Research Journal of Chemical Sciences ISSN 2231-606X
Vol. **13(3),** 52-59, October **(2023)** *Res. J. Chem. Sci.*

Review Paper

Natural products as corrosion inhibitors for mild steel in H_2SO_4 solution: A Review

R.T. Vashi
Chemistry Department, Navyug Science College, Rander Road, Surat, India
vashirajendra@yahoo.co.in

Available online at: www.isca.in, www.isca.me
Received 19th May 2023, revised 7th June 2023, accepted 11th August 2023

Abstract

Corrosion is the destructive attack of a metal by chemical or electrochemical reaction with its environment. A percentage of inhibition was carried out by various methods like WL and electrochemical methods such as PDP and EIS. Natural products were used for the protection of MS materials from corrosion as they are biodegradable and eco-friendly. Nature of surface films produce on metal was studied by using various techniques like FT-IR, UV-Visible spectroscopy, EDX, AFM, GC-MS and SEM. The present review paper covers the research works (Period 2017-2018) carried out by various researchers on corrosion inhibition of MS in H_2SO_4 solutions by using a Natural products as green inhibitors.

Keywords: Mild Steel, H_2SO_4, Corrosion, Natural Products, PDP, EIS, SEM.

Introduction

Corrosion is a spontaneous physical or chemical attack on materials due to an interfacial reaction in an ambient environment resulting in the loss of material properties. A severe corrosion condition may result in tremendous economic loss as well as environmental impact to some extent. MS is also considered low carbon steel as it is defined as having a low carbon content of 0.05–0.25%. MS are extensively used in various industrial and manufacturing plants, including petroleum, chemical, petrochemical, construction, pulp and paper, and transportation industries due to its remarkable features such as low cost, easy availability, high strength, durability etc. MS corrosion occurs inevitably during the process of cleaning boilers, acid pickling, and de-scaling. H_2SO_4 is a strong acid and is used as a cleaner for rust, algae and scale from condensers and cooling tower[1]. Sulfuric acid corrodes majority of most alloys and metals.

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[2]. The use of natural products as corrosion inhibitors is of enormous interest because they are environmentally friendly, ecologically acceptable, readily available, inexpensive and can be used nowadays as 'Green Inhibitors'. The inhibitor molecules can also adsorb on the mild steel surface based on donor-acceptor interactions between π-electrons of the aromatic/heterocyclic ring and vacant d-orbitals of surface iron.

In this review study indicates only Langmuir, Temkin, Freundlich adsorption isotherm found. The efficiency of these natural products extracts as corrosion inhibitors was calculated by using WL and electrochemical tests, such as PDP and EIS measurements. The objective of this review is to summarize the results regarding inhibition of MS corrosion in H_2SO_4 solution by using various Natural products.

Phytoconstituents present in Natural products

In this review, most of the Natural products found to contain active phytochemicals such as tannins, amino acids, flavonoids, saponins, phenols and alkaloid which have corrosion inhibitive effect.

Natural products used for inhibition of corrosion of MS in H_2SO_4 solutions are presented in Table-1.

Table-1: Natural products as corrosion inhibitor for MS in H_2SO_4 solution.

Inhibitor	Solution	Techniques(s)	Findings	$I.E._{max}$ (in %)	Ref. No.
Acacia cyanophylla	1 M H_2SO_4	WL, EIS, AFM, FT-IR	Langmuir adsorption isotherm	83.80 WL, 86.95 EIS	3
Adansonia digitata (Baobab)	0.5 M H_2SO_4	WL with time, EIS, PDP and SEM	Mixed inhibitor. Langmuir isotherm.	74.52 WL, 73.82 PDP, 73.97 EIS	4

