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Corrosion Inhibition of Aluminium in Hydrochloric Acid Using *Bacopa monnieri* Leaves Extract as Green Inhibitor

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Abstract : The inhibitive action of *Bacopa monnieri*(Brahmi)leaves extract on corrosion of aluminium in hydrochloric acid solution was investigated through weight loss, potentiodynamic polarization and electrochemical impedance spectroscopic (EIS) methods.The effect of inhibitor concentrations on different acid concentrations was investigated. The present study revealed that the percentage of inhibition efficiency (I.E.) is enhanced with increase of inhibitor concentration and decrease with rise in temperature. The inhibitive action of the extract is discussed in view of adsorption of *Bacopa monnieri* molecule on the metal surface. It was found that the adsorption follows Langmuir adsorption isotherm. Tafel plots of polarization study indicate that the *Bacopa monnieri* leaves extract acts as a mixed type inhibitor. Maximum I.E. of *Bacopa monnieri* leaves extract was found 91.85% at 1.2 g/L inhibitor concentration in 0.75M HCl solution.
Key words : Corrosion, Aluminium, *Bacopa monnieri*, HCl, inhibition, Polarization, EIS.

1. Introduction :

The study of aluminium corrosion in different acidic and alkaline environments has attracted considerable attention in view of important application of the metal. Aluminium is highly electropositive and resistance to corrosion because of hard, tough film of oxide it forms on the surface[1]. Hydrochloric acid is commonly used for pickling and etching purposes due to its low cost as compared to other acids. The corrosion rate of metals can be reduced with the application of inhibitors in acid solutions[2,3]. Plant extracts have attracted attention in the field of corrosion inhibition as they are a source of non-toxic, eco-friendly, bio-degradable and of potentially low cost inhibitors for preventing metal corrosion[4].Amongst others, extracts of some plants such as *Azadirachta indica*[5,6], *Lawsonia inermis*[7,8] , *Oscimum sanctum*[9] and *Bacopa monnieri*[10,11] have been reported to inhibit the of corrosion rate of metals. *Bacopa monniera* which commonly known as brahmi, belongs to the Scrophulariaceae family. *Bacopa monnieri* is perennial, creeping herb native to wetlands regions of Southern India, Australia, Africa, Asia, and North and South America. It is used for skin disorders, as an antipileptic , antipyretic and analgesic[12,13].The present study was undertaken to investigate the inhibitive action of *Bacopa monnieri* leaves extract on the aluminum metal in HCl solution by using weight loss, polarization measurements and electrochemical impedance spectroscopy techniques.

2. Materials and Methods :

2.1. Preparation of samples and solutions

The aluminium specimens with a chemical composition of 99.54 % Al, 0.090 % Si, 0.320 % Fe, 0.0012 % Cu, 0.0034 % Mn, 0.0014 % Mg, 0.0042 % Cr, 0.0046 % Ni, 0.0020 % Zn, 0.0079 % Ti, 0.0005 %

Pb, and 0.0026 % Sn are used in the present study. The metal sheet, test specimens of size 5.0 x 2.50 x 0.198cm having an effective area of 0.279 $dm^2$are used. Hydrochloric acid was used as corrosive solution having concentration of 0.75, 1.0 and 1.25 M prepared by diluting analytical grade of HCl purchased from Merck using double distilled water.

2.2. Preparation of Extract

Bacopa monnieri leaves was collected, dried and blended to powder form. 10 gm powder was refluxed in 400 ml double distilled water for 5h. The refluxed solution was allowed to standfor 8h, filtered and stored[1].Inhibitor concentration ranging from 0.6 g/L to 1.2 g/L are prepared from stock solution.

2.3. Weight loss measurement

For weight loss experiment, an aluminium coupons are each suspended completely in 0.75, 1.0 and 1.25 M HCl solutions without and with different concentrations of *Bacopa monnieri* with the help of glass hooks at 301 ± 1 K for 24 h (1 day). The volume of solution kept 230 mL. the coupons are retrieved after 24 h, washed by distilled water, dried well and reweighed. From the weight loss data, corrosion rate in mg/dm^2dwas calculated.

2.4. Temperature effect

To study the effect of temperature on corrosion rate, the aluminium coupons are completely immersed in 230 mL of 0.75 M HCl solution without and with different concentrations of *Bacopa monnieri* leaves extracts at 313, 323 and 333 K for 2 h.

