



J. Environ. Nanotechnol.
Volume 3, No.1 pp.01-08
ISSN (Print) : 2279-0748
ISSN (Online) : 2319-5541
doi : 10.13074/jent.2014.01.132041

Datura Metal as a Potential Corrosion Inhibitor for Aluminium in 1M HCL solution

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Received: 12.07.2013 Accepted: 01.01.2014

Abstract

The corrosion inhibition of commercial Al in 1M HCl medium by Datura metal (DM) leaves extract was carried out using chemical and electrochemical techniques. Effect of temperature, immersion time and acid concentration were discussed. The inhibition efficiency increases with increase in concentration. The inhibition efficiencies were also evaluated using hydrogen evolution technique at 303K. Using potentiodynamic polarisation technique the extract proved that mixed type inhibitor. EIS showed that the charge transfer controls the corrosion process. These results revealed that Datura metal (DM) was an effective inhibitor and the inhibition efficiencies obtained from weight loss, hydrogen evolution measurements, potentiodynamic polarisation and ac impedance technique were in good agreement. Phytochemical analyses show that the leaves extract containing alkaloid and steroids. These are responsible for inhibition. The inhibitive action of the extract was discussed using Langmuir adsorption isotherm. Thermodynamic and kinetic parameters such as activation energy, free energy of adsorption, change in entropy and change in enthalpy were also calculated. The negative value of the free energy of adsorption indicates that the adsorption of inhibitor molecules was spontaneous. Physical adsorption proposed from activation parameters.

Keywords: Adsorption isotherm; Aluminium; Corrosion inhibitor; Datura metal.

1. INTRODUCTION

Aluminium is one of the world 3rd most commonly used metal due to its lightness coupled with its strength, conductivity, barrier properties and its excellence. It is widely used in many industries, reaction vessels, pipes, machinery and aeronautics because of cheaper cost and high conductance in nature. Aluminium oxide seals the core aluminium from any further reaction. The corrosion resistance of aluminium depends upon this protective oxide film, which is

stable when the pH is between about 4.5 and 8.5 (Sharma, 2002) below and above the pH it starts corrosion. In the present study, HCl is used as a medium. HCl solutions are used for pickling, chemical or electrochemical etching for Al foil. Most of the corrosion inhibitors are synthetic chemicals, pollute the environment and also toxic in nature. In order to avoid chemicals, the field of research shifted to eco-friendly corrosion inhibitors.

The naturally occurring plant products are environmentally compatible, non-polluting, less toxic, easily available, bio-degradable and are cheap corrosion inhibitors. Therefore, it is desirable source as green inhibitor for Al (El-Etre *et al.* 2003; Jothi and

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Ravichandran, 2013; Gunavathy and Murugavel, 2013). A large number of plant organic compounds containing heteroatom such as S, N, O and extended conjugation in their molecule is considered as good corrosion inhibitor. Plant inhibitors are incredibly rich sources of natural chemical compounds (e.g. alkaloids, terpenoid, steroid, flavonoid, triterpenes, phenolic compounds and tannins) were known for inhibitive action. By using weight loss method, hydrogen evolution method, potentiodynamic polarisation and electrochemical impedance spectroscopy method were used to study the corrosion rate and its inhibition efficiency of Datura metal (DM) leaves extract for Al in 1M HCl solution. Datura metal Linn. is a family Solanaceae and its common name is called as Ummetta, Umattai, Dhustura and Devil's trumpet. The phytochemical screening of the leaves of Datura metal revealed that alkaloids, tripenoid, steroid, flavonoid, triterpenes, phenolic compounds and tannins were present.



Fig. 1:

2. MATERIALS & METHODS

2.1 Materials

2.1.1 Preparation of leaves extract

The leaves were washed with water, shade dried for a few days and then the dry leaves were ground well and made into fine powder. 20 g of the sample was

dissolved in 200 ml of double distilled water and refluxed in 250 ml R.B. flask for one hour and kept aside for overnight. The refluxed solution was filtered carefully and the filtrate was evaporated over the waterbath until solid mass was obtained. The solid mass was ground well. Then the accurate amount of inhibitor was weighed and dissolved in acid solution.

2.1.2 Preparation of the specimens

The experiment was performed with Al pieces of size 5 x 1 x 0.15 cm for weight loss measurement and for hydrogen evolution method. Aluminium was in the form of cylindrical rod. The rod is embedded using Teflon holder with flat surface was the only exposed area at bottom of the rod with 0.2826 cm² was used for electrochemical study.

