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Corrosion Inhibition Efficacy of Extracts of Plant *Cordia dichotoma* for Iron and Aluminium in Presence of Acidic Media

R. Khandelwal, S.K. Arora, S.P. Mathur*
Material research laboratory
Govt. College, Ajmer (Raj.)
Email: rakhi.gweca @gmail.com
arorask_67@ yahoo.co.in

ABSTRACT

The corrosion inhibitive effects of alcoholic extracts of different parts of plant *cordia dichotoma* on iron and aluminium has been studied in presences of hydrochloric and sulphuric acid by mass loss and thermometric methods. The experiments were carried out at 299 ± 0.2 K in presence of different concentrations of dry fruit, leaves and stem extracts of *Cordia dichotoma*. The results reveal that the alcoholic extracts of *Cordia dichotoma* is a better corrosion inhibitor than that of toxic chemicals. The fruit extract is more potent than leaves and stem extracts to inhibit the corrosion rate. The results show that the inhibition efficacy of fruit extract is higher for aluminium as compared to the iron, but an average inhibition efficacy of fruit extract is higher for iron than aluminium.

The fruit extract show maximum inhibition efficiency up to 95 % in 0.5 N HCl for iron and for aluminium it is 97% in 1.0 N H_2SO_4. The leave extract shows maximum inhibition efficiency up to 89.88 % in 0.5 N HCl for iron and for aluminium it is 89.47 % in 0.5 N H_2SO_4. The Stem extract shows maximum inhibition efficiency up to 92.59 % in 0.5 N HCl for iron and for aluminium it is 88.98 % in 0.5 N HCl.

Introduction

Metallic corrosion is a very common but serious problem causing considerable revenue loss throughout the whole world[1]. Iron is selected for many mechanical and engineering purposes due to its properties such as strength, ease of fabrication, and cost. Aluminium also found to have variety of engineering applications because of lightness, strength, thermal and electrical conductivities, heat and light reflectivity and hygienic and non toxic qualities[2], but both the metals and their alloys suffer from a serious corrosion problem. Corrosion commonly occurs at metal surfaces in the presence of oxygen and moisture involving two electrochemical reactions. Oxidation takes place at anodic site and reduction occurs at cathodic site. In acidic medium hydrogen evolution reaction predominates. Aluminium is a reactive metal according to electrochemical series (E^o = -1.66 V), yet it is unreactive in moisture due to the formation of a stable oxide film on its surface. Aluminium is not attacked by pure water but dissolves in aqueous acids with liberation of hydrogen gas.

Corrosion inhibitors reduce or prevent the corrosion reactions. They are adsorbed on metal surface and form a barrier to oxygen and moisture by complexing with metal ions or by removing corrodent from the corrosive environment. Some of the inhibitors facilitate formation of passivating film on the metal surface[3]. There are varieties of synthetic chemical corrosion inhibitors available but our present focus is on the naturally occurring green inhibitors which are eco-friendly, less expensive and having no side effects. Numerous naturally occurring substances such as Prosopis juliflora, [4] Eugenia jambolans,[5] Lawsonia extract,[6] Opuntia extract,[7] Swertia aungustifolia,[8] Ficus religeosa,[9] Heena,[10] Datura stromonium,[11] and Calotropis plants [12] have been evaluated as potential green corrosion inhibitors. In the present paper we aimed to study by mass loss method and thermometric method, the effect of naturally occurring plant *Cordia dichotoma* on corrosion rate of the iron and aluminium in different concentrations of acid solutions.

Description of inhibitor - *Cordia dichotoma* belongs to family Boraginaceae. Its common name is Indian cherry, lasura. Its chemical constitutions are mono and polysachharides, Beta-sitosterol, flavonol glycoside, taxifotin, 3-rhamnoside, 3-5-dirhamnoside, distylin, 3-xyloside, allantoin. It is Astringent, anthelmintic, diuretic, demulcent and expectorant (fruit) and useful in the cough, chest diseases hence it relief from severe colic pain. Probably the alkaloid - Allantoin (Fig. 1) present in *Cordia dicotoma* is effective for corrosion inhibition activity in acidic media for iron and aluminium.

