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Alocasia Odora Extract as Environmentally Benign Corrosion Inhibitor for Aluminum in HCl

Haque J.[1], Zulaikha M. A.[1,2], Wan Nik W. B.[1*], Daoudi W.[3], Berdimurodov E.[4], Zulkifli F.[1]**

[1]Faculty of Ocean Engineering Technology and Informatics, Universiti Malaysia Terengganu, 21030 Kuala Nerus, Terengganu, Malaysia
[2]Shin Yang Shipyard Sdn. Bhd.
Survey Lot 2125, Kuala Baram, Sungai Tujuh, 98009 Miri, Sarawak, Malaysia
[3]Laboratory of Materials and Environment, Dept of Chemistry, Multidisciplinary Faculty of Nador, University of Mohamed 1, 60700 Nador, Morocco
[4]Faculty of Chemistry, National University of Uzbekistan, Tashkent, 100034, Uzbekistan,
**Corresponding author, Email address: niksani@umt.edu.my*
***Corresponding author, Email address: fakhratulz@umt.edu.my*

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Abstract: The development of effective and environmentally benign corrosion inhibitors is now a major objective in the subject of corrosion. The impact of corrosion on aluminum in HCl has been investigated using leaves extract of Alocasia Odora (A. Odora) from Araceae family. Alocasia Odora is indigenous to Southeast Asia. The selected plant extract is nontoxic with LD50 > 0.5 g/kg (oral mice) and biodegradable. The leaf extract of Alocasia Odora was easily dissolved in HCl solution. The corrosion inhibition studies of plant extract were carried out using weight loss, potentiodynamic polarization and electrochemical impedance spectroscopic (EIS). The A. Odora retards the rate of dissolution by 94% and exhibit excellent corrosion inhibition aluminum in HCl. The plant extract also shows potential inhibition (69%) at a longer immersion time (48h). The polarization curves showed that A. Odora acts as the cathodic inhibitor. A big capacitive loop present in the EIS at high frequencies, followed by a big inductive loop at low frequency values compared to blank, suggesting the effective corrosion inhibition in the presence of inhibitor. SEM results also depicted that the surface of aluminum with A. Odora extract smoother compared to blank.

Keywords: *Leaf extract; Corrosion inhibitor; Electrochemical; Adsorption, SEM.*

1. Introduction

The development of simple and affordable methods to preserve the metal surface via a barrier layer is necessary due to the significant financial loss and safety concerns caused by the widespread corrosion of metals. Since water is the primary cause of metal corrosion, most solutions concentrate on ways to prevent water from entering the system. Bio-mimicked surfaces are now being applied to metal surfaces to reduce corrosion. They were inspired by naturally occurring surfaces (Peethan *et al.,* 2022). Aluminium's resistance to corrosion in varied settings depends on the creation of a dense, adherent passive oxide coating. Along with its combination of low weight, attractive look, mechanical strength,

and excellent thermal and electrical conductivity, aluminium lends itself to an excessive number of technical applications (Deng & Li, 2012; Wan Nik *et al*., 2011; Gelfgat *et al.,* 2019).

Practically, corrosion inhibitors are applied to the aluminium surface to prevent corrosion in a medium. Corrosion inhibitors are organic or inorganic substances that are employed sparingly to significantly slow the corrosion rate (Rani & Basu, 2012; Wan Nik *et al*., 2010; Benmohamed *et al.,* 2022). According to Ennouri *et al*., (2017), he stated that numerous studies have been conducted on the corrosion of aluminium and its alloys in acidic solutions. The usage and production of inorganic and industrially manufactured organic inhibitors are avoided despite the fact that they are effective inhibitors due to the rising concern of environmental regulatory authorities of various nations, citing the risk they provide to the environment and human health (Mobin *et al.,* 2020).

Nowadays, it has been advised for widespread use of the mixed compounds recovered from plant components such as roots, tree trunks, branches, bark, leaves, flowers, fruits, peels, and seeds as green corrosion inhibitors to replace the harmful and expensive substances (Nik *et al.,* 2017; Chahul *et al*., 2019; Haque *et al.,* 2021; Mouden *et al*., 2021). Leaf extracts are preferred among these ingredients because of their ease of collection and recycling, availability, cost-effectiveness, direct processing and, above all, their ability to offer excellent protection even at low concentrations (Ikhmal *et al.,* 2019; Hajar *et al.,* 2016; Beda *et al.,* 2017; Fouda *et al*., 2021; Ikhmal *et al.,* 2018; Hossain *et al*., 2021 & 2022; Eziuka *et al.,* 2023; Ezeh *et al.,* 2023). Leaf extracts contain a multitude of multifunctional groups of benefit to the steel surface immersed in an aqueous corrosive environment. These include groups containing amino, carboxyl, amide, hydroxyl, phenyl, sulfur and phosphorus heteroatoms. As a result, Trung *et al.,* 2021 stated that leaf extract was being extensively used in several industrial applications as a green corrosion inhibitor.

