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Corrosion Inhibition of Aluminium by Extract of Aerial Parts of *Phyllanthus niruri* in Hydrochloric acid and Study of Extract as Antibacterial

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Abstract: *Corrosion is a natural electro-chemical phenomenon, in which metals revert to their natural and lower energy state, which can be simply defined as rust. It is a result of the inherent tendency of metals to revert to their more stable compound such as oxides, hydroxide and sulphides. It can be defined as the deterioration of metals and its alloys by chemical or electrochemical means. Metals are found in the form of ores in nature. In the refining process, energy is added to the ore to produce the metal. It is the same energy process that provides the driving force causing the metal to revert back to the more stable compound ores. As we know aluminium is one of the most widely used metals. When it combines with other metals by thermo-mechanical processing, aluminium alloys display a marked improvement in mechanical properties and in presence of various corrosive agents including corrosive gases, mineral acids etc. aluminium and its alloys get corroded and they loss their properties. In the present study the alkaloid extract of aerial parts of Phyllanthus niruri was investigated as green corrosion inhibitor for aluminium metal in different concentration (1N, 2N, and 3N) of Hydrochloric acid at 301K temperature by using weight loss and thermometric methods. The study showed that with increasing concentration of extract, inhibition efficiency increases whereas the rate of corrosion decreases. The study revealed that extract of Phyllanthus niruri is an efficient natural corrosion inhibitor in acidic medium. The leaves extract showed maximum inhibition efficiency (95.03%) in lowest concentration (1N) of HCl and stem extract is a better inhibitor than leaves and flower extract of the plant. The absorption of plant extract on the metal surface obeyed the Langmuir isotherm. The present investigation it may also be interpreted that Phyllanthus niruri plant has good antibacterial activity and may be used as an alternative food preservative in food industries.*

Keywords*: Weight loss, Thermometric, Surface coverage, Inhibition efficiency, Corrosion rate, Phyllanthus niruri.*

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I. Introduction

The interaction between the metal surface and its environment, which degrades the metallic properties of metals through chemical or electrochemical attack, by its environment is called corrosion (1). According to IUPAC, Corrosion is an irreversible interfacial reaction of material with its environment which results in its consumption or dissolution into the material of a component of the environment. Thus "corrosion is a process of reverse extraction of metal". Corrosion is a natural phenomenon, which is destructive in nature. Corrosion affects almost all metals. The problems may be made clear by the approximate estimate of loss of metals due to corrosion as 2 to 3 billion dollars per annum all over the world.

Aluminium is a silvery-white, nonmagnetic, durable, malleable, light weight, good conductor of heat and electricity and ductile metal belonging to the boron family. It is the third most abundant element on the earth after oxygen and silicon. Generally it exists in a stable + 3 oxidation state and almost all compounds of aluminium (+3) are colourless (2). Aluminium is almost 100% recyclable without loss of its properties. It is used in alloyed form which improves its mechanical properties. Nowadays aluminium is used in Packaging (3) (cans, foil, frame), transportation (4) (tube, sheet and casting), food and beverage containers, construction (5) (doors, windows, siding, building wire, sheathing, roofing, etc.) cooking utensils, household items, coins, transistors, LED lighting, CPUs, photographic equipments, musical instruments, etc. Pure aluminium naturally forms a thin surface layer of aluminium oxide on contact with oxygen in the environment through oxidation, which creates a physical barrier to corrosion or further oxidation in various environmental conditions which are considered as passivation. But some aluminium alloys do not form an oxide layer and in results they get corroded adversely in acidic environment and loss their chemical and physical properties (6-10).

Heterocyclic compound containing O,N,S and P atoms have been used as corrosion inhibitors in acidic media because the compound contains aromatic rings with free π-electron pair for sharing with metal and donating their electron to the surface of metal and hence forms a coordinate bond between metal and inhibitor that reduces the corrosion rate. In this process inhibitor acts as nucleophile while metal acts as electrophile centres. The electron density, orientation, size and shape of molecule play significant role in the effectiveness of inhibition. In this mechanism, the inhibitor got adsorbed on the active centre of the metal surface and blocked the active corrosion site on the metal surface due to the liberation of H^+ ions and dissociation of metal is stopped in the acidic media, consequently the corrosion of metal stops. Schiff bases are good corrosion inhibitors [(11)] and Mannich bases are also investigated as good corrosion inhibitor [(12)].All the above organic compounds are good corrosion inhibitors but these are very costly, pollutant, toxic and harmful. That's why we need eco-friendly inhibitors.

Natural occurring plant products are easily available, less toxic, economic, eco-friendly and biodegradable so they are widely used as corrosion inhibitors without any harmful effects [(13-15)].

Number of natural products extracted from plant is also found effective corrosion inhibitor like: *Argemone Mexicana*, *Olea europaea* [(16)], *Withania somnifera*, *Holly basil* [(17)] etc.