2.5. Electrochemical measurements

Electrochemical measurements are carried out by using an electrochemical work station (CHl608C-series, U. S. Model with CH instruments). In electrochemical experiments Ag/AgCl was used as a reference electrode, platinum as an auxiliary electrode and aluminium metal was used as a working electrode. For polarization study, aluminium specimens having an area of 1 cm^2exposed to 230 ml of 0.75 M HCl in absence and presence of *Bacopa monnieri* leaves extract and allowed to establish a steady state open circuit potential (OCP) for about 30 minutes. Test coupons are then polarized by the application of potential drift of -250 mV cathodically and +250 mV anodically with respect to the OCP at a scan rate of 5.0 mV/s.The potentiodynamic polarization plots (Tafel curves) are developed simultaneously. Anodic and cathodic polarization curves give anodic and cathodic Tafel lines correspondingly. The intersect point of Tafel lines gives the corrosion potential (E_{corr}) and corrosion current (i_{corr})[14]. The electrochemical impedance studies are carried out in the same setup using potentiodynamic polarization studies described above. Impedance studies were carried out at steady state open circuit potential (OCP). A small amplitude (5.0 mV) sinusoidal AC voltage, in wide frequency range 1 Hz to $10^5 H_z$ was applied over the system.

3. Results and Discussion:

3.1. Weight loss experiments

The corrosion rate of aluminium in 0.75, 1.0 and 1.25 M of HCl solution without and with 0.6, 0.8, 1.0 and 1.2 g/L concentration of *Bacopa monnieri* leaves extract at 301 ± 1 K for an exposure period of 24 h was calculated from the weight loss data using following equation:

$$\text{CR (mg/dm}^2\text{d)} = \frac{\text{Weight loss (gm)} \times 1000}{\text{(metal surface area)dm}^2 \times \text{day}} \ldots (1)$$

The Inhibition efficiency (I.E.) was calculated by using following formula:

$$\text{IE (\%)} = \frac{W_{uninh} - W_{inh}}{W_{uninh}} \text{ x } 100 \ldots (2)$$

Where, W_{uninh} = weight loss without inhibitor, W_{inh} = weight loss with inhibitor.

The degree of surface coverage (Θ) for different concentration of the inhibitor in acidic media have been evaluated from weight loss experiments using this equation:

$$\theta = \frac{W_{uninh} - W_{inh}}{W_{uninh}} \ldots (3)$$

Table 1:Corrosion rate for aluminium in various HCl concentrations in the absence and presence of different concentrations of *Bacopa monnieri* leaves extract at 301 ± 1 K.

Inhibitior concentration (g/L)	Acid concentration					
	0.75 M		1.0 M		1.25 M	
	CR (mg/dm²d)	I.E. (%)	CR (mg/dm²d)	I.E. (%)	CR (mg/dm²d)	I.E. (%)
Blank	2195.20	-	6163.74	-	8283.87	-
0.6	743.65	66.12	2577.76	58.17	3968.53	52.09
0.8	654.27	70.19	2216.66	64.03	3643.18	56.02
1.0	429.03	80.45	1358.59	77.95	2824.45	65.90
1.2	178.76	91.85	657.84	89.32	1151.23	86.10

3.2. Effect of acid concentration

Results showed in Table 1 indicates that as the concentration of acid increase corrosion rate increases while I.E. decreases.

3.3. Effect of inhibitor concentration

At constant acid concentration, I.E. increases with increase in *Bacopa monnieri* leaves extract concentrations (Figure 2). The maximum I.E. (91.85%) has been observed at 1.2 g/L inhibitor concentrations in 0.75 M HCl at 24 h at 301 K . At constant inhibitor concentration, the I.E. was decreases as the acid concentration increases (Table 1). The results indicates that with increase in inhibitor concentration from 0.6 to 1.2 g/L the corrosion rate was decreases from 743.65 to 178.76 mg/dm^{2}d in 0.75 M HCl (Table 1 and Figure 1).

Table 2:Corrosion rate (log ρ) of aluminium in 0.75 N HCl in absence and presence of *Bacopa monnieri* leaves extract for an immersion period of 24 hr at 301 ± 1 K.