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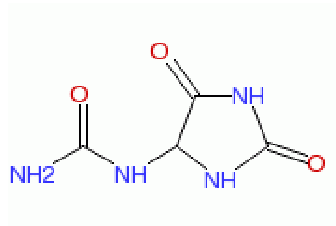

(Fig. 1)

Allantoin

Experimental

MASS LOSS MEASUREMENTS:

Specimen preparation

Rectangular specimens of iron of dimensions 2.54 X 1.52 X .02 cm with a small hole of about 2mm diameter near the upper edge were employed for the determination of mass loss measurements. Surface preparation is of prime importance in mass loss method. To achieve uniform surface, a substantial thickness of metal surface should be removed by milling and turning by use of emery papers or by mean of mechanical grinding.

Test solution preparation

The solutions of 0.5N and 1N were prepared using doubly distilled water. The Fruit leave and stem extracts of *Cordia dichotoma* was obtained by dried, then finely powdered and extracted with boiling ethanol.

To observe the influence of various parameters like inhibitor concentration, acid concentration and time, the corrosion inhibition efficiency (η) of the compounds has been calculated by mass loss method using following equation.[13]

$$\eta \% = (\Delta M\mu - \Delta Mi) / \Delta M\mu \times 100$$

Where $\Delta M\mu$ is mass loss without inhibitor and ΔMi is mass loss with inhibitor. The degree of surface coverage (θ) can be calculated as –

$$(\theta) = (\Delta M\mu - \Delta Mi) / \Delta M\mu$$

The Corrosion rate in mili meter penetration year (mmpy) [14] can be obtained by following equation.

$$\text{Corrosion rate (mmpy)} = (\text{Mass loss} \times 87.6) / \text{area} \times \text{time} \times \text{metal density}$$

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Where mass loss is expressed in mg, area is expressed in square cms of metal surface exposed, time is expressed in hours of exposure and metal density is expressed in gms/ cm^3.

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THERMOMETRIC MEASUREMENTS:

For the thermometric study[15] the specimens of size 2.54 X 1.52 X .03 cm were immersed in 50ml of acid solution. The test was carried out in 0.5N HCl and 0.5 N H_2SO_4 solutions. The inhibition studies were carried out in the concentrations 0.12%, 0.24%, 0.36% 0.48% and 0.60% of the extract of natural products. Thermometer bulb and the test specimen were completely immersed in the test solution, which was kept in Dewar flask. The change in temperature was recorded at the successive intervals of 5 minutes with the help of thermometer with a precision of 0.1°C . The results were used to calculate reaction number (R.N.) and inhibition efficiency (η %). In this method, the variation of temperature is followed as a function of time. The test specimen is immersed in a cell containing the test solution along with the thermometer bulb, which is immersed in a dewar flask. Initially the temperature of the system remains constant. Reaction number can be calculated by the following equation-

$$RN = (Tm - Ti) / t$$

Where Tm and Ti are initial and maximum temperatures respectively and t is the time in minutes to attain Tm in the experiment. Drastic corrosion is indicated by higher reaction number. The inhibitor efficiency can be calculated as –

$$\eta \% = (R_{Nfree} - R_{Ni}) / R_{Nfree} \times 100$$

Where R_{Nfree} and R_{Ni} are reaction number in blank and inhibited system respectively. The results are presented in Table 5 and 6.

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Table 1
Mass loss and inhibition efficiency (η) for Iron and Aluminium in 0.5 N HCl (18hrs) with given inhibitor addition at 299 ± 0.2 K