In this study, *Alocasia Odora* (A.Odora) leaf extract has been selected as a corrosion inhibitor for aluminium alloy. *A.Odora* is naturally found in tropical southeast Asia. *Araceae* family plant is well known as to have a medical property such as antimicrobial and antifungal to treat light injury (Ongpoy & Ongpoy, 2017). This plant's leaf is selected as an inhibitor because the alkaloids compound (GAO *et al*., 2022) in the leaf extract could retard the process of corrosion of aluminium in 0.5 M HCl solution.

2.1 Material and method

Aluminum (Al) coupons of 2.45 cm x 2.45 cm x 0.3 cm and 1.0 cm^2 in the exposed area were chosen for the electrochemical investigation and the measurement of weight loss, respectively. The Al coupon was completely cleaned of any clinging contaminants with different grades of emery paper ranging from 120 to 1200. Then, the polished metal was washed with water followed by acetone degreasing (Krishnaveni & Ravichandran, 2014). The specimen had a composition of 0.40% Si, 0.80% Mg, 0.25% Zn, 0.15% Cu, 0.7% Fe, 0.15% Ti, 0.15% Mn, and Al balance.

2.2 Leave extract (Corrosion inhibitor)

At a village in Jenagor, Kuala Berang, Terengganu, fresh *A. Odora* leaves were collected **(Figure 1),** washed with water to remove ash and dirt, dried for two days at 60 °C, and then milled into powder. 100 g of the powder was mixed and stirred in 1000 ml of 80% (by volume) Ethanol, C_2H_5OH by using an orbital shaker for 72 hours with 200 ppm. Next, the mixture was evaporated by a rotary evaporator for one hour with 100 ppm at 20 °C of vapor temperature and 35 °C of bath temperature. Filtering the evaporated solution 60 ml of dark green residue (Deng & Li, 2012). The residue was collected and stored in a chiller. The extract is utilised at a concentration of 1.5-8.0 g/L.

Figure 1. *Alocasia Odora* plant

2.3 Corrosion studies

2.3.1 Weight loss study

Before being immersed in 100 ml of 0.5 M to 5 M HCl solution, clean, dry metal samples were carefully weighed on an electronic balance, in the presence and absence of different concentrations of A. Odora extract (0.5 to 8.0 g/L), in order to perform gravimetric calculations. The aluminum coupons were removed after immersion for 3 to 48 hours, then washed with distilled water and acetone, dried and accurately weighed to determine the corrosion rate of inhibited and non-inhibited samples. Each experiment was carried out in duplicate to obtain reproducible results. The corrosion rate (C_R) and inhibition efficiency (η %) of aluminum were calculated using the following formulas.

$$C_R = \frac{W}{At} \qquad \textbf{Eqn.1}$$

$$\eta\,(\%) = \frac{C_R - C_{R(i)}}{C_R} \times 100 \qquad \textbf{Eqn.2}$$

where A is the surface area of the specimen (14.945 cm^2), W is weight loss (mg), t is exposure time (h), $C_{R(i)}$ and C_R are the corrosion rates with and without inhibitor, respectively.

2.3.2 Electrochemical study

The electrochemical experiment was carried out using a Gamry potentiostat/galvanostat (Model 300) connected to three-electrode cells at room temperature. The working electrode was made of aluminum with a surface area of 1 cm^2. The counter and reference electrodes were made of platinum and Ag/AgCl respectively. Results were analyzed using Gamry software: Echem Analyst version 5.0. Before starting the experiment, the open circuit potential (OCP) was measured as a function of time for 200 seconds to obtain a stable OCP, which corresponds to the corrosion potential. Alternating signals with an amplitude of 10 mV peak-to-peak were used at PCO to perform impedance measurements over a frequency range from 100,000 to 0.01 Hz. Potentiodynamic polarization curves were obtained by automatically changing the electrode potential from -0.25 V to +0.25 V relative to PCO, with a sweep rate of 0.5 mV/s. Extrapolation of anodic and cathodic curves from linear Tafel plots is necessary to determine corrosion current densities (i_{corr}) (Chaubey *et al*., 2018).

