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Itraconazole Drug as Corrosion Inhibitor for Aluminium in 0.7 M Hydrochloric Acid Medium

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Abstract: Corrosion poses a significant challenge in various industrial applications, especially in acidic environments. This study explores the potential of Itraconazole, a widely used antifungal drug, as a corrosion inhibitor for aluminium in a corrosive 0.7M hydrochloric acid at 303, 313, and 323K. The corrosion inhibition efficiency of Itraconazole was investigated through weight loss measurements. The findings suggest that Itraconazole exhibits notable corrosion inhibition properties for aluminium in 0.7M HCl, showcasing its potential as an eco-friendly and cost-effective alternative to traditional corrosion inhibitors. The highest inhibition efficiency of the drug (77.1%) was observed when using 0.4g/L of drug concentration in 0.7M HCl at 303K while the lowest inhibition efficiency (58.8%) was observed when using 0.1g/L of drug concentration in 0.7M HCl at 323K. FT-IR spectra showed that the inhibition mechanism was an adsorption process through the functional groups present in the drug onto the metal surface. Langmuir isotherm provided more accurate description of the adsorption behavior of the Itraconazole drug because of its R^2 values obtained were relatively closer to unity when compared to Freundlich model.

1. Introduction

Corrosion of metals, particularly aluminum, in aggressive acidic environments remains a significant concern in various industrial applications (Verma *et al.*, 2018). Hydrochloric acid (HCl) is a common corrosive medium that can lead to accelerated deterioration of metal surfaces, affecting the structural integrity and performance of materials (Galai *et al.*, 2016; Tebbji *et al.*, 2007). To mitigate such

corrosion challenges, the exploration of novel and effective corrosion inhibitors is crucial. In recent years, the utilization of pharmaceutical compounds as corrosion inhibitors has gained attention due to their diverse chemical structures and inherent biocompatibility (Galai *et al.*, 2016; James *et al.*, 2011). According to several researchers, corrosion is a significant issue in various industries, including the transportation, construction, and chemical sectors, leading to substantial economic losses and safety concerns (Al-Amiery *et al.*, 2024; Iorhuna *et al.*, 2023; Iorhuna and Nyijime, 2022;). Aluminum, being a widely used material, is susceptible to corrosion, especially in acidic environments such as hydrochloric acid (HCl) solutions.

Itraconazole, a triazole antifungal drug, is one such pharmaceutical compound that exhibits potential beyond its primary medical applications. The unique molecular structure of Itraconazole, characterized by multiple heterocyclic rings containing nitrogen atoms, suggests its possible role as a corrosion inhibitor (Quiang *et al.*, 2017). The selection of Itraconazole as a potential corrosion inhibitor is grounded in its chemical structure, which may facilitate adsorption onto the metal surface, forming a protective barrier against corrosive agents (Gang *et al.*, 2018). Numerous attempts have been made to use antifungal drugs as corrosion inhibitors (Lyapun *et al.*, 2023; Fernandes *et al.*, 2019; Damej *et al.*, 2016; Zarrouk *et al.*, 2012; El Issami *et al.*, 2007). In other words, Itraconazole belongs to 1,2,4-Triazole containing a five-membered, π-excessive, aromatic nitrogen heterocycle, which is widely used as corrosion inhibitors for various metallic materials in aggressive media (Belal *et al.*, 2023; Quraishi and Jamal, 2020; Belghiti *et al.*, 2016; Akalezi & Oguzie, (2016); Salghi *et al.*, 2013; Fox & Bradely *et al.*, 1980). Aluminum, widely used in industries ranging from aerospace to construction, is susceptible to aggressive corrosion in acidic environments. The corrosive attack on aluminum surfaces can result in the release of harmful by-products, compromising the structural integrity and longevity of the metal. Hence, the development of effective corrosion inhibitors is crucial for preserving the integrity of aluminum structures and extending their service life (Al-Amiery *et al.*, 2023; Tan *et al.*, 2018).