Aegle marmelos	0.5 M H_2SO_4	WL, PDP, EIS, SEM, AFM, FT-IR, UV-Vis, QCC	Mixed- type of inhibitor. Langmuir isotherm	83.08 WL, 83.08 PDP, 81.85 EIS	5
Agave Americana	0.5 M H_2SO_4	PDP, EIS, SEM, FT-IR	Mixed type inhibitor. Langmuir adsorption isotherm	81.87 PDP	6
Alkana tinctorial	0.5 M H_2SO_4	WL with temp., EIS, SEM, AFM, FT-IR, UV-Vis, QCC	Mixed inhibitor. Langmuir isotherm	90.46 WL 91.63 PDP, 91.42 EIS	7
Alstonia boonei	0.5 M H_2SO_4	WL with temp.	Langmuir isotherm.	83.0 WL	8
Alternanthera bettzickiana	5.0 M H_2SO_4	WL with time and temp.	Langmuir and Temkin isotherm	Leaves92.07WL, Stem89.95WL, Root 88.41WL	9
Armoracia rusticana	0.5 M H_2SO_4	WL, PDP, EIS, SEM, AFM, FT-IR, UV-Vis SE, ATR, QCC	Mixed-type inhibitor. Langmuir adsorption isotherm	93.27 WL 95.74 PDP, 94.04 EIS	10
Artemisia vulgaris	1 M H_2SO_4	WL with time and temp., SEM, EDX	Langmuir adsorption isotherm	71.80 WL	11
Asparagus racemosus	0.5 M H_2SO_4	WL, EIS, PDP, SEM, UV, AFM, FT-IR, QCC	Mixed inhibitor. Langmuir isotherm.	88.85 WL, 93.25 PDP, 90.56 EIS	12
Aster koraiensis	1 M H_2SO_4	WL with temp., PDP, SEM, EDX, FT-IR	Mixed-type of inhibitor. Langmuir isotherm.	90.53 WL	13
Azadirachta indica	1 NH_2SO_4	WL with temp., GC-MS, SEM	Langmuir isotherm.	65.6 WL	14
Bauhinia tomentosa	1 N H_2SO_4	WL with temp., PDP, EIS, SEM, UV-Vis. and FT-IR	Mixed inhibitor. Temkin adsorption isotherm	86.63 WL, 84.17 PDP, 88.47 EIS	15
Black Liquor	0.5 M H_2SO_4	WL with time, PDP, EIS, SEM	Cathodic-type of inhibitor	76.0WL, 95.0 PDP, 89.0 EIS	16
Bombax ceiba Flower	0.5 M H_2SO_4	WL, PDP, SEM.	Mixed inhibitor. Langmuir isotherm	90.84 WL, 96.29 PDP	17
Butea monosperma	0.5 M H_2SO_4	WL, PDP, EIS, SEM, IR, AFM UV	Good inhibitor	87.45 WL, 97.94 PDP, 98.41 EIS	18
Clerodendrum Splendens	0.7, 1.2 and 2.2 M H_2SO_4	WL with temp. & time, SEM, FT-IR	Langmuir adsorption isotherm	87.58 WL	19
Corchorus olitorius	0.5 M H_2SO_4	WL with time and temp., PDP, EIS, GC-MS, SEM	Mixed-type of inhibitor. El-Awady adsorption isotherm.	93.0 WL	20
Coreopsis tinctoria (*C. tinctoria*)	0.5 M H_2SO_4	WL, PDP, EIS, EDX, AAS, SEM, RS	Langmuir adsorption isotherm.	80.62 WL	21
Coriandrum Sativum. L.	0.5 M H_2SO_4	OCP, PDP, EIS, FT-IR, SEM	Mixed inhibitor. Langmuir, Freundlich and Temkin adsorption isotherms.	96.7 PDP, 96.4 EIS	22
Cuscuta reflexa	0.5 M H_2SO_4	WL, PDP, EIS, UV, FT-IR, AFM, SEM and DFT	Mixed-type of inhibitor. Langmuir adsorption isotherm.	95.47 PDP, 93.05 EIS	23
Cyclea peltata	1 N H_2SO_4	WL	Langumir, Temkin and El-awady adsorption isotherm.	91.89 WL	24