Inhibitor	Inhibitor Concentration(g/L)	CR (ρ) (mg/dm²d)	log ρ	I.E. (%)	Surface coverage (θ)	C/θ
Blank	-	2195.20	3.34	-	-	-
Bacopa monnieri (Brahmi)	0.6	743.65	2.87	66.12	0.66	0.91
	0.8	654.27	2.81	70.19	0.70	1.14
	1.0	429.03	2.63	80.45	0.80	1.25
	1.2	178.76	2.25	91.85	0.91	1.32

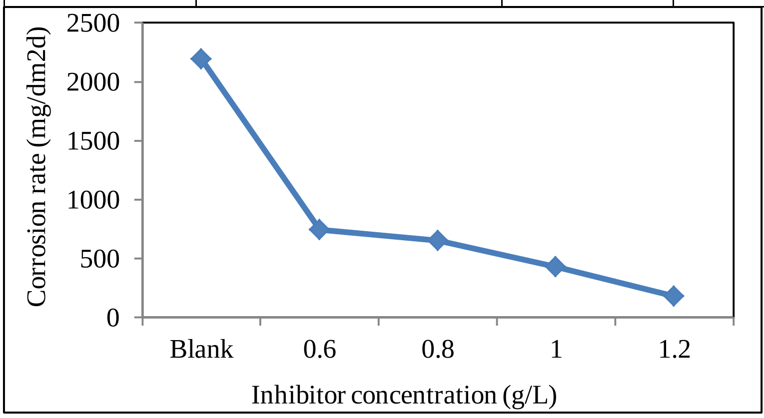


Figure 1:Corrosion rate of aluminium corrosion in 0.75 M HCl solution in absence and presence of different concentration of *Bacopa monnieri* leaves extract for an immersion period of 24 h.

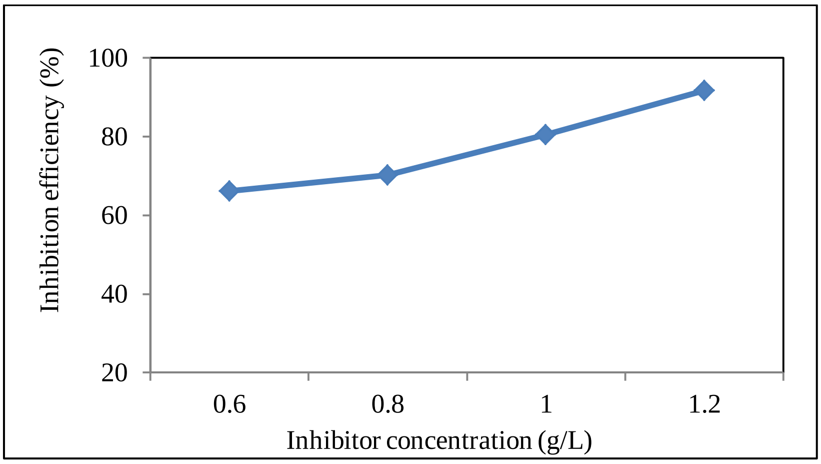


Figure 2:I.E. of aluminium corrosion in 0.75 M HCl solution in presence of different concentration of *Bacopa monnieri* leaves extract for an immersion period of 24 h.

3.4. Effect of temperature

The importance of temperature variation in corrosion study involving the use of inhibitors is to determine the mode of inhibitor adsorption on the metal surface. The results presented in Table 3 shows that as temperature increases corrosion rate increase while I.E. was decrease (Figure 4). The maximum I.E. (91.99 %) was found at 1.2 g/L *Bacopa monnieri* extract concentration at 24 h at 313 K. The value of energy of activation (E_a) has been calculated with the help of following Arrhenius equation[15].

$$\log\frac{\rho_2}{\rho_1} = \frac{E_a}{2.303R}\left(\frac{1}{T_1}-\frac{1}{T_2}\right) \ldots \quad (4)$$

where, ρ_1 and ρ_2 are the corrosion rate at temperature T_1 and T_2 respectively.

Table 3:Temperature effect on corrosion rate (CR), inhibition efficiency(I.E.) and activation energy (E_a) for aluminium in 0.75 M HCl in absence and presence of *Bacopa monnieri* leaves extract for an immersion period of 2 h.