This study is aimed at investigating the inhibiting effect of Itraconazole on aluminum corrosion in 0.7 M HCl through weight loss measurement analyses. This study will not only contribute to the understanding of Itraconazole as a corrosion inhibitor but also explores the potential of pharmaceutical compounds in corrosion protection strategies.

2. Materials and Methods

2.1 Sample collection and preparation of drug solution

The Itraconazole tablets were collected from Robi Pharmacy in Makurdi local government of Benue State, Nigeria and transported to the General Research Laboratory, Department of Chemistry College of Physical Sciences, Joseph Sarwaun Tarka University, Makurdi (JOSTUM) for further use as inhibitor. The molecular structure is presented in figure 1.

The itraconazole tablets were crushed using mortar and pestle to be able to dissolve quickly while forming a solution of it with corrodent. 0.1, 0.2, 0.3 and 0.4 gL^{-1} of the drug in 0.7 M HCl were prepared by weighing 0.1, 0.2 0.3 and 0.4 g of the powdered drug and dissolving each in 1litre of 0.7 M HCl separately.

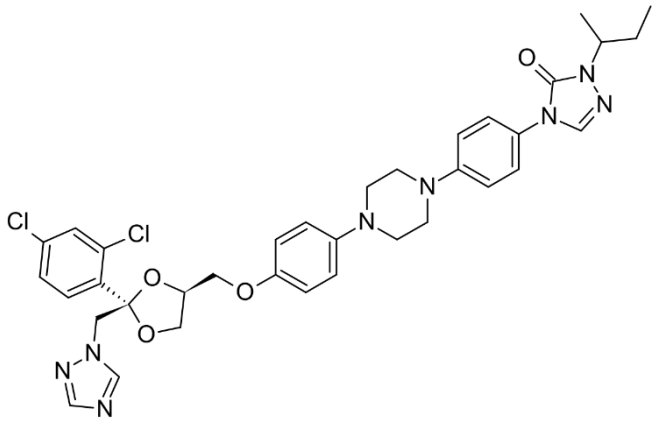

Figure 1. Molecular structure of Itraconazole Molecule

2.2 Preparation of corrodent solutions

The corrodent solutions were prepared using 0.7M HCl

Preparation of 1000cm^3 of 0.7M HCL:

$$\text{Concentration (C1) of stock solution} = \frac{\%Purity\ X\ density\ X\ 10}{Molar\ mass\ of\ HCl} \quad (1)$$

Whereby % purity of HCl = 36.25

Density of HCl = 1.18 gcm^3

Molar Mass of HCl = 36.46gmol^{-1}

Therefore $C_1 = \frac{36.25\ X\ 1.18\ X\ 10}{36.46} = 11.7M$

That means the concentration of HCl from which 0.7M was prepared is 11.7M

The volume of 11.7M HCl measured in preparing 1000cm^3 of 0.7M HCl was calculated from the relationship below:

$$C_1V_1 = C_2V_2 \quad (2)$$

C_1 = Concentration of stock solution = 11.7

V_1 == volume of stock solution to be measured

C_2 = concentration to be prepared = 0.7M

V_2 = volume to be prepared = 1000cm^3

$$V_1 = \frac{C_2V_2}{C_1} = \frac{0.7X1000}{11.7} = 59.8cm^3$$

Therefore, 59.8 cm^3 of stock solution was measured and poured into a 1000 cm^3 volumetric flask. The flask was then filled up to the mark with distilled water to prepare a 0.7M HCl solution.

2.3 Preparation of aluminum coupons

In this work, a commercially available grade of aluminum (purity = 98% Al) was identified and obtained locally. Aluminum sheets were mechanically cut into coupons with diameters of 2 cm x3 cm x 0.1 cm. A tiny hole about 5 mm in diameter was drilled near the upper edge of the coupons to aid in holding them with grass hooks and suspending them in the corrosive liquid. The vouchers were used without any additional polishing.