2.2 Methods

Two different methods have been used for studying the inhibition efficiency for corrosion of aluminium by Datura metal leaves extract are chemical method (weight loss and hydrogen evolution method) and electrochemical techniques (potentiodynamic polarisation and ac impedance technique).

2.3 Chemical methods

2.3.1 Weight loss measurements

The experiment was performed with aluminium pieces which were polished with emery sheets and then washed with double distilled water, cleaned, degreased with acetone and etched well. Accurate weight of the metal pieces was found from digital balance. 100 ml of each inhibitor concentration ranging from (i.e.) 0.3, 0.6, 0.9, 1.2 and 1.5 g L⁻¹ were taken in separate 100 ml open beakers and the metal pieces of accurately known weight were fully dipped in standing position at room temperature. It was removed from the solutions after 1h, 2h, 3h and washed with double distilled water, dried and then polished with emery paper grade from 300 to 2000. Again the metal pieces were washed with double distilled water, cleaned, dried and weighed.

2.3.2 Gasometry

A 100ml solution of 1M HCl solution was introduced into the reaction vessel connected to a burette through a delivery tube. The initial volume of air in the burette was recorded. An Al specimen is dropped into the solution of HCl and the reaction vessel quickly closed to prevent any escape of H₂ gas. The volume of H₂ gas evolved from the corrosion reaction was monitored by the volume change in the level of the paraffin oil in the graduated burette. The change in volume was recorded every 3 minute of period. The experiment was performed at different concentrations (0.3, 0.6, 0.9, 1.2 and 1.5 g L⁻¹).

2.4 Potentiodynamic polarisation and EIS measurements

Electrochemical experiments was carried out in the conventional three electrode cell with a platinum counter electrode (CE), a saturated calomel electrode (SCE), used as reference electrode and Al as working electrode (WE). In order to minimise ohmic contribution, the Luggin capillary was placed close to the working electrode. The working electrode used in electrochemical measurements were polished with emery papers in the range (300-2000) grit, rinsed with double distilled water, acetone and dried in air. The working electrode was immersed in the test solution before 30 minutes to attain the steady state. The electrochemical measurements were performed in the test solution after measurement of the open - circuit potential (E_{ocp}). EIS measurement was carried out in the 100 kHz -10 MHz frequency range. The polarisation curves were carried out from cathodic potential of -0.2 V to +0.2 V with respect to the open circuit potential at a sweep rate of 1mVs⁻¹. The Tafel segments of the anodic and cathodic curves were extrapolated to corrosion potential (E_{corr}) to obtain the corrosion current densities (i_{corr}).

3. RESULTS & DISCUSSION

3.1 Weight loss measurements

The corrosion of aluminium in 1M HCl in the absence and presence of different concentrations of Datura metel (DM) in the room temperature was studied using weight loss technique.

The degree of surface coverage (θ) was calculated from the weight loss measurement and percentage of inhibitor efficiency using the following equations (Jones, 1996)

$$\text{Surface Coverage } (\theta) = \frac{W_{\text{uninh}} - W_{\text{inh}}}{W_{\text{uninh}}} \quad (1)$$

$$\% \text{ I.E.} = \frac{W_{\text{uninh}} - W_{\text{inh}}}{W_{\text{uninh}}} \times 100 \quad (2)$$

Where W_{inh} and W_{uninh} are weight loss of aluminium in the presence and absence of inhibitor.

Table 1. Corrosion rate, inhibitor efficiency and surface coverage of inhibitor at various concentration of 1M HCl at 303K.

	Weight loss (g/cm ²)	CR (mm/y)	% IE	Surface coverage (θ)
Blank	0.0101	437	-	-
0.3	0.0035	151	65	0.65
0.6	0.0022	95	78	0.78
0.9	0.0016	69	84	0.84
1.2	0.0014	61	86	0.86
1.5	0.0010	43	90	0.90

The calculated values of corrosion rate (mppy), inhibition efficiency (%IE) and surface coverage for aluminium corrosion in 1M HCl in the presence of different concentration of the leaves extract from the weight loss measurements are presented in the Table 1 indicates that the weight loss decreased with increase in inhibitor concentrations. The corrosion rate decreased with increase in concentration. This trend may results from the fact that inhibition efficiency and surface coverage increased with the increase in concentrations. Thus the surface is efficiently separated from the medium (Al-Andis et al. 1995). The above factors indicated that the inhibitor solution inhibits the corrosion of Al in 1M HCl solution

3.2 Hydrogen evolution technique

The plot indicates that slow rate of hydrogen evolution at the inhibition of the reaction and after sometime, the volume of gas evolved H_2 gas varies linearly with reaction time.