1.1 PLANT DESCRIPTION

Phyllanthus niruri is a field weed which is often found in tropical and subtropical regions of Asia, America, and China. Taxonomically, the annual herb Phyllanthus niruri belongs to the family Phyllanthaceae. The whole plant has been used as medicine in the Ayurveda System from ancient time. *Phyllanthus niruri* is an annual herb which grows in the forest after first showers of monsoon in Jharkhand, Bihar, Chhattisgarh, Rajasthan etc. states of India. However, it has also been reported to grow commonly in coastal areas. In Indian states it usually grows during second week of June and starts bearing fruits up to mid-July. *Phyllanthus niruri* is an erect annual herb, growing 40 – 60 cm height having ascending herbaceous branching; it is quite glabrous and branching at the base. The genus *Phyllanthus* means "leaf and flower" because the flower and fruit can be associated with the leaf. It is a plumose leaf that carries flower and fruit [(18-20)].

Phyllanthus niruri has extensive medicinal properties and has long history in the health care system of tropical countries. The plant is known in traditional health care systems. The plant is commonly known as "Chanca pedra" (or) "stone breaker". Phyllanthus niruri is used as a folk medicine for treating kidney stones, gallbladder stones, liver related diseases such as liver cancer & jaundice, apart from these it is also administered for diuretic, hypoglycemic and hypertension cases and it also shows anti-inflammatory, anti-tumor, antinociceptive and anti-oxidant properties [(21-22)].

As *Phyllanthus niruri* is medicinally important shrub and it has variety of phytochemicals and their pharmacological properties. The active phytochemicals, flavonoids, alkaloids, terpenoids, lignans, polyphenols, tannins, coumarins and saponins, have been identified from various parts of *Phyllanthus niruri* [(23-26)].The main chemical constituents of plant extract are given as bellow:

Alkaloid - 4-Methoxy-nor-securinine, nirurin, ent-norsecurin etc.
Benzenoid - Gallic acid, Corilagin etc.
Coumarins - Ellagic acid, ethyl brevifolin carboxylate etc.
Flavonoids - Quercetin, rutin, astragalin, quercetin, isoquercitrin, eridictyol-7-rhamnopyranoside, fisetin , nirurin etc.
Lignin - Phyllanthin, hypophyllanthin, niranthin, nirtetralin, phyltetralin, hinokinin etc.
Lipid - Ricinoleic acid
Phytallate -Phyllester
Sterol - Estradiol, ß-sitosterol, isopropyl-24-cholesterol
Tannin - Geranin
Triterpene - Lupeol acetate, phyllanthenol, phyllanthenone etc.

II. Materials And Methods

2.1 Experimental

Commercially available aluminium sheet was used for this study. Square shaped specimen of aluminium sheet having dimension 2.5 × 2.5 cm^2 containing small hole of about 2mm diameter near the upper edge were used for experiments.

2,2 Plant Extract

Phyllanthus niruri was identified with help of subject expert and its aerial part was collected from theme park educational botanical garden, Regional Institute of Education, Ajmer. The whole plant including stem, leaves and flower were air dried at room temperature, and then grinded to make powder. The extract of stem, leaves, and flower were obtained by refluxing the dried powder of the aerial parts of plant in soxhlet unit in ethanol with refluxing for sufficient time.

2.3 Chemical Used

1N, 2N, 3N solutions of Hydrochloric acid were prepared for experimental study and these acid solutions were used for corrosion analysis. In this study we have used 0.1, 0.3, 0.5 and 0.7 percentage composition solutions of stem, leaves and flower extract as inhibitors.

2.4 METHOD

2.4 a. Weight loss method: Each specimen was suspended by a V-shaped glass hook made of fine capillary and plunged into a beaker containing 100 mL of the test solution at room temperature. After the sufficient exposure, test specimens were washed with running tap water and dried with hot air dryer. Duplicate experiments were performed in each case and mean value of weight loss was calculated. The percentage inhibition efficiency (η %) was calculated [27-28] by using following equation:

$$\eta\% = 100\left[\frac{(\Delta W_u - \Delta W_i)}{\Delta W_u}\right]$$

Where ΔW_u and ΔW_i are the weight loss of the metals in absence and presence of inhibitor solution, respectively. The degree of surface coverage (θ) was calculated by using following equation:

$$\theta = \frac{(\Delta W_u - \Delta W_i)}{\Delta W_u}$$

Langmuir adsorption isotherm-

$\log[\theta / 1-\theta] = \log A + \log C - (\theta / 2.3RT)$

Should give a straight line of unit gradient for the plot [θ / 1- θ] versus log C, where θ is surface coverage, A is a temperature independent constant and C is concentration of inhibitor. The corresponding plot (for aluminium Figs 1b, 2b, and 3b) is linear but the gradients are not equal to unity as would be expected for the Langmuir adsorption isotherm equation.

2.4 b. Thermometric method:

Inhibition efficiencies were determined by using thermometric method technique. This involved the immersion of single specimen of area $13cm^2$(both sides) in reaction chamber containing 100 mL solution of acid at an initial temperature of 301K. Experiments were carried out in 1N, 2N, 3N, Hydrochloric acid solutions in absence and presence of different concentration of inhibitor viz. 0.1%, 0.3%, and 0.5% and 0.7%. Thermometer bulb and specimen were completely immersed in test solution in a beaker. The beaker was kept in a thermally insulated chamber. Temperature changes were measured at definite intervals using thermometer with a precision of 0.1^0C. The temperature increased slowly at first, then rapidly and attained a maximum value which was recorded.

