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Bitter Leaf (*Vernonia amygdalina*) as a Green Corrosion Inhibitor for Protection of Metals and Alloys-A Review

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Abstract:

Corrosion is the deterioration of a metal by a chemical attack or reaction with its environment. Bitter Leaf (*Vernonia amygdalina*) can control the corrosion of various metals and alloys, such as aluminum, carbon steel, mild steel and stainless steel. Various techniques like the weight loss (WL) method and electrochemical methods such as potentiodynamic polarization (PDP) and electrochemical impedance spectroscopy (EIS), have been used to evaluate the corrosion inhibition efficiency (I.E.) of *Vernonia amygdalina*. The protective film has been analyzed by Fourier-transform infrared spectroscopy (FT-IR), Gas chromatography mass spectroscopy (GC-MS), UV-visible spectroscopy (UV-Vis.) and scanning electron microscopy (SEM) methods. Adsorption of *Vernonia amygdalina* on metal surfaces obeys the Langmuir, Frumkin, Freundlich, Temkin or Flory-Huggins depending on the nature of metal and the corrosive environment. A polarization study reveals that *Vernonia amygdalina* can function as an anodic or mixed type of inhibitor.

1. Introduction

Corrosion of metals/alloys, which can be defined as the deterioration or disintegration of materials due to their reaction with the environment, has continued to receive attention in the technological world. It is a constant and continuous problem, often difficult to eliminate completely. Recently, a study revealed that corrosion causes economic losses of about 2.5 trillion US dollars annually, constituting almost 3.4% of the worldwide GDP (Verma *et al.*, 2018). As corrosion is associated with economic and safety issues, it should be highly addressed by researchers worldwide. Metals and alloys are exposed to hostile environments during their industrial usage, including manufacturing, processing, and transportation, accelerating their degradation. Acid solutions are extensively used in industry, the most important of which are acid pickling, industrial acid cleaning, acid descaling and oil well acidizing. The commonly used acids are hydrochloric acid, sulphuric acid, nitric acid, etc. Since acids are aggressive, inhibitors are usually used to minimize the corrosive attack on metallic materials. There are numerous methods for controlling the corrosion of metals but the use of inhibitors is still one of the best methods of protecting metals against corrosion (Ita *et al.*, 2004; Chetouani *et al.*, 2004; Wang *et al.*, 2007).

A corrosion inhibitor is generally referred to as a chemical substance that, when applied in small quantities to a corrosive medium, reduces the rate of corrosion of a metal or a metal alloy (Roberge *et al.*, 2000; Dafali *et al.*, 2002; Zarrouk *et al.*, 2012; Zarrok *et al*., 2012). The use of chemical inhibitors for prevention of corrosion of metals has been giving researchers a lot of concern in view of its toxicity and poisoning of living organisms in the environment. In view of this harmful effect of chemical inhibitor, the use of plant extracts as an inhibitor of corrosion is being considered. Green corrosion inhibitors are biodegradable and do not contain heavy metals or other toxic compounds. The use of plant extracts to prevent corrosion has become important because they are environmentally acceptable, less toxic, readily available and renewable source for a wide range of needed inhibitors (Alinnor and Ukiwe, 2012; Elmsellem *et al.*, 2014; Abdallah *et al.*, 2021; Hernandez *et al.*, 2019; Abdallah *et al.*, 2021; Loukili *et al.*, 2021; Hbika *et al.*, 2023). These inhibitor molecules comprise heterocyclic compounds with polar functional groups (e.g., N, S, O, and P) and conjugated double bonds with different aromatic systems. Basically, these substances adsorb on the metal surface to block the destruction reaction with aggressive media. Corrosion inhibitors frequently work as anodic, cathodic or mixed inhibitors (Pacheco *et al*., 2011, Liu and Shi, 2009) by adsorbing themselves on the metallic surface (physical adsorption) by forming a film layer on the surface. Temperature is an important factor influencing the phenomenon of corrosion on metal surfaces. Similarly, the immersion time is another factor that could modify inhibition efficiency (I.E%).

Vernonia amygdalina (VA) is a member of the Asteraceae family and its genus is Vernonia which is a small shrub that grows in the tropical Africa and Asia (Nwabanne and Okafor, 2012, Alinnor and Ukiwe, 2012, Ijeh *et al*., 2011). The full binomial name is *Vernonia amygdalina* Del. VA is comonly called "bitter leaf" because of its bitter taste. VA is commonly called ewuro and edidot or bitter leaf in Ibadan and Cross River State in South West and South – South Nigeria respectively (Bonsi *et al.*, 1995). It is planted through the root system in the first quarter of the year and can be harvested after one year when it is fully grown and matured (Erasto *et al.*, 2007). VA leaves are medium to dark green, usually measuring 10-15 cm in length and 4-5 cm in weidth. No seeds are produced and the tree has therefore to be distributed through cutting. VA is an annual, erect, branched and hairy herb, having a height of 30 -120 cm. (Alinnor and Ukiwe, 2012). The leaves of the plant are simple and alternate. The plant is geographically distributed in West African countries including Nigeria. Vernonia amygdalina leaf is shown in **Figure 1**.