Table 2 shows that the on introduction of the leaves extract the volume of hydrogen evolved was decreased and further decreased as the concentration of the extract increases.

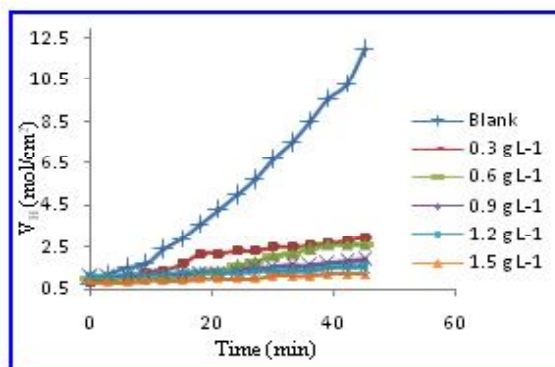


Fig. 2: Variation of volume of H_2 evolved against time for Al corrosion in 1M HCl solution in the absence and presence of different concentration of leaves extract at 303K.

3.3 Potentiodynamic polarisation method

Potentiodynamic polarisation curve for aluminium in 1M HCl solution with various concentration of Datura metel (DM) leaves extract at 303K are shown in Table 3, Fig. 3 shows that, the increase of concentrations of the inhibitor leads to displacement of the anodic and cathodic curves to low current densities and decreased the corrosion rate. At higher concentrations of the inhibitor was associated

Table 3. Potentiodynamic polarisation parameters for aluminium in 1M HCl containing various concentrations of the leaves extract.

C ($g L^{-1}$)	$-E_{corr}$ (mV)	i_{corr} ($m A/cm$)	Tafel slope (mV/dec)		CR (mm/y)	I.E %
			β_a	β_c		
Blank	744	14.7	199	247	569	-
0.3	741	5.2	176	201	201	65
0.6	752	3.2	170	261	81	78
0.9	754	2.7	167	226	70	82
1.2	759	2.4	157	208	61	84
1.5	762	1.3	146	200	32	91

with the formation of an adsorbed thin layer on Al surface or thin layer form non- porous oxide film leads to minimise the passing current on Al surface. Fig 3 and Table 3 also indicate that, E_{corr} shiftsto lesscathodic direction (i.e. the negative values). DM leaves extract act as mixed type of inhibitor and gets adsorbed at both the active sites. The addition of DM leaves extract decreased markedly the corrosion current density, i_{corr} with respect to inhibitor concentrations.

Table 2. Variation of volume of hydrogen evolution with concentration (g L⁻¹) of DM leaves extract for aluminium in 1M HCl solution.

C (g L ⁻¹)	Volume of hydrogen evolution (m l/m in)	Inhibitor Efficiency (%)
Blank	12	-
0.3	3	75
0.6	2.6	78
0.9	1.9	84
1.2	1.5	88
1.5	1.2	90

3.4 Electrochemical impedance spectroscopy

Fig. 4 shows that the effect of DM leaves extract on Al sample in 1M HCl solution after the immersion of 30 min. The Nyquist plots was regarded as one part of a semicircle. These data revealed that the impedance diagrams in most of the cases is not a perfect semi- circle and this has been attributed to frequency dispersion and the half circle indicates that the corrosion of Al is mainly controlled by the charge transfer process and the presence of DM leaves extract in solution does not affect the mechanism of dissolution of Al.

Electrochemical parameters such as R_{ct} and C_{dl} established from the Nyquist diagrams, without and with the addition of different concentrations of DM leaves extract in 1M HCl solution are listed in Table 4. The decreased value of C_{dl} can result from the decrease of the local dielectric constant and/ or the

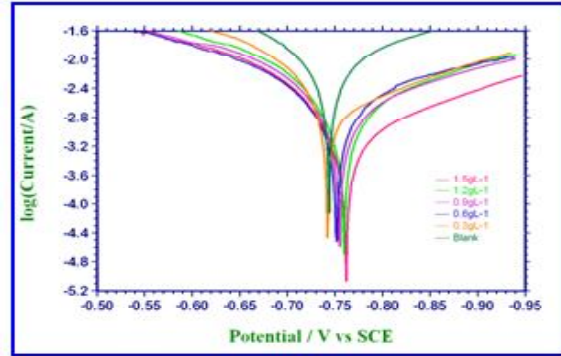


Fig. 3: Polarisation curves for Al sample corrosion in different concentrations of 1M HCl solution at 303K for DM leaves extract.

