed to precipitate with the addition of ethyl alcohol. The precipitates were removed by centrifugation and were dried after being washed with alcohol and ether. Different concentrations of the inhibitors were prepared by dissolving 0.1g, 0.2g, 0.3g, 0.4g and 0.5g of the gum extracts in 1L of 2.5 M H_2SO_4 for thermometric analysis. The same portions of the extracts were also dissolved in 0.1 M H_2SO_4 for use in gravimetric analysis.

Corrosion Inhibition Studies

Gravimetric Method

Gravimetric study using DO and CA gums exudates was done by dipping a previously weighed metal (mild steel) coupon into 20 ml of the test solution maintained at 303 and 333 K in a thermo stated bath. The weight loss was determined by retrieving the coupons at 24 hours intervals progressively for 168 hours (7 days). Prior to measurement, each coupon was immersed in a solution of 20 % sodium hydroxide containing 200 g/l of zinc dust to terminate the corrosion reaction and then rinsed in acetone before drying. The difference in weight was taken as the weight loss of the mild steel. From the average weight loss (mean of three replicate analyses) results, the inhibition efficiency (%I) of the inhibitor, the degree of surface coverage (θ) and the corrosion rate of mild steel (CR) were calculated using the following equations,

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

$$\Theta = 1 - W_1/W_2 \quad (2)$$

$$CR\ (gh^{-1}cm^{-2}) = \Delta W/At \quad (3)$$

Where W_1 and W_2 are the weight losses (g) for mild steel in the presence and absence of the inhibitor in H_2SO_4 solution, ϴ is the degree of surface coverage of the inhibitor, A is the area of the mild steel coupon (in cm^2), t is the period of immersion (in hours) and W is the difference in weight before and after immersion.

Thermometric Method

Thermometric analysis was equally carried out also as reported in literature [10, 11].

From the average of three replicate measurements obtained for the rise in temperature of the system per minute, the reaction number (RN) was calculated using Equation (4)

$$RN(°C/Min) = \frac{T_m - T_i}{t} \quad \dots \quad 4$$

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, using equation (5)

$$\%I = \frac{RN_{aq} - RN_{wi}}{RN_{aq}} \quad \dots \quad 5$$

Where RNaq is the reaction number in the absence of inhibitors (blank solution) and RNwi is the reaction number for 2.5 M H_2SO_4 containing studied inhibitor.

Results and Discussion

Weight Loss, Corrosion Rate and Inhibition Efficiency

Measurements of weight loss of mild steel in acidic medium in the presence and absence of DO and CA exudates gums of various concentrations were made after 168 hrs of immersion at temperature range of 303 – 333 K. The results obtained are as expressed in Fig. 1 and Table 1. Fig1, which is the variations of weight loss against time for mild steel in 0.1 M H_2SO_4 containing (a) DO and (b) CA exudates gums at 303 K clearly shows a reduction in weight loss of the metal coupons in the presence of these inhibitors compared to the uninhibited solution (blank). The figure further revealed that the loss in weight of the coupons maintains an inverse proportional relationship with the concentration of the inhibitors. At higher temperature, similar trend was observed although with greater values of weight loss. Table 1 which is the calculated values of corrosion rate in (mm/yr) for mild steel in 0.1 M H_2SO_4 in the absence and presence of DO and CA exudates gums at various concentrations show a reduction in the corrosion rates of the solutions with the exudates compared to the blank (H_2SO_4). From the table, it is equally obvious that the corrosion rate increases with increase in temperature and decreases as the concentration of the exudates gums increases.

Thus, the extent of corrosion inhibition depends on the quantity of DO and CA exudates gums present. The exudates gums derived from DO and CA can actually be said to have inhibited the rate of corrosion of mild steel in the acidic medium due to the reduction in weight loss and corrosion rates in the presence of these additives. The inhibition efficiencies of DO and CA as also presented in Table 1 show an increase with increase in the concentrations of exudates. Table 1 equally shows that inhibition efficiencies of

DO and CA exudates gums decrease with increase in temperature, thus physical adsorption mechanism is suggested for both inhibitors. Preliminary phytochemical screening of DO and CA revealed the presence of Flavonoids, Tannins, Anthran h quinone, Cardiac glycoside, Alkaloids, Triterpenoids Saponins and reducing sugars [12, 13, 14].