2.4 Surface study

To evaluate the interaction of *A. Odora* extraction component with dissolved Al in 0.5 M HCl, the UV-visible spectra were taken using a Shimadzu UV-1800 spectrophotometer with a wavelength range restricted between 200 and 700 nm. With the aid of a scanning electron microscope (SEM, JOELJSM 6390), the impact of an inhibitor on the corrosion of aluminium was investigated. Investigations were made on the surface analysis of the Al for inhibited and uninhibited samples with 3 and 24 hour immersion time.

3. Results and Discussion

3.1 Weight loss

3.1.1 Effect of *A. Odora* extract concentrations

Table 1 demonstrates that the addition of *A.Odora* extract significantly lowers the corrosion rate of aluminium alloys. This behaviour may be explained by the inhibitor's increased surface coverage as a result of adsorption on the aluminium surface. With of A.Odora extract, inhibition efficiencies η % rise to 94.32% at 8.0 g/L. In other research, Khadraoui *et al.,* 2016 stated that they used 1.5 g/L of Mentha pulegium extract to protect aluminium alloys against corrosion in 1 M HCl solution with an inhibition efficiency of more than 60%. From **Table 1**, the results show that the inhibition efficiency escalate over the increment of *A.Odora* extract. 8.0 g/L of A. Odora extract is considered as optimum concentration as in past results found that the increment of corrosion rate slowed after reaching 8.0 g/L.

Table 1. Weight loss results of concentrations of *A. Odora* extract on inhabitation efficiency (η) on Al in 0.5 M HCl after immersion time 3h

Inhibitor Conc. (g/L)	Weight loss (mg)	Corrosion rate (mg/ cm^2.h)	η (%)	Surface coverage (θ)
0.0	223.1	4.98	-	
2.0	27.0	0.60	87.99	0.8799
4.0	23.0	0.51	89.79	0.8979
6.0	18.5	0.41	91.81	0.9181
8.0	12.9	0.29	94.32	0.9432

3.1.2 Effect of immersion time

The effect of several intervals of immersion time towards the inhibition efficiency of 4.0 to 8.0 g/L of A. Odora extract were studied. **Table 2** shows the highest inhibition efficiency 94% found at 3 hours immersion time at 8.0 g/L of A. Odora. While the lowest inhibition efficiency (11%) was obtained during 24h immersion time with 4.0 g/L of A. Odora. In all the immersion time, the inhibition efficiency of the leaves extract decreases over time and then increases again. The decreasing inhibition effect at 6.0 g/L may be due to the protective layer interaction with the corrosive species in the solution and degrading, which would reduce the effectiveness of the inhibition (Ghahremani *et al.,* 2021).

3.1.3 Effect of acid concentration

Increasing the concentration of HCl can increase the rate of aluminium corrosion. However, the addition of a green inhibitor can counteract the corrosive effects of HCl by forming a protective film on the aluminium surface. The inhibition efficiency of the aluminium can be influenced by the concentration of HCl, as a higher concentration of HCl can lead to the degradation of the protective film, reducing its inhibition efficiency (Branzoi *et al*., 2002). Based on the result obtained in **Figure 2**, the inhibition efficiency of Al decreased extremely over the increase of HCl's concentration with the lowest result is -45% when 5.0 M HCl was used. Hence the A. Odora extract is an effective corrosion inhibitor for Al metal up to 1 M HCl solution.

Table 2. Effect of inhibition efficiency on Al corrosion in interval immersion time

Inhibitor Conc. (g/L)	***η (%)***			
	3 h	6 h	24 h	48 h
4.0	92	51	11	51
6.0	90	57	19	65
8.0	94	66	30	69

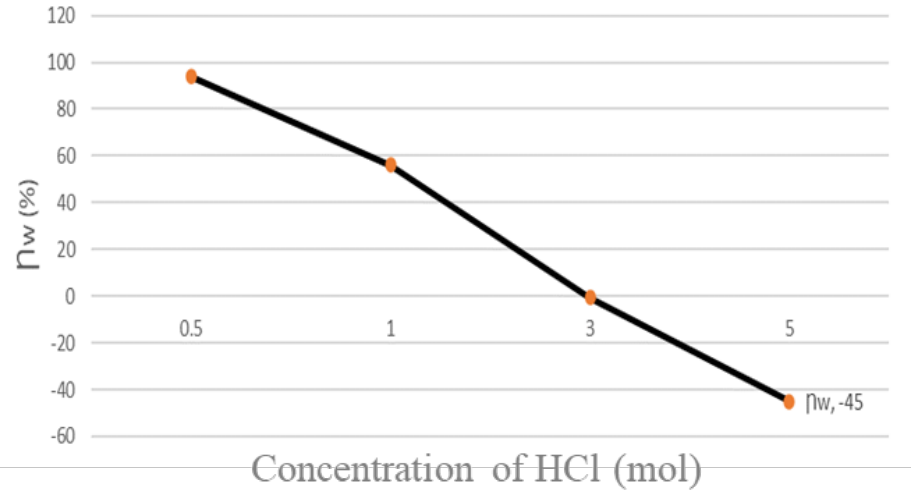


Figure 2. Effect of concentration of HCl on Al corrosion in the presence of 8.0 g/L of *A. Odora* extract