Figure 1. Vernonia amygdalina plant (Debi *et al.*, 2013)

Traditional uses of Vernonia amygdalina

The leaves are used either as a vegetable (macerated leaves in soups) or aqueous extracts to treat various illnesses (Opata *et al.*, 2006). Extracts from the leaves and roots are also used in the indigenous treatment and management of several diseases such as malaria and diabetes (Mohammed *et al.*, 2015, Yedjou *et al.*, 2008), anticancer (Joseph *et al.*, 2021), antimicrobial (Erasto *et al.*, 2006), antileishmanial (Tadesse *et al.*, 1993), antifertility (Steenkamp *et al.*, 2003), anti-inflammatory, antipyretic (Asante *et al.*, 2019), analgesic (Njan *et al.*, 2008), appetizer (Iwu et al. 1986), laxative (Igile *et al.*, 1994), oxytocin (Kamatenesi-Mugisha *et al.*, 2003), antifungal (Wedge *et al.*, 2000), anticancer (Edo *et al.*, 2023) and wound healing (Giday *et al.*, 2003). Among all these reported traditional usages, its usage as antidiabetic agent is widely acclaimed (Mohammed *et al.*, 2015, Akah *and* Okafor, 1992). It is also used in the treatment of a variety of ailments ranging from emesis, nausea, diabetes, loss of appetite, dysentery and other gastrointestinal tract problems (Mohammed *et al.*, 2015). Recently, *Vernonia amygdalina* is also been used as a green corrosion inhibitor for protection of metals and alloys.

2. Methodology

Corrosion inhibition of different metals and alloys in various medium by VA as an inhibitor was shown in **Table1**.

Table 1. Corrosion inhibition of metals and alloys in different media by Venonia amygdalina as an inhibitor.

Metal / Alloy	**Medium + Additive**	**Techniques used**	**Findings**	**I.E. max. (in %)**	**Reference**
Aluminium	1.0 M NaOH	WL with temperature, PDP, SEM, FT-IR, RSM.	Mixed type of inhibitor. Langmuir, and Frumkin adsorption isotherms.	80.54 WL	Omotioma *et al.*, 2015
Aluminium	0.5 N H_2SO_4	WL with temperature.	Langmuir adsorption isotherm.	62.92 WL	Meena et al., 2023
Aluminium	1 M HCl	WL with temperature.	Langmuir adsorption isotherm.	--	Dauda *et al.*, 2013
AA1100 Al-Alloy	0.5 M HCl	WL with time.	Langmuir adsorption isotherm.	99.0 WL	Oluseyi *et al.*, 2012
AA1100 Al-Alloy	0.5 M HCl	WL with time.	Langmuir adsorption isotherm.	100.0 WL	Ajanaku et al., 2010
AA1060 Al- Alloy	1 M HCl	WL with temperature.	Langmuir adsorption isotherm.	99.1 WL	Nwagbo *et al.*, 2023
AA1060 Al- Alloy	1 M HCl	WL with temperature.	Langmuir and Flory – Huggins adsorption isotherms.	99.10 WL	Alinnor *et al.*, 2012
Al-Si Alloy	0.5 M NaOH	WL with time.	Physical adsorption	87.0 WL	Ayeni *et al.*, 2012
AA7075 Al- Alloy	0.5 M HCl and 0.5M NaOH	WL with time.	Good corrosion inhibitor.	88.9 WL in HCl, 82.1 WL in NaOH	Kasim *et al.*, 2014
Aluminium alloy	0.5 M HCl	WL with time and temperature.	Anodic-type of inhibitor.	62.0 WL	Abdulwahab *et al.*, 2012
AI-1.0 Wt. % Zn alloy	0.5 and 1.0 M HCl	WL with time.	Good inhibitor.	--	Ogbu *et al.*, 2017
Aluminium alloy	2.0 M HCl	HE, SEM.	Temkin adsorption isotherm.	100.0 HE	Omotosho *et al.*, 2012
2S and 3S Al Alloy	0.1 M HNO_3 and 0.1 M HCl	WL	---	49.5 WL in 0.1 M HCl and 72.5 WL in HNO_3	Gregory *et al.*, 2003

