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Comparative Study of Corrosion Inhibition Efficiency of *Phyllanthus niruri* for Tin in Different Solutions of HCl and H_2SO_4.

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Abstract: *The corrosion inhibition of tin in different solutions (1N, 2N, 3N) of HCl and H_2SO_4 by the leaves extract of Phyllanthus niruri has studied by using weight loss and thermometric methods at 303K temperature inhibition was found to increase with increasing concentration of the extract. Results indicate that Phyllnthus niruri leave extract was an efficient natural corrosion inhibitor in the acidic media. In case of 3N HCl maximum inhibition efficiency (96.96%) was noted at 0.7% in inhibition concentration and in 3N H_2SO_4, it was found 91.30% efficiency at the same concentration of inhibitor. At lower concentration of inhibitor, better inhibition was observed in H_2SO_4 medium as compared to HCl. The decreased corrosion rate of metal was due to absorption of plant extract on metal surface. Absorption of Phyllanthus niruri depends on its chemical compositions which showed the presence of various compounds like alkaloids, flavonoids, steroids, tannins etc. which has O, N and S atoms with lone pair electrons to form co-ordinate bonding with metal.*

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

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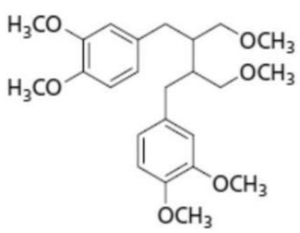
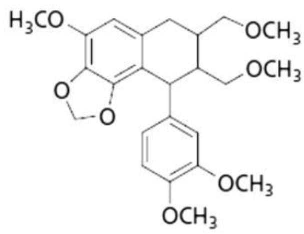
I. INTRODUCTIO1N

Corrosion is a natural destructive process. In this, metal is destructed gradually by chemical or electrochemical reaction with their surrounding environment [1-2]. Many metals are affected by this destructive process. Tin and its alloys with copper have been used from more than thousand years for making tools and weapons [3-4], bronze is a common example which has been used. Now a days tin and its alloys are used to make cans for packaging food and other commercial purposes[5]. Tin does not corrode in pure water even in moisture. It forms a stable oxide film on its surface but in acidic media viz. hydrochloric acid, sulphuric acid, and nitric acid, it gets corroded[4-5]. Naturally occurring plant product are easily available, less toxic, economic, eco-friendly and biodegradable so they are widely used as corrosion inhibitors without any side effects[6-8].

Phyllanthus niruri plant was selected for this study. It is very common in climatic condition of Rajasthan as well as in India and it is known as bhui amla or jangli amla. It is bitter in taste and is used to cure liver and kidney disorder. The plant has different classes of organic compound[9] like, alkaloids, flavanoids, lactose, steroids terpenoids, ligame, tannins etc., main alkaloids are phyllanthin, hypophyllanthin, phyllanthol, phyllanthenol etc.



Phyllanthin



Hypophyllanthin

Heterocyclic compounds containing O, N and S atoms have been used as corrosion inhibitors[10-15] in acid media. They are found to have higher basicity and electron density. O, N and S are the active centres in adsorption process on the metal surface. The electron density, orientation, size and shape of the molecule play significant role in the effectiveness of inhibition. They are used as corrosion inhibitor since they got adsorbed on the metallic surface and blocked the active corrosion site due to that liberation H^+ and dissolution metal ion is stopped in acid media.

II. MATERIALS AND METHODS

2.1 Preparation of Leaves Extract:

Fresh *Phyllanthus niruri* plants were collected from Theme Park Botanical garden of RIE, Ajmer. The leaves of the plant were air dried at room temperature, then grinded to make powder. The extract of powder leaves of *Phyllatnus niruri* obtained by refluxing the dried leaves in soxhlet unit in ethanol solvent with refluxing by heating for sufficient time (3-4 hrs.).

2.2 Metal used:-

Tin foil was used for all experiments. Specimens of tin foil were prepared by cutting the sheet of pure tin. Specimens were prepared in square shaped having dimension of 2.0 cm × 1.5 cm with a small hole of about 2mm diameter near the upper edge.