The inhibitive effect of these exudates gums is attributed to the presence of some of these phytochemicals. These compounds contain oxygen and nitrogen atoms which are the centers of adsorption. Therefore, the assumption that these exudates gums derive their inhibitive action through adsorption of these phytochemical component molecules on the metal surface is germane. A barrier is said to be created between the metal and the corrosive medium by this adsorption process, leading to inhibition of corrosion. Thus, inhibition efficiency increases as the surface area of the metal covered by the adsorbed molecule increases. The two inhibitors DO and CA can be said to posses relatively the same inhibition potentials, since there is no significant difference in percentage inhibition efficiency between the two inhibitors ($P \leq 0.05$).

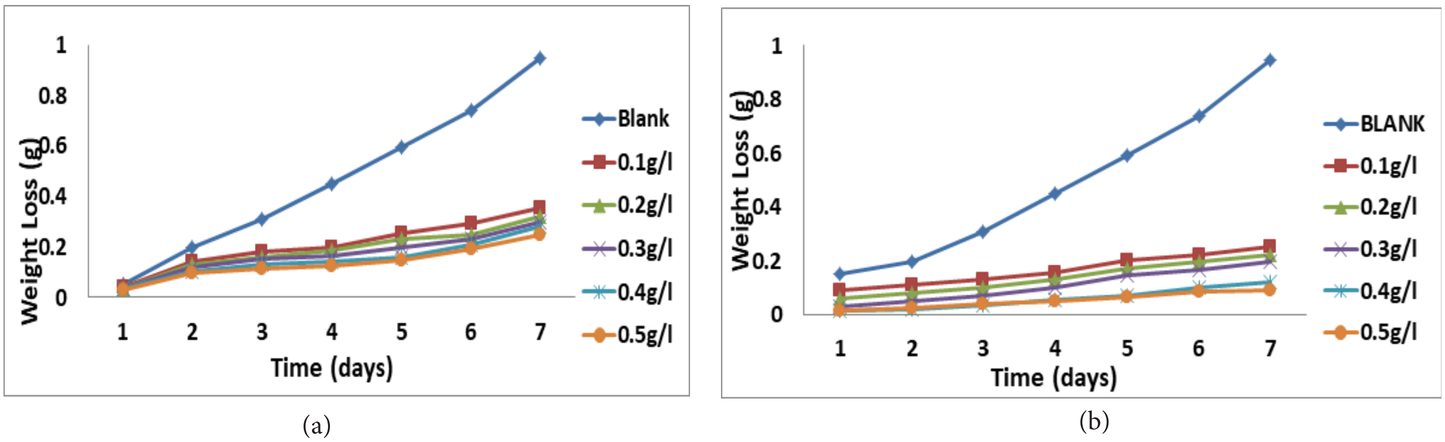


Figure 1: Variations of Weight Loss of Mild Steel with Time for The Corrosion of Mild Steel in 0.1 M H_2SO_4 Containing Various Concentrations of (a) DO and (b) CO Exudates Gums at 303 K.

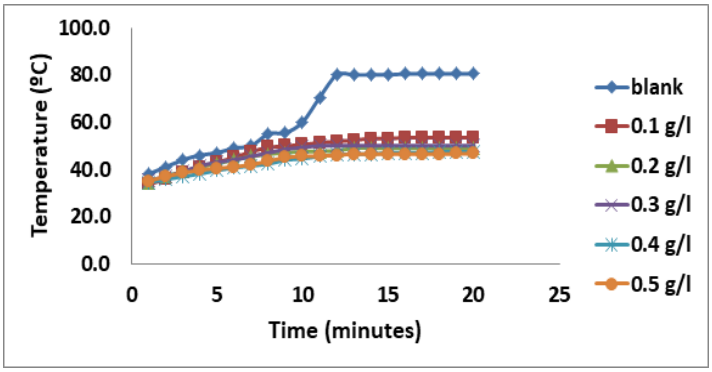
Table 1: Calculated Values of Corrosion Rate (mm/yr), Inhibition Efficiency (% I), and Degree of Surface Coverage (Ɵ), for Exudates Gums from Weight Loss Data