Delonix regia	0.1 M H_2SO_4	PDP, SEM, FT-IR, GC-MS	Mixed-type of inhibitor. Langmuir adsorption isotherm.	88.65 PDP	25
*Eriobotrya japonica Lindl. (*Loquat*)*	0.5 M H_2SO_4	WL with temp. & time, PDP, EIS, FT-IR, HPLC, SEM	Cathodic inhibitor. Langmuir adsorption isotherm	96.2 WL, 96.3 PDP, 94.3 EIS	26
Ervatamia coronaria	1 M H_2SO_4	WL, EIS, PDP, SEM, XRD	Mixed-type of inhibitor. Langmuir adsorption isotherm.	89.92 WL, 79.71 PDP, 71.91 EIS	27
Fenugreek	1 M H_2SO_4	WL with temp., SEM, UV-Vis	Langmuir adsorption isotherm.	89.06 WL	28
Ficus racemosa (FR)	0.5 M H_2SO_4	WL with temp., PDP, EIS	Mixed inhibitor. Langmuir isotherm.	--	29
Ficus religiosa	0.5 M H_2SO_4	WL, PDP, EIS, SEM, AFM, FT-IR and UV-Vis, QCC	Mixed inhibitor. Langmuir isotherm.	88.29 WL, 92.26 PDP, 91.80 EIS	30
Gardenia aqualla	1 M H_2SO_4	WL with temp.	Langmuir adsorption isotherm.	89.5 WL	31
Gongronema latifolium	5 M H_2SO_4	WL with temp. and time, GM	Langmuir isotherm.	96.5 WL	32
Ground nut	1 M H_2SO_4	WL with time and temp., RSM, SEM	Langmuir adsorption isotherm	85.9 WL	33
Gum Tragacan powder	1N H_2SO_4	WL with temp, PDP, EIS	Mixed-type of inhibitor. Langmuir adsorption isotherm.	69.05 WL, 70.97 PDP, 86.89 EIS	34
Houttuynia cordata	0.5 M H_2SO_4	WL with temp., PDP, EIS, SEM.	Mixed but predominantly cathodic inhibitor. Langmuir isotherm	89.39 WL, 94.4 PDP, 94.8 EIS	35
Imperrata cylindrica	1 M H_2SO_4	WL with time and temp.	Good inhibitor.	76.58 WL	36
Ipomoea batatas	1.0 M H_2SO_4	WL and GM	Langmuir adsorption isotherm	61.1 WL, 52.8 GM	37
Jatropha tanjorensis	1 M H_2SO_4	WL with temp., HE.	Langmuir adsorption isotherm.	89.73 WL, 51.91 HE	38
Lannea coromandelica	1 M H_2SO_4	WL with temp., PDP, EIS, XRD, FT-IR, SEM and AFM	Mixed-type of inhibitor. Langmuir isotherm.	89.0 WL, 88.5 PDP, 93.8 EIS	39
Linum Usitatissimum	1 M H_2SO_4	WL with time and temp., PDP, EIS, SEM, FT-IR, LU Chromatogram	Mixed-type of inhibitor. Langmuir isotherm.	95.5 WL, 96.5 PDP, 96.5 EIS	40
Luffa Aegyptiaca	0.5 M H_2SO_4	WL with time and temp., Colorometric, PDP, EIS, SE, FT-IR, Optical profilometry	Mixed-type of inhibitor. Langmuir and El-Awade isotherms.	94.29 WL, 92.84 PDP, 93.82 EIS	41
Manihot esculenta (Casava)	0.5 M H_2SO_4	WL with temp., PDP, EDX, SEM, FT-IR	Mixed-type of inhibitor. Langmuir isotherm.	93.2 WL, 83.52 PDP	42
Mentha spicata	2.0 M H_2SO_4	WL temp. & time, SEM, UV-Vis, FT-IR	Langmuir adsorption isotherm.	81.22 WL, 82.58 RN	43
Microdesmis puberula	1 M H_2SO_4	WL with temp., HE	Freundlich adsorption isotherm.	83.25 WL, 82.82 HE	44

