

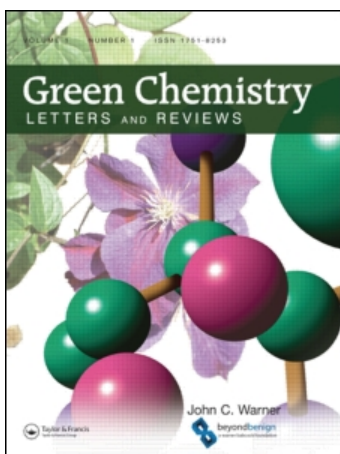
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Nnabuk Eddy Okon^a

^a Department of Chemistry, Ahmadu Bello University, Zaria, Kaduna State, Nigeria

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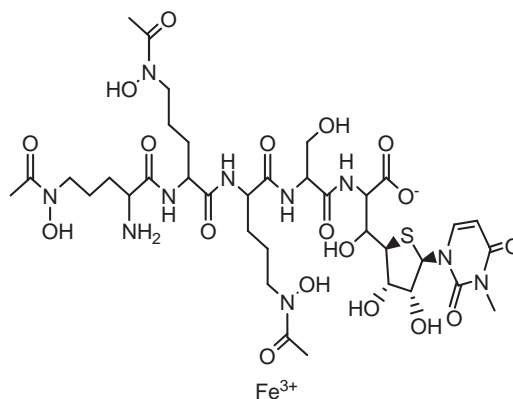
Fermentation product of *Streptomyces griseus* (albomycin) as a green inhibitor for the corrosion of zinc in H₂SO₄

Nnabuk Okon Eddy*

Department of Chemistry, Ahmadu Bello University, Kaduna State, Zaria +234, Nigeria

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Inhibitive and adsorption properties of albomycin as a green inhibitor for the corrosion of zinc in H₂SO₄ solutions were studied using weight loss and hydrogen evolution methods. The results obtained, indicate that albomycin is a good adsorption inhibitor for the corrosion of zinc in H₂SO₄ solution. The inhibition potentials of albomycin for the corrosion of zinc in H₂SO₄ solutions are attributed to the adsorption of the inhibitor on the surface of zinc and its inhibition efficiency increases with increase in the concentration of the inhibitor but decreases with increasing temperature. The range obtained for the values of activation energy and free energy of adsorption were within the limit expected for the mechanism of physical adsorption. Also, the adsorption of albomycin on zinc surface was spontaneous, exothermic, and supported the Langmuir adsorption model.



Optimized structure of 2-[[2-[[5-[acetyl(hydroxy)amino]-2-[[5-[acetyl(hydroxy)amino]-2-[[5-[acetyl(hydroxy)amino]-2-aminopentanoyl]amino] pentanoyl] amino] pentanoyl]amino]-3-hydroxypropanoyl]amino]-3-[(2R,3S,4R,5R)-3,4-dihydroxy-5-(3-methyl-2,4-dioxypyrimidin-1-yl)thiolan-2-yl]-3-hydroxypropanoic acid; iron(3+) (albomycin)

Keywords: corrosion; zinc; green inhibitor; albomycin

Introduction

Corrosion inhibitors are needed in the oil, fertilizer, and metallurgical industries because when metals (such as zinc, mild steel, aluminium, etc.) come in contact with aggressive solutions (such as acids, bases, and salts), the rate at which they corrode is reduced by the adsorption of these inhibitor on their surfaces (1–6).

Numerous studies have been carried out on the inhibition of the corrosion of metals in acidic and

alkaline media (7–10). Most of these studies are centered on the use of organic compounds as inhibitors. These organic compounds have suitable functional groups, conjugated systems, or centers for pi-bond, which facilitate the adsorption of the inhibitor on the metal surface. It has also been established that the presence of aromatic groups and hetero atoms also enhance the adsorption potentials of these groups of inhibitors (11,12). Based on their structures, some of the inhibitors that have been used

