Available online at www.elixirpublishers.com (Elixir International Journal)

Corrosion and Dye

Elixir Corrosion & Dye 116 (2018) 49989-49993

ISSN: 2229-712X

Corrosion Inhibition of Tin by Alkaloid Extract of Aerial Part of Phyllanthus Niruri in HCl and H_2SO_4 Solutions

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ARTICLE INFO

Article history:
Received: 21 January 2018;
Received in revised form:
24 February 2018;
Accepted: 3 March 2018;

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

ABSTRACT

The corrosion inhibition of tin by *Phyllanthus niruri* extract in different solutions of HCl (1N, 2N, 3N) and H_2SO_4 (1N, 2N, 3N) was investigated by weight loss and thermometric methods at 301K temperature. From the study it was found that the inhibition efficiency increases with the increase in the concentration of extract in HCl and H_2SO_4 solutions. Results indicate that *Phyllanthus niruri* extract was an efficient natural corrosion inhibitor in acidic media. The stem extract of the *Phyllanthus niruri* is better than leave extract of the plant. Absorption of *Phyllanthus niruri* depends on its chemical compositions which showed the presence of various compounds like alkaloids, flavanoids, steroids, tannins etc. which has O, N and S atoms with lone pair electrons to form co-ordinate bonding with metal.

Introduction

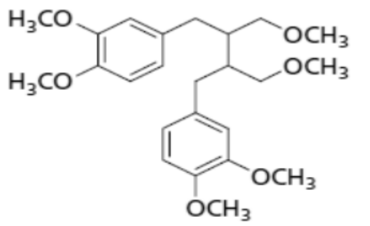
Corrosion is a natural destructive process. In this, metal is destructed gradually by chemical or electrochemical reaction with their surrounding environment [1-3]. Many metals are affected by this destructive process. Tin and its alloys with copper have been used from more than thousand years for making tools and weapons [4-5], 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[6].

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[5-6].

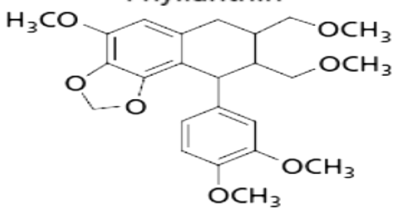
Naturally occurring plant product are easily available, less toxic, economic, eco-friendly and biodegradable so they are widely used as corrosion inhibitors without any side effects[7-9].

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 bhumi amla or jangli amla.

It is bitter in taste and it is used in liver and kidney disorder, and the most is very useful in medicine. The plant has different classes of organic compound[10] like, alkaloids, flavanoids, lactose, steroids, terpenoids, ligame, tannins etc. Main alkaloids are phyllanthin, hypophyllanthin, phyllanthol, phyllanthenol etc.



Phyllanthin



Hypophyllanthin

Heterocyclic compounds containing[11-16] O, N and S atoms have been used as corrosion inhibitors 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 of H^+ and dissolution metal ion is stopped in acid media.

Materials And Methods

Preparation of Leaves Extract:

Fresh *Phyllanthus niruri* plants were collected from Theme Park Botanical garden of RIE, Ajmer. The plant

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(including leaves, stem and roots) was air dried at room temperature, then grinded to make powder. The extract of powder leaves and powder stem of *Phyllanthus niruri* were obtained by refluxing the dried leaves and stem in soxhlet unit in ethanol solvent with refluxing by heating for sufficient time (8-10) hrs.).

Metal used:-

Tin foil was used for all experiments. Specimens of tin foil were prepared by cutting the sheet of pure tin. Specimens was 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.