Inhibitor concentration. (g/L)	Temperature						Energy of activation (E_a) (kJ/mol)			**(E_a) from Arrhenius plot (kJ/mol)**
	313 K		**323 K**		**333 K**		**313-323 K**	**323-333 K**	**Mean**	
	CR (mg/dm^{2}d)	**I.E. (%)**	**CR (mg/dm^{2}d)**	**I.E. (%)**	**CR (mg/dm^{2}d)**	**I.E. (%)**				
Blank	21451.44	-	26642.76	-	43417.92	-	18.21	43.64	30.92	30.40
0.6	4719.24	78.00	9438.60	64.57	18534.12	57.31	58.26	60.34	59.30	59.27
0.8	3732.48	82.60	8494.80	68.11	15745.44	63.73	69.12	55.17	62.14	62.45
1.0	2831.52	86.80	7121.88	73.26	11026.08	74.60	77.52	45.45	61.48	62.24
1.2	1716.12	91.99	5577.36	79.06	8580.60	80.23	99.04	50.62	74.83	75.91

Results given in Table-3, indicates that the mean values of E_a are found higher in inhibited acid ranging from 59.30 to 74.83 kJ/mol than the E_a values for uninhibited acid 30.92 kJ/mol. The higher values of E_a indicate physical adsorption of the inhibitor on the metal surface and the adsorption of inhibitor causes an increase in the E_a of the process[16].Results of Table-3 indicates that as temperature increases, rate of corrosion increase while percentage of I.E. decreases. The value of Ea were also calculated from the slope of the Arrhenius plot of log p versus 1/T x 1000 (Fig.3).The mean value of E_a was 30.92 kJ/mol in uninhibited acid and the value calculated from the slope of the Arrhenius plot was found 30.40 kJ/mol, which was found almost similar (± 1.0 kJ/mol).

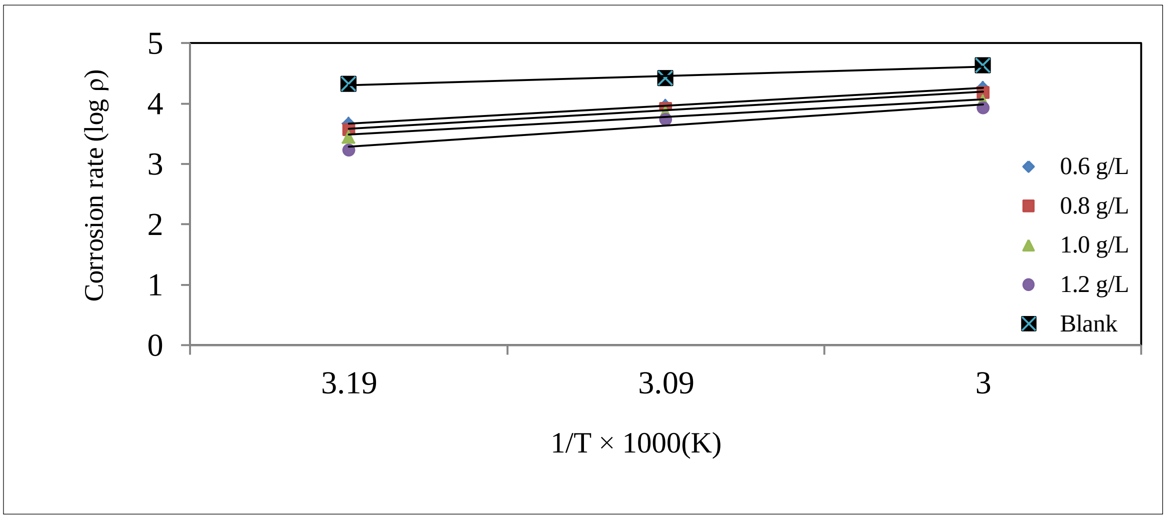


Figure 3:Arrhenius plot for corrosion of Al in 0.75 M HCl in absence and presence of different concentration of *Bacopa monnieri* leaves extract for an immersion period of 2 h.