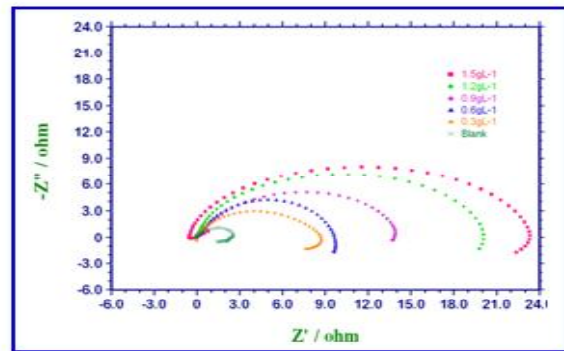


Fig. 4: Nyquist plot for Al sample corrosion in different concentrations of 1M HCl solution at 303K for Datura metal leaves extract.

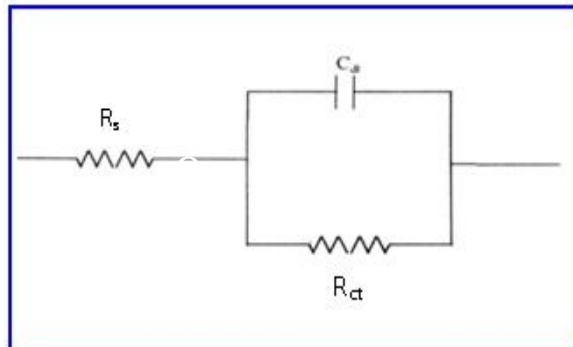


Fig. 5: The equivalent circuit model used to fit the experimental results.

Table 4. Electrochemical impedance parameters and the corresponding inhibition efficiencies for aluminium in 1M HCl solution in the absence and in presence of different concentrations of DM leaves extract at 303K.

C(g/cm ²)	R _{ct} ? cm ²	C _{dl} (μ F/cm ²)	Inhibition efficiency (%)
Blank	2.0	12.74	-
0.3	6	15.89	65
0.6	8.6	5.144	78
0.9	11.6	3.986	83
1.2	16.3	3.400	88
1.5	20.6	3.217	90

increase of thickness of the electrical double layers, suggested that the inhibitor molecules function by adsorption at the metal/solution interface. The high (R_{ct}) values, are generally associated with slow corroding system (Bosch, 2001). It is seen from the Table 4 that the increased in the concentration of the DM leaves extract increases the value of R_{ct} and decreased the corrosion of Al in 1M HCl solution and considerably corrosion was reduced due to the adsorption of the DM leaves extract component on Al surface.

3.5 Adsorption isotherm

Attempts were made to fit the plot of θ values to various isotherm including Freundlich, Temkin, El-Awady and Langmuir isotherm. According to this Langmuir isotherm, the surface coverage (θ) is related to inhibitor concentration.

$$\frac{C_{inh}}{\theta} = \frac{1}{K_{ads}} + C_{inh} \quad (3)$$

The ratio of concentration to surface coverage (C/θ) against concentration displayed a straightline for the inhibitor in Fig. 6. It showed that the linear plots with high correlation coefficient (0.99) and slope of about unity clearly revealed that the surface adsorption process of DM leaves extract on the Al surface obeys

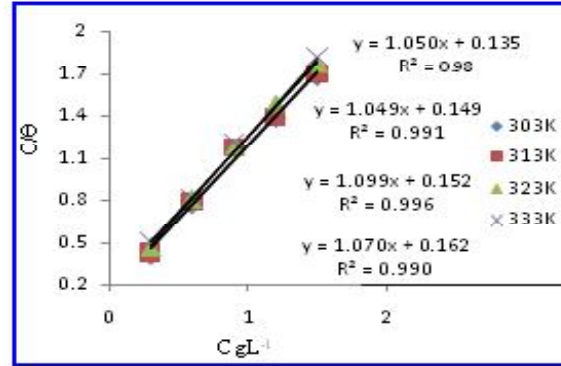


Fig. 6: Langmuir adsorption isotherm plot for aluminium corrosion in 1M HCl for DM leaves extract at different temperature.

the Langmuir adsorption isotherm. Therefore, one can infer that physisorption occurred.