Chemicals used:-

1N, 2N, 3N different solutions of HCl and H_2SO_4 were prepared using analytical grade concentration 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%)

Methods

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 [17-18] 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 [19-21] by this equation

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

Thermometric method:

Inhibition efficiencies were determined by using thermometric method technique. This involved the immersion of single specimen of area $6cm^2$(both sides) in reaction chamber containing 50 mL solution of acid at an initial temperature of 301K.Tests were carried out in 1N, 2N, 3N, 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 -0.1^0C. The temperature increased slowly at first, then rapidly and attained a maximum value before failing the maximum temperature was recorded.

The reaction number(R N) is calculated by this equation:

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

Table 1.Inhibition efficiencies (η%) for Tin in HCl (1N, 2N, 3N) solutions with P*hyllanthus niruri* extract of aerial part.

Temperature: 301K ± 0.1K **Area of specimen: 6 cm^2**

Inhibitor Concentration(%)		Corrosion rate (CR) (mm/yr)			Inhibition efficiency (IE) (η%)			Surface coverage(θ)		
		3N HCl	2N HCl	1N HCl	1N HCl	2N HCl	3N HCl	1N HCl	2N HCl	3N HCl
Blank	Stem extract	0.00268	0.00149	0.00137						
	Leave extract	0.0398	0.00165	0.00161						
0.1	Stem extract	0.00048	0.00026	0.00038	71.64	82.19	81.81	0.7164	0.8219	0.8181
	Leave extract	0.00080	0.00046	0.00048	70.00	72.15	79.79	0.7000	0.7215	0.7979
0.3	Stem extract	0.00038	0.00022	0.00026	80.59	84.93	85.71	0.8059	0.8493	0.8571
	Leave extract	0.00048	0.00039	0.00043	72.72	75.94	87.87	0.7272	0.7594	0.8787
0.5	Stem extract	0.00017	0.00014	0.00022	83.58	90.41	93.50	0.8358	0.9041	0.9350
	Leave extract	0.00020	0.00027	0.00039	75.32	83.54	94.94	0.7532	0.8354	0.9494
0.7	Stem extract	0.00006	0.00010	0.00018	86.56	94.44	97.49	0.8656	0.9444	0.9740
	Leave extract	0.00012	0.00018	0.00027	83.11	88.60	96.96	0.8311	0.8860	0.9696

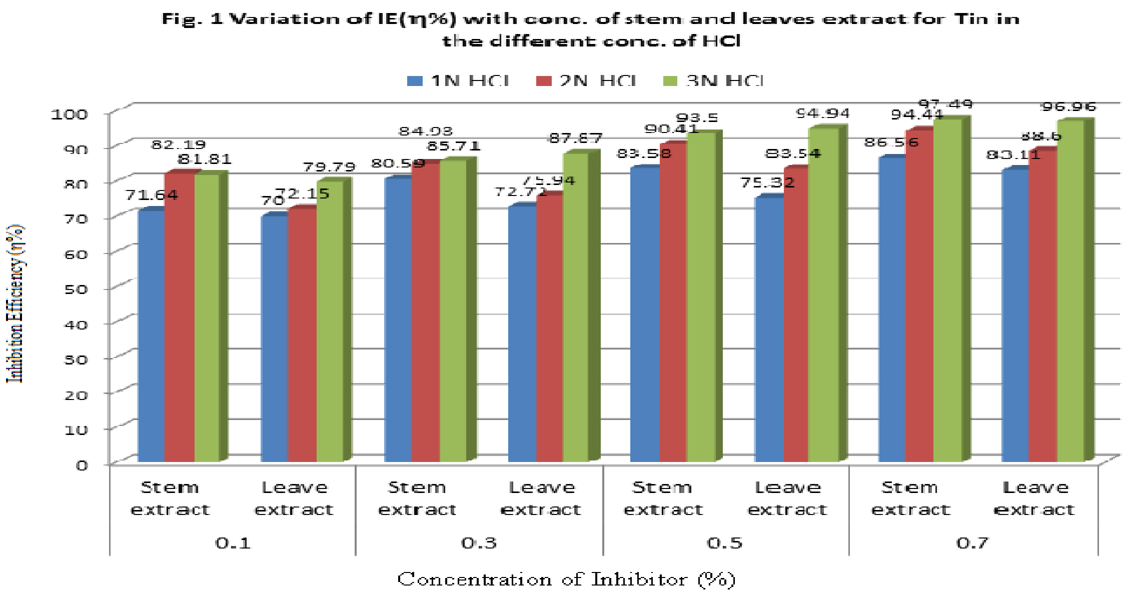

Fig 1. Variation of IE(η%) with conc. of stem and leaves extract for Tin in the different conc. of HCl.

