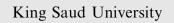
Arabian Journal of Chemistry (2016) 9, S1584-S1589



### **ORIGINAL ARTICLE**



## Arabian Journal of Chemistry

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# *Piper longum* extract as green corrosion inhibitor for aluminium in NaOH solution



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Received 11 July 2011; accepted 21 April 2012 Available online 2 May 2012

#### **KEYWORDS**

Aluminium; Corrosion inhibition; Plant extract; Alkaline solution; Weight loss method Abstract The inhibiting action of *Piper longum* seed extracts was investigated as the corrosion inhibitors of aluminium in 1 M NaOH solution using potentiodynamic polarization, electrochemical impedance spectroscopy (EIS) methods, and weight loss measurements. The results revealed that *P. longum* extract was an effective inhibitor, and the inhibition efficiencies obtained from polarization and weight loss experiments were in good agreement. Using the potentiodynamic polarization technique, the extract was proving to have a mixed-type character for aluminium in alkaline solution by suppressing both anodic and cathodic reactions on the metal surface. The adsorption isotherm model was determined according to fitted results obtained from the weight loss experiments performed on the aluminium specimen in 1 M NaOH solution at 308 K. Thermodynamic parameters such as activation energy and free energy of adsorption were calculated and given in kJ mg<sup>-1</sup>.
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#### 1. Introduction

Aluminium is the second most used metal after iron; it is used in a large number of applications by itself and is used in a wide range of alloys. Because of the low atomic mass and the negative value of the standard electrode potential, aluminium potentially attracts as an anode material for power sources with high energy densities (Emregul and Abbas Aksut, 2008).

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Peer review under responsibility of King Saud University.



The corrosion behaviour of pure aluminium and its alloys in aqueous alkaline solutions has been extensively studied in the development of the aluminium anode for the aluminium/ air battery (Bocksite et al., 1963; Macdonald et al., 1988; Chu and Savinel, 1991; Abdel-Gaber et al., 2008). Corrosion of aluminium causes many problems viz., (1) it passivates the cathode active material, (2) its solid products increase the electrical resistance, (3) its soluble products contaminate the electrolyte and increase the self-discharge rate, and (4) the dissolved Al<sup>3+</sup> ions migrate to the counter anode and reductively deposit (Zhang and Jow, 2002). Despite the fact that aluminium/air battery is an eco-friendly system and the energy density of this system is excellent. It is not greatly used in practice due to severe hydrogen evolution problems resulting from corrosion of the aluminium electrode. Thus, commercial application of Al and its alloys requires control of the hydrogen gas evolution which must necessarily be achieved without compromising the eco-friendly nature of the system.

http://dx.doi.org/10.1016/j.arabjc.2012.04.029

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In efforts to mitigate aluminium corrosion, the main tactic is to separate the metal from corrosive environments. This can be achieved using corrosion inhibitors. Recently, the use of chemical inhibitors has been limited due to environmental regulations; plant extracts have again become important because they are the environmental and renewable source for a wide range of needed inhibitors. Plant extracts are viewed as an incredibly rich source of naturally synthesized chemical compounds that can be extracted by simple procedures with low cost. A lot of natural products were previously used as corrosion inhibitors for different metals in various environments (Zhang and Jow, 2002; El Hosary et al., 1972; El Hosary and Saleh, 1993; Avwiri and Igho, 2003; Gunasekaran and Chauhan, 2004; El-Etre, 1998; El-Etre et al., 2005; Abdel-Gaber et al., 2006; Oguzie, 2007; Singh et al., 2011) and their optimum concentrations were reported. The obtained data showed that plant extracts could serve as effective corrosion inhibitors. It is well established that corrosion inhibition occurs via adsorption of their molecules on the corroding metal surface and efficiency of inhibition depends on the mechanical, structural and chemical characteristics of the adsorption layers formed under particular conditions.

As a contribution to the current interest on eco-friendly, green, corrosion inhibitors, and the present study investigates the inhibiting effect of seed extracts of *Piper longum* on the aluminium corrosion in 1 M NaOH solution using the weight loss and polarization techniques. The plant, which is popularly known in India as "Pippali", belongs to the family Piperaceae and various Piper species are widely distributed in the tropical and subtropical regions of the world, throughout the Indian subcontinent, Sri Lanka, Middle Eastern countries and the Americas (Yende et al., 2010). The effect of temperature as well as kinetic and activation parameters that govern metal corrosion have been evaluated.

#### 2. Experimental section

Aluminium (Al-1060) strips were used for weight loss as well as electrochemical studies. The aluminium specimens were machined into test electrodes of dimension 8 cm × 1 cm and embedded in PVC holder by epoxy resin (araldite) leaving one surface of an area of 1 cm<sup>2</sup> for electrochemical measurements. The exposed surface was abraded with silicon carbide abrasive paper from 400 to 1200, degreased with acetone, rinsed in distilled water, and dried in the air. The corrosive medium was 1 M NaOH solution prepared from analytical-reagent-grade NaOH (MERCK) and bidistilled water. Stock solution of *P. longum* was extracted by reflux of 100 g of the dry materials in 500 mL bidistilled water for 5 h. The refluxed solution was filtered to remove any contamination. The concentration of the stock solution was calculated in terms of mg L<sup>-1</sup>.