Inhibitor concentration (%)	Iron			Aluminium		
	Δ M, (gm)	η %	Corrosion rate (mmpy)	Δ M, (gm)	η %	Corrosion rate (mmpy)
Fruit extract						
Unhibited	0.02		0.0012	0.121		0.0218
0.12	0.010	50.00	0.0006	0.024	80.16	0.0043
0.24	0.009	55.00	0.0005	0.021	82.64	0.0037
0.36	0.007	65.00	0.0004	0.020	83.47	0.0036
0.48	0.004	80.00	0.0002	0.019	84.30	0.0034
0.60	0.001	95.00	0.0001	0.010	91.74	0.0018
Leaves extract						
Unhibited	0.089		0.0055	0.121		0.0218
0.12	0.033	62.92	0.0020	0.25	79.34	0.0450
0.24	0.027	69.66	0.0016	0.022	81.82	0.0039
0.36	0.021	76.40	0.0012	0.021	82.64	0.0037
0.48	0.014	84.26	0.0008	0.019	84.30	0.0034
0.60	0.009	89.88	0.0005	0.013	89.47	0.0023
Stem extract						
Unhibited	0.054		0.0033	0.121		0.0218
0.12	0.028	48.15	0.0017	0.027	77.68	0.0048
0.24	0.024	55.56	0.0014	0.024	80.16	0.0043
0.36	0.023	57.41	0.0014	0.023	80.99	0.0041
0.48	0.020	62.96	0.0012	0.022	81.82	0.0039
0.60	0.004	92.59	0.0002	0.014	88.98	0.0025

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Table 2

Mass loss and inhibition efficiency (η) for Iron and Aluminium in 1.0 N HCl (18hrs), with given inhibitor addition at 299 ± 0.2 K

Inhibitor concentration (%)	Iron			Aluminium		
	Δ M, (gm)	η %	Corrosion rate(mmpy)	Δ M, (gm)	η %	Corrosion rate (mmpy)
Fruit extract						
Unhibited	0.074		0.0045	0.132		0.0237
0.12	0.026	64.86	0.0016	0.032	75.76	0.0057
0.24	0.024	67.57	0.0014	0.030	77.27	0.0054
0.36	0.022	70.27	0.0013	0.028	78.79	0.0050
0.48	0.021	71.62	0.0012	0.024	81.82	0.0043
0.60	0.013	82.43	0.0008	0.017	87.12	0.0030
Leaves extract						
Unhibited	0.074		0.0045	0.132		0.0237
0.12	0.040	45.95	0.0024	0.035	73.48	0.0063
0.24	0.035	52.70	0.0021	0.031	76.52	0.0055
0.36	0.032	56.76	0.0019	0.030	77.27	0.0054
0.48	0.030	59.46	0.0018	0.027	79.55	0.0048
0.60	0.029	60.81	0.0017	0.019	85.61	0.0034
Stem extract						
Unhibited	0.074		0.0045	0.132		0.0237
0.12	0.054	27.03	0.0033	0.036	72.73	0.0064
0.24	0.050	32.43	0.0030	0.032	75.76	0.0057
0.36	0.049	33.78	0.0030	0.031	76.52	0.0055
0.48	0.045	39.19	0.0027	0.030	77.27	0.0054
0.60	0.041	44.59	0.0025	0.021	84.09	0.0037

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Table 3

Mass loss and inhibition efficiency (η) for Iron and Aluminium in 0.5 N H_2SO_4 (18hrs), with given inhibitor addition at 299 ± 0.2 K

Inhibitor concentration (%)	Iron			Aluminium		
	Δ M, (gm)	η %	Corrosion rate(mmpy)	Δ M, (gm)	η %	Corrosion rate (mmpy)
Fruit extract						
Unhibited	0.398		0.0245	0.038		0.0068
0.12	0.115	71.10	0.0071	0.021	44.74	0.0037
0.24	0.098	75.38	0.0060	0.012	68.42	0.0021
0.36	0.089	77.64	0.0055	0.009	76.32	0.0016
0.48	0.065	83.67	0.0040	0.007	81.58	0.0012
0.60	0.020	94.97	0.0012	0.002	94.74	0.0003
Leaves extract						
Unhibited	0.398		0.0245	0.038		0.0068
0.12	0.116	70.85	0.0071	0.023	39.47	0.0041
0.24	0.099	75.12	0.0061	0.013	65.79	0.0023
0.36	0.092	76.88	0.0056	0.012	68.42	0.0021
0.48	0.080	79.90	0.0049	0.009	76.32	0.0016
0.60	0.041	89.70	0.0025	0.004	89.47	0.0007
Stem extract						
Unhibited	0.398		0.0245	0.038		0.0068
0.12	0.118	70.35	0.0072	0.025	34.21	0.0045
0.24	0.102	74.37	0.0063	0.015	60.53	0.0027
0.36	0.094	76.38	0.0058	0.013	65.79	0.0023
0.48	0.082	79.40	0.0050	0.010	73.68	0.0018
0.60	0.051	87.18	0.0031	0.005	86.84	0.0009