The reaction number(R N) is calculated [29-32] by using following equation:

$$RN = \frac{T_m - T_i}{t}$$

Where T_m and T_i are maximum and initial temperatures, respectively and t is the time in minutes required to attain maximum temperature. The percentage inhibition efficiencies (%) were obtained by using following equation:

$$\% = \frac{(RN_f - RN_i)}{RN_f} \times 100$$

Where RN_f, and RNi are reaction numbers in the absence and in the presence of inhibitor respectively.

The corrosion rate (CR) in mm/year can be calculated [33—35] by using the following equation:

$$\text{R corr.} = \frac{(\Delta W \times 87.6)}{(A \times T \times d)}$$

Where ΔW is weight loss in mg, A is surface area of specimen in cm^2; t is time of exposure in hrs. and d is density of metal in g /cm^3.

III. Results And Discussion

The corrosion rate of aluminium in nitric acid solution of different strength were studied by using weight loss and thermometric method in absence and presence of stem, leaves and flower extracts of *Phyllanthus niruri* plant at 301K temperature and percentage inhibition efficiencies was calculated by using both methods.

Weight loss data, percentage inhibition efficiency, corrosion rate and surface coverage for aluminium metal in 1N , 2N, and 3N Hydrochloric acid solutions with different concentration of inhibitors are given in Table-1,2, and 3, respectively. The corresponding graphs are for inhibition efficiency and Langmuir adsorption isotherm shown in Figs.1a-b, 2a-b, 3a-b.

The data was utilized for calculation of reaction number and percentage of inhibition efficiencies are given in Table- 4 for , stem, leaves and flower extract in 1N, 2N and 3N HCl acid solutions and their corresponding graphs are shown in Figs 4a, 4b and 4c, respectively

Table - 1
Weight loss data (Δw), inhibition efficiency (η%), surface coverage (θ) and corrosion rate for Aluminium in 1N HCl acid solution with various concentration of aerial parts of *Phyllanthus niruri* plant

Temperature: 301K±0.1^{0}C Time of Exposure: 0.75 hrs Area of Specimen: 6cm^2

Inhibitor Concentration (%)	Weight Loss (Δw) in g	Corrosion Rate in (mm/yr)	Inhibition Efficiency (η%)	Surface Coverage (θ)	$\log\left(\frac{\theta}{1-\theta}\right)$
Stems					
Uninhibited	0.2560	1.8457			
0.1	0.0312	0.2249	87.81	0.8781	0.8575
0.3	0.0251	0.1809	90.19	0.9019	0.9634
0.5	0.0181	0.1304	92.92	0.9292	1.1180
0.7	0.0127	0.0915	95.03	0.9503	1.1207
Leaves					
0.1	0.0431	0.3107	83.16	0.8316	0.6935
0.3	0.0361	0.2602	85.89	0.8589	0.7844
0.5	0.0291	0.2098	88.63	0.8863	0.8918
0.7	0.0239	0.1723	90.66	0.9066	0.9870
Flowers					
0.1	0.0542	0.3907	78.82	0.7882	0.5707
0.3	0.0471	0.3395	81.60	0.8160	0.6468
0.5	0.0401	0.2891	84.33	0.8433	0.7309
0.7	0.0351	0.2530	86.28	0.8628	0.7985

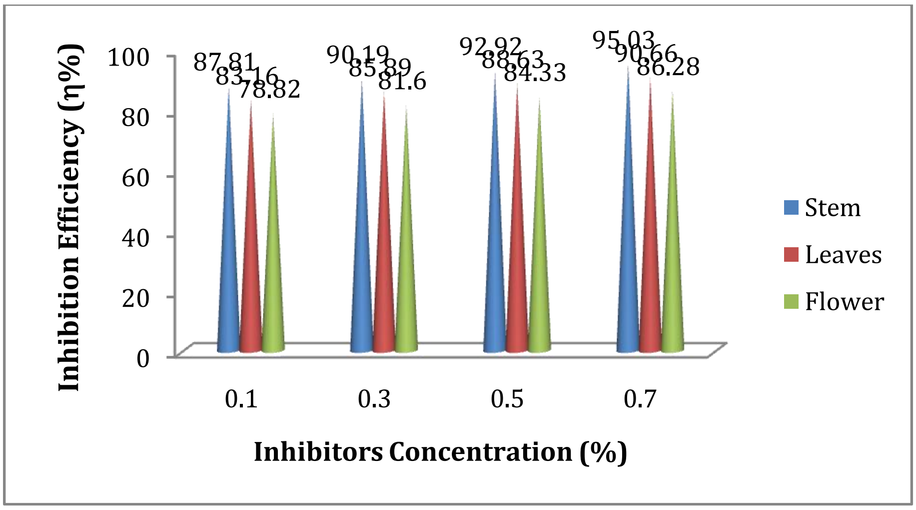

Fig. 1 (a) Variation of inhibition efficiency with concentration of in 1N HCl acid solution