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:

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

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

The corrosion rate (CR) in mm/year can be calculated by the following equation[22-26].

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

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.

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 and stem extract of *Phyllanthus niruri* plant at 301K temperature and percentage inhibition efficiency were calculated in both the conditions.

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. Weight loss, percentage inhibition efficiency, surface coverage and corrosion rate for different concentration of HCl solution with different concentration of inhibition are given in table 1.

We can see and observed from the table-1 that inhibition efficiency decreases with increasing strength of hydrochloric acid and inhibition efficiency increases with increasing concentration of extract in strength of each acid solution.

The maximum efficiency has been observed in highest concentration of HCl i.e. 3N HCl with highest concentration of inhibitor i.e. of 0.7% (97.49) for stem extract. Whereas it is (96.96) for leaves extract in same HCl concentration. The corrosion rate has been observed maximum in blank solution and it decreased with the increasing concentration of inhibitor in HCl solution of different strength. Corresponding variation of inhibition efficiencies with concentration of inhibitor are shown in figure -1 for different concentration of HCl solution.

Table 2.Inhibition efficiency (η%) for Tin in H_2SO_4 solution with extract of *Phyllanthus niruri*

Temperature: 301K ± 0.1°C Area of specimen: 6 cm^2

Inhibitor Concentration(%)		Corrosion rate (CR)(mm/yr)			Inhibition efficiency (IE) (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
Blank	Stem extract	0.00118	0.00053	0.00163						
	Leave extract	0.00144	0.00067	0.00026						
0.1	Stem extract	0.00046	0.00028	0.00013	50.00	47.50	61.01	0.5000	0.475	0.6101
	Leave extract	0.00060	0.00043	0.00017	34.48	35.41	57.97	0.3448	0.3541	0.5797
0.3	Stem extract	0.00030	0.00025	0.00010	52.94	52.50	74.57	0.5294	0.525	0.7457
	Leave extract	0.00043	0.00040	0.00014	44.82	39.58	64.56	0.4482	0.3958	0.6456
0.5	Stem extract	0.00026	0.00014	0.00009	67.64	72.50	77.96	0.6764	0.725	0.7996
	Leave extract	0.00039	0.00029	0.00014	44.86	56.25	72.46	0.4486	0.5625	0.7246
0.7	Stem extract	0.00008	0.00009	0.00007	70.00	82.50	93.22	0.7000	0.825	0.9322
	Leave extract	0.00012	0.00018	0.00010	62.06	72.91	91.30	0.6206	0.7291	0.9130