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Table 4
Mass loss and inhibition efficiency (η) Iron and Aluminium in 1.0 N H_2SO_4 (18hrs), with given inhibitor addition at 299 ± 0.2 K

Inhibitor concentration (%)	Iron			Aluminium		
	Δ M, (gm)	η %	Corrosion rate (mmpy)	Δ M, (gm)	η %	Corrosion rate (mmpy)
Fruit extract						
Unhibited	0.485		0.0299	0.043		0.0077
0.12	0.398	17.94	0.0245	0.035	18.60	0.0063
0.24	0.258	46.80	0.0159	0.022	48.83	0.0039
0.36	0.218	55.05	0.0134	0.013	69.76	0.0023
0.48	0.118	75.67	0.0072	0.010	76.74	0.0018
0.60	0.110	77.32	0.0067	0.001	97.67	0.0001
Leaves extract						
Unhibited	0.485		0.0299	0.043		0.0077
0.12	0.394	18.76	0.0243	0.028	34.88	0.0050
0.24	0.263	45.77	0.0162	0.019	55.81	0.0034
0.36	0.221	54.43	0.0136	0.011	74.42	0.0019
0.48	0.121	75.05	0.0074	0.009	79.07	0.0016
0.60	0.113	76.70	0.0069	0.003	93.02	0.0005
Stem extract						
Unhibited	0.485		0.0299	0.043		0.0077
0.12	0.399	17.73	0.0246	0.026	39.53	0.0046
0.24	0.271	44.12	0.0167	0.018	58.14	0.0032
0.36	0.225	53.61	0.0139	0.011	74.42	0.0019
0.48	0.131	72.99	0.0080	0.009	79.07	0.0016
0.60	0.120	75.26	0.0074	0.004	90.70	0.0007

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Table 5

Reaction Number (RN) and Inhibition efficiency (η %) for Iron and Aluminium in 0.5 N HCl solution with given inhibitor addition at 299 ± 0.2 K

Inhibitor concentration (%)	Iron		Aluminium	
	RN	η %	RN	η %
Unhibited	0.0750	-	0.0600	-
Fruit extract				
0.12	0.0178	76.27	0.0148	75.33
0.24	0.0149	80.13	0.0120	80.00
0.36	0.0110	85.33	0.0104	82.67
0.48	0.0105	86.00	0.0088	85.33
0.60	0.0100	86.67	0.0079	86.83
Leave extract				
0.12	0.0189	74.80	0.0159	73.5
0.24	0.0162	78.4	0.0122	79.67
0.36	0.0121	83.87	0.0109	81.83
0.48	0.0109	85.47	0.0092	84.67
0.60	0.0104	86.13	0.0088	85.33
Stem extract				
0.12	0.0192	74.4	0.0162	73.00
0.24	0.0168	77.6	0.0134	77.67
0.36	0.0128	82.93	0.0121	79.83
0.48	0.0122	83.73	0.0095	84.17
0.60	0.0118	84.27	0.0090	85.00

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Table 6

Reaction Number (RN) and Inhibition efficiency (η %) for Iron and Aluminium in 0.5 N H_2SO_4 solution with given inhibitor addition at 299 ± 0.2 K