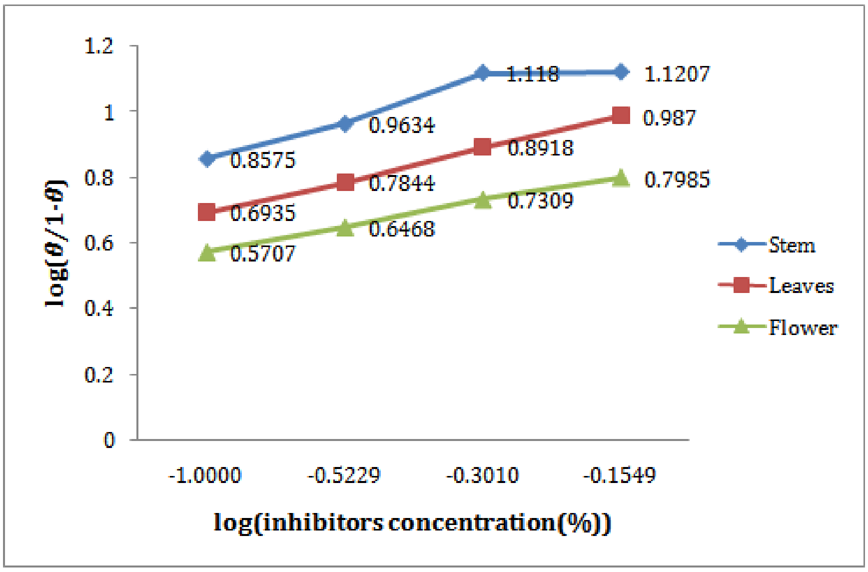

Fig. 1 (b) Langmuir adsorption isotherm for Aluminium in 1N HCl acid solution

Table - 2

Weight loss data (Δw), inhibition efficiency (η%), surface coverage (θ) and corrosion rate for Aluminium in 2N HCl acid solution with various concentrations of aerial parts of *Phyllanthus niruri* plant

Temperature: 301K±0.1^{0}C Time of Exposure: 0.25 hrs Area of Specimen: 6cm^2

Inhibitor Concentration (%)	Weight Loss (Δw) in g	Corrosion Rate in (mm/yr)	Inhibition Efficiency (I.E.) (η%)	Surface Coverage (θ)	$\log\left(\frac{\theta}{1-\theta}\right)$
Stems					
Uninhibited	0.2585	5.5912			
0.1	0.0395	0.8543	84.17	0.8471	0.7435
0.3	0.0305	0.6597	88.20	0.8820	0.8735
0.5	0.0255	0.5515	90.13	0.9013	0.9605
0.7	0.0195	0.4217	92.45	0.9245	1.0879
Leaves					
0.1	0.0483	1.0447	81.31	0.8131	0.6385
0.3	0.0407	0.8803	84.25	0.8425	0.7282
0.5	0.0345	0.7462	86.65	0.8665	0.8122
0.7	0.0285	0.6164	88.97	0.8897	0.9066
Flowers					
0.1	0.0573	1.2393	77.83	0.7783	0.5453
0.3	0.0509	1.1009	80.30	0.8030	0.6102
0.5	0.0443	0.9581	82.86	0.8286	0.6843
0.7	0.0395	0.8543	84.71	0.8471	0.7435

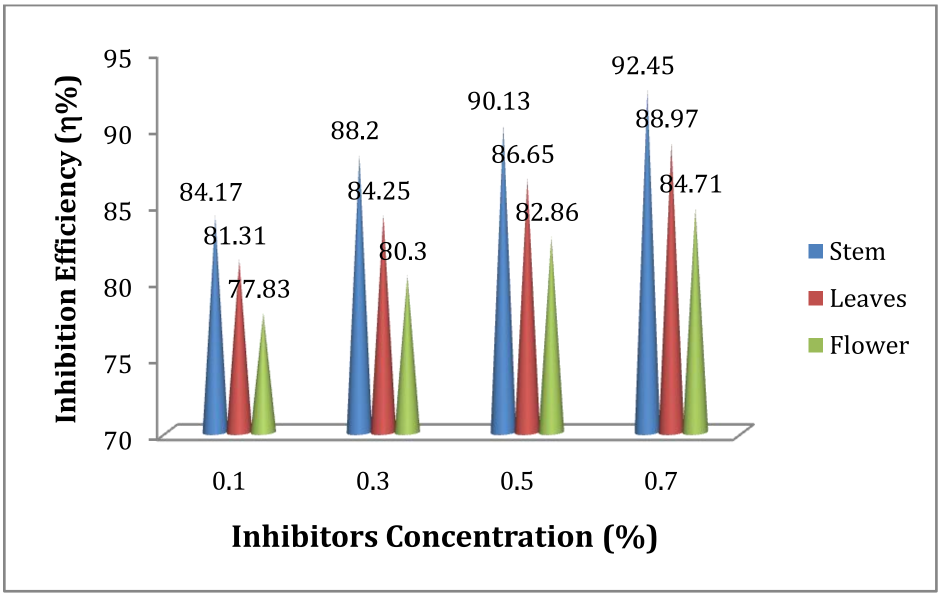

Fig. 2 (a) Variation of inhibition efficiency with concentration of inhibitors forAluminium in 2N HCl acid solution