3.2 Electrochemical study

3.2.1 Tafel polarization study

Tafel polarization curves corresponding to the concentration of *A. Odora* extract for aluminum in a 0.5 M HCl solution are shown in **Figure 3**. This study involves adjusting the potential of the working electrode while recording the current generated as a function of time or potential. The cathodic curves show lower current densities in the presence of A. Odora extract, as shown in **Figure 3** resulting in a significant reduction in the corrosion rate. However, A. Odora extract slightly slows down the anodic reaction of the corrosion process. Electrochemical parameters such as corrosion potential (E_{corr}),

corrosion current density (i_{corr}) and cathodic Tafel constants ($-\beta_c$) are obtained from polarization measurements presented in **Table 3**. Using i_{corr} data, the percentage of inhibition (η %) was calculated as follows:

$$\eta(\%) = \frac{i^0_{corr} - i_{corr}}{i^0_{corr}} \times 100 \qquad \text{Eqn.3}$$

where i^0_{corr} and i_{corr} are the corrosion current density without and with the inhibitor, respectively.

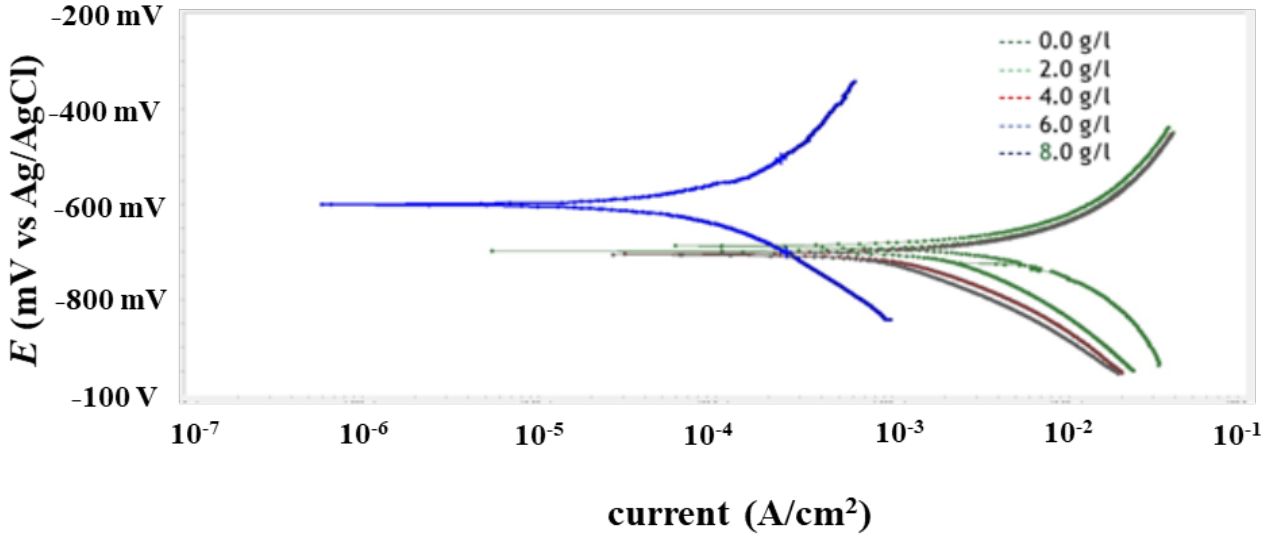


Figure 3. Tafel curves for Al / 0.5 M HCl at different extract concentrations

The straight Tafel lines generated by the cathodic polarisation curves show that the hydrogen evolution process is activation regulated. So, extrapolating the cathodic linear area back to the corrosion potential yields correct estimates of the corrosion current density values. For aluminium in HCl, various organic inhibitors have shown a similar fitting technique. However, the anodic domain makes it challenging to identify the linear Tafel zone since A. Odora extract significantly slows down the anodic reaction of the corrosion process in HCl solution. This finding suggests that *A. Odora* extract primarily functions as an inhibitor of the cathodic kind. The parallel cathodic polarisation curves further imply that the mechanism of the hydrogen evolution process is unaffected in the presence of A. Odora extract.