Inhibitor concentration (%)	Iron		Aluminium	
	RN	η %	RN	η %
Unhibited	0.0810	-	0.0510	-
Fruit extract				
0.12	0.0192	76.29	0.0122	76.08
0.24	0.0161	80.12	0.0101	80.20
0.36	0.0129	84.07	0.0092	81.96
0.48	0.0112	86.17	0.0082	83.92
0.60	0.0098	87.90	0.0070	86.27
Leave extract				
0.12	0.0194	76.05	0.0125	75.49
0.24	0.0165	79.63	0.0118	76.86
0.36	0.0135	83.33	0.0098	80.78
0.48	0.0119	85.31	0.0087	82.94
0.60	0.0102	87.40	0.0077	84.90
Stem extract				
0.12	0.0198	75.55	0.0128	74.90
0.24	0.0175	78.39	0.0120	76.47
0.36	0.0139	82.84	0.0101	78.23
0.48	0.0122	84.94	0.0092	81.96
0.60	0.0109	86.54	0.0081	84.11

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Result and Discussion

Mass Loss Method

Mass loss, inhibition efficiency and corrosion rate for iron and aluminium at different concentrations of HCl and inhibitors are given in Table 1 and 2. And for H_2SO_4 and inhibitor are given in Table 3 and 4. It is observed that the inhibition efficiency (%) increases with the increase in inhibitor concentration for both the metals iron and aluminium. The inhibition efficiency of alcoholic extracts of fruit, leaves and stem of *Cordia dichotoma* was calculated, and it is observed that the fruit extract shows the maximum inhibition efficiency.

In 0.5 N HCl the fruit extract shows the maximum inhibition efficiency 95 % for iron and 91.47 % for aluminium. The leave extract exhibits maximum inhibition efficiency 89.88 for iron and 89.47 for aluminium. And the stem extract shows maximum inhibition efficiency 92.59 for iron and 88.98 for aluminium.

In 1.0 N HCl all the three extracts show maximum inhibition efficiency at maximum inhibitor concentration. It is also observed that in 1.0 N HCl inhibition efficiencies are higher for aluminium as compared to iron. The fruit extract shows the maximum inhibition efficiency 87.12 % for aluminium 82.43 % for iron. The leave extract shows maximum inhibition efficiency 85.61 for aluminium 60.81 for iron. And the stem extract exhibits maximum inhibition efficiency 84.09 for aluminium 44.59 for iron.

In 0.5 N H_2SO_4 the fruit extract shows the maximum inhibition efficiency 94.97 % for Iron and 94.74 % for aluminium. The leave extract shows maximum inhibition efficiency 89.70 for iron and 89.47 for aluminium, and the stem extract shows maximum inhibition efficiency 87.18 for iron and 86.84 for aluminium.

In 1.0 N H_2SO_4 the fruit, leaves and stem extracts exhibit maximum inhibition efficiency 77.32, 76.70, 75.26 for iron while 97.67, 93.02, and 90.70 for aluminium.

The efficiencies of inhibitors expressed as the relative reduction in corrosion rate can be quantitatively related to the amount of adsorbed inhibitors on the metal surface. It is assumed that the corrosion reactions are prevented from occurring over the active sites of the metal surface covered by adsorbed inhibitor, whereas the corrosion reactions occurs normally on the inhibitors free area. The inhibition efficiency is then

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directly proportional to the fraction of surface covered with adsorbed inhibitor. This assumption has been applied to deduce the effect of concentration on adsorption of inhibitors.

Generally the adsorption of organic molecules on metallic surface involves O, N, and S atoms. In the case of plant extracts of *Cordia dichotoma*, the N and O of the alkaloids may be responsible for adsorption. This process may block the active sits on metal surface, hence decreasing the rate of corrosion. Due to the higher electron density the N atom of the alkaloid acts as the reaction centre, resulting in the formation of a monolayer on the metal surface. Organic inhibitors with active portions generally contain large hydrocarbon chains or rings with positively charged amine N group at the one end. In acids and water the terminal primary, secondary and tertiary amines groups take additional hydrogen that gives them a net cationic charge. The polar amine group is adsorbed on the metal and hydrocarbon portion forms an oily water repellent surface film. The molecular dissymmetry helps these materials to act as surfactants and can stabilize emulsions of oil and water. The Organic corrosion inhibitor may function by –