*Email: nabukeddy@yahoo.com

for the corrosion of zinc are quinoline, aniline, ephedrine, narcotine, brucine, stryctuine, piperazine, caffeine, barbitone, sparfloracin, some surfactant, benzotriazole, norfloxacin, thiourea, quinolones, azithromycin, and pyridine derivatives (13–17). Attempts have also been made to use some naturally occurring substances as inhibitors for the corrosion of metals including zinc (18–20). Inhibitors from this source are called green corrosion inhibitors because they are environmentally friendly and do not contain heavy metals or other toxic compounds (21–39). However, one of the major problems associated with the use of natural products as corrosion inhibitors, is the isolation of the active constituents responsible for the inhibition process. In the light of this, the present study seeks to investigate the possibility of using the fermentation product of *Streptomyces griseus* (albomycin) as an inhibitor for the corrosion of zinc in 0.01–0.04 M H_2SO_4 . Albomycin is an antibiotics, often used for the treatments of some infections. It can be produced from *S. griseus* in concentrations of approximately 1 mg/l. The production depends on the phosphate, iron, and ornithine concentrations in the medium. In optimized conditions, the production of albomycin could be increased to 25 mg/l in a feed-batch fermentation. Isolation and purification could be achieved by reversed-phase, size-exclusion chromatography, and preparative high-performance liquid chromatography (40). The optimized structure of albomycin is as shown below. From the structure, it is expected that albomycin may be a good inhibitor for the corrosion of zinc in H_2SO_4 solutions because it has hetero atoms in its structure and contains some functional groups that may facilitate its electron donating ability. Therefore, the objective of the present study is to investigate the adsorption and inhibitive properties of albomycin for the corrosion of zinc in H_2SO_4 .

Results and discussions

Effect of concentration of H_2SO_4 /albomycin

Figure 1 shows the variation of weight loss of zinc with time for the corrosion of zinc in 0.01 M H_2SO_4 containing various concentrations of albomycin at 303 K. From Figure 1, it is evident that the weight loss of zinc increases with increase in the period of contact but decreases with increase in the concentration of albomycin. Also weight loss of zinc for the blank solution is higher than those obtained for solutions of H_2SO_4 containing various concentrations of albomycin. Therefore, albomycin is an adsorption inhibitor for the corrosion of zinc in H_2SO_4 solutions. At higher temperatures and concentrations of H_2SO_4 (plots not shown), weight loss of zinc was also found to increase

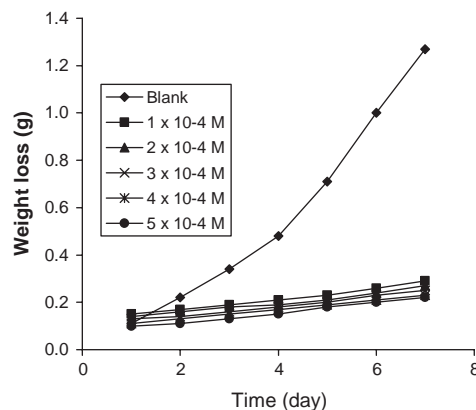


Figure 1. Variation of weight loss with time for the corrosion of zinc in 0.01 M tetraoxosulphate (VI) acid containing various concentration of albomycin at 303 K.

with increase in temperature but decreases with increase in the concentration of H_2SO_4 . This indicates that the rate of corrosion of zinc increases with increase in temperature and with increasing concentration of H_2SO_4 . To further assert the above findings, the inhibition efficiencies of various concentrations of albomycin in H_2SO_4 solutions and the corrosion rates of zinc in similar media were calculated. The results of the calculations are presented in Table 1. From the results obtained, the corrosion rates of zinc were found to increase with increasing temperature and with increase in the concentration of H_2SO_4 but decreases with increase in the concentrations of albomycin. Hence, albomycin retarded the corrosion of zinc in H_2SO_4 solutions. On the other hand, at a given concentration of H_2SO_4 , the inhibition efficiency of albomycin increases with increasing concentration but decreases with increasing temperature implying that albomycin is an adsorption inhibitor for the corrosion of zinc in H_2SO_4 and that the adsorption of albomycin on zinc surface supports the mechanism of physical adsorption.

Effect of temperature

The Arrhenius equation was used to calculate the activation energy for the corrosion of zinc in H_2SO_4 solutions. The concept of Arrhenius theory is provoked by the fact that the corrosion of zinc in H_2SO_4 requires a minimum energy input and that the corrosion reaction can not proceed until the energy barrier is overcome. From the Arrhenius equation, if the corrosion rate of zinc varies with temperature, then we have the following relation (41,42):

$$CR = A \exp(-E_a/RT), \quad (1)$$

where CR is the corrosion rate of zinc, A is the Arrhenius or pre-exponential factor, E_a is the activation energy,

Table 1. Corrosion rate (in $\text{gem}^{-2} \text{h}^{-1}$) of zinc and inhibition efficiencies (%) of albomycin in various concentrations of H_2SO_4 .