Table 3. Potentiodynamic polarization parameters for Al in 0.5 M HCl in the absence and presence of different extract concentrations

Inhibitor Conc. (g/L)	E_{corr} (mV/SCE)	β_a (mV/dec)	$-\beta_c$ (mV/dec)	i_{corr} (mA/cm^2)	η (%)
0.0	-687	0.3533	0.399	11.5	
2.0	-698	0.2195	1.0E+15	10.4	9.56
4.0	-704	0.1481	1.0E+15	5.17	55.04
6.0	-706	0.1229	13950.00	3.61	68.61
8.0	-600	0.2340	2.44E-01	0.12	98.97

The results reveal that the i_{corr} value is greater in 0.5 M HCl, but when the concentration of *A. Odora* extract increased, the i_{corr} value decreases and the ɳ % value increases. This may be because an inhibitor molecule has adhered to the metal/acid interface. At 2.0 g/L, the largest i_{corr} value drop (10.4 mA/cm^{-2}) and maximum ɳ (98.97 %) are seen, confirming that *A. Odora* extract is a more effective inhibitor for aluminium in acid solution. It can be seen from the table that in the presence of *A. Odora* extract E_{corr} values move to a more negative direction with somewhat altered values and also exhibit a significant change in the value of the cathodic Tafel constant at various concentrations. This finding indicates that *A.Odora* extract may be configured as a cathodic-type inhibitor (Chaubey et al., 2018).

3.2.2 Electrochemical impedance measurement

Figure 4a, the Nyquist plots obtained of Al in 0.5 M HCl in the absence and presence of different doses of *A.Odora* extract. A capacitive time constant at high frequencies and an inductive time constant at low frequencies make up the Nyquist plot. Uninhibited systems show a modest diameter, but when of A. Odora extract concentration rises, and the diameter grows owing to rising resistance. The charge transfer mechanism based on either direct electron transfer at the metal surface or conduction of electrons through the surface coating is where the capacitive time constant arises. It is generally agreed upon that the dielectric characteristics of a surface layer account for the first time constant about an electric double layer and charge transfer resistance. However, adsorbed charged intermediates induce an inductive time constant to appear. More prominent spectra are seen when the intermediates are heavily adsorbed. According to Chaubey *et al*., (2018), the phenomena may be caused by the relaxation of adsorbed species like H_{ads}^{+}. However, other publications asserted that intermediates of relaxation deposited on the electrode surface include inhibitor species, oxygen ions, or Cl^{-}.

Additionally, inductive behaviour may be seen in the pitting active state and be explained by changes in the salt film's properties and surface area, as well as by the re-dissolution of the oxide layer's surface at low frequencies. No changes in the form of the spectra are seen with the addition of A. Odora extract, and they remain the same at all concentrations that have been tested, thus demonstrating that the corrosion process was not altered by the addition of the inhibitor. An analogous circuit is used to examine the data (depicted in **Figure 4b**). R_s stands for solution resistance, R_{ct} for charge transfer resistance, R_L and L for inductive elements for constant phase element in this circuit. The following equation may be used to determine the polarisation resistance R_p:

$$R_P = \frac{R_t \times R_L}{R_t + R_L} \qquad \textbf{Eqn. 4}$$

Then the inhibition efficiency (ɳ %) is calculated from R_P using the following relation:

$$\eta(\%) = \frac{R_{P(inh)} \times R_P}{R_{P(inh)}} \times 100 \qquad \textbf{Eqn. 5}$$

respectively, R_P and $R_{P\ (inh)}$ represent the charge transfer resistance in the absence and presence of an inhibitor.

When the charged metal surface and the solution are considered, the double layer is referred to as an electrical capacitor. Referring to **Table 4**, the explanation for this is the creation of a protective layer on the electrode surface. The inhibitor molecule adsorbs on the metal surface and reduces its electrical capacity by displacing the water molecules and adsorbed ions on the surface. Due to the of *A. Odora*

extract molecule's electrostatic adsorption on the metal surface, this protective layer's thickness grows as the inhibitor concentration rises (Khadraoui *et al*., 2016). Comparing the corrosion inhibition performance of A. Odora extract with earlier reported works: synthesized corrosion inhibitors (I.B. Obot *et al*., 2009; Sumayah Bashir *et al.,* 2020; Hongyu Cen *et al.,* 2019; Tarek A. Yousef *et al.,* 2022; Abdulbasit *et al*., 2023) as well as plant extracts (Charitha *et al*., 2017, Hossain *et al*., 2021 & 2022), was found that A. Odora extract is most effective and cost-effective corrosion inhibitor for Al metal in 0.5 M HCl.