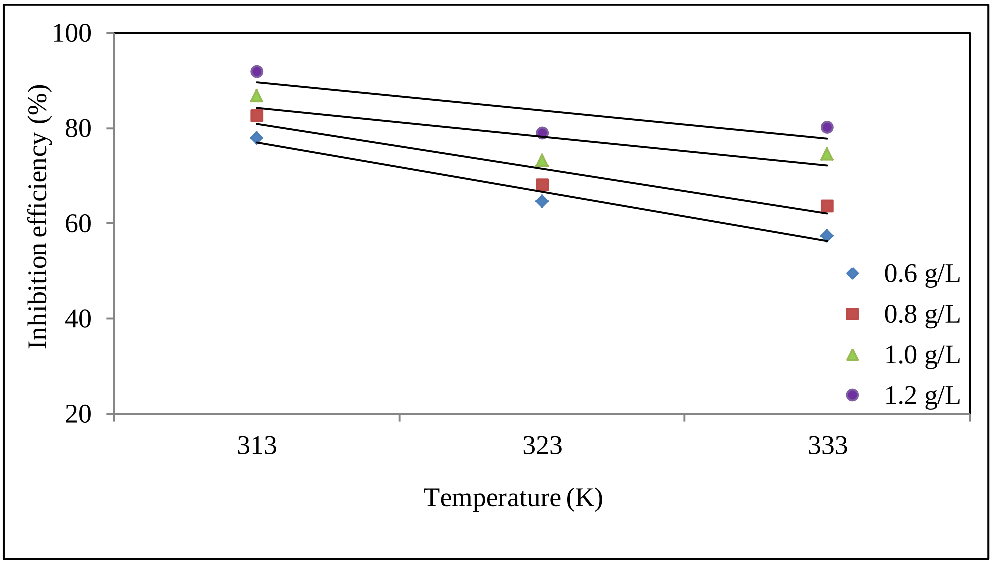


Figure 4: Effect of temperature on I.E. for aluminium corrosion in 0.75 M HCl at different concentration of *Bacopa monnieri* leaves extract for immersion period of 2 h.

The value of heat of adsorption (Q_{ads}) were calculated by following equation[17]:

$$Q_{ads} = 2.303R\left[\log\left(\frac{\theta_2}{1-\theta_2}\right) - \log\left(\frac{\theta_1}{1-\theta_1}\right)\right] \times \left[\frac{T_1T_2}{T_2-T_1}\right] \qquad \ldots (5)$$

Where, Θ_1 and Θ_2 are the fraction of the metal surface covered by the inhibitor at temperature T_1 and T_2 respectively. Result calculated from eq.(5), it is evident that in all cases, the Q_{ads} values are negative and ranging from -02.45 to -93.53 kJ/mol. The negative values of Q_{ads} shows that the adsorption process and hence the I.E. decreases with rise in temperature supporting the physisorption mechanism[18].

3.5. Adsorption Isotherm

Basic information on the interaction between inhibitors and a metal surface can be provided using the adsorption isotherm[19].The surface coverage ‘Θ’suggest that a chemical bond is formed between the metal atoms and the inhibitor molecules. Langmuir isotherm was given by a plot of C/Θ Vs Care shown in Figure 5 to be linear with slope value equal to unity indicates that the inhibitor obeys Langmuir adsorption isotherm. It was found that the best suitable adsorption isotherm for the studied inhibitor on the aluminium surface is the Langmuir equation[20] which was defined as follows:

$$\frac{C}{\theta} = \frac{1}{K_{ads}} + C \quad \ldots (6)$$

Where, K_{ads} is equilibrium constant of the adsorption process and C is inhibitor concentration.

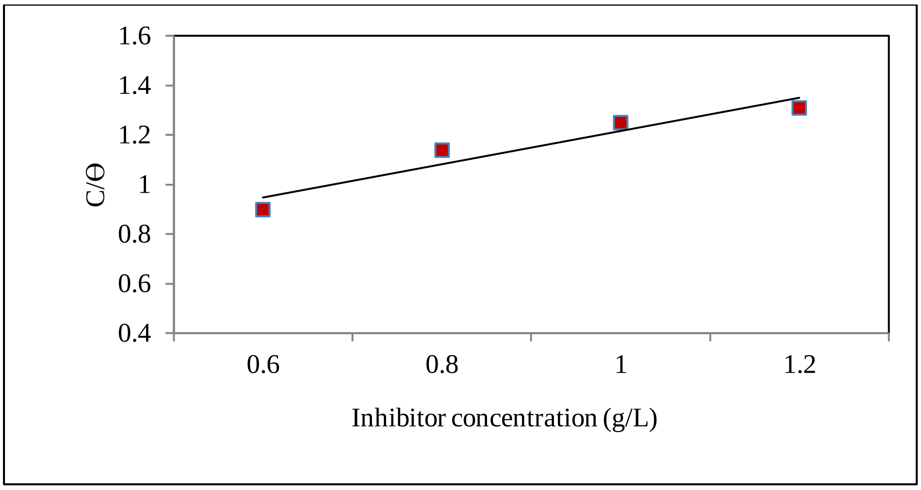


Figure 5: Langmuir adsorption isotherm for corrosion of aluminium in 0.75 M HCl solution containing different concentration of *Bacopa monnieri* leaves extract for an immersion period of 24 h.