Concentration of exudates (g/L)	Corrosion rate (mm/yr)10-5, inhibition efficiency (% I) and degree of surface coverage (Ɵ)					
	303 K			333 K		
DO Exudate Blank	47.12			55.56		
0.1	16.67[a]	(64.56)[b]	[0.64][c]	21.02	(55.09)	[0.55]
0.2	16.32	(65.30)	[0.65]	20.78	(57.31)	[0.57]
0.3	15.87	(80.90)	[0.80]	19.56	(64.21)	[0.64]
0.4	15.21	(85.44)	[0.85]	18.79	(67.43)	[0.67]
0.5	14.67	(90.01)	[0.90]	17.32	(70.45)	[0.70]
Total		**(386.21)**			**(314.49)**	
CA Exudate Blank						
0.1	18.92[a]	(59.71)[b]	[0.59][c]	23.58	(53.22)	[0.53]
0.2	15.31	(61.62)	[0.61]	21.08	(56.01)	[0.56]
0.3	14.12	(73.21)	[0.73]	18.54	(61.36)	[0.61]
0.4	13.61	(86.92)	[0.86]	16.21	(65.14)	[0.65]
0.5	13.02	(90.08)	[0.90]	15.93	(70.28)	[0.70]
Total		**(371.54)**			**(306.01)**	

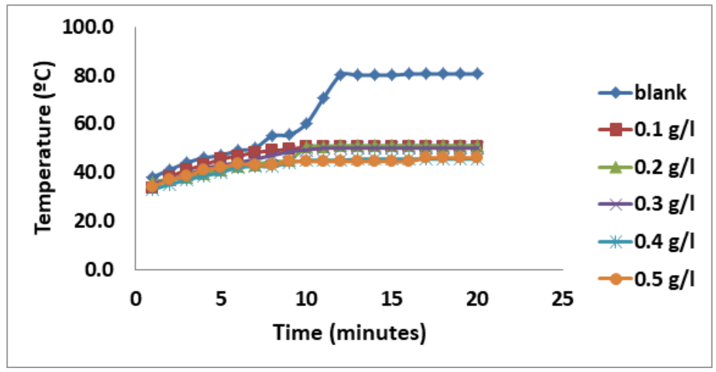
(a) Corrosion rate obtained from equation (1); (b) Inhibition efficiency (%I) obtained using equation (2); (c) Degree of surface coverage (Ɵ) obtained using equation (3); DO = *Daniella oliveri*; CA =*Commiphora Africana.*

Thermometric Studies

Thermometric analysis has been widely applied and has been considered valuable in studying corrosion patterns of metals and alloys in various corroding media. The inhibition efficiency of many surface-active agents has also been evaluated using this technique. Results obtained from thermometric method compared favorably with weight loss method, which is a well-established method. Fig .2 shows variations of temperature with time due to the corrosion reaction of mild steel in 2.5 M H_2SO_4 solution in the presence and absence of DO and CA exudates gums. The plot shows a gradual dissolution of the mild steel as shown in the slight increase in temperature in the first few minutes of immersion of the mild steel coupons in the test solution. It could be observed that there was an instant destruction of the pre-immersion oxide film and so there was no incubation period. The temperature of the blank system rose sharply, while those of the inhibited systems maintained a gradual and systematic rise due to exothermic corrosion reaction until a maximum value TM, when the change in temperature became constant with time. This could be attributed to the systems establishing a dynamic equilibrium. The figure equally revealed that the maximum temperature attained decreased with the introduction of DO and CA exudates gums, indicating that the two gums inhibit the corrosion of mild steel in acidic medium, probably by adsorption on the metal surface. The degree of coverage of the metal by the adsorbed molecules determines the extent of inhibition [15, 16, 17]. Adsorption is strongest at higher concentration of exudates as shown by the decrease in maximum temperature (TM) attained.



(a)



(b)

Figure 2: Variations of Temperature with Time for The Corrosion of Mild Steel in 2.5 M H_2SO_4 Containing Various Concentrations of (a) DO Exudates and (b) CA Exudates.

The calculated values of reaction number (RN) and inhibition efficiency (percentage reduction in reaction number) for the various systems are shown in Table 2. The table shows a reduction in the reaction number in the presence of the exudates compared to the blank solution. The inhibition efficiency equally increases with increase in the concentrations of the two exudates under study. However, values of inhibition efficiencies of both DO and CA were not significantly different, thus DO and CA can be said to have the same inhibition potentials. This assertion is equally corroborated by weight loss measurements.