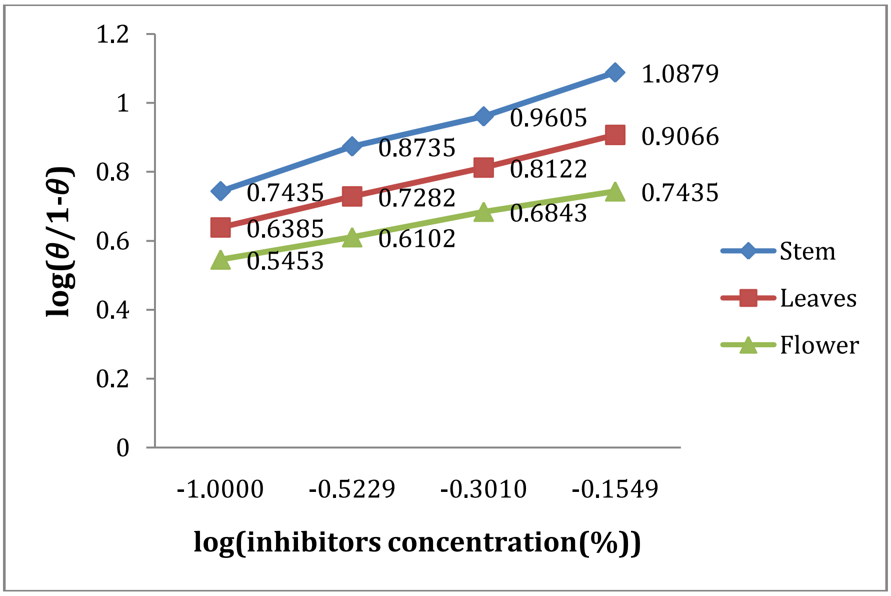

Fig. 2 (b) Langmuir adsorption isotherm for Aluminium in 2N HCl acid solution

Table - 3
Weight loss data (Δw), inhibition efficiency (η%), surface coverage (θ) and corrosion rate for Aluminium in 3N HCl acid solution with various concentration of aerial parts of *Phyllanthus niruri* plant

Temperature: 301K±0.1^{0}C Time of Exposure: 0.133 hrs Area of Specimen: 6cm^2

Inhibitor Concentration (%)	Weight Loss (Δw) in g	Corrosion Rate in (mm/yr)	Inhibition Efficiency (I.E.) (η%)	Surface Coverage (θ)	$\log\left(\frac{\theta}{1-\theta}\right)$
Stems					
Uninhibited	0.2657	10.5797			
0.1	0.0523	2.0844	80.71	0.8071	0.6215
0.3	0.0429	1.7101	83.83	0.8383	0.7146
0.5	0.0338	1.3478	87.26	0.8726	0.8356
0.7	0.02674	1.0647	89.93	0.8993	0.9508
Leaves					
0.1	0.0813	3.2372	69.40	0.6940	0.3556
0.3	0.0703	2.7992	73.54	0.7354	0.4439
0.5	0.0623	2.4806	76.55	0.7655	0.5137
0.7	0.0539	2.1462	79.13	0.7913	0.5788
Flowers					
0.1	0.0965	3.8424	63.68	0.6368	0.2438
0.3	0.0868	3.4562	67.33	0.6733	0.3140
0.5	0.0759	3.0222	71.43	0.7143	0.3979
0.7	0.0641	2.5523	75.87	0.7587	0.5788

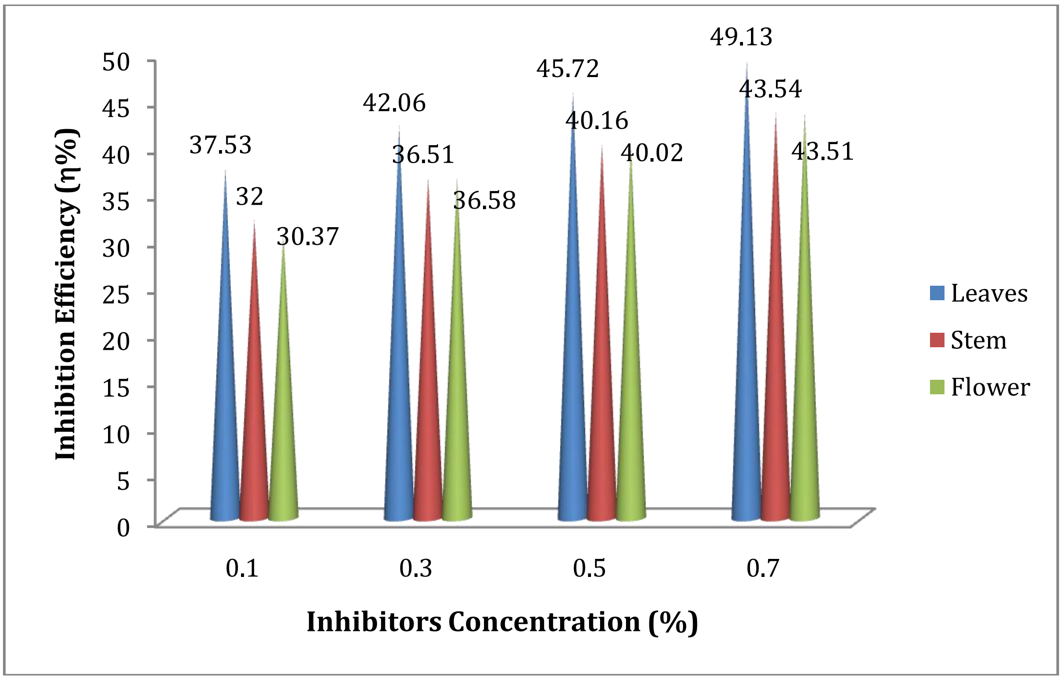

Fig. 3 (a) Variation of inhibition efficiency with concentration of Inhibitors for Aluminium in 3N HCl acid solution