*Microdesmis puberul*a	1 M H_2SO_4	WL with temp.	Langmuir adsorption isotherm.	82.05 WL	45
Mimosa pudica	2.0 M H_2SO_4	WL with temp., PDP, SEM, EDX	Frendulich isotherm.	72.89 WL	46
Moringa oleifera	1 N H_2SO_4	WL with time, PDP, EIS, SEM, EDX, FT-IR	Mixed inhibitor.	96.00 WL, 56.57 PDP, 64.00 EIS	47
Musa Sapientum	2.0 M H_2SO_4	WL	--	--	48
Myristica fragrance	0.5 M H_2SO_4	WL, EIS, PDP, UV, IR, SEM, NMR, AFM and QCC	Mixed inhibitor.	83.27 WL, 87.42 PDP, 87.81 EIS	49
Myristica fragrance	0.5 M H_2SO_4	WL, EIS, SEM, UV-Vis	Langmuir adsorption isotherm	87.81 WL	50
Ocimum Canum	0.5 M H_2SO_4	WL with temp., GM	Langmuir isotherm.	89.2GM, 89.0 WL.	51
Ocimum gratissimum (OG)	0.5 M H_2SO_4	WL, SEM, PDP, EIS, FT-IR	Mixed-type of inhibitor. Langmuir isotherm.	77.2 PP, 96.2 EIS	52
Opuntia cochenillifera	0.5 M H_2SO_4	WL, EIS, PDP, SEM, FT-IR, EDX	Mixedinhibitor. Langmuir isotherm.	94.41 WL, 83.79 PDP, 85.18 EIS	53
Pedalium murex L.	0.5 M H_2SO_4	WL, PDP, EIS	Mixed inhibitor predomentely Cathodic. Langmuir isotherm.	90.47 WL, 87.51 PDP, 89.33 EIS	54
Piper Guineense Squeezed	2 M H_2SO_4	WL with time and temp.	---	42.0 WL	55
Pongamia Pinnata	1 N H_2SO_4	WL with time and temp., PDP, EIS, EDX, SEM, FT-IR, GC-MS	Mixed-type of inhibitor. Langmuir, Temkin and Freundlich isotherms.	---	56
Poupartiabirrea back	0.5 M H_2SO_4	WL with time	Efficient inhibitor.	70.0 WL	57
Rice Husk	1 M H_2SO_4	WL with time, XRD.	Langmuir adsorption isotherms.	96.60 WL	58
Saraca ashoka	0.5 M H_2SO_4	WL, EIS, PDP, SEM, UV–Vis., FT-IR, AFM, QCC	Mixed-type of inhibitor. Langmuir isotherm.	89.98 WL, 95.48 PDP, 93.09 EIS	59
Sida acuta	0.7, 1.2, 2.2 M H_2SO_4	WL with time, ANN, FT-IR	--	96.37 WL	60
Sida cordifolia	0.5 M H_2SO_4	WL, PDP, EIS, AFM, SEM	-	--	61
Solenostemon Monostachyus	1 M H_2SO_4	WL with time and temp., HE	Freundlich isotherms.	80.35 WL, 70.53 HE	62
Spathodea Campanulata	1 N H_2SO_4	WL with temp., PDP, SEM	Mixed inhibitor. Langmuir, Temkin and Freundlich isotherms.	82.74 WL	63
Telfairia occidentalis Rind	1 M H_2SO_4	WL with temp., HE, SEM	Langmuir adsorption isotherms.	89.01 WL, 90.94 HE.	64
Tephrosia villosa	0.5 M H_2SO_4	WL with temp., PDP, EIS	Mixed-type of inhibitor predominantly cathodic. Langmuir and Temkin adsorption isotherms.	97.64 WL, 94.12 PDP, 94.06 EIS	65
Tribulus terrestris	0.5, 0.75, 1.0 and 1.5 M H_2SO_4	WL with temp.	Langmuir adsorption isotherms.	0.5N 92.81WL	66

Valeriana wallichii	0.5 M H_2SO_4	WL, PDP, EIS, AFM, SEM, UV-Vis., FT-IR, QCC	Mixed inhibitor. Langmuir isotherm.	93.74 PDP	67
Viscum album	5.0 M H_2SO_4	WL with temp., GM	Temkin isotherm.	99.99 WL	68
Carcia papaya (CP) *and Azadirachta indica* (AZ) *(Neem)* Leaf	1 and 2 M H_2SO_4	WL with time	--	CP79.99WL, AZ75.49WL, CP+AZ 92.20WL	69
Musa paradisiacal (MP*), Azadiratcha indica*(AZ*), Citrullus lanatus*(CL*)*	0.5 M H_2SO_4	WL with time	--	MP68.73WL, AZ98.56WL, CL94.06 WL	70

Solutions: In this Review, H_2SO_4 solution was used as corrosive solutions.

Type of plant materials used as green inhibitors: Different parts of plant such as, Leaves[3,8,13,15,19, 24,26,28,29,31,33,36-39,41,42,47,52, 54,60-63,65,69], Fruits[5,12,30,66], Flower[17], Root[44,67], Stem[20,45,53], Peels [48], Seeds[4,22,40,] and Plant extract[51] were used as corrosion inhibitor.