Carbon Steel	3 M HCl	WL with time.	Langmuir and Frumkin adsorption isotherms.	96.22 WL	Emmanuel *et al.*, 2023
Mild Carbon Steel	1 M HCl	WL, SEM, FT-IR.	Langmuir adsorption isotherm.	78.12 WL	Olawale *et al.*, 2017
Mild Steel	1.5 M H_2SO_4	WL with time and temperature, HE, FT-IR, GC-MS.	Langmuir adsorption isotherm.	90.0 WL	Awe *et al.*, 2015
Mild Steel	0.2 M H_2SO_4	WL, OCP, ANOVA	--	60.68 WL	Loto *et al.*, 2013a
Mild Steel	0.5 to 2.0 M H_2SO_4	WL	I.E. increases with increase in the concentration of the inhibitor.	--	Oriji *et al.*, 2021
Mild Steel	HCl	WL,TM, PDP, GC-MS.	Mixed-type of inhibitor.	85.4 WL	Onukwuli *et al.*, 2019
Mild steel	1.0 M H_2SO_4	WL with temperature.	I.E. increases with increase in the temperature.	66.0 WL	Okediran *et al.*, 2023.
Mild Steel	1 M HCl and 0.5 M H_2SO_4	WL with time, PDP, EIS, SEM, DFT, FT-IR, HOMO-LUMO.	Mixed-type of inhibitor. Langmuir adsorption isotherm.	98.0 WL in 1 M HCl & 96.0 WL in 0.5 M H_2SO_4	Adindu *et al.*, 2016
Mild Steel	1 M HCl	WL with time, PDP.	Anodic inhibitor.	95.14 WL	Olisakwe *et al.*, 2023
Mild Steel	1 M HCl	WL with time and temperature, FT-IR, SEM.	Efficient inhibitor.	82.22 WL	Oyedeko *et al*, 2021
Mild Steel	1 M HCl	WL with temperature.	Efficient inhibitor.	84.0 WL	Okediran *et al.*, 2022
Mild Steel	2 M H_2SO_4	GM with time.	Freundlich adsorption isotherm.	100.0 GM	Ajayi *et al.*, 2011
Mild Steel	1 M H_2SO_4	WL with time and temperature, SEM.	Efficient inhibitor.	85.0 WL	Achebe *et al.*, 2015
Mild Steel	1.5 M H_2SO_4	WL with time.	Langmuir adsorption isotherm.	76.0 WL	Udoisoh *et al.*, 2024
Mild Steel	0. 2 M H_2SO_4	WL with time and temperature.	Langmuir, Temkin, Frumkin and Flory-Huggins adsorption isotherms.	38.59 WL	Nwabanne *et al.*, 2012
Mild Steel	3.5 M NaCl	WL with time, OCP, ANOVA.	Efficient inhibitor.	90.08 WL	Loto *et al.*, 2013b
Mild Steel	0.5 M HCl and H_2SO_4	WL with temperature, OCP.	Efficient inhibitor.	--	Loto *et al.*, 2003
Mild Steel	0.1- 0.5 M HCl and HNO_3	WL with time and temperature.	Langmuir adsorption isotherm.	94.13 WL in 0.5 M HCl & 93.94 WL in 0.5 M HNO_3	Ndibe *et al.*, 2011

Mild Steel	1.0 M H_2SO_4	WL with time, PDP, FT-IR, QCC	Mixed-type of inhibitor.	89.11 WL	Daniel *et al.*, 2021
Mild Steel	1.0 to 2.5 M H_2SO_4	WL with time and temperature, GM, TM, FT-IR.	Langmuir adsorption isotherm.	95.75 WL	Odiongenyi *et al.*, 2009
Mild Steel	Sea water	WL with time.	Efficient inhibitor.	--	Tuaweri *et al.*, 2017
Mild Steel	0.4 M HNO_3	WL with time and temperature.	Langmuir, Temkin, Frumkin and Flory-Huggins adsorption isotherms	50.74 WL	Nwabanne *et al.*, 2011
Mild Steel	1.0 M HCl	WL with time and temperature.	Langmuir and Temkin adsorption isotherms.	52.55 WL	Umarani *et al.*, 2021
Mild Steel	3.5 % NaCl	WL with temperature, FT-IR.	Efficient inhibitor.	75.0 WL	Debi *et al.*, 2013
Stainless Steel	2.5 M H_2SO_4	WL with time.	Efficient inhibitor.	69.0 WL	Obiukwu *et al.*, 2013

Abbrevations: EFM: electrochemical frequency modulation, **ENA**: electrochemical noise analysis, **DFT**: density function theory, **GM**: gasometric method, **HE**: hydrogen evolution, **HOMO:** highest occupied molecular orbital, **LUMO**: lowest unoccupied molecular orbital, **MDS**: molecular dynamics simulation, **MD**: molecular dynamics, **MCD**: mulliken charge distributions, **OCP:** open circuit potential, **RSM:** response surface method, **QCC:** quantum chemical calculation.

Gas chromatography mass spectrometry (GC-MS) study

Onukwuli et al. (Onukwuli and Omotioma, 2019) studied of corrosion inhibition of Mild steel in HCl solution by VA as corrosion inhibitor. They carried out GC-MS spectra of the VA shown in **Figure 2** which indicates various levels of peaks.