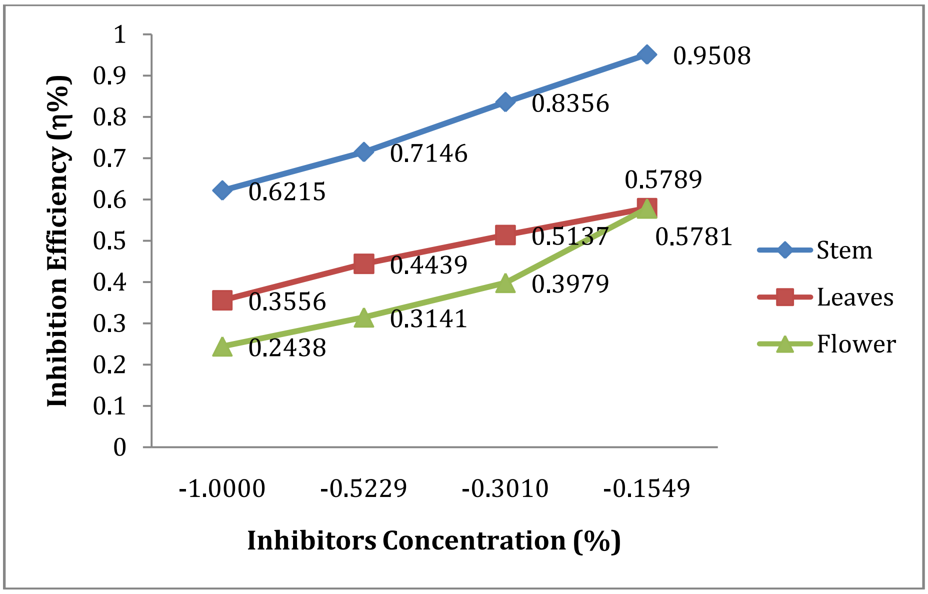

Fig. 3 (b) Langmuir adsorption isotherm for Aluminium in 3N HCl acid solution

Table – 4
Reaction number (RN) and inhibition efficiency (%) for Aluminium in 1N, 2N and 3N HCl acid with various concentrations of inhibitors

Temperature : 301K±0.1^{0}C **Area of Specimen : 6cm^2**

Inhibitor Concentration (%)	1N HCl		2N HCl		3N HCl	
	RN Reaction Number	I.E.(%) Inhibition Efficiency	RN Reaction Number	I.E.(%) Inhibition Efficiency	RN Reaction Number	I.E.(%) Inhibition Efficiency
			Stems			
Uninhibited	0.6243		0.8927		0.9761	
0.1	0.1734	72.22	0.3092	65.36	0.3360	61.52
0.3	0.1264	79.55	0.2692	69.84	0.3145	67.77
0.5	0.1039	83.55	0.2466	72.37	0.2939	71.93
0.7	0.0739	88.16	0.1983	78.16	0.2307	76.36

Leaves						
0.1	0.2035	67.40	0.3486	60.94	0.4003	58.98
0.3	0.1565	74.93	0.3188	64.28	0.3743	61.65
0.5	0.1369	78.07	0.2751	69.15	0.3369	65.48
0.7	0.1033	83.45	0.2449	75.92	0.2981	69.46
Flowers						
0.1	0.2413	61.34	0.4181	47.46	0.5377	45.32
0.3	0.1869	70.06	0.3459	61.25	0.4432	54.59
0.5	0.1539	75.34	0.3093	65.35	0.3769	61.38
0.7	0.1303	79.12	0.2695	69.81	0.3103	68.81

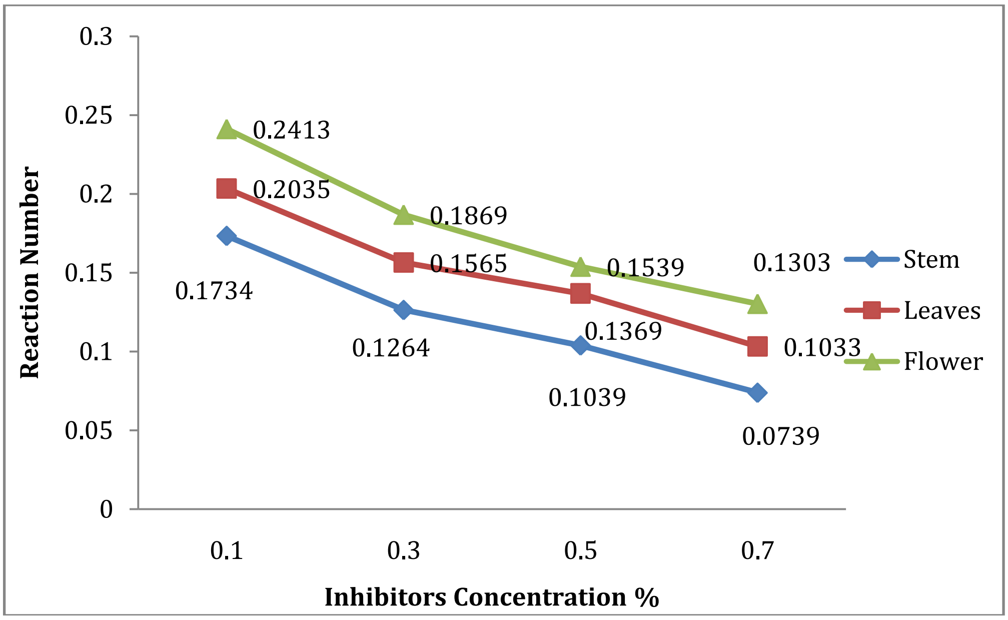

Fig. 4 (a) Variation of Reaction Number with concentration of inhibitors for Aluminium in 1N HCl acid solution