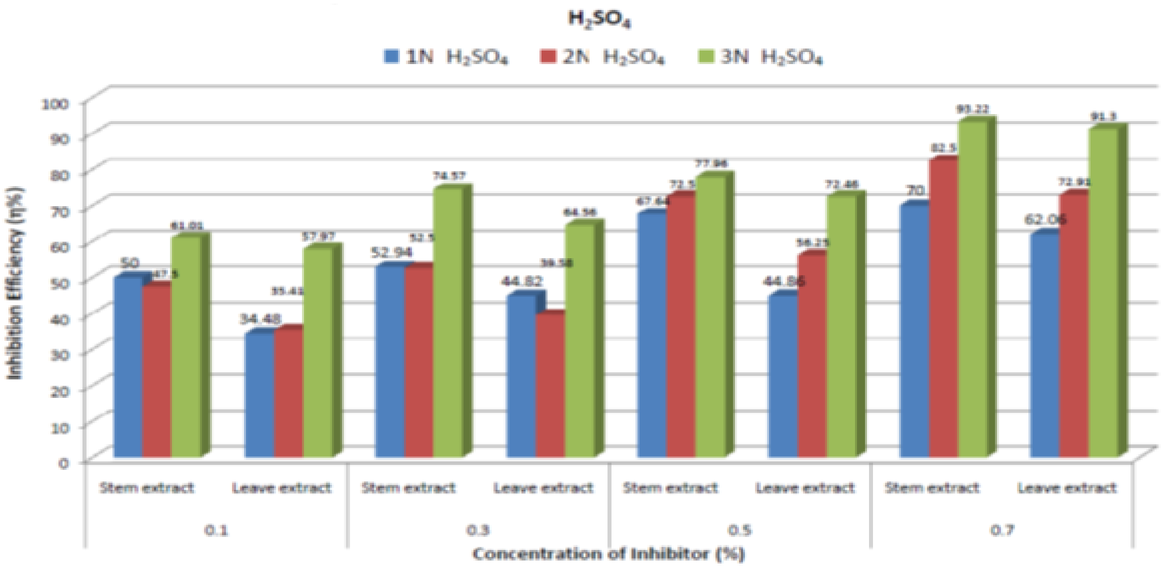

Fig 2.Variation of IE(%)with conc. of stem and leaves extract for tin in the different conc. of H_2SO_4.

Table -2 shows results of H_2SO_4solution from the table it is observed that inhibition efficiency increases with increasing strength of H_2SO_4solution and it is also increases with increasing concentration of extract in strength of each acid solution. The maximum inhibition efficiency has been observed in highest and concentration i.e. 3N H_2SO_4 with highest concentration of inhibitor i.e. 0.7% (93.22) for stem extract, whereas it is (91.30) for leaves extract in same acid concentration. The corrosion has been observed maximum in blank solution and it decreases with increasing concentration of inhibitor in H_2SO_4 solution of different strength. Corresponding variation of inhibition efficiencies with concentration of inhibitor are shown in figure -2 for different concentration of H_2SO_4 solution.

Table 3. Reaction number and Inhibition efficiency (η%) for Tin in HCl solution with *Phyllanthus niruri* extract

Temperature: 301K ± 0.1°C Area of specimen: 6 cm^2

Inhibitor Concentration(%)		3N HCl		2N HCl		1N HCl	
		R N	I E (η%)	R N	I E (η%)	R N	I E (η%)
Blank	Stem Extract	0.035		0.056		0.058	
	Leave Extract	0.036		0.059		0.065	
0.1	Stem Extract	0.020	42.85	0.026	53.57	0.030	48.27
	Leave Extract	0.018	50.00	0.029	50.84	0.035	46.15
0.3	Stem Extract	0.015	57.14	0.021	62.50	0.028	51.72
	Leave Extract	0.014	61.11	0.027	54.23	0.031	52.30
0.5	Stem Extract	0.010	71.42	0.017	69.64	0.025	56.89
	Leave Extract	0.012	66.66	0.024	59.32	0.027	58.46
0.7	Stem Extract	0.005	85.71	0.013	76.78	0.018	68.96
	Leave Extract	0.009	75.00	0.017	71.18	0.021	67.69