2.3 Chemicals used:-

1N, 2N, 3N different solutions of HCl and H_2SO_4 were prepared using analytical grade reagents and these acid solutions were used for corrosion analyses and for preparation of inhibitor solutions of different composition. (i.e. 0.1%, 0.3%, 0.5% and 0.7%)

III. METHODS

3.1 Weight loss method:-

Each specimen was suspended by a V-shaped glass hook made of fine capillary and plunged into a beaker containing 50 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 was calculated **[16-17]** by this 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 by inhibitor was calculated **[18]** by this equation:

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

3.2 Thermometric method:

Inhibition efficiencies were also determined by using thermometric method, this involved the immersion of single specimen of area 6cm^2 in reaction chamber containing 50mL solution of acid at an initial temperature of 303K.Tests were carried out in 1 N, 2 N, 3 N, acid solutions and 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 intervals of 5 minutes using thermometer with a precision of ± 0.1k. The temperature increased slowly at first, then rapidly and attained a maximum value, before falling the maximum temperature was recorded.

The reaction number(R N) is calculated[19-20] by this equation:

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

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

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

Where RN_f and RN_i are the reaction number in the absence, and in the presence of inhibitor, respectively.

The corrosion rate (CR) in mm/year can be calculated[21-24] by the following equation:

$$\text{Corrosion rate (mm/yr.)} = \frac{(\Delta W \times 87.6)}{(A \times T \times d)}$$

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.

IV. RESULTS AND DISCUSSIONS

The corrosion rate of tin in HCl (1N, 2N, 3N) and H_2SO_4 (1N, 2N, 3N) solutions were studied by weight loss method in absence and presence of leaves extract of *Phyllanthus niruri* plant at 303K temperature and percentage inhibition efficiency was calculated. Weight loss data, percentage inhibition efficiency, corrosion rate and surface coverage for different concentration of HCl and H_2SO_4 solution with different concentration of inhibitor are given in table-1 and table-2 and corresponding graphs for both acids are shown in fig.-1 and fig.2.The data was utilized for calculation of reaction number and percentage of inhibition efficiencies are given in table -3, and 4 for both acid HCl and H_2SO_4 and corresponding graphs are shown in fig.-3 and fig.-4.

Table 1

Corrosion rate, % inhibition eficiency and surface coverage data obtained from weight loss method for tin in HCl (1N, 2N, 3N) in the absence and with different concentrations of inhibitors.

Temperature: 303K ±0.1K **Area of specimen: 6 cm^2**

Inhibitor concentration %	Corrosion rate (mm/yr)			% Inhibition efficiency (η%)			Surface coverage (θ)		
	3N HCl	2N HCl	1N HCl	1N HCl	2N HCl	3N HCl	1N HCl	2N HCl	3N HCl
Blank/ Uninhibited	0.00398	0.00165	0.00161						
0.1	0.00080	0.00046	0.00048	70.00	72.15	79.79	0.7000	0.7215	0.7979
0.3	0.00048	0.00039	0.00043	72.72	75.94	87.87	0.7272	0.7594	0.8787
0.5	0.00020	0.00027	0.00039	75.32	83.54	94.94	0.7532	0.8354	0.9494
0.7	0.00012	0.00018	0.00027	83.11	88.60	96.96	0.8311	0.8860	0.9696

Table 2

Corrosion rate, % inhibition efficiency and surface coverage data obtained from weight loss method for tin in H_2SO_4 (1N, 2N, 3N) in the absence and with various concentration of inhibitors.

Temperature: 303K ± 0.1K **Area of specimen: 6 cm^2**

Inhibitor concentration %	Corrosion rate (mm/yr)			% Inhibition efficiency (η%)			Surface coverage (θ)		
	3N H_2SO_4	2N H_2SO_4	1N H_2SO_4	1N H_2SO_4	2N H_2SO_4	3N H_2SO_4	1N H_2SO_4	2N H_2SO_4	3N H_2SO_4
Uninhibited	0.00144	0.00067	0.00026						
0.1	0.00060	0.00043	0.00017	34.48	35.41	57.97	0.3448	0.3541	0.5797
0.3	0.00043	0.00040	0.00014	44.82	39.58	64.56	0.4482	0.3958	0.6456
0.5	0.00039	0.00029	0.00014	44.86	56.25	72.46	0.4486	0.5625	0.7246
0.7	0.00012	0.00018	0.00010	62.06	72.91	91.30	0.6206	0.7291	0.9130

Table- 1, 2 showed the variation of corrosion rate (mm/yr.), percent inhibition efficiency (η %) and surface coverage (θ) with varying concentration of inhibitor. From the data it was observed that corrosion rate was decreased in presence of inhibitor.