Systems	Corrosion rate $\times 10^2$			Inhibition efficiency (%)		
	303	313	323	303	313	323
0.01 M H_2SO_4	37.8	38.7	40.8	–	–	–
1×10^{-4} M Alb+0.01 M H_2SO_4	8.63	21.13	13.39	77.17	45.40	67.17
2×10^{-4} M Alb+0.01 M H_2SO_4	8.04	18.75	20.24	78.74	51.55	50.40
3×10^{-4} M Alb+0.01 M H_2SO_4	7.44	17.56	19.35	80.32	54.63	52.59
4×10^{-4} M Alb+0.01 M H_2SO_4	6.85	14.58	16.37	81.89	62.32	59.88
5×10^{-4} M Alb+0.01 M H_2SO_4	6.55	10.12	13.39	82.68	73.85	67.17
0.02 M H_2SO_4	41.7	44.3	46.1	–	–	–
1×10^{-4} M Alb+0.02 M H_2SO_4	11.90	22.02	23.51	71.45	50.28	49.00
2×10^{-4} M Alb+0.02 M H_2SO_4	11.01	19.64	21.43	73.59	55.66	53.52
3×10^{-4} M Alb+0.02 M H_2SO_4	10.12	18.45	20.24	75.73	58.35	56.10
4×10^{-4} M Alb+0.02 M H_2SO_4	9.82	15.48	17.86	76.45	65.07	61.26
5×10^{-4} M Alb+0.02 M H_2SO_4	9.52	12.80	14.88	77.16	71.11	67.72
0.03 M H_2SO_4	43.2	46.4	49.4	–	–	–
1×10^{-4} M Alb+0.03 M H_2SO_4	18.15	26.19	28.27	57.98	43.56	42.77
2×10^{-4} M Alb+0.03 M H_2SO_4	16.07	21.43	23.81	62.80	53.82	51.80
3×10^{-4} M Alb+0.03 M H_2SO_4	14.88	19.64	20.83	65.55	57.67	57.83
4×10^{-4} M Alb+0.03 M H_2SO_4	13.10	17.56	18.75	69.69	62.16	62.04
5×10^{-4} M Alb+0.03 M H_2SO_4	70.38	13.99	16.37	70.30	69.85	66.86
0.04 M H_2SO_4	51.5	58.93	65.48	–	–	–
1×10^{-4} M Alb+0.04 M H_2SO_4	21.13	27.38	29.76	58.97	53.51	54.56
2×10^{-4} M Alb+0.04 M H_2SO_4	18.45	24.70	28.27	64.17	58.06	56.83
3×10^{-4} M Alb+0.04 M H_2SO_4	17.26	23.51	26.79	66.48	60.08	59.11
4×10^{-4} M Alb+0.04 M H_2SO_4	16.67	23.21	25.30	67.64	60.59	61.38
5×10^{-4} M Alb+0.04 M H_2SO_4	14.88	22.32	24.40	71.10	62.10	62.74

R is the gas constant, and T is the temperature. Transforming Equation (1) to a logarithm function, Equation (2) is obtained:

$$\log \text{CR} = \log A - E_a/2.303 RT. \quad (2)$$

Using Equation (2), plots of $\log \text{CR}$ versus $1/T$ were linear as shown in Figure 2 (a–d). Values of E_a calculated from the slopes of lines on the plots are presented in Table 2. The results obtained, indicated that the activation energies are within the limit expected for the mechanism of physical adsorption. This is because, E_a values greater than 80 KJ/mol are usually accepted as an evidence for the mechanism of chemical adsorption, whereas E_a value less than 80 KJ/mol supports the mechanism of physical adsorption. The results also revealed that the activation energies tend to decrease with increase in the concentration of albomycin as opposed to the inhibition efficiencies which increased with increase in the concentration of the inhibitor. Therefore, the adsorption of albomycin on zinc surface is diffusion controlled. Consequently, the rate of adsorption of albomycin on zinc surface is not controlled by the reaction process itself but by the rate at which albomycin molecules can get into similar

reaction cage. However, the cage presents a potential barrier, higher than that required for the formation of the activated complex.