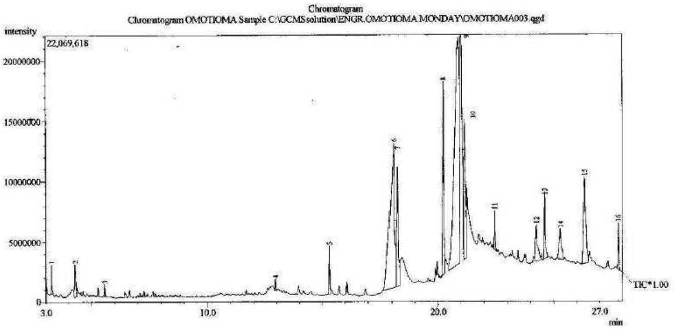


Figure 2. GC-MS chromatogram of the bitter leaves extract (Onukwuli and Omotioma, 2019).

The peaks represent various compounds as determined by GC-MS spectra. The analysis revealed the presence of C_6H_8O (96 g/mole: 2,4- Hexadienal; Sorbaldehyde , n-Hex-2,4-dienal, Hexa-2,4-dienal; Sorbic aldehyde 1,3-Pentadiene-1-carboxaldehyde; 2,4-Hexadien-1-al; 2,4-Hexadienal; trans,trans-2,4-Hexadienal; 3-Propyleneacrolein); C_7H_{12} (96 g/mole: 3,4- Heptadiene; 1,3-Diethylallene); $C_{10}H_{18}O_2$ (170 g/mole: 2-Decenoic acid). Other compounds present in the extract include $C_9H_{16}O_2$ (156 g/mole: 2- Nonenoic acid; trans-2-Nonenoic acid; Nonylenic acid, 2-Nonenylic acid); $C_{12}H_{14}N_2O_6$ (282g/mole: Phenol, 2-(1-methylpropyl)-4,6-dinitro-, acetate; Acetic acid, 2-(sec-butyl)-4,6-dinitrophenyl ester); $C_{12}H_{18}O$ (178 g/mole: Benzene ethanol; 2-(3-Isopropylphenyl)-1-propanol); $C_{18}H_{34}O_2$ (282 g/mole: Oleic Acid, 9-Octadecenoic acid; cis-9-Octadecenoic Acid); $C_{18}H_{34}O$ (266 g/mole: 13-Octadecenal; cis-13-Octadecenal) and $C_{16}H_{30}O$ (238g/mol: cis-9- Hexadecenal; 9-Hexadecenal (Onukwuli and Omotioma, 2019).

Potentiodynamic polarization (PDP) Study

The polarization curves for mild steel in (a) 1 M HCl and (b) 0.5 M H_2SO_4 in the absence and presence of different concentrations VA are given in **Figure 3** (Adindu *et al*., 2016).

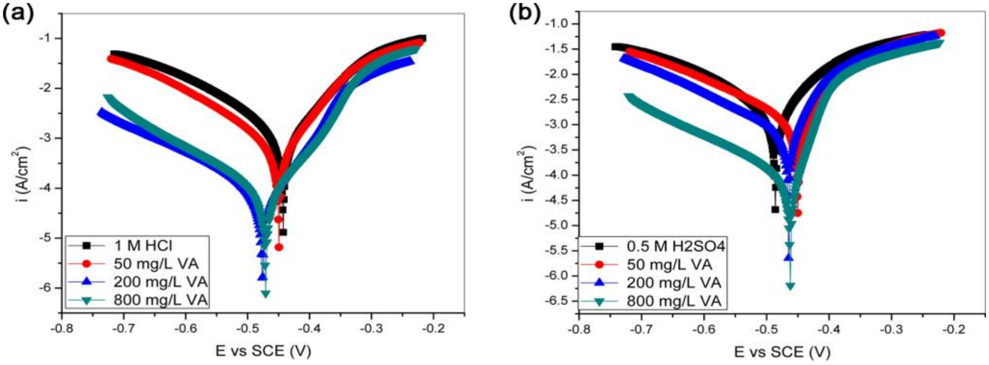


Figure 3. Potentiodynamic polarization curves for mild steel corrosion in (a) 1 M HCl and (b) 0.5 M H_2SO_4 solutions without and with VA ethanol extract (Adindu *et al*., 2016).

The results show that VA modified both the anodic and cathodic reactions, and displaced the corrosion potential (E_{corr}) slightly towards cathodic values in 1 M HCl and slightly towards the anodic values in 0.5 M H_2SO_4 while reducing both the cathodic and anodic current densities inhibitor in both 1 M HCl and 0.5 M H_2SO_4 (Noor and Al-Moubaraki, 2008).

Electrochemical Impedance Spectroscopy (EIS) study

The effect of various concentrations of VA on Nyquist diagrams for mild steel corrosion in 1 M HCl solution is shown in **Figure 4** (Adindu *et al*., 2016). The Nyquist plots show single capacitive semi-circles in the high frequency region, corresponding to just one time constant as observed in the Bode plots.