2.4 Weight loss measurement

An analytical weighing balance was used throughout this experiment to weigh aluminum samples both before and after they were submerged in the acidic medium. Coupons that had been previously cleaned and weighed were placed in 100 milliliter beakers along with test solutions containing 0.7 M of hydrochloric acid, both with and without the inhibitor. After being closed, the beakers were placed in a water bath with a thermostat set to maintain 303, 313, and 323 K. After being removed from the test solutions for four hours at one-hour intervals, the coupons were reweighed after being cleaned with distilled water and a soft brush and then dried in acetone. Weight loss was assumed based on the variations in weight (Iorhuna *et al.*, 2022). Based on the outcomes of the weight reduction trial, the percentage inhibition efficiency, From the weight loss experiment results, the percentage (%)inhibition efficiency, the degree of surface coverage (Ɵ) and the corrosion rate of the aluminum (CR) were calculated using equations shown below;

$$\%IE = (1 - \frac{W_i}{W_f}) \times 100 \quad (3)$$

$$Ɵ = 1-\frac{W_i}{W_f} \quad (4)$$

$$CR\ (g/hcm^{-2}) = \frac{\Delta W}{At} \quad (5)$$

Where W_i and W_f are the initial and final weight loss in grams (g) of aluminum in the absence and presence of inhibitor in HCl solutions respectively, ΔW is the change weight loss (g), A is the area of one face of the specimen (cm^2) and t is the period of immersion in hour (h).

3. Results and Discussion

3.1 Weight Loss experiment

Weight loss experiments were performed to determine the extent of corrosion inhibition by itraconazole drugs. These experiments involve immersing aluminum samples in the 0.7M HCl medium, both with and without itraconazole inhibitors, for a predetermined period and presented in Table 1. The corrosion inhibition of aluminum was carried out at various temperatures of 303k, 313k and 323k respectively in different concentrations of the drug solution ranging from 0.1g to 0.4g at the interval of 1 hour for four hours in 0.7M concentration of HCl. The obtained results show the decrease in weight loss as the concentration of the inhibitor increased from 0.1g to 0.4g. If we assume that 100 mg (0.1 g) of Itraconazole in the tablet of 1g; we can calculate the molarity of Itraconazole by using the following relation:

M = m*0.1 / 705.64(g/mol)*1L

The corrosion rate also decreases as the concentration of the inhibition increases but increases as the temperature rises. The inhibition efficiency (IE) of the inhibition on the surface of the aluminum decreases with temperature rise but increases with the inhibitor concentration from 0.1 t0 0.4g/L. This inhibition efficiency could be a result of the increase in the absorption of the itraconazole drug

on the aluminum boundary as the concentration increases. The results presented in Table 1 show that the weight loss of both inhibited and uninhibited aluminum increases with an increase in temperature across all concentrations of the aggressive solution. Similar results were obtained in previous studies (James and Akarenta, 2014; Ajanaku *et al.*, 2015)

Table 1. Corrosion rate and inhibition efficiency obtained from weight loss measurements of inhibitor in 0.7M HCl containing various concentrations of Itraconazole drug at different temperatures

Temp. (K)	Conc. (g/dm^3)	Conc. (mol/dm^3)	ΔW	CR	θ	%IE
303	0.0	0.0	0.302	0.0083	-	-
	0.1	1.417 E-5	0.099	0.0027	0.674	67.4
	0.2	2.834 E-5	0.085	0.0023	0.722	72.2
	0.3	4.251 E-5	0.078	0.0021	0.746	74.6
	0.4	5.668 E-5	0.069	0.0019	0.771	77.1
313	0.0	0.0	0.285	0.0079	-	-
	0.1	1.417 E-5	0.108	0.0030	0.620	62.0
	0.2	2.834 E-5	0.091	0.0025	0.683	68.3
	0.3	4.251 E-5	0.085	0.0023	0.708	70.8
	0.4	5.668 E-5	0.079	0.0021	0.734	73.4
323	0.0	0.0	0.252	0.007	-	-
	0.1	1.417 E-5	0.106	0.0029	0.585	58.5
	0.2	2.834 E-5	0.099	0.0027	0.614	61.4
	0.3	4.251 E-5	0.081	0.0022	0.685	68.5
	0.4	5.668 E-5	0.071	0.0019	0.728	72.8