On the other hands, results obtained from gasometric experiments were 62.45%, 73.34%, 81.01%, 82.35%, and 86.34% for albomycin concentrations of 0.0001 M, 0.0002 M, 0.0003 M, 0.0004 M, and 0.0005 M, respectively. These results are comparable to those obtained from gravimetric experiments.

Thermodynamic/adsorption considerations

Thermodynamic principles can be used to predict the feasibility of a chemical reaction such as that of corrosion. In order to explore these principles, the transition state equation (which can be written as Equation (3)) was adopted (43)

$$\text{CR} = R/Nh \exp(\Delta S_{\text{ads}}/R) \exp(-\Delta H_{\text{ads}}/RT), \quad (3)$$

where CR is the corrosion rate of zinc, R is the gas constant, N is the Avogadro's number, h is the Planck constant, T is the temperature, ΔS_{ads} and ΔH_{ads} are the entropy and enthalpy of adsorption of the inhibitor on zinc surface, respectively. Transforming Equation (3) into a linear form, Equation (4) is obtained

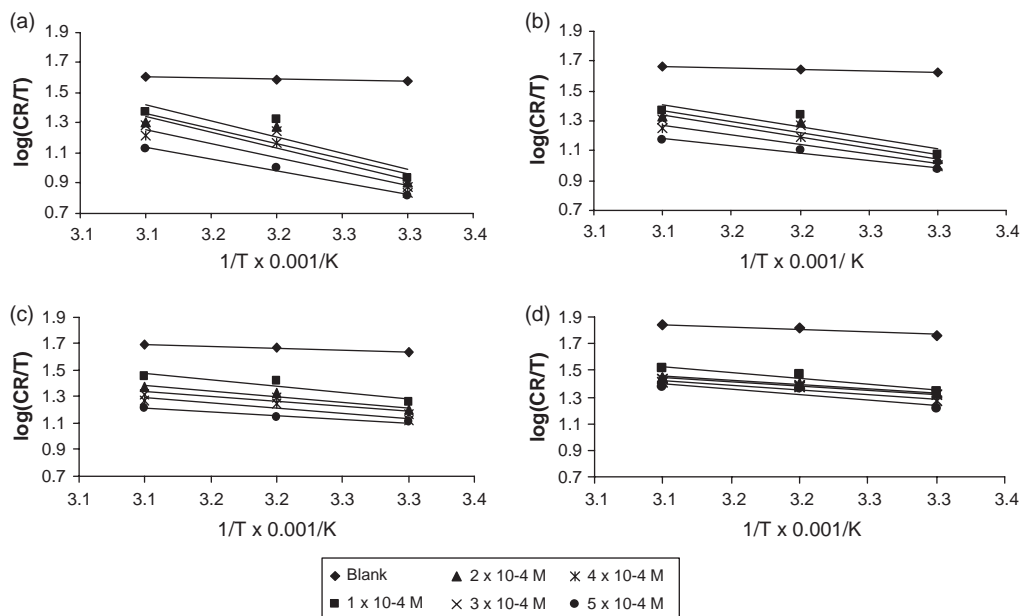


Figure 2. Variation of $\log(\text{CR}/T)$ with $1/T$ for the inhibition of zinc corrosion [in (a) 0.01 M; (b) 0.02 M; (c) 0.03 M; and (d) 0.04 M H_2SO_4] by albomycin.

Table 2. Activation energies and some thermodynamic parameters for the inhibition of the corrosion of zinc by various concentrations of albomycin.