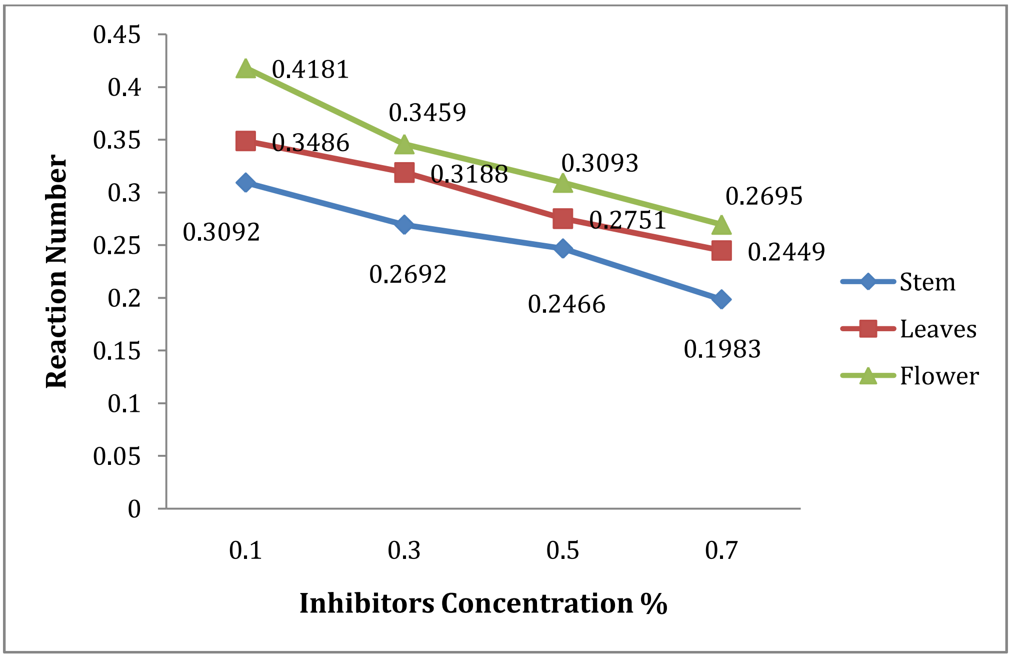

Fig. 4 (b) Variation of Reaction Number with concentration of inhibitors for Aluminium in 2N HCl acid solution

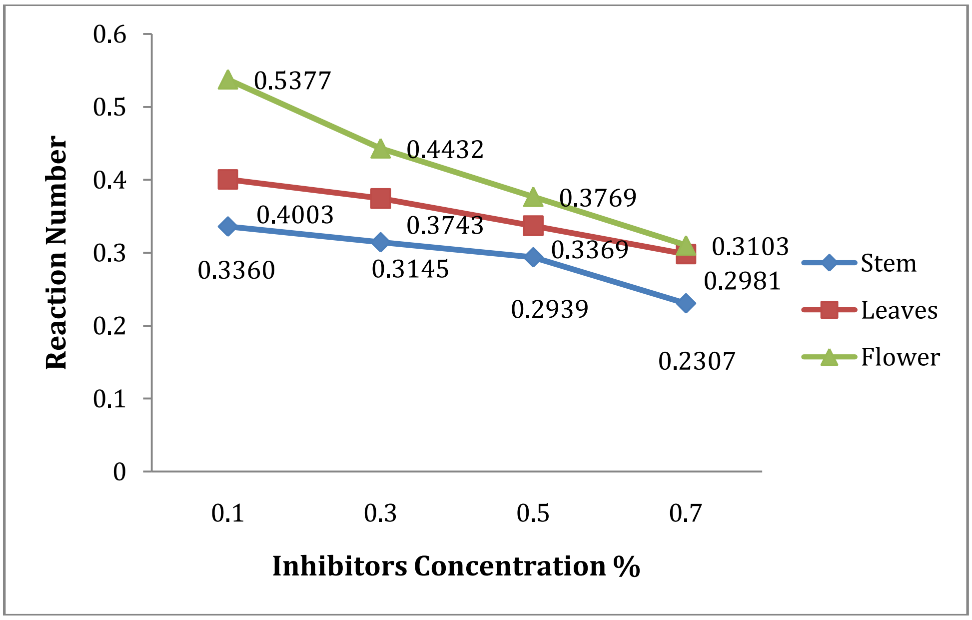

Fig. 4 (c) Variation of Reaction Number with concentration of inhibitors for Aluminium in 3N HCl acid solution

We can observe from the Table -1, 2 and 3 that inhibition efficiency decreases with increasing strength of hydrochloric acid and it increases with increasing concentration of inhibitor. The maximum inhibition efficiency (95.03%) was observed in lowest concentration of HCl i.e. 1N with highest concentration of stem extract i.e. of 0.7% and for leaves is 88.97 %, whereas for flower is 84.71 in the same strength of hydrochloric acid acid at the same concentration of inhibitor.