Fourier-transform infrared spectroscopy (FT-IR) study

Odiongenyi et al. (Odiongenyi *et al.*, 2009) studied the inhibitive effect of VA on the corrosion of MS in 1.0 to 2.5 M H_2SO_4. In this study, the VA extracts were characterized using FT-IR spectra was shown in **Figure 5**. From **Figure 5**, it is seen that the extract exhibited broad adsorption band at 3402.06 cm^{-1} (peak height = 56.880 cm), indicating the presence of alcohol or phenol functional group (i.e. –OH). An adsorption band was also found at 1046.01 cm^{-1} (peak height = 87.459 cm), suggesting the presence of –CO stretch. The adsorption band at 633.61 cm^{-1} (peak height = 92.300 cm) suggests the presence of carbon-carbon triple bond (i.e., alkyne type of compound).

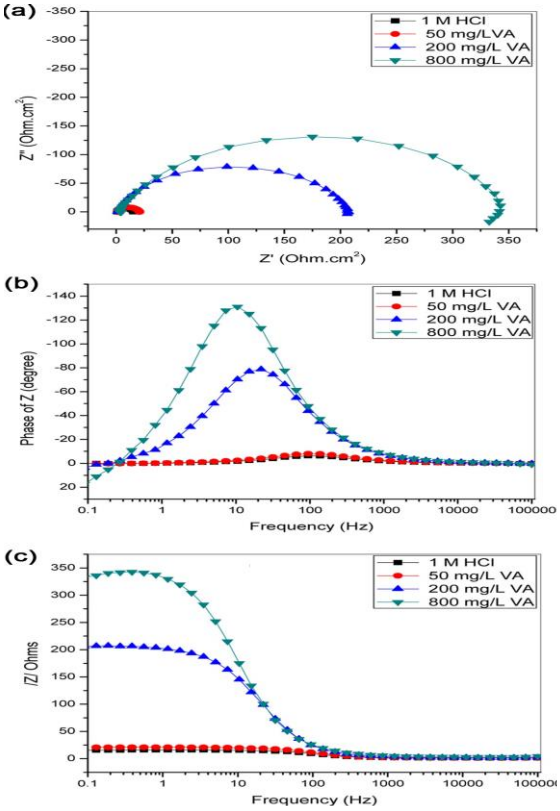


Figure 4. EIS of mild steel corrosion in 1 M HCl solution without and with VA ethanol extract (a) Nyquist (b) Bode phase and (c) Bode modulus plots (Adindu *et al*., 2016).

However, in the presence of the inhibitor, IR spectrum of the corrosion product revealed that the –OH stretch (3407.06 cm^{-1}) was shifted to 3435.22 cm^{-1} and the C=O stretch (1046.01 cm^{-1}) was shifted to 1632.74 cm^{-1}, while C-H bend (633.61 cm^{-1}) was missing, indicating that there is interaction between the ethanol extract of *vernonia amygdalina* and the surface of mild steel (Odiongenyi *et al*., 2009).

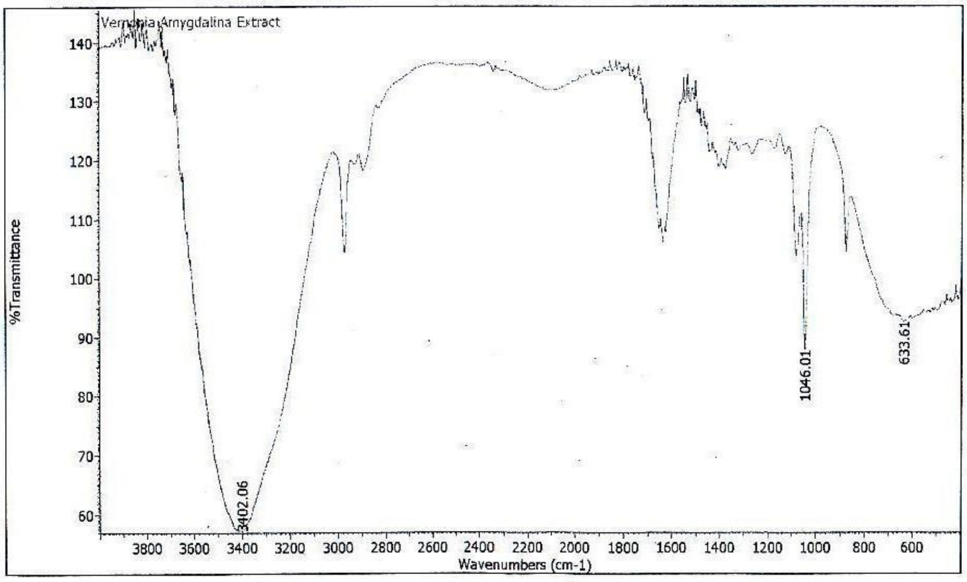


Figure 5. FTIR spectrum of ethanol extract of *Vernonia amydalina* (VA) alone (Odiongenyi *et al*., 2009)

Scanning Electron Microscopy (SEM) Study

The surface morphology of the corroded samples in the presence and absence of VAE was examined (Olawale *et al*., 2017) is shown in **Figure 6** shows that the mild steel sample immersed in 0.1 M HCl without inhibitor corroded more than the mild steel sample exposed to degradation in the presence of VAE inhibitor.