1. Chemisorption on the metallic surface.
2. Neutralizing the corrodent
3. Adsorbing the corrodent

They offer large coverage due to the long hydrocarbon chain and by the presence of –NH groups. Being hydrophilic in nature, the –NH group counteracted the effects of chain length and ensured higher solubility. It has been observed that the fruit extract of *Cordia dicotoma* has maximum inhibition efficiency as compared to leaves and stem extracts. This may be attributed to the presence of alkaloids. In the fruit extract the electron repelling hydroxyl group on the alkaloid is present; as a result the electron density at the N atom becomes more than the any other additives. This process increased the adsorptivity of the fruit extract on the corroding site of the metal. This explains the higher inhibition efficacy displayed by the fruit extract for 0.6 % concentration.

Thermometric Method

Inhibition efficacies were also determined using thermometric method. Temperature change for iron and aluminium in o.5 N HCl and 0.5 N H_2SO_4 were recorded both in presence and absence of inhibitor. Results summarized in Table 5 for HCl and

Table 6 for H_2SO_4 show a good agreement with the results obtained by mass loss method. The maximum inhibition efficiency was obtained with the highest concentration of inhibitor. The reaction number decreases with increase in the inhibitor concentration.

Conclusion

It is anticipated that the proposed investigation on green corrosion inhibitor may be useful for preventing losses caused due to corrosion. The extracts of *Cordia dichotoma* are found to be an effective inhibitor for both the metals iron and aluminium in acidic media giving IE up to 95.00% in HCl, 97.67 % in H_2SO_4 It is also concluded that at lower acid concentrations extracts of *Cordia dichotoma* are more potent to inhibit corrosion in iron but at higher acid concentrations extracts are more effective inhibitors for aluminium.

On the basis of the results it is concluded that the alcoholic extracts of *Cordia dichotoma* may de use as ecofriendly corrosion inhibitors without any toxic effect and pollution.

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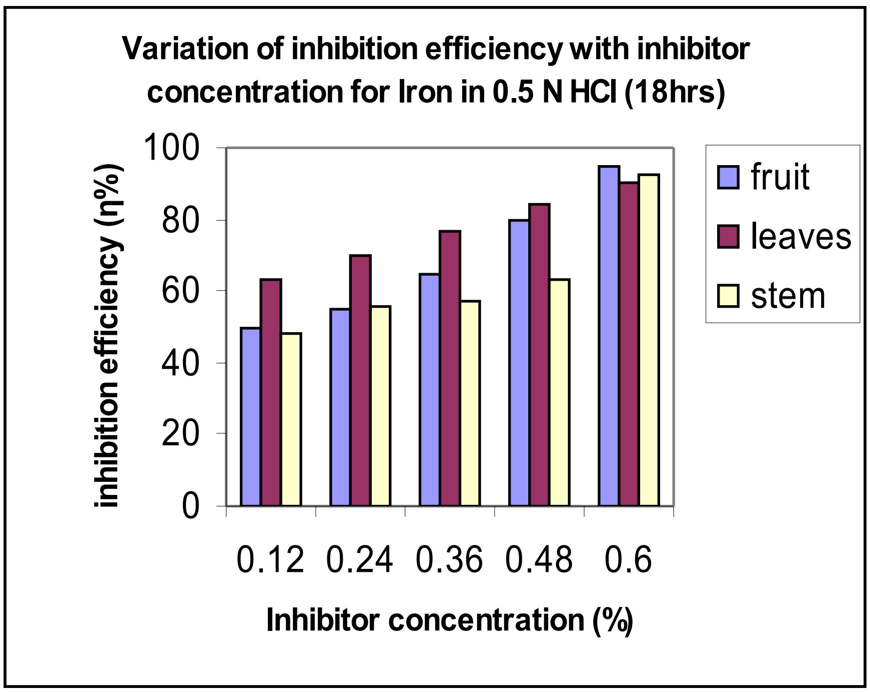