3.3 FT-IR Spectra Analysis:

The FT-IR results are analyzed to determine the molecular interactions and chemical bonding between the Itraconazole drugs and the aluminum surface. FT-IR spectroscopy is a powerful analytical technique that provides information about functional groups present in the sample. The FT-IR spectra obtained before and after the corrosion experiments reveal changes in the peak intensities and shifts in the wave numbers, indicating the formation of a protective film on the aluminum surface. The shifts in adsorption bands reveal the adsorption of different phytochemical components of Itraconazole drug on the aluminium surface. The surface analysis of the inhibited corroded aluminium using FT-IR reveals that the peak between 3372.87 to 3350.87 cm^{-1} is O–H stretch of alcohol, and the band at 2650.86 to 2072.85 cm^{-1} is assumed to be C=O bond. The broad peak at 1544 to 1510 cm^{-1} is ascribed to be C–O stretched. As can be observed from Figure 2a-b, there are no significant changes in the adsorption band between the uninhibited and inhibited aluminium coupons (Li *et al.*, 2014).

It's possible that the combination of various components in itraconazole tablets could exhibit synergistic effects in inhibiting aluminum corrosion. Different compounds may work together to enhance inhibition efficiency beyond what could be achieved with individual components alone.

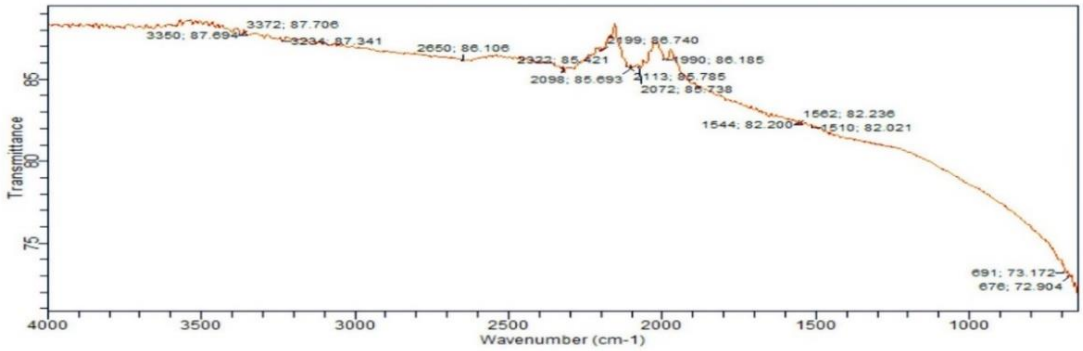

Figure 2a. FT-IR spectra of uninhibited aluminium coupon in 0.7M HCl solution

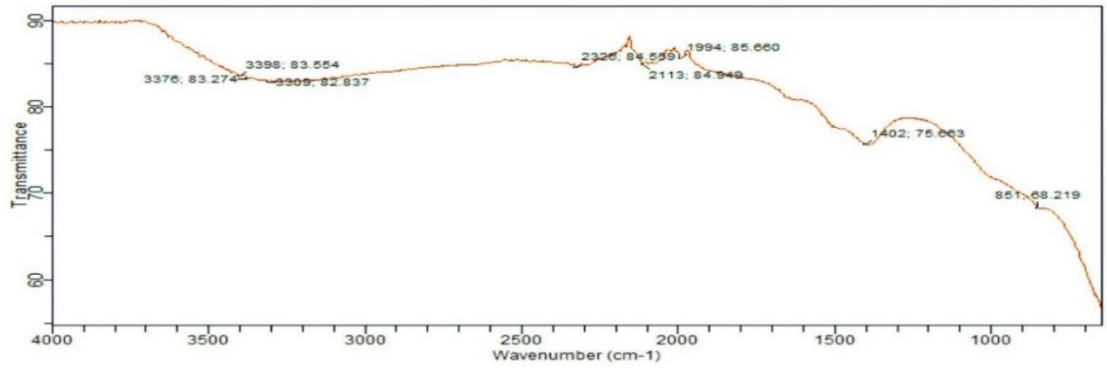

Figure 2b. FT-IR spectra of inhibited aluminium coupons in Itraconazole drug in 0.7M HCl

3.4 Adsorption Isotherm Analysis

Adsorption isotherm studies are essential in understanding the relationship between the amount of adsorbate (substance being adsorbed) and the concentration of the adsorbate in a given system, typically at a constant temperature. Two of the adsorption isotherms studied is the Langmuir and Freundlich isotherms.