The corrosion rate was found dependent on the concentration of inhibitor, with increase in concentration the corrosion rate lowered gradually. Figure- 1 and 2 shown the variation of percentage inhibition efficiency with concentration of inhibitor and it was found that inhibition efficiencies increases with the increasing the concentration of inhibitor, in both the case of HCl and H_2SO_4.

In case of 3N HCl, maximum inhibition efficiency (96.96%) was noted at 0.7% concentration of inhibitor, shown fig-1, whereas in 3N H_2SO_4, it was found 91.30% at the same concentration of inhibitor, shown in fig.2, at the lower concentration of inhibitor, better inhibition was observed in H_2SO_4 medium as compared to HCl.

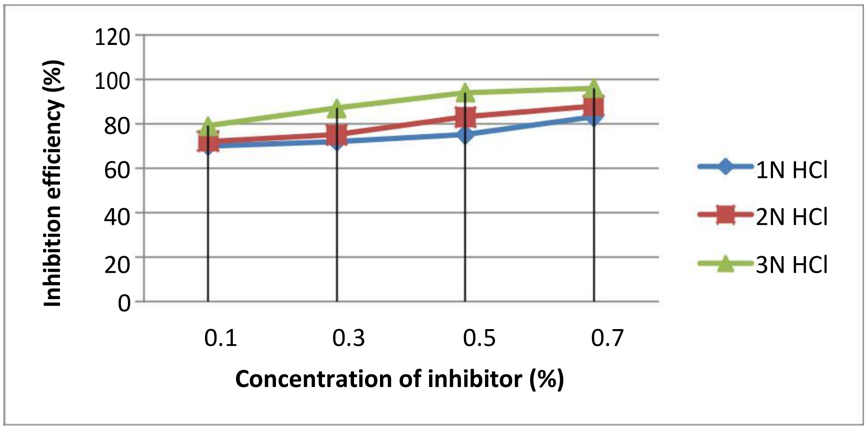


Fig.1

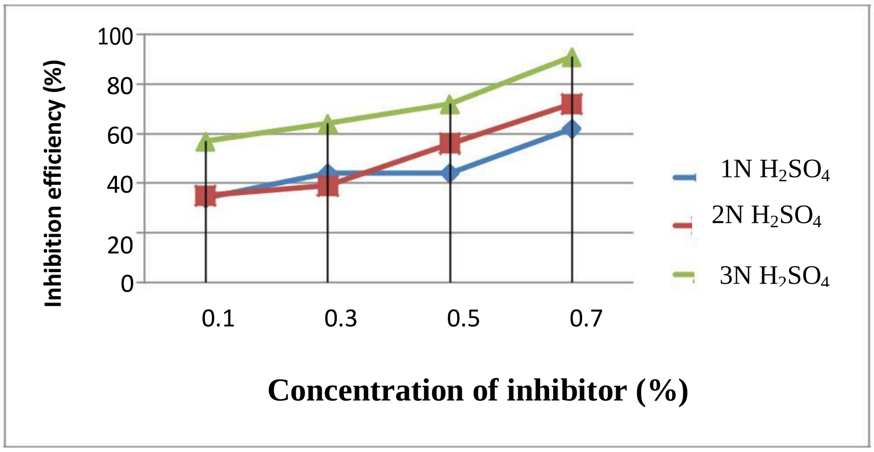


Fig. 2

Table 3

Reaction number and inhibition efficiencies (η %) for tin in HCl of different solutions (3N, 2N, 1N) in the absence and in presence of various concentration of inhibitor.

Temperature: 303K ± 0.1K **Area of specimen : 6 cm^2**

Conc. of Inhibitor (in %)	3N HCl		2N HCl		1N HCl	
	RN	IE (η %)	RN	IE (η %)	RN	IE (η %)
Blank/ Uninhibited	0.036	-	0.059	-	0.065	-
0.1	0.018	50.00	0.029	50.84	0.035	46.15
0.3	0.014	61.11	0.027	54.23	0.031	52.30
0.5	0.012	66.66	0.024	59.32	0.027	58.46
0.7	0.009	75.00	0.017	71.18	0.021	67.69

Table-4

Reaction number and inhibition efficiencies (η %) for tin in H_2SO_4 of different solutions (3N, 2N, 1N) in the absence and in presence of various concentration of inhibitor.