	E_a (J mol ⁻¹)	R^2	ΔH_{ads} (J mol ⁻¹)	ΔS_{ads} (J mol ⁻¹)	R^{2*}
C (M)			0.01 M H_2SO_4		
Blank	3.15	0.9532	0.52	-232.39	0.9934
0.0001	41.46	0.8251	-38.80	-98.06	0.8661
0.0002	38.39	0.8117	-37.08	-105.00	0.7892
0.0003	39.74	0.8263	-35.73	-108.81	0.8049
0.0004	36.22	0.8477	-33.56	-117.46	0.8274
0.0005	29.73	0.9846	-27.07	-139.93	0.9617
C (M)			0.02 M H_2SO_4		
Blank	4.23	0.9835	1.51	-228.22	0.9116
0.0001	28.82	0.8214	-26.16	-138.83	0.7912
0.0002	28.31	0.8465	-25.64	-139.11	0.8188
0.0003	27.69	0.8482	-25.03	-141.88	0.8244
0.0004	24.87	0.9168	-22.21	-152.38	0.8984
0.0005	18.57	0.9658	-15.91	-173.66	0.9547
C (M)			0.03 M H_2SO_4		
Blank	2.94	0.9271	2.92	-223.30	0.9959
0.0001	18.42	0.8747	-15.77	-168.53	0.8376
0.0002	16.35	0.9330	-13.69	-176.60	0.9080
0.0003	14.91	0.8765	-12.25	-182.98	0.8245
0.0004	13.99	0.8818	-11.33	-183.35	0.8556
0.0005	10.23	0.9750	-7.57	-184.97	0.9540
C (M)			0.04 M H_2SO_4		
Blank	9.99	0.9950	-7.34	284.56	0.9913
0.0001	16.49	0.9320	-11.68	-181.28	0.8844
0.0002	15.58	0.9816	-15.08	-170.87	0.9421
0.0003	12.95	0.9918	-14.69	-169.63	0.9308
0.0004	12.10	0.9131	-15.62	-172.91	0.8631
0.0005	11.37	0.8254	-17.90	-163.14	0.8484

Note: C = concentration of clarithromycin; R^2 = degree of linearity for the Arrhenius plot; R^{2*} = degree of linearity for the transition state plot.

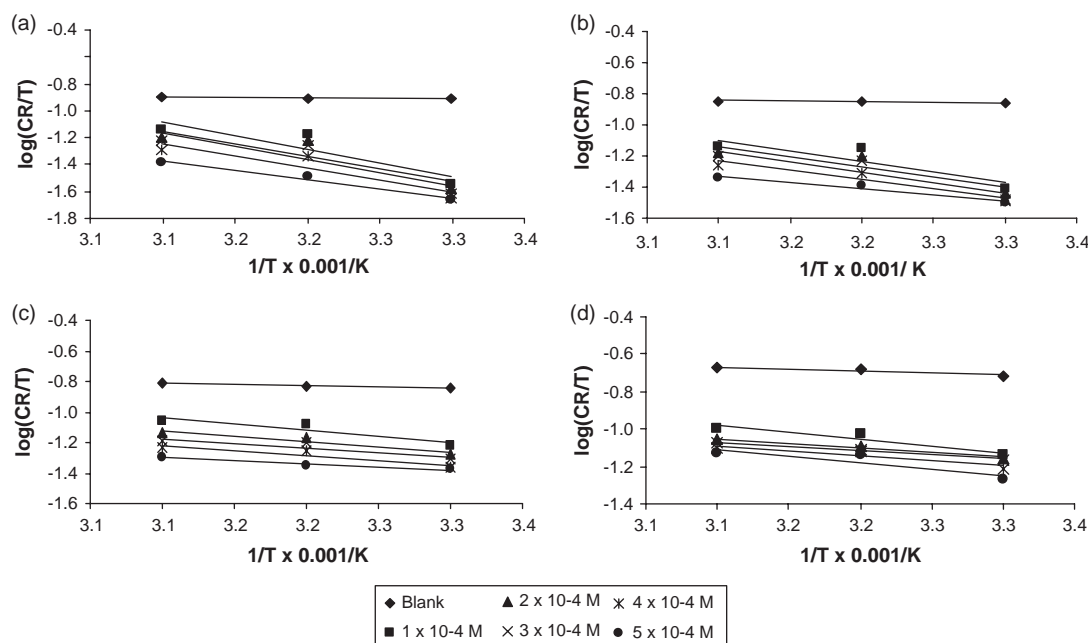


Figure 3. Variation of $\log(CR/T)$ with $1/T$ for the inhibition of zinc corrosion [in (a) 0.01 M; (b) 0.02 M; (c) 0.03 M; and (d) 0.04 M H_2SO_4] by albomycin.

$$\log(CR/T) = \log(R/Nh) + \Delta S_{\text{ads}}/2.303R - \Delta H_{\text{ads}}/2.303RT. \quad (4)$$