Langmuir Isotherm: The Langmuir isotherm assumes monolayer adsorption on a homogeneous surface with a finite number of identical sites.

$$\frac{C_{inh}}{\theta} = \frac{1}{K_{ads}} + C_{inh} \qquad (6)$$

Where K_{ads} is the equilibrium constant of adsorption, C_{inh} is the concentration of inhibitor and 1 is the corrective factor for the linearized Langmuir equation with a slope not equal to unity (Langmuir, 1918).

By plotting a graph of $\frac{C_{inh}}{\theta}$ against C_{inh}, (Figure 3a), the value of K_{ads} can be determined from the y-intercept value of the plot and is related to the standard free energy of adsorption (ΔG°_{ads}) (Mashuga *et al.*, 2017):

$$\Delta G^{\circ}_{ads} = - RT \ln (55.5\ K_{ads}) \qquad (7)$$

The free enthalpy is calculated on the assumption that the tablet of 1g contains 100 mg (0.1 g) of Itraconazole.

Freundlich Isotherm: The Freundlich isotherm is an empirical model describing heterogeneous surfaces and multilayer adsorption (Figure 3b).
The equation is given by:

$$\log\Theta = n\log C + \log K_{ads} \quad (8)$$

where: K_{ads} is adsorption constant the K_{ads} value can be calculated from the plot of logΘ vs log C with the intercept line on the logΘ axis.

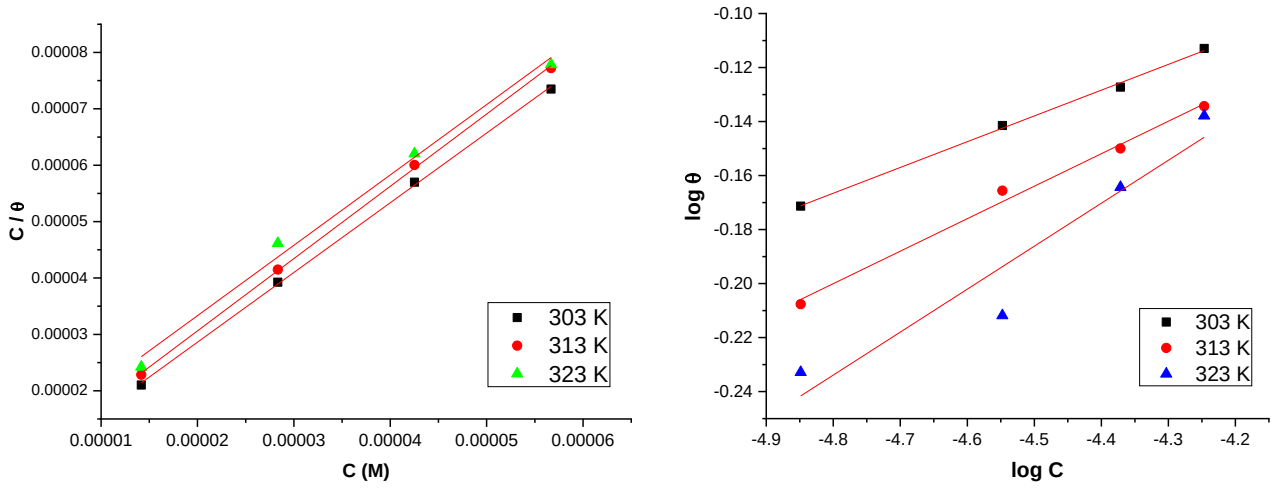

Figure 3. Plots of (a) Langmuir and (b) Freundlich isotherm for the adsorption of Itraconazole drug on the Aluminium surface

From Table 2, it can be observed that Itraconazole drug obeys the Langmuir model for all the temperatures studied for aluminium metal. The obtained data indicate that Langmuir isotherm is valid for this system. The unity value of the correlation coefficient obtained in the Langmuir isotherm is an indication that the adsorption of the drug onto aluminium surface best fits into the theory of Langmuir isotherm.