Corrosion rate increases with increasing reaction number that depends on temperature variations and we can observe from Table - 4, the corrosion rate was maximum in blank experimental solution of 3N HCl i.e. 0.9761 and it decreased with the increasing concentration of inhibitor in HCl solution of different strength. The corrosion rate was minimum in 1N HCl i.e. 0.6293 with highest concentration of stem inhibitors i.e. 0.7%. Corresponding curves for the variation of reaction number with concentration of inhibitor in 1N, 2N, and 3N HCl solutions are shown in Figs 4a, 4b and 4c, respectively.

3.1Antibacterial Activity of Extract of *Phyllanthus niruri*:

Aerial parts (Stem, leaves and flower) extract of *Phyllanthus niruri* investigated in vitro for antibacterial activity against gram positive, i.e. *Staphylococcus* and gram negative, i.e. *Escherichia coli* bacteria strain by using paper disc method. Effect of ethanol solvent on bacterial strains also studied.

The antibacterial activity of different aerial part of *Phyllanthus niruri* was analysed by using liquid inhibition test and inhibition zone was measured for each sample.

At low concentration, inhibition zone was too small to measure so higher concentration of plant extract was used. The observed results are shown in Fig. 5.a and 5.b. Summaries of results are also given in table 5. During the study it is found that all the extract of plant showed good inhibition activity at 1000µg/mL.

Zone of inhibition obtained for leaves, stem and flower extract against gram positive and gram negative bacteria are shown in Table 5. Zone of inhibition in the presence of stem extract was obtained 21 mm and 20 mm against gram positive and gram negative bacteria, respectively. In the presence of leaves extract it was obtained 20 mm and 24 mm whereas it was obtained 13 mm and 17 mm in the presence of flower extract against gram positive and gram negative bacteria, respectively. Antibacterial activity was also compared with standard drugs imipenem and linezolid. Inhibition zone was measured 14 mm and17 mm against gram positive bacteria and 26 mm and 12 mm against gram negative bacteria for imipenem and linezolid, respectively. The results revealed that the different parts of *Phyllanthus niruri* showed good inhibition against bacteria.

Table. 5
ANTIBACTERIAL ACTIVITY FOR AREIAL PARTS EXTRACT OF *PHYLLANTHUS NIRURI* ON GRAM POSITIVE AND GRAM NEGATIVE BACTERIA

S. No.	compounds	Gram positive bacteria (Inhibition zone in mm.)	Gram negative bacteria (Inhibition zone in mm)
1.	Solvent	0	0
2.	Stem Extract	21	20

3.	Leave Extract	20	24
4.	Flower Extract	13	17
5.	Imipenem	14	26
6.	Linezoilid	17	12

Fig. – 5.a Effect on gram positive bacteria

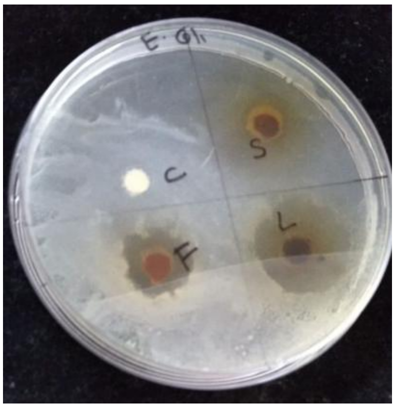

Fig. 5.b Effect on gram negative bacteria

IV. Conclusion

The study of aerial part of *Phyllanthus niruri* revealed that it would be good corrosion inhibitor for aluminium in different strength of hydrochloric acid solutions. The result of the study shows that the inhibition inefficiency is maximum (95.03%) for highest concentration (0.7%) of stem extract in lowest strength of hydrochloric acid (1N) and inhibition efficiency is minimum (86.28 %) for lowest concentration of flower extract (0.1%) in 1N HCl. Results indicate that the inhibition efficiency increases with the increase in the concentration of inhibitor in HCl acid solutions and it was concluded that extract of stem is better corrosion inhibitor than the leaves and flower extract of *Phyllanthus niruri.* In this phenomenon the adsorption reaction is depends on chemical composition of inhibitors that contains various heterocyclic compounds like alkaloids, flavonoids, steroids and tannins etc. which has higher electronegative atoms like O, N and S having with lone pair electrons. These atoms forms co-ordinate bond with metal and stopped the discharge of H^+ ions and dissolution of metal ion in acidic media. Hence the corrosion of metal reduces in the presence of inhibitors. Antibacterial activity study of aerial part extract revealed that *Phyllanthus niruri* has higher antibacterial activity against gram negative bacteria than gram positive bacteria. Leaves extract showed highest antibacterial activity against gram positive bacteria i.e. 20 mm and against gram negative bacteria i.e. 24 mm. Hence, from the present investigation it may also be interpreted that Phyllanthus niruri plant had good antibacterial activity and may be used as an alternative food preservative in food industries.

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