From the intercepts of the straight lines on the C/Θ axis, K_{ads} can be calculated . This value of K_{ads} is also related to free energy of adsorption ΔG_a^o, as given by following equation[21,22]

$$\Delta G^o_{ads} = -RT \ln (55.5\ K_{ads}) \quad \ldots (7)$$

Where, R is the gas constant, T is the absolute temperature (K) and the value 55.5 in the above equation is the concentration of water in solution in Molar[23], K_{ads} is the equilibrium constant of the adsorption/desorption process. The sign ofΔG_a^o was negative which reflects that the adsorption of these inhibitors is spontaneous process. The mean ΔG_a^o value is close to -13 kJ/mol indicates that the adsorption mechanism of *Bacopa monnieri* on aluminium in 0.75 M HCl at the studied temperatures is physisorption with adsorptive layer having electrostatic character[24]. This is concluded on the fact that the values of ΔG^o_{ads} -20 kJ/mol are consistant with physisorption, while those around –40 kJ/mol or higher are associated with chemisorption[25]. The enthalpy of adsorption (ΔH_a^o) and entropy of adsorption (ΔS_a^o) are calculated using the equations (8) and (9).

$$\Delta H^o_a = E_a - RT \quad \ldots (8)$$

$$\Delta S^o_a = \Delta H^o_a - \Delta G^o_a / T \ldots (9)$$

The results revealed that ΔH_a° values are positive and ranging from 45.42 to 99.01 kJ/mol indicating the endothermic nature of the reaction suggests that higher temperature favors the corrosion process. Positive value of ΔS_a° , lie between 0.18 to 0.36 kJ/mol K indicate the affinity of the adsorbent for the inhibitor and the corrosion process is entropically favorable.

3.6. Kinetic parameters : Rate constant (k) and Half-life ($t_{1/2}$) :

As concentration of inhibitor increases rate constant 'k' decreases whereas the half-life values are increases with increasing concentrations of inhibitor. The result are in good agreement withthe result obtain by and Muthukumar and Chandrasekara [26]. Corrosion rate constant 'k' increases with increase in acid concentration (Table 4).

Table 4:Kinetic data for the corrosion of aluminium in various concentration of HCl containing *Bacopa monnieri* leaves extract.

Inhibitor	Inhibitor concentration (g/L)	Acid concentration					
		0.75 M		1.0 M		1.25 M	
		Rate const. ($k\times10^{-3}$)	Half life ($t_{1/2}$)	Rate const. ($k\times10^{-3}$)	Half life ($t_{1/2}$)	Rate const. ($k\times10^{-3}$)	Half life ($t_{1/2}$)
Blank	**0.0**	99.48	07.00	307.53	22.53	439.41	157.71
Bacopa monnieri	**0.6**	32.59	21.65	117.57	58.97	186.53	03.71
	0.8	28.58	24.75	99.76	07.00	169.71	04.08
	1.0	18.64	38.50	59.59	11.74	128.65	05.38
	1.2	07.72	89.76	28.36	24.75	50.35	13.86

3.7. Potentiodynamic polarization measurements

From Table 5, it was observed that the addition of *Bacopa monnieri* leaves extract in HCl solution, the significant decrease in the corrosion current density (i_{corr}) and decrease in the corrosion rate with respect to the blank. In general,an inhibitor is anodic or a cathodic if the variation E_{corr} against the blank is higher or above 85 mV[27,28]. In this study, the displacement of E_{corr} was 19 mV which is less than a 85 mV (Table 5),which suggest that the *Bacopa monnieri* function as a mixed-type of inhibitor. Figure 6 shows potentiodynamic polarization curves.

Table 5: Potentiodynamic polarization parameters for aluminium in 0.75 M HCl and in absence and presence of 1.2 g/L *Bacopa monnieri* leaves extract.

System	E_{corr} (V)	I_{corr} ($\mu A/cm^2$)	Tafel slope V/ decade		IE (%) calculated from	
			Anodic β_a	Cathodic β_c	Polarization method	Weight loss method
Blank	-0.895	9156	6.706	5.621	-	-
Bacopa monnieri	-0.876	2296	8.234	6.371	74.92	91.85

Inhibition efficiency (I.E.) from (i_{corr}) was calculated using following equation [29]:

$$\text{I.E.}\ (\%) = \frac{i_{corr(uninh)} - i_{corr(inh)}}{i_{corr(uninh)}} \times 100 \qquad \ldots(10)$$