Table 2. Calculated values of slopes, R^2 and K_{ads} for the Langmuir and Freundlich isotherm temperature in 0.7M HCl

Isotherms	Temp. (K)	Slope	R^2	K_{ads} (kJ/mol)	ΔG°_{ads} (kJ/mol)
Langmuir	303	1.90	0.997	80971.66	-38.59
	313	2.32	0.999	78186.08	-39.77
	323	2.39	0.998	79239.30	-41.08
Freundlich	303		0.987	1.9567	-11.81
	313		0.996	2.3848	-12.71
	323		0.996	3.3870	-14.06

As we retained the Langmuir isotherm adsorption, it's clear that the negative values of ΔG°_{ads} obtained indicate that the adsorption process of these compounds on the metal surface is spontaneous

according to the chemical adsorption. This assumption may be consolidated by the possible formation of Itraconazole-aluminium ion complex since the formation of other complexes with Zn(II), Co(II), Ni(II), Cu(II)… ions are confirmed by several authors (Azevedo-França *et al.*, 2020; Bagihalli *et al.*, 2009) Furthermore, certain compounds in itraconazole, such as starch or sucrose, might have the ability to adsorb onto the aluminum surface there by forming a monolayer or multilayer film, they can inhibit the adsorption of aggressive ions or molecules responsible for initiating corrosion reactions. Also, the values of ΔG°_{ads} deduced from Freundlich kind are lower that those determined in the case of Langmuir one.

These findings can be interpreted by the molecular structure: (±)-1-[(RS)-sec-butyl]-4-[p-[4-[p-[[(2R,4S)-rel-2-(2,4-dichlorophenyl)-2-(1H-1,2,4-triazol-1-ylmethyl)-1,3-dioxolan-4-yl]methoxy] phenyl]-1-piperazinyl]phenyl]-D2-1,2,4-triazolin-5-one, containing various function groups as triazole, piperazine … widely used as efficient inhibitors of metals in acidic media (Haque *et al.*, 2024; Merimi *et al.*, 2022; Bouklah *et al.*, 2013). In other words, we assist to the synergistic intramolecular effect of these attractive centers as discussed in literature (Batah *et al.*, 2024; Goni *et al.*, 2024;). Furthermore, the existing data on the … paramaters reveals that Itraconazole exhibited an excellent inhibitory effect on corrosion of various metals. Gong *et al.*, 2017 obtained the higher value of E_{HOMO} - 4.11 eV- indicating that the itraconazole molecule has a strong electron donating nature, and the electrons given are easily combined with cuprous ions to develop a stable adsorption film on the copper surface, thereby effectively inhibiting copper corrosion. Also, the obtained energy gap ($\Delta E = E_{LUMO} - E_{HOMO}$) = 2.41 eV) regarded as a key parameter when discussing the molecular properties of corrosion inhibitors; means that itraconazole exhibits excellent corrosion inhibition (Obot *et al.*, 2012; Madkour & Elshamy, (2016)).

Conclusion

From the result of this study, the following conclusions can be drawn: The observed trend in weight loss indicates that Itraconazole has a noticeable inhibitory effect on aluminum corrosion. The progressive reduction in weight loss with increasing exposure time and temperature suggests an enhanced protective capability of Itraconazole over time and at elevated temperatures. The inhibition efficiency of the Itraconazole drug solution on the surface of aluminum in the 0.7M HCl increases as the concentration of the drug solution increases, reaching a value of 77.2% at 0.4 mol/dm^3. The adsorption isotherm of the Itraconazole solution on the aluminum surface in 0.7M HCl follows the Langmuir isotherm. The adsorption was spontaneous and obeyed the Langmuir adsorption isotherm.

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