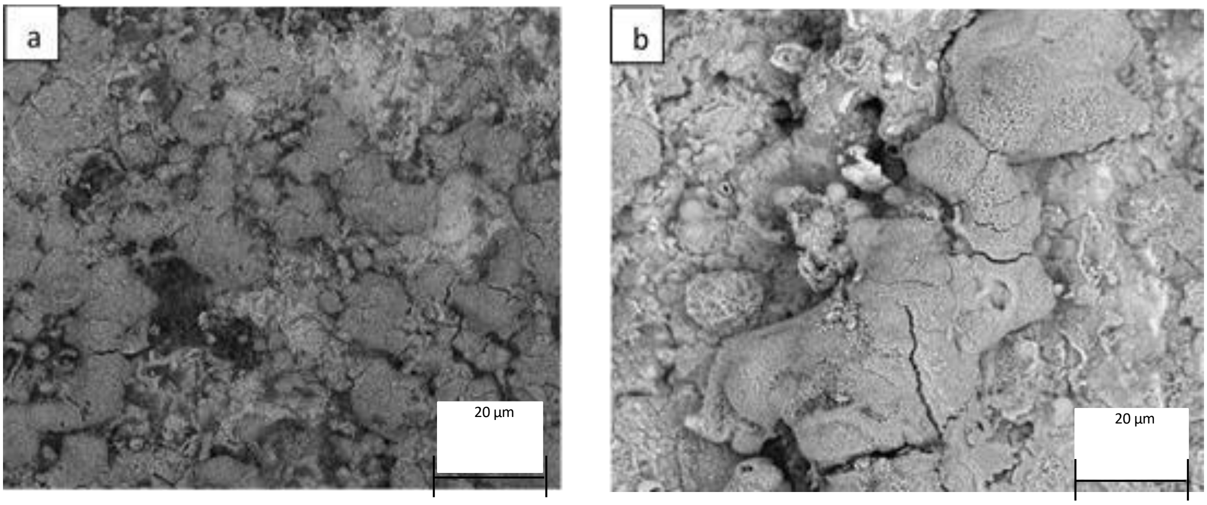


Figure 6. SEM images of mild steel coupon in 1 M HCl solution after 360 h of total immersion (a) in absence and (b) in presence of 0.8 g/L VAE inhibitor (b*)* (Olawale *et al*., 2017)

The white patches on the surface of the samples represent the protective oxide. Typically, its formation was partly dependent on the inhibitor chemical composition and molecular structure, and the substrate charged surface (*i.e.* the oxide affinity for the metal surface) (Singh *et al*., 2012).

Computational study

Adindu et al. (Adindu *et al*., 2016) studied the corrosion protection of mild steel in 1 M HCl and 0.5 M H_2SO_4 solutions by ethanol extract of VA. HOMO and LUMO orbitals shown in **Figure 7.** The experimental results already show evidence that the corrosion inhibiting efficacy of VA ethanol extract was achieved via adsorption of some organic constituents of VA on the surfaces of the corroding mild steel specimens. Such adsorption has been shown to involve the overlap of frontier orbitals: the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) and can be modeled by means of quantum chemical calculations and molecular dynamic simulations (MDS) in the frame work of density function theory (DFT).