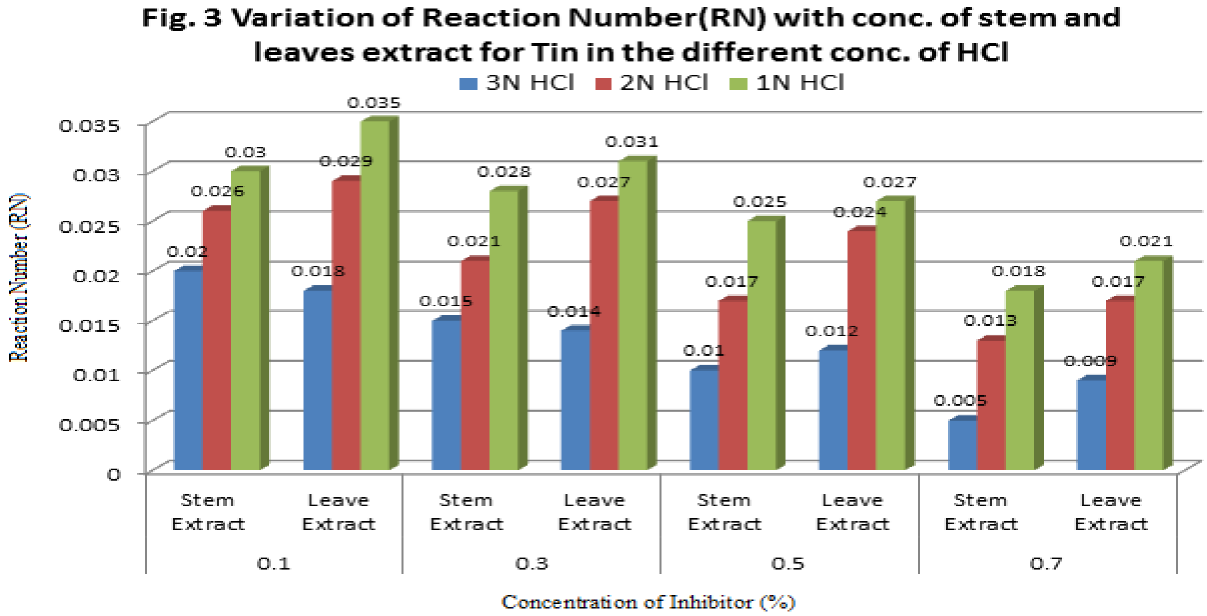

Fig 3. Variation of Reaction Number(RN) with conc. of stem and leaves extract for Tin in the different conc. of HCl.

Table shows that the corresponding data of reaction number (RN) with concentration of inhibitor in 1N, 2N and 3N HCl solution.

Table -3 indicates that reaction number increases with increasing strength of HCl solution as well as it decreases with increasing concentration of inhibitor in each solution.

Inhibition efficiency increases with as it decreases with increasing strength of HCl solution corresponding curves for the variation in reaction number with concentration of inhibitor are in figure-3 for different concentration of HCl solution.

Table 4. Reaction number (N) and Inhibition efficiency (η%) for Tin in H_2SO_4 solution with *Phyllanthus niruri* extract

Temperature: 301K ± 0.1°C Area of specimen: 6 cm^2

Inhibitor Concentration(%)		3N H_2SO_4		2N H_2SO_4		1N H_2SO_4	
		RN	I E (η%)	R N	I E (η%)	R N	I E (η%)
Blank	Stem extract	0.032	-	0.0083	-	0.010	
	Leave Extract	0.033		0.099		0.011	
0.1	Stem extract	0.029	9.37	0.0069	16,86	0.0060	40.00
	Leave Extract	0.033	61.62	0.042	57.57	0.046	24.59
0.3	Stem extract	0.022	31.25	0.0055	33.73	0.0046	54.00
	Leave Extract	0.030	65.11	0.039	60.60	0.042	31.14
0.5	Stem extract	0.017	46.87	0.0020	75.90	0.0041	59.00
	Leave Extract	0.027	68.60	0.035	64.64	0.039	36.06
0.7	Stem extract	0.009	72.87	0.0006	92.03	0.0029	73.00
	Leave Extract	0.024	72.09	0.029	70.70	0.032	47.54