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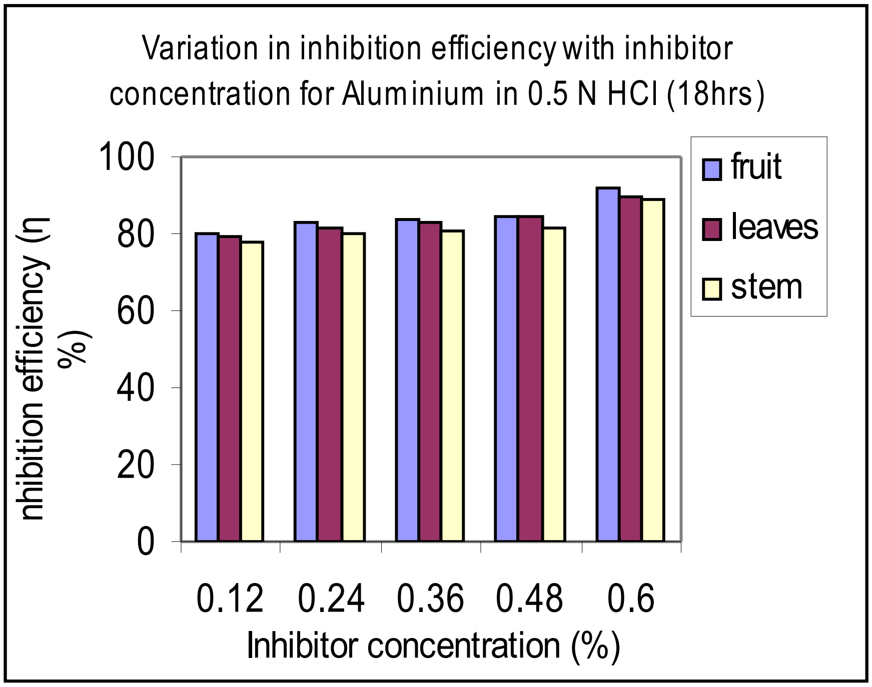

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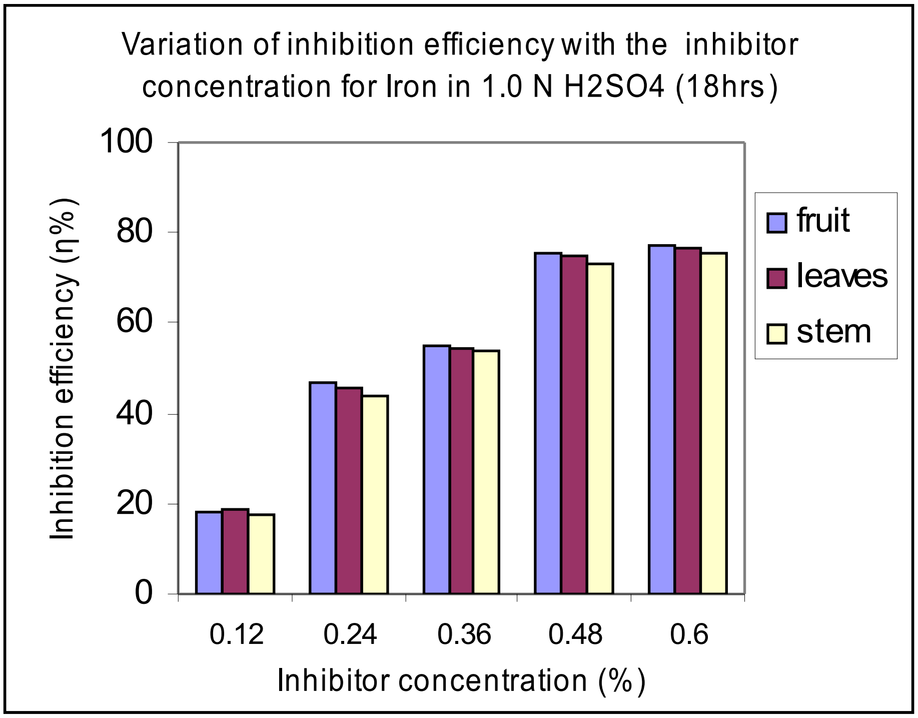

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