Table 2: Values of Reaction Number and Percentage Inhibition Efficiency for Corrosion of Mild Steel in 2.5 M H_2SO_4 Containing Exudates Gums from DO and CA from thermometric method

Concentrations of exudates (g/L)	Reaction number (RN) ($^0Cmin^{-1}$)	% Inhibition efficiency
DO Exudate Blank	2.13	
0.1	1.00	52.94
0.2	0.75	64.71
0.3	0.73	65.65
0.4	0.65	69.41
0.5	0.55	72.71
Total		**325.42**
CA Exudate		
0.1	0.84	64.57
0.2	0.74	65.18
0.3	0.63	68.71
0.4	0.56	70.59
0.5	0.70	71.76
Total		**340.81**

Thermodynamic and Adsorption Considerations

The mechanisms of organo-electrochemical reactions are better understood using adsorption isotherms [18]. The degree of surface coverage (ϴ) is almost indispensable in discussing isotherms. The degree of surface coverage values obtained from weight loss measurements using equation (2) for DO and CA exudates gums were fitted into different adsorption models, Langmuir and Temkin adsorption models were found to suit the adsorption mechanisms of the two inhibitors [19].

Temkin adsorption isotherm model has the form: -

exp(- 2aϴ) = KC … (6)

$$\exp(-2a\Theta) = KC \quad \ldots \quad (6)$$

Where C is the concentration of the inhibitor in the bulk electrolyte, K is the adsorption equilibrium constant, ϴ is the degree of surface coverage of the inhibitor and a is the interaction parameter. If equation 6 is rearranged and logarithm taken, equation 7is obtained

$$\theta = \frac{-\mathrm{LnK}}{2a} - \frac{\mathrm{LnC}}{2a} \quad \ldots \quad 7$$

From equation 7, a plot of θ versus log C should be linear, if assumptions of Temkin isotherm are valid. Fig 3 shows the plots of θ against logC from equation 7. The plots were linear, showing that Temkin adsorption isotherm applies to the adsorption of DO and CA on the surface of mild steel.

For Langmuir adsorption Isotherm, the following assumptions are established.

$$C/\theta = 1/K + C \quad \ldots \quad (8)$$

$$\mathrm{Log}\,(C/\theta) = \mathrm{LogC} - \mathrm{LogK} \quad \ldots \quad (9)$$

A plot of Log (C/θ) versus LogC from equation 9 yielded straight lines as shown in fig 4 with intercept equal to -Logk.

Values of the free energy of adsorption (ΔG^o_{ads}) of DO and CA on the surface of mild steel were calculated using equation (10) and are as recorded in Table 3.

$$\Delta G^o_{ads} = -2.303RT\log(55.5K) \quad \ldots \quad (10)$$

The values of free energy are negatively less than the threshold value of -40 $kJmol^{-1}$ required for chemical adsorption, therefore, the adsorption of exudates gums of DO and CA on mild steel surface is spontaneous and support the mechanism of physical adsorption.