Temperature : 303K±0.1K **Area of specimen : 6 cm^2**

Conc. of Inhibitor (in %)	3N H_2SO_4		2N H_2SO_4		1N H_2SO_4	
	RN	IE(η %)	RN	IE(η %)	RN	IE(η %)
Blank/ Uninhibited	0.086	-	0.099	-	0.011	-
0.1	0.033	61.62	0.042	57.57	0.046	24.59
0.3	0.030	65.11	0.039	60.60	0.042	31.14
0.5	0.027	68.60	0.035	64.64	0.039	36.06
0.7	0.024	72.09	0.029	70.70	0.032	47.54

Table -3,4 given, the variation in reaction number and inhibition efficiencies with varying the concentration of inhibitors and it was observed that the reaction number decreased and inhibition efficiency increased with increasing concentration of inhibitors for HCl as well as H_2SO_4 . In case hydrochloric acid , maximum efficiency (75 %) and minimum reaction number (0.009) were noted for 3N HCl at 0.7 % inhibitor concentration , in of case 3N H_2SO_4, reaction number and inhibition efficiency were 0.024 and 72.09% respectively at same concentration of inhibitors as shown in fig-3, 4

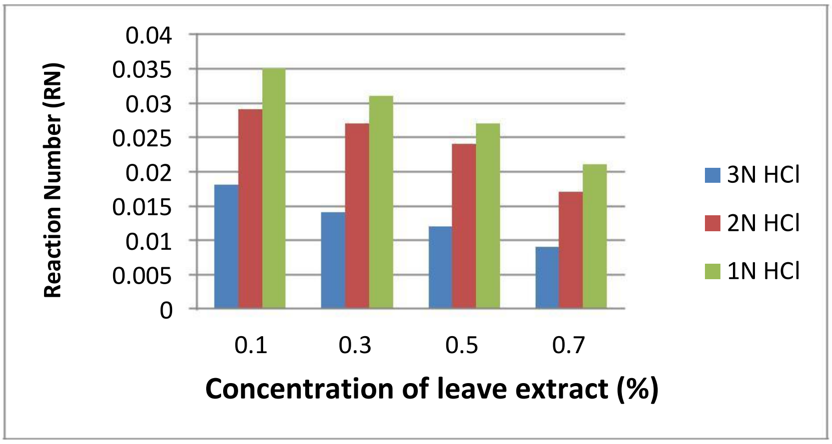


Fig-3(a): Variation of reaction number with concentration of leave extract of *Phyllanthus niruri* in HCl of different solutions (3N, 2N, 1N)

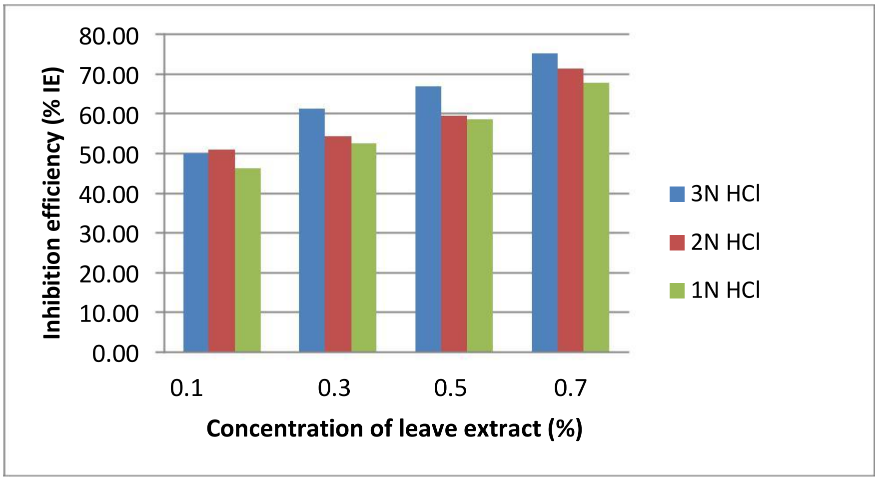


Fig-3(b): Variation of inhibition efficiency with different concentrations of leave extract of *Phyllanthus niruri* in HCl of different solutions (3N, 2N, 1N)