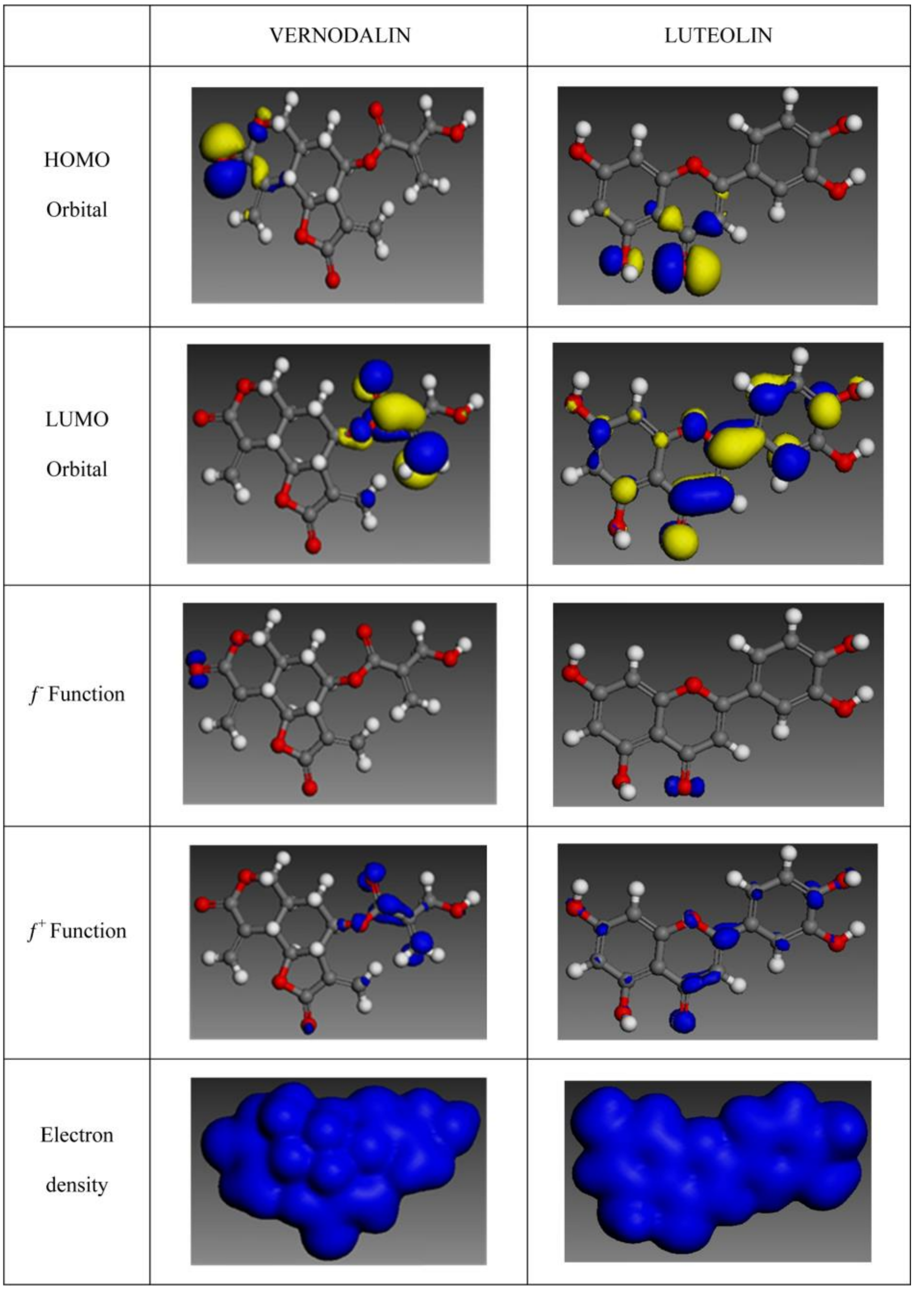


Figure 7. Electronic properties of vernodalin, and luteolin (Adindu *et al*., 2016).

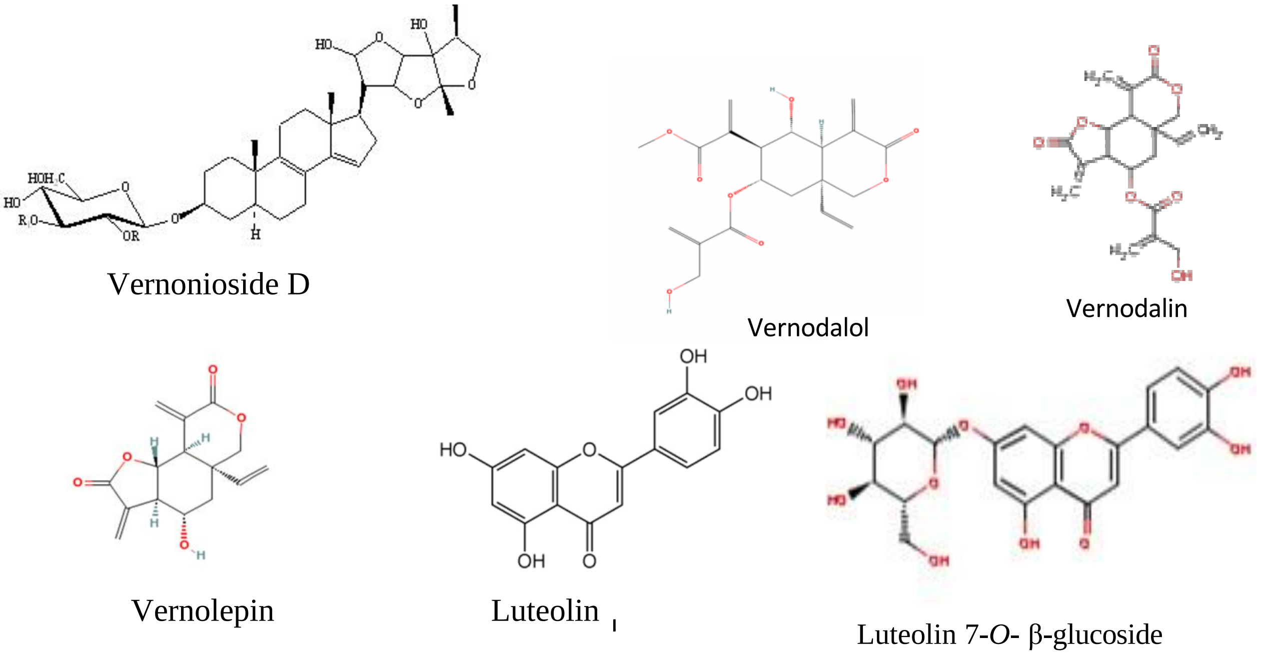
Figure 7 shows the HOMO and LUMO orbitals, Fukui functions for nucleophilic (f^+) and electrophilic (f^-) attack and electron density for vernodalin and luteolin respectively. The f^- functions of the two constituents correspond with the HOMO locations, showing that these are the sites where the constituents can be adsorbed on the mild steel surface, and f^+ correspond with the LUMO locations, showing sites through which the constituents could interact with the non-bonding electrons in the metal. The electron density shows charge distribution saturated all around each molecule; which is suitable for flat-lying adsorption orientations. High values of E_{LUMO} indicate better opportunity for the constituents to donate electrons to the mild steel surface. Accordingly, E_{HOMO} for vernodalin shows that it has a better disposition to donate electron to the mild steel surface than luteolin.