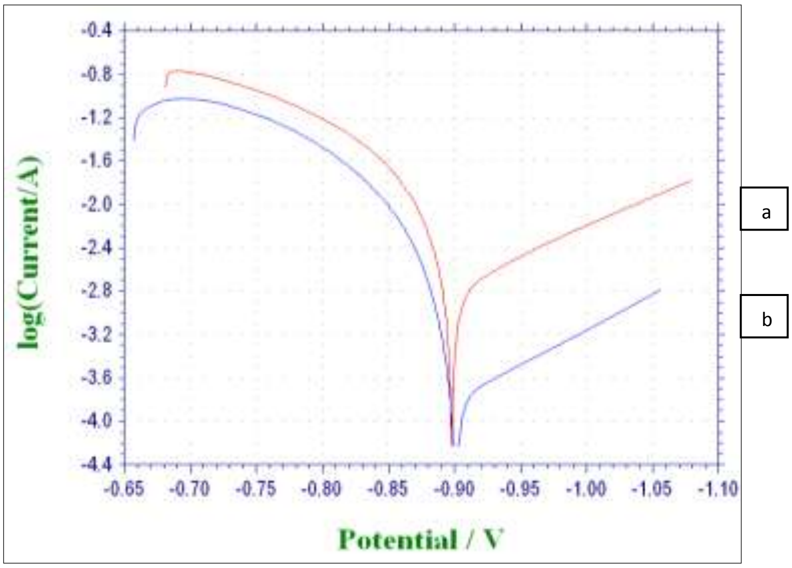


Figure 6:Potentiodynamic polarization curves for aluminium in (a) 0.75 M HCl and (b) 0.75 M HCl containing 1.2 g/L *Bacopa monnieri* leaves extract.

3.8. Electrochemical impendence spectroscopy (EIS) measurement

Nyquist plots for aluminium in 0.75 M HCl solution obtained in absence and presence of *Bacopa monnieri* extract was examined by EIS method at room temperature were shown in Figure 7 and EIS parameters were presented in Table 6. It was observed from Figure 7 that the impedance diagram is almost semicircular in appearance. The semicircle nature of the plots indicates that corrosion process was mainly charge transfer controlled[30].To obtain the double layer capacitance (C_{dl}), the frequency at which the imaginary component of the impedance is maximum was found as presented in the following equation[31]:

$$C_{dl} = \frac{1}{2\pi F_{max} R_{ct}} \quad (11)$$

Where, f_{max} is the frequency at maximum height of the semicircle on the imaginary axis and R_{ct} is the charge transfer resistance[32].

Table 6 :EIS parameters for the corrosion of aluminium in 0.75 M HCl in absence and presence of 1.2 g/L *Bacopa monnieri* leaves extract.

System	R_{ct} (Ω cm^2)	C_{dl} ($\mu F/cm^2$)	IE (%) calculated from	
			EIS method	Weight loss method
Blank	84	45.130	-	-
Bacopa monnieri	400	1.990	95.59	91.85

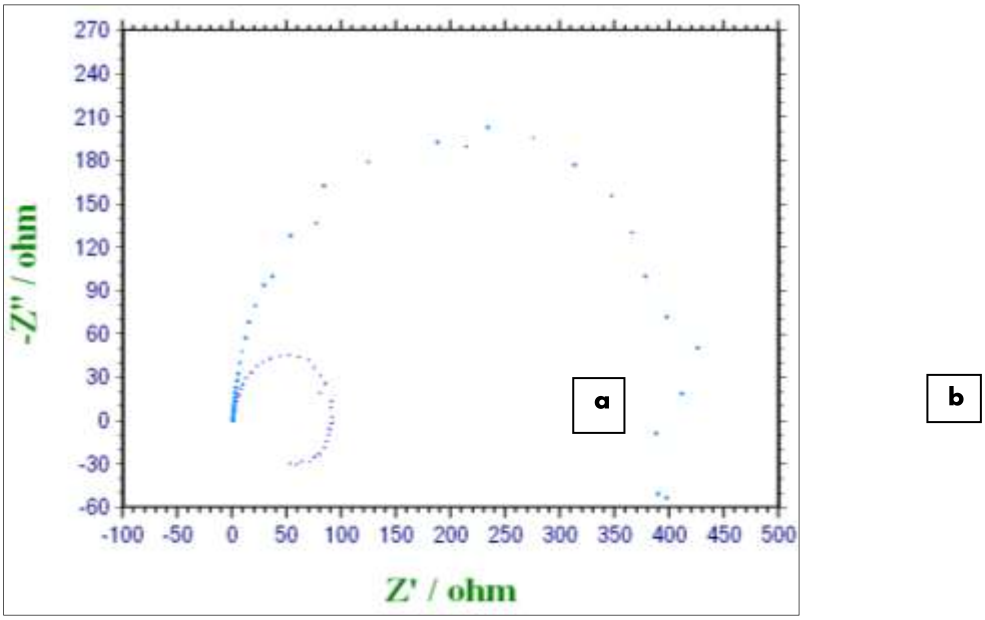


Figure 7:Nyquist plot for aluminium in (a) 0.75 M HCl alone and (b) 0.75 M HCl containing 1.2 g/L *Bacopa monnieri* leaves extract.