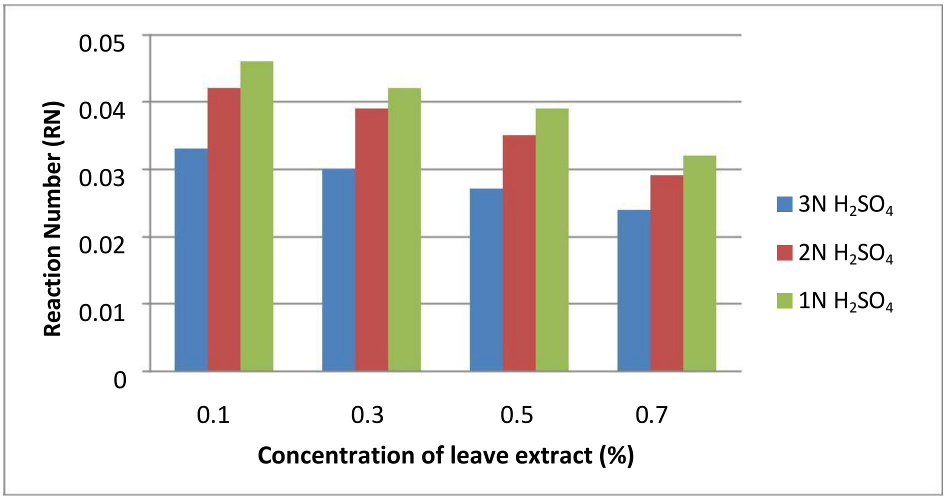


Fig-4(a): Variation of reaction number with concentration of leave extract of *Phyllanthus niruri* in H_2SO_4 of different solutions (3N, 2N, 1N)

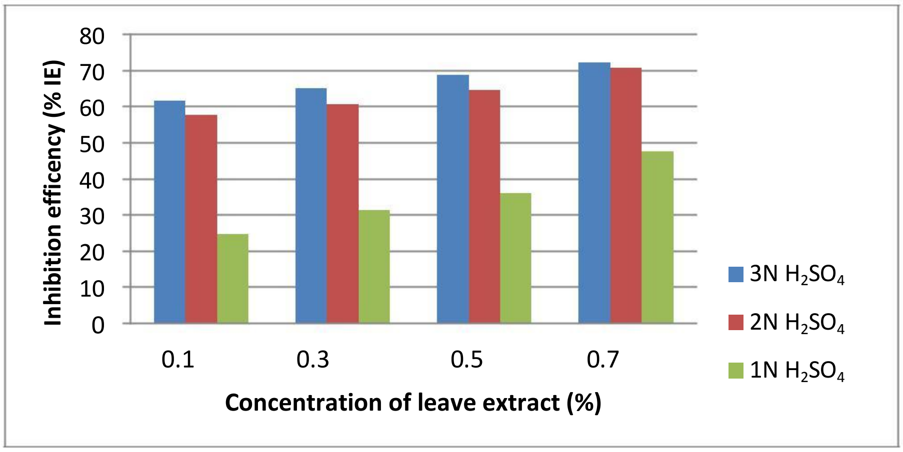


Fig-4 (b): Variation of inhabitant efficiency with different concentrations of leave extract of *Phyllanthus niruri* in H_2SO_4 of different solutions (3N, 2N, 1N)

The decreasing in corrosion rate and increasing inhibition efficiency was attributed to the fact that the adsorption of inhibitor on the metal surface, due to absorption of inhibitor, the corrosion sites of metal surface gets blocked and absorbed film of inhibitor acts as physical barrier between metal surface and corrosion medium.

V. CONCLUSION

The study of leave extract of *Phyllanthus niruri* has showed that it would be better corrosion inhibitor for tin in different solutions of HCl and H_2SO_4 and inhibition efficiency of leave extract increased with increasing extract concentration . Data and results from the tables and graphs are indicating that leave extract of *Phyllanthus niruri* is an efficient natural corrosion inhibitor in the acidic media and by both the method, weight loss and thermometric. It was found that inhibition efficiency of leave extract is more in hydrochloric acid than the sulphuric acid , by weight loss method, it was observed that the maximum inhibition efficiency (96.96%) for 3N HCl at 0.7% inhibition concentration whereas in 3N H_2SO_4 , it was reported 91.30% efficiency at the same concentration of inhibitor and by thermometric method, maximum efficiencies (75%) and minimum reaction number (0.009) were noted at 0.7 % concentration for 3N HCl and in case of H_2SO_4 , 72.09% and 0,024 were noted, respectively, at the same concentration of inhibitor and at the lower concentration of inhibitor, better inhibition was observed in H_2SO_4 medium as compared to HCl.

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