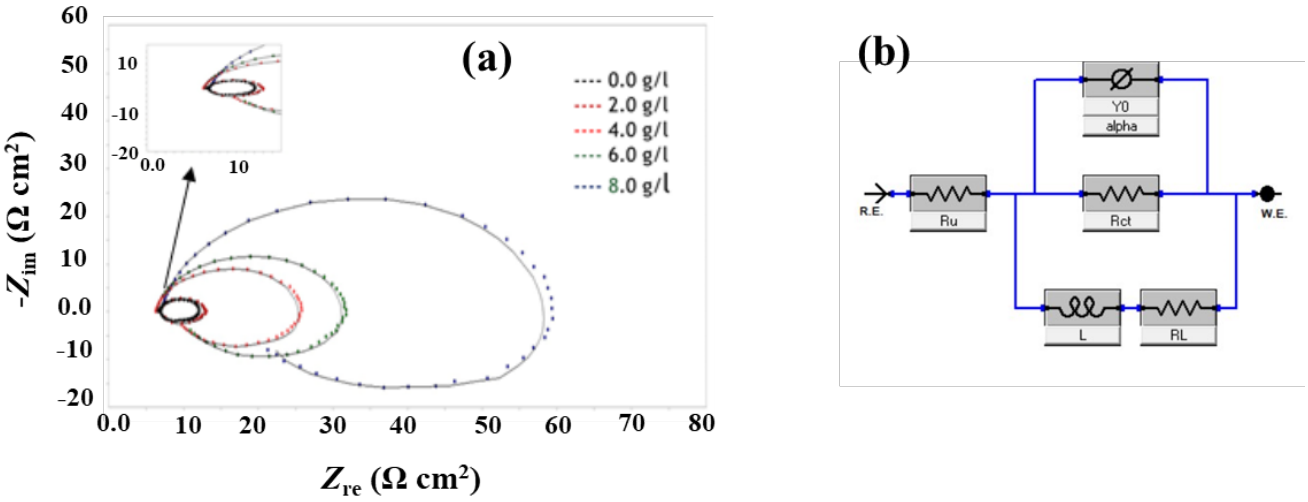


Figure 4. Nyquist plot of Al in 0.5 M HCl in the absence and presence of *A. Odora* extract (a) and equivalent circuit used for the analysis of impedance spectra (b)

Table 4. Electrochemical impedance parameters for Al in 0.5 M HCl in the absence and presence of different concentrations of *A. Odora* extract

Inhibitor Conc. (g/L)	R_u (Ωcm^2)	R_{ct} (Ωcm^2)	R_L (Ωcm^2)	R_p (Ωcm^2)	ɳ (%)
0.0	6.729	5.132	0.43	0.40	
2.0	6.835	5.888	0.59	0.53	25.17
4.0	6.33	19.18	4.06	3.35	88.09
6.0	6.364	24.95	5.28	4.36	90.84
8.0	6.98	51.88	19.83	14.35	97.22

3.3 Adsorption isotherm

The adsorption isotherm analysis signifies about the interaction of the corrosion inhibitor with the metal surface. Langmuire adsorption isotherm is the most important isotherm for analysis of organic corrosion inhibitor performance (Haque *et al.,* 2020) that plotted between the experimental surface coverage (θ) versus inhibitor concentrations (C), as following the Langmuir equation:

$$\frac{C}{\theta} = \frac{1}{K_{ads}} + C$$ **Eqn. 6**

The K_{ads} stands for adsorption constant which was calculated the form the intercept. The obtained Langmuir isotherm found a well-fitted plot with a regression coefficient ($R^2 = 0.9911$) **(Figure 5)**. The result reveals that *A. Odora* extract adsorbs on Al surface and forms monolayer protective film that reduces the corrosion process. The richness of *A. Odora* extract in various components bioactives as discussed in literature, three major compounds are: alocasin A **(1)** (Zhu et al. 2012), hyrtiosin B **(2)** (Elsbaey *et al.* 2016; Zhu *et al.* 2012), and hyrtiosulawesine **(3)** (Zhu *et al.* 2012; Ha *et al.* 2022) and others at various contents explained the excellent inhibitory effect against the Al corrosion in HCl solution. Several researchers introduced the synergistic intermolecular effect to adsorb easily at the metal surface and then avoided the arrival of aggressive H^+ ions (Lrhoul *et al.* 2023; Mu'azu *et al.* 2022; El Ouasif *et al.* 2016; Bammou *et al.* 2010).