The value of K_{ads} can be calculated from the intercepts of the straight lines from Fig. 6. Table 5 shows that the positive values of adsorption equilibrium constant imply a better adsorption. The negative values of indicates that the adsorption of the inhibitors on the Al surface is spontaneous (Obot, 2009).

3.3. Thermodynamic / Corrosion kinetic parameters

Activation energy can be obtained by modified Arrhenius equation as follows

$$\log CR = \left(\frac{-E_a}{2.303 R} \right) \left(\frac{1}{T} \right) + \log A \quad (4)$$

Weight loss measurement was used to obtain the corrosion rate values of Al in the absence and presence of 1 M HCl of leaves extract at different temperature of 303-313K. These values were plotted as shown in Fig.7. The values of activation energy of corrosion were determined from the slope of log CR versus 1/T plot. Table 6 and Fig.7 showed that the addition of inhibitor modify the value of Ea is attributed to the adsorption of inhibitor on Al surface and this adsorption makes energy barriers and this energy barrier

Table 5. Parameters of linear regression from Langmuir isotherm plot for aluminium corrosion in 1M HCl containing extracts of DM leaves extract.

Temp (K)	K_{ads}	G_{ads} kJ/mol
303	7.41	15.16
313	6.71	15.41
323	6.58	15.85
333	6.17	16.16

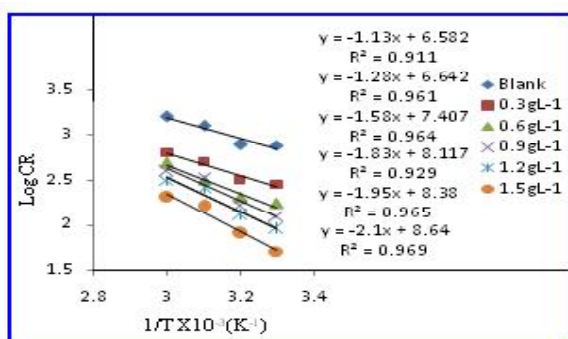


Fig. 7: Arrhenius plot for Al corrosion in 1M HCl for blank and different concentration of DM leaves extract.

Table 6. Calculated values of thermodynamic parameters for aluminium corrosion in 1M HCl solution in the absence and presence of different concentrations of DM leaves extract

C ($g L^{-1}$)	E_a kJ/mol	ΔH^* kJ/mol	$-\Delta S^*$ J/mol/K
Blank	21.64	26.69	106.52
0.3	24.51	29.43	104.98
0.6	30.25	30.83	102.07
0.9	35.04	32.95	97.31
1.2	37.34	38.56	78.89
1.5	40.21	42.09	73.50

of corrosion reaction increases as the concentration of the inhibitor was increased and hence % I.E. increased. It suggested that the process was activation controlled. Since the activation energy value of DM leaves extract for 1M HCl solution for inhibitor systems support the fact that the inhibitor was physically adsorbed on Al surface. A transition state complex is decay in to products after forming the high energy (Khadom, 2009). The mathematical form of transition state theory is shown as below

$$CR = \frac{RT}{Nh} e \left(\frac{\Delta H^*}{RT} \right) e \left(\frac{\Delta S^*}{R} \right) \quad (5)$$

Where ΔH^* and ΔS^* are the enthalpy and the entropy of activation, N is Avogadro number (6.022×10^{23} molecule mol^{-1}) and h is the Planck's constant (6.626×10^{-34} J s mol^{-1}) and CR is the corrosion rate. The positive values of ΔH^* reflect the endothermic nature of the aluminium dissolution process in the 1M HCl. The negative sign of ΔS^* showed an increase in the system order.

4. CONCLUSION

The leaves extract of DM act as good and efficient inhibitors for corrosion of aluminium in 1M HCl solution. In the weight loss method, the inhibitor efficiencies increased with increase in extracts concentration. The value of % inhibitor efficiency obtained from hydrogen evolution measurements are in good agreement with those obtained from weight loss technique. Potentiodynamic polarisation studies revealed that the extracts act through mixed mode of inhibition. The impedance method revealed that activation process mainly controls the corrosion of Al. From the effect of temperature, the activation parameters for the corrosion process (E_a , ΔH^* and ΔS^*) were calculated and indicates that corrosion is much reduced in the presence of inhibitor. The adsorption of the leaves extract on the aluminium surface was found to obey Langmuir adsorption isotherm.

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