From Equation (4), the plots of $\log(CR/T)$ versus $1/T$ should produce straight lines, whose slopes and intercepts should be equal to $\Delta H_{\text{ads}}/2.303R$ and $(\log(R/Nh) + \Delta S_{\text{ads}}/2.303R)$, respectively. Figure 3 (a–d) show the transition state plots for the inhibition of the corrosion of zinc in 0.01–0.04 M H_2SO_4 solutions, respectively. Values of ΔH_{ads} and ΔS_{ads} calculated from the slopes and intercepts of the transition state plots are presented in Table 2. From the results obtained, ΔH_{ads} and ΔS_{ads} values are negative indicating that the adsorption of albomycin on zinc surface is exothermic and is accompanied by increasing degree of orderliness of the inhibitor's molecules.

The relationship between the amount of albomycin adsorbed per unit area of zinc surface and the concentration of albomycin is given by adsorption isotherm. Values of degrees of surface coverage obtained from Equation (8) were used to fit curves for different adsorption isotherms. The tests indicated that the adsorption of albomycin on zinc surface is best described by Langmuir adsorption isotherm. The validity of Langmuir adsorption isotherm can be expressed as follows (44)

$$\log(C/\theta) = \log C - \log K, \quad (5)$$

where C is the concentration (in moles/dm³) of the inhibitor in the bulk electrolyte, θ is the degree of

surface coverage of the inhibitor, and K is the adsorption equilibrium constant. Figure 4 shows that the plots of $\log(C/\theta)$ versus $\log C$ were linear. Therefore the adsorption of albomycin on zinc surface is consistent with the Langmuir adsorption model. Values of adsorption parameters deduced from Langmuir isotherms are presented in Table 3. From the results obtained, values of R^2 and the slopes are very close to unity indicating strong adherence of the adsorption process to the assumptions establishing the Langmuir adsorption model.

The equilibrium constant of adsorption (indicated in Equation (5) above) is related to the free energy of adsorption as follows (45)

$$\Delta G_{\text{ads}}^0 = -2.303RT \log(55.5K), \quad (6)$$

where ΔG_{ads}^0 is the free energy of adsorption of the inhibitor, R is the gas constant, T is the temperature, and 55.5 is the molar concentration of the acid in the solution. Calculated values of ΔG_{ads}^0 (Table 3) are within the limit of values expected for a physical adsorption process, hence the adsorption of albomycin on zinc surface is spontaneous and follows the mechanism of physical adsorption.

Experimental

Materials

The material used for the study was zinc sheet of composition (w%); Pb (0.001), Fe (0.002), Cd

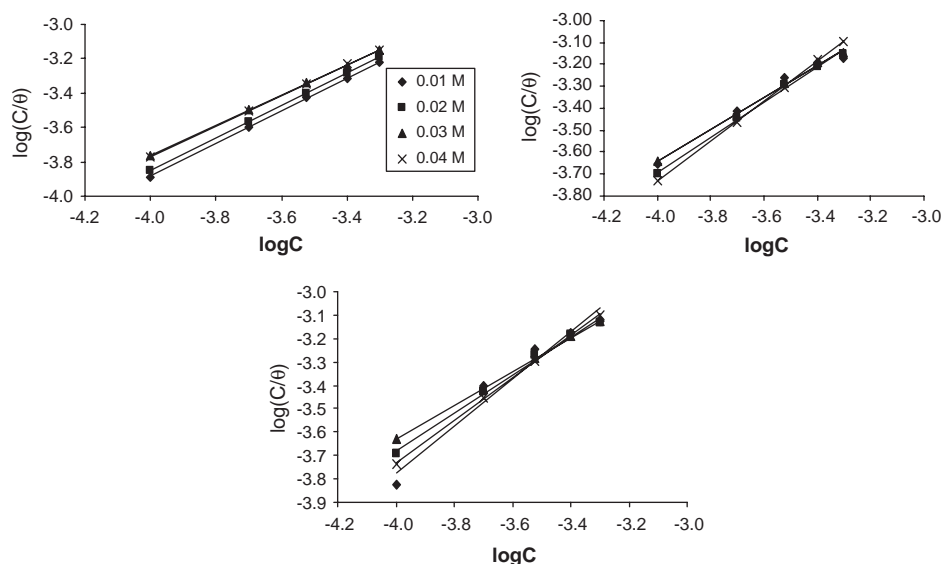


Figure 4. Langmuir isotherm for the adsorption of albomycin on zinc surface in various concentrations of H_2SO_4 at (a) 303 K; (b) 313 K; and (c) 323 K.