Extracts used: Various solvents like Distilled water[14,18,22,28,49, 54], Double Distilled water[3,15,24,39,65], Ethanol[5,7,9,12,20,23,30-32,42,44, 46-48,51,52,60,64,68,69], Methanol[4,11,43,57] and NaOH[58] were used for the preparation of plant extract.

Techniques: Various techniques like WL[3,5,10,12,17,18,21,23,24,27,30, 37,48-50,52-54,59,61,67], WL with time[4,9,11,16,19,20,26,32,33,36,40,41,43,47,55-57, 58,60,62,69,70], WL with temp.[7-9,11,13-15,19,20,26,28,29,31,35 36,38-46, 51, 52,55,56,62,63-66,149], GM[32,37,51,68], HE[38,44,62,64], PDP[4-6,10,12,13,15,16-18,20-27,29,30,34,35,39-41,43,46,47,49,53,54,56,59,61,63,65,67] and EIS[3-7,10,12,15,16,18,20-23, 26,27,29,30,34,35,40,41,47,49,50,52-54,56,59,61,65,67] were employed for calculation of percentage inhibition efficiency. Other techniques like OCP[22], SE[10,41], RS [21], Colorometry[41], XRD[27,39,58], AFM[3,5,7, 10,12,18,23,30,39,49,59,61,67], RSM[33], DFT[23], HPLC[26], NMR[49], AAS[21,60], ATR[10], ANN[60], QCC[5,7,10,12,30,49,59,67] were also used.

Techniues for surface film analysis: Film formed on metal surface were studied by various techniques, like SEM[4-7,10-15,16-23, 25-28,30,33,35,39,40,42,43,46,47,49,50,52,53,56,59,61,63,64,67], FT-IR[3,5-7,10,12,13,15,18, 19,22,23,25,26,30,39-43,47,49,52,53,56,59,60,67], UV-Vis[5,7,10,12,15,18,23,28,30,43,49,50, 59,67], GC-MS[14,20,25,56] and EDX[11,13,21,42,46,47,53,56].

Adsorption isotherms*:* In this review adsorption isotherms like Langmuir[3-14,17,19,21-35,37-43,45,50,51-53,56,58,64-67], Freundlich[22,44,46,56,62, 63], Temkin[9,15,22,24,56,63,65,68] and El-Awaday[20,24, 41] were suggested.

Conclusion

This review paper has summarized the research works carried out by various researchers on corrosion of MS in H_2SO_4 and it's inhibition by using different natural plant extracts and techniques. Corrosion inhibition efficiency depends on many variables such as corrosion inhibitor, inhibitor concentrations, temp. and immersion time. In fact, plant extracts have numerous photochemical constituents that are capable to easily be adsorbed onto metals, and thus inhibit their corrosion. Percentage I.E. of a corrosion inhibitor was evaluated by WL, PDP and EIS techniques. Other techniques such as SEM, FT-IR, AFM, GC-MS and EDX were also used to study film formed on surface of metal. Results obtained from weight loss data were in good agreement with PDP and EIS methods.

Abbreviations: AAS: atomic absorption spectroscopy, AFM: atomic force microscope, ANN: artificial neural network, ATR: alternate total reflection spectroscopy, CR: corrosion rate, DFT: density functional theory, EDS: energy-dispersive spectroscopy, EIS: electrochemical impedance spectroscope, FT-IR: fourier-transform infrared spectroscope, GC-MS: gas chromatography mass spectrometry, GM: gasometry method, H_2SO_4: sulfuric acid, HE: hydrogen evolution, HPLC: High pressure liquid chromatography, IEmax (%): maximum inhibition efficiency, KI: potassium iodide, MS: mild steel, NaCl: sodium chloride, NaOH: sodium hydroxide, NMR: nuclear magnetic resonance, OCP: open circuit potential, PDP: potentiodynamic polarization, QCC: quantum chemical calculations, QMC: Quantum mechanical calculations. RSM: response surface methodology, RS: Raman spectroscopy, RN: reaction number, SE: synergistic effect, SEM: scanning electron microscopy, UV-vis: ultraviolet-visible spectroscope or ultraviolet-visible spectrophotometry, WL: weight loss, XRD: X-raydiffraction.

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