Phytochemical constituents of Vernonia amygdalina

Bitter leaf extract contains phytochemicals such as flavonoids, alkaloids, tannins and phenols (Altemimi *et al*.,2017; Ullah *et al.*, 2020, Nwabanne and Okafor, 2012). Flavonoids include luteolin, luteolin 7-O-β-glucosides and luteolin 7-O-glucuronide (Jisaka *et al.*, 1992). Some of the identified sesquiterpene lactones are vernolide, vernodalol (Erasto *et al*., 2006). Other phytochemicals present in the leaves of VA are terpenes, coumarins, phenolic acids, lignans, xanthones and anthraquinones (Tona *et al.*, 2004). VA leaves also contain vernolepin, vernodalin and hydroxyvernolide (Igile *et al*., 1994). In a study by Oboh et al., (Oboh *et al.*, 2006) it was revealed that VA leaf has high protein (33.3%), fat (10.1%), crude fibre (29.2%), ash (11.7%), mineral (Na, K, Ca, Mg, Zn, & Fe), phytate (1015.4 mg/100g) and tannin (0.6%) content, while it contains low cyanide (1.1 mg/kg).

Mechanism of corrosion inhibition by bitter leaf (*Vernonia amygdalina)*

Ijeh and Ejike 2011 (Ijeh and Ejike, 2011) found some phytochemicals abundantly in the leaves of VA shown in **Figure 8.**



Vernonioside D

Vernodalol

Vernodalin

Vernolepin

Luteolin

Luteolin 7-*O*- β-glucoside

Figure 8. Structures of some phytochemicals in the leaves of *Vernonia amygdalina* (Ijeh and Ejike, 2011)

The inhibitory effect of the bitter leaf extract can be attributed to the presence of various phytochemicals, such as tannins, flavonoids, alkaloids, cardiac glycosides, and saponins (Udoisoh *et al.*, 2024, Umarani and Ngobiri, 2021). These compounds contain functional groups characterized by high electron density and basicity, including oxygen (O), nitrogen (N), and sulfur (S). The electrondonating nature of these functional groups facilitates the formation of coordinate bonds with the metal surface, leading to the adsorption of inhibitor molecules (Alara *et al*., 2019). Particularly, tannins, with their multiple hydroxyl and carboxyl groups, contribute to the development of a dense and stable protective layer on the metal surface, preventing corrosive ion access (Koopmann *et al*., 2020, Das *et al*., 2020). This observation aligns with the findings reported by (Raja *et al*., 2022; Chen *et al*., 2015, El Ibrahimi and Berdimurodov, 2023, Chigondo and Chigondo, 2016) who documented similar adsorption behaviour of plant extracts on metal corrosion in acidic environments.

Conclusion

In this review, various research works on the corrosion inhibition of different metals and alloys in different acidic, neutral, seawater, and alkaline media by *Vernonia amygdalina* as green inhibitor were presented. Langmuir, Freundlich, Frumkin, Flory-Huggins and Temkin adsorption isotherms were observed. *Vernonia amygdalina* extract behaved as an anodic or mixed-type inhibitor. The maximum inhibition efficiency for *Vernonia amygdalina* was found to be 100.0 (WL data). The results obtained from weight loss data were in good agreement with the results obtained from the PDP and EIS methods. Various techniques such as SEM, FT-IR, GC-MS, DFT, UV-Vis. etc. were used to study the corrosion mechanism.

Disclosure statement

Conflict of Interest: The authors declare that there are no conflicts of interest.

Compliance with Ethical Standards: This article does not contain any studies involving human or animal subjects.

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