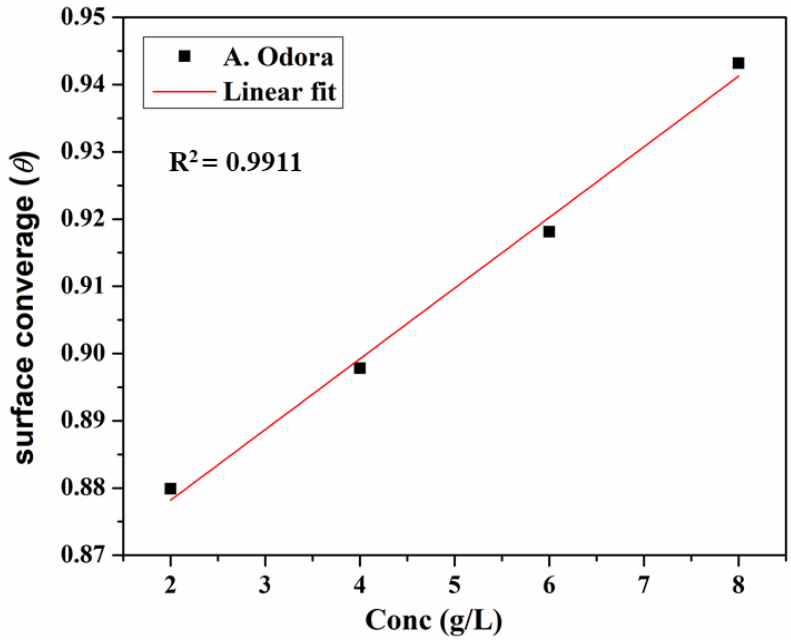
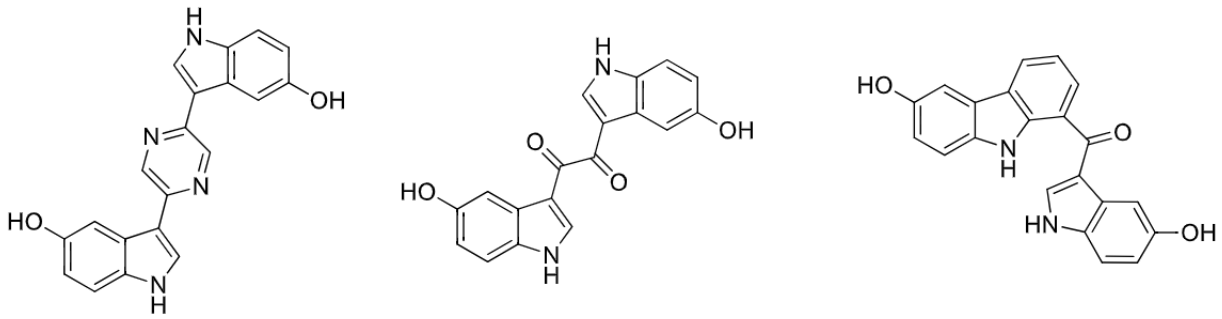


Figure 5. Langmuir isotherm of Al / 0.5 M HCl at different concentrations of *A. Odora* extract 307 K



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The molecular structure of these biomolecules incited researchers to synthesis hyrtiosulawesine, alkaloide, and their derivatives which also favors the formation of metal complexes (Szabó *et al.* 2019; Zhu *et al.* 2012). Hyrtiosulawesine showed antiproliferative activity against human nasopharyngeal carcinoma epithelial as well as antioxidant activities (Zhu *et al.* 2012; Arbain *et al.* 2022). The presence

of heteroatoms like O, N near aromatic rings and ketones, plays a major role in adsorption phenomenon at the metal surface (Alamiery *et al.* 2021; Loukili *et al.* 2022).

4.5 Surface analysis

To determine the extract molecules' corrosion inhibition effect on the metal surface, scanning electron microscope pictures were captured (Bangera *et al*., 2023; Haque *et al*., 2023). The Al surface immersed in HCl with the presence of 8.0 g/L *A. Odora* extract (**Figure 6b)** appeared to be smoother and more uniform compared to the absence of inhibitor for both 3 hours **(Figure 6a)** indicating that A.Odora extract was able to slow down the corrosion process.