(0.001), Cu (0.003), and the rest zinc. The sample 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, dipped in acetone, and allowed to dry in the air before they were preserved in a desiccator. All reagents used for the study were Analar grade and double distilled water was used for their preparation.

The inhibitor (albomycin) was supplied by LIVE-MOORE Pharmaceuticals, Ikot Ekpene, Akwa Ibom State, Nigeria and was used without further purification. The concentration range used for the inhibitor was 0.0001–0.0005 M.

Gravimetric method

In the weight loss experiment, the pre-cleaned zinc coupon was dipped in 200 ml of the test solution maintained at 303 K in a thermostated bath. The weight loss was determined by retrieving the coupons at 24 hours interval progressively for 168 hours (7 days). Prior to measurement, each coupon was washed in 5% chromic acid solution (containing 1% silver nitrate) and rinsed in deionized water (46). The difference in weight was taken as the weight loss of zinc. The experiments were also carried at 313 and 323 K as described above.

Table 3. Langmuir adsorption parameters and free energies of adsorption of albomycin on the surface of zinc.

Temperature (K)	Slope	$\log K$	ΔG_{ads} (KJ/mol)	R^2
		Concentration of $\text{H}_2\text{SO}_4 = 0.01$ M		
303	0.9562	-0.0608	-9.74	0.9999
313	0.7242	-0.7424	-5.98	0.9818
323	1.0049	0.2464	-12.28	0.9578
		Concentration of $\text{H}_2\text{SO}_4 = 0.02$ M		
303	0.9508	-0.0506	-9.80	1.0000
313	0.7953	-0.5093	-7.38	0.9949
323	0.8128	-0.4282	-8.11	0.9950
		Concentration of $\text{H}_2\text{SO}_4 = 0.03$ M		
303	0.8753	-0.2606	-8.58	0.9997
313	0.7252	-0.7388	-6.00	0.9971
323	0.726	-0.7272	-6.26	0.9999
		Concentration of $\text{H}_2\text{SO}_4 = 0.04$ M		
303	0.8915	-0.2056	-8.90	0.9997
313	0.9102	-0.0913	-9.88	0.9997
323	0.912	-0.0850	-10.24	0.9997

From the weight loss measurements, the inhibition efficiency (%*I*) of the inhibitor, degree of surface coverage (θ), and the corrosion rate (CR) of zinc were calculated using Equations (7), (8), and (9), respectively (46).

$$\%I = (1 - W_1/W_2) \times 100 \quad (7)$$

$$\theta = 1 - W_1/W_2 \quad (8)$$

$$CR = W/At \quad (9)$$

where W_1 and W_2 are the weight losses (in g) for zinc in the presence and absence of the inhibitor in H_2SO_4 solutions, θ is the degree of surface coverage of the inhibitor, A is the area of the zinc coupon (in cm^2), t is the period of immersion (in hours), W is the weight loss of zinc after time t , and CR is the corrosion rate of zinc in $gh^{-1} cm^{-2}$

Gasometric method

Gasometric experiments were carried out at 303 K using a gasometer, as described in the literature (47). The gasometric experiments were conducted at 303 K for the blank (3 M H_2SO_4) and for H_2SO_4 solutions containing various concentrations of albomycin. From the volume of hydrogen gas evolved per minute, inhibition efficiencies of various concentrations of the inhibitor were calculated using Equation (10)

$$\%I = \left(1 - \frac{V_{H_2}^1}{V_{H_2}^0}\right) \times 100, \quad (10)$$

where $V_{H_2}^1$ and $V_{H_2}^0$ are the volumes of hydrogen gas evolved at time “ t ” for inhibited and uninhibited solutions, respectively.

Conclusion

Albomycin is a good inhibitor for the corrosion of zinc in H_2SO_4 . The inhibitory action of albomycin is affected by the period of contact with the acid, concentration of the acid/inhibitor, and temperature. Therefore, the performance of albomycin as an inhibitor can be optimized by taking advantage of time, concentration, and temperature. Also thermodynamic and adsorption models can adequately be used to predict the feasibility of the inhibitor.

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