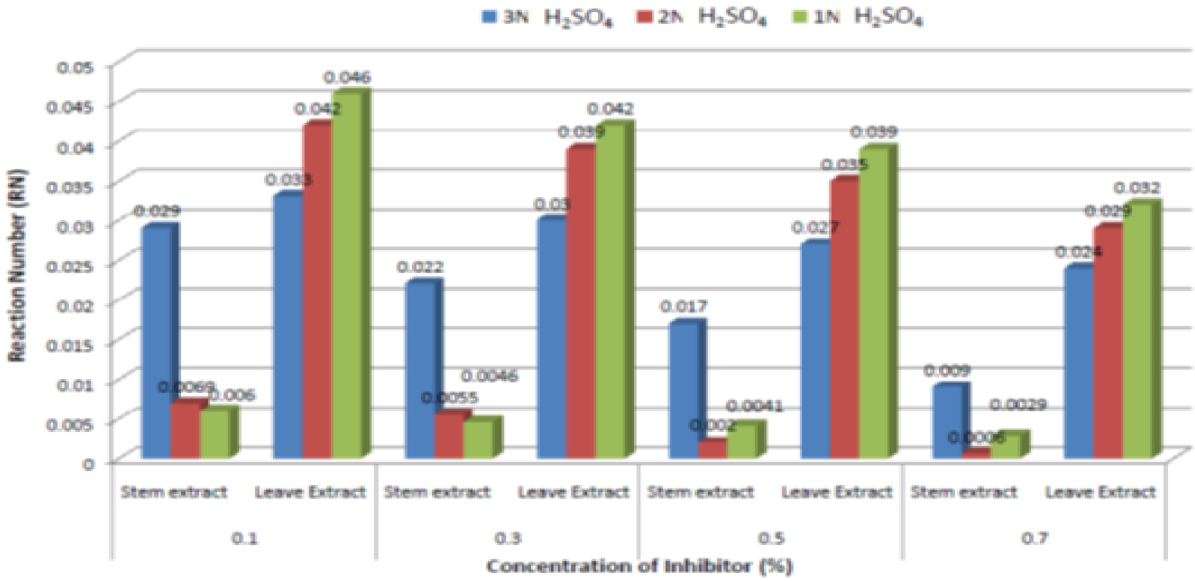

Fig. 4 Variation of Reaction Number(RN) with conc. of stem and leaves extract for Tin in the different conc. of H_2SO_4.

Result for H_2SO_4solution of reaction number shown in table- 4 indicate that reaction number increases with increasing strength of acid solution as well as it decreases with increasing concentration of inhibitor of each solution.

Inhibition efficiency increases with increasing concentration of inhibitor in each acid solution as well as increases with increasing strength of H_2SO_4 solution.

Corresponding curves for the variation in reaction number with concentration of inhibitor are shown in fig.-4 for different concentration of H_2SO_4 solution.

It is clear in both cases of acids the inhibition efficiency is maximum at higher concentration. This is because of fact that in the case of H_2SO_4 oxygen atom formed a protective layer. On the metal surface, which essentially block the discharge of H^+ and dissolution of metal ion in acid media so they are reduce the corrosion rate of tin with inhibitor.

Conclusion

The study of aerial part of *Phyllanthus niruri* has showed that it would be better corrosion inhibitor for tin in different solution of HCl and H_2SO_4. From the study it was found that the inhibition efficiency increases with the increase in the concentration of extract in HCl and H_2SO_4 solutions. Results indicate that *Phyllanthus niruri* extract was an efficient natural corrosion inhibitor in acidic media. The maximum inhibition efficiency shown by stem extract was 97.49 for 0.7% concentration for 3N HCl solution, whereas it was observed 93.22 for 3N H_2SO_4in the same concentration of inhibitor .It was conclude that extract of stem is better corrosion inhibitor than the leaves.

Acknowledgement

One of author (O. P. Meena) is grateful to Regional Institute of Education (NCERT), Ajmer as employee.

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