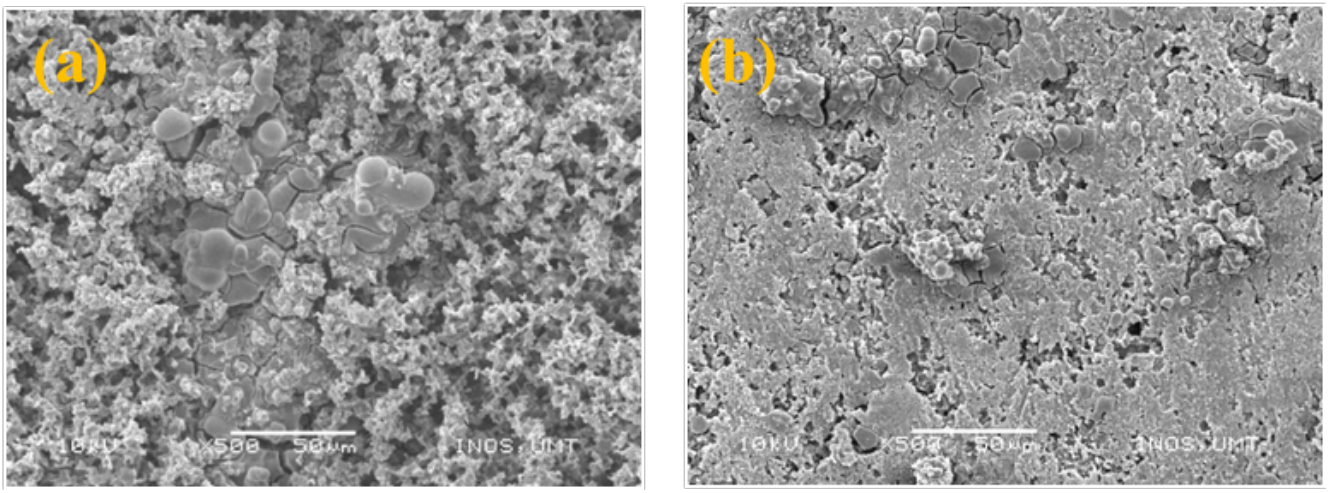

Figure 6. SEM image of Al surface (a) immersed in 0.5 M HCl for 3 hours (b) immersed in 0.5 M HCl in the presence of 8.0 g/L *A. Odora* extract for 3 hours

UV Visible spectroscopy was analyzed of *A. Odora* inhibition in 0.5 M HCl before and after immersion of Al metal. The spectra of before and after immersion of Al show superimposing each other **(Figure 7),** revealing that the A. Odora extract does not interact with dissolved Al^{+3} ions.

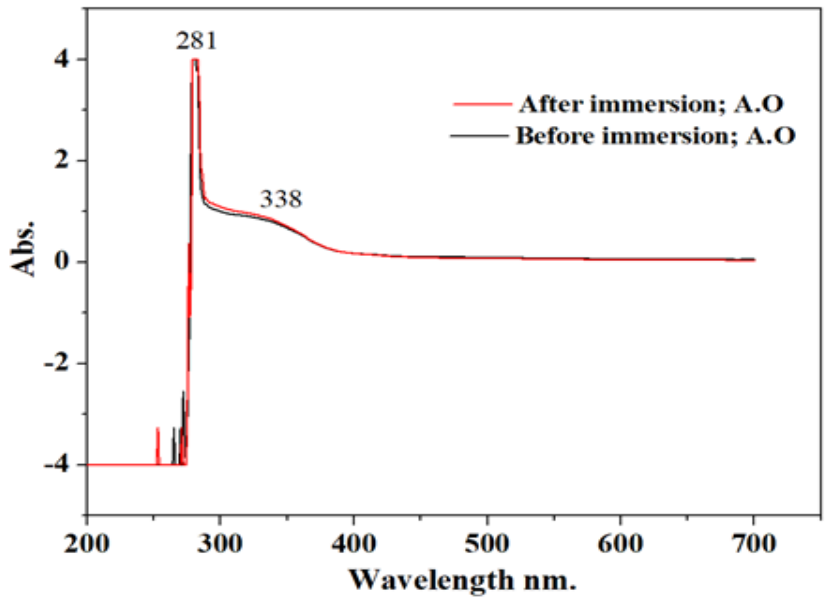

Figure 7. UV visible analysis of *A. Odora* inhibition in 0.5 M HCl before and after immersion of Al metal.

Conclusion

1. The *A. Odora* extract's effectiveness as an inhibitor increased with concentration, peaking at 94.32% at 8.0 g/L.

2. Tafel polarization indicates that *A. Odora* extract is arranged as cathodic type inhibitor.

3. EIS study confirms that an increase in R_{ct}. According to EIS, which is explained by a fall in the local dielectric constant and/or a rise in electrical double layer thickness brought on by the adsorption inhibitor molecules at the metal/solution interface.

4. The SEM analysis showed the formation of a protective film on the aluminium surface in the inhibited system.

5. The adsorption of *A. Odora* extract on Al surface follows the Langmuir adsorption isotherm.

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