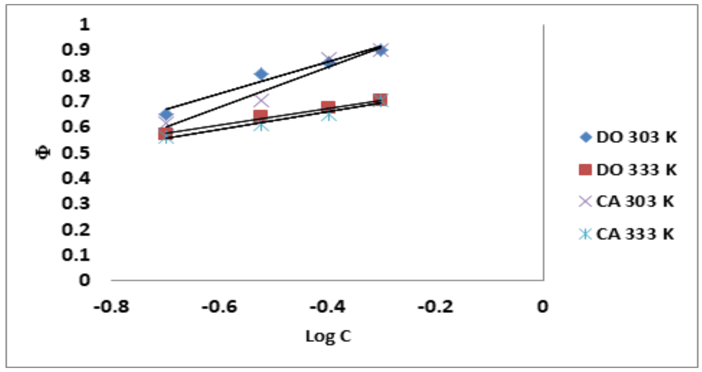


Figure 3: Temkin Plots for The Adsorption of DO and CA on Mild Steel Surface

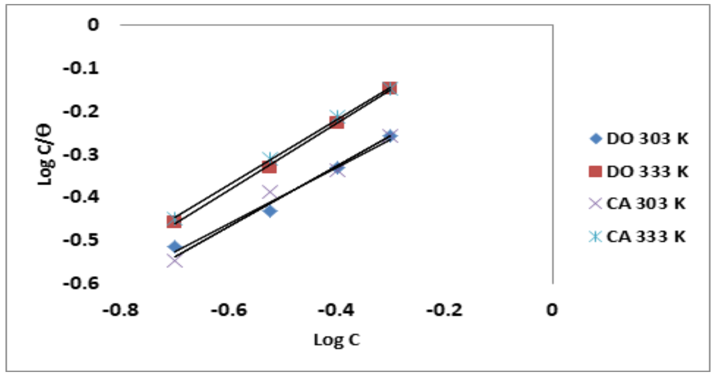


Figure 4: Langmuir Plots for The Adsorption of DO and CA on Mild Steel Surface

From Figs 3 and 4 above, linear plots were obtained for DO and CA exudates gums with very good correlation coefficients (R^2), indicating that the experimental data fit well into Temkin and Langmuir adsorption isotherms. Values of Temkin and Langmuir adsorption parameters deduced from the slopes and intercepts of the plots in Figs 3 and 4 are presented in Tables 3 and 4 respectively. The values of molecular interaction parameter 'a' from Table 3 are negative in all cases for both DO and CA exudates, thus repulsion could be said to exist in the adsorption layer [20]. The strength between the adsorbate and adsorbent is always denoted by K. Large values of K imply more efficient adsorption and hence better inhibition efficiency. The decrease in the value of K with increasing temperature as observed in Table 4 suggests that the inhibitors are physically adsorbed on the surface of mild steel and are better inhibitors at lower temperature [21].

Table 3: Temkin Adsorption Parameters for The Adsorption of DO and CA on Mild Steel Surface

	Temp	Log K	Slope	A	ΔG^o_{ads} (KJ/mol)	R^2
DO	303K	1.9251	0.5714	-2.0152	-21.21	0.958
	333K	1.4001	0.2857	-4.0305	-20.05	0.991
	Temp	**Log K**	**Slope**	**A**	ΔG^o_{ads} **(KJ/mol)**	R^2
CA	303K	1.4637	0.7857	-1.4656	-18.62	0.958
	333K	2.5462	0.3142	-3.6649	-27.35	0.982

Table 4: Langmuir Adsorption Parameters for The Adsorption of DO and CA on Mild Steel Surface

	Temp	Log K	Slope	R^2
DO	303K	0.08	0.5833	0.983
	333K	-0.08	0.7286	0.998
	Temp	**Log K**	**Slope**	R^2
CA	303K	0.05	0.7142	0.980
	333K	-0.08	0.7757	0.999

Table 5: Activation Energy, Ea (kJ/mol) for Mild Steel Corrosion in The Presence of DO and CA Exudates in 0.1 M H_2SO_4

Conc. of Blank/Inhibitor mol/dm³	Ea (KJ/mol)		
	Blank	DO	CA
0.1	29.29	70.41	61.03
0.2	25.67	62.92	88.65
0.3	23.52	63.23	79.44
0.4	21.31	61.44	48.44

Ea values for both blank solutions and solutions containing different concentrations of DO and CA were calculated using the modified Arrhenius equation (11).

$$\log\frac{CR_2}{CR_1} = \frac{E_a}{2.303R}\left[\frac{1}{T_1} - \frac{1}{T_2}\right] \quad \dots \quad 11$$

Where Ea is the activation energy, CR_1 and CR_2 are values of corrosion rates at temperatures T_1 and T_2 and R is the universal gas constant. Calculated values of Ea are shown in Table 5.

It is seen from the table, that Ea values are higher in the presence of the inhibitors compared to the blank. The higher values of the activation energy of the process in the inhibitors' presence when compared to that in their absence is attributed to physisorption, while the opposite is the case with chemisorption's [22, 23]. Also, the higher values of activation energy in the presence of the inhibitors confirm that corrosion has actually been impeded, since greater energy now needs to be attained for corrosion reaction to occur.

Conclusion

1. DO and CA exudates gums are good corrosion inhibitors for the inhibition of mild steel corrosion in acidic medium, with both of them having relatively the same inhibition potentials.
2. The inhibition efficiency of DO and CA maintain direct proportional relationship with the concentrations of the inhibitors and inversely proportional to temperature.
3. The values of ΔG^O_{ads} are negative, suggesting strong adsorption of the inhibitors' molecules on the mild steel surface.
4. Exudates gums from DO and CA were found to obey both Temkin and Langmuir adsorption models.
5. Thermodynamic parameters revealed that the adsorption process for DO and CA is spontaneous, exothermic and support physical mechanism.

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