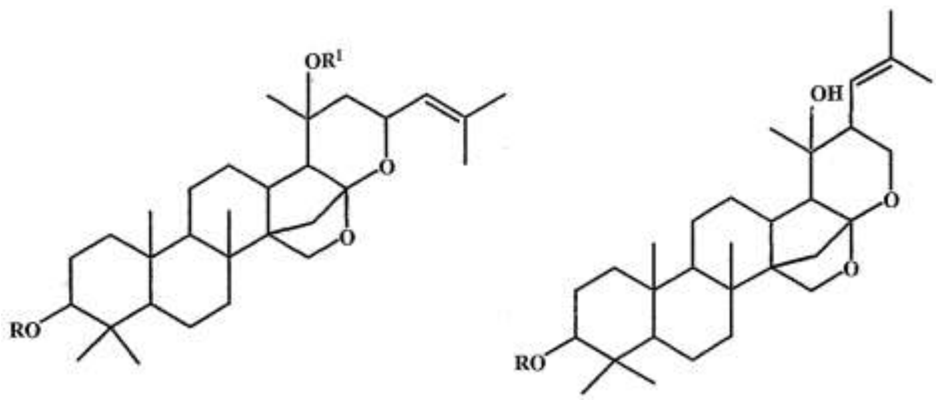
Inhibition efficiency (I.E.) from EIS method was calculated using following equation:

$$IE\ (\%) = \frac{Cdl(uninhi.) - Cdl(inhi.)}{Cdl\ (uninhi.)} x\ 100 (12)$$

The results suggest that the inhibitor acts by the formation of a physical protective layer on the surface that retards the charge transfer process and therefore inhibit the corrosion reaction, leading to increase in R_{ct} values. Moreover, the adsorbed inhibitor species decrease the electrical capacity of electrical double layer values at the electrode/solution interface and therefore decrease the value of C_{dl}[33].

4. Mechanism of inhibition by *Bacopa monnieri* leaves extract:

Bacopa monnieri contain Saponins, Monnierin, Hersaponin, Bacoside -A, Bacoside –B, Brahmine and other chemicals like Stigmastanol, β -Sitosterol and Stigmasterol[10,34-36].*Bacopa monnieri* contained alkaloid brahmine, nicotinine, and herpestine[37,38] .*Bacopa monnieri* also containedbetulinicacid,D-mannitol[39].Bacoside-A[3-(α-L-arabinopyransoyl)-O-β-D-glucopyranoside-10, 20-dihydroxy-16-keto-dammer-24-ene] and Triterpenoid Saponins were isolated from *Bacopa monnieri* . According to Singh and Quraishi[11] the active constituents of *Bacopa monnieri* leaves extract includes Bacoside-A and Bacoside-B whose structures are given below in Fig.8.



Bacoside-A Bacoside-B
Figure 8: Structure of Bacoside- A and Bacoside- B

The I.E. of *Bacopa monnieri* leaves extract can be explained on the basis of the adsorption of the inhibitor mainly via hetero atoms (viz., N) present in different constituents of extract in addition to the availability of π-electrons in the aromatic system[40].*Bacopamonnieri* found as a mixed type inhibitor. The results are in good agreement with thework of Singh and Quraishi[11].

5.Conclusion :

The present study shows that *Bacopa monnieri* was found to be a good eco-friendly inhibitor for the corrosion control of aluminium in HCl solution. Corrosion rate increases as acid concentration increases. The inhibition efficiency increases with increase in *Bacopa monnieri* eaves extract concentration. The maximum I.E. was found 91.85 % at 1.2g/L inhibitor concentration in 0.75 M HCl solution. *Bacopa monnieri* adsorbed on metal surface follows Langmuir adsorption isotherm. Tafel plot indicates *Bacopa monnieri* acts as a mixed type inhibitor. AC impedance spectra reveal that a protective film is formed on the metal surface. All three techniques give almost identical values of I.E. for aluminium in HCl solution.

6. Acknowledgement

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