Electrochemical experiments were carried out in a conventional three-electrode glass cell of capacity 100 mL, using a GAMRY POTENTIOSTAT/GALVANOSTAT PCI 4 electrochemical workstation. A saturated calomel electrode (SCE) equipped with a Luggin capillary and a platinum foil of 1 cm  $\times$  1 cm were used as reference and counter electrode, respectively. All the potentials reported are with reference to SCE. Before measurement, the working electrode was immersed in test solution for approximately 30 min until a steady open-circuit potential (OCP) was reached. EIS measurement was carried out in the 100 kHz–10 mHz frequency range at OCP. The sinusoidal potential perturbation was 10 mV in amplitude and the cell temperature was maintained at  $308 \pm 1$  K using a thermostatic water-bath. The electrochemical experimental data were collected and analysed by electrochemical software Echem Analyst ver. 5.5. The polarization curves were carried out from cathodic potential of -0.25 V to anodic potential of +0.25 V with respect to the open circuit potential at a sweep rate of  $1 \text{ mV s}^{-1}$ . The linear Tafel segments of the anodic and cathodic curves were extrapolated to corrosion potential ( $E_{\text{corr}}$ ) to obtain the corrosion current densities ( $I_{\text{corr}}$ ). In each measurement, a fresh working electrode was used. Several runs were performed for each measurement to obtain reproducible data.

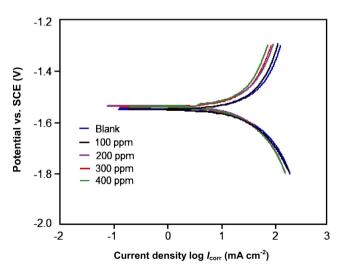
The aluminium specimens of  $2.5 \text{ cm} \times 2 \text{ cm} \times 0.05 \text{ cm}$  sizes were used for weight loss measurements and were abraded with a series of silicon carbide abrasive paper (grade 600– 800–1000–1200) and degreased with acetone, rinsed in distilled water, and dried in the air, and weighed accurately. Aluminium specimens were immersed in 100 mL of 1 M NaOH with and without addition of different concentrations of *P. longum* seed extract. The temperature of the corrosive system was controlled by an air thermostat. After 1 h immersion, the aluminium specimens were carefully washed in double-distilled water, dried, and then weighed. The weight loss data were gained from the average value of three parallel samples in 1 M NaOH with *P. longum* extract at different concentrations.

#### 3. Results and discussion

#### 3.1. Electrochemical measurements

#### 3.1.1. Polarization measurements

Polarization curves of Al in 1 M NaOH solution without and with different concentrations of *P. longum* extract are shown in Fig. 1. It could be observed that both the cathodic and anodic reactions were suppressed with the addition of *P. longum*, which suggested that the inhibitor exerted an efficient inhibitory effect both on anodic dissolution of metal and on cathodic



**Figure 1** Polarization curves for Al in 1 M NaOH in presence of different concentrations of *P. longum* seed extract.

Inhibitor (mg L <sup>-1</sup> )	$E_{\rm corr}$ (V/SCE)	$I_{\rm corr}  ({\rm mA  cm}^{-2})$	$\beta_a (V dec^{-1})$	$\beta_{\rm c} ({\rm V dec}^{-1})$	$IE_P$ (%)
1 M NaOH	-1.55	274	1.189	0.61	-
50	-1.53	112	1.211	0.79	59
100	-1.54	107	1.024	0.76	61
200	-1.54	47	0.972	0.75	83
300	-1.54	37	0.834	0.71	86
400	-1.51	31	0.652	0.65	89

 Table 1
 Electrochemical polarization parameters and the corresponding inhibition efficiencies for Al in 1 M NaOH solution in the absence and presence of different concentrations of *Piper longum* seed extract.

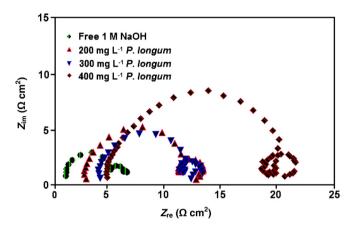


Figure 2 Nyquist plots for aluminium in 1 M NaOH with various concentrations of *P. longum* seed extract.

hydrogen reduction reaction. Electrochemical parameters such as  $E_{\text{corr}}$ ,  $I_{\text{corr}}$ , and anodic and cathodic Tafel slopes ( $\beta_a$ ,  $\beta_c$ ) obtained from the polarization measurements are listed in Table 1. The inhibition efficiency ( $IE_P$ %) was calculated by following equation (Tao et al., 2011):

$$IE_P\% = \frac{I_{\rm corr} - I_{\rm corr(i)}}{I_{\rm corr}} \times 100$$
(1)

 $I_{\text{corr}}$  and  $I_{\text{corr}(i)}$  signify the corrosion current density in the absence and presence of inhibitors, respectively.

It is evident from Table 1 that values of  $\beta_c$  decreases with increasing extract concentration, which indicated that the P. longum extract was adsorbed on the metal surface and the addition of the inhibitor hindered the alkali attack on the aluminium electrode. Therefore, the inhibitor molecules did not change the hydrogen evolution reaction mechanism. In anodic domain, the value of  $\beta_a$  decreases with the presence of *P. longum*. The shift in the anodic Tafel slope  $\beta_a$  might be attributed to the modification of anodic dissolution process due to the inhibitor modules adsorption on the active sites. Compared to the free NaOH solution, the cathodic and anodic curves of the working electrode in the alkaline solution containing the P. longum shifted obviously to the direction of current reduction, as it could be seen from these polarization results; the inhibition efficiency  $(IE_P\%)$  increased with extract concentration reaching a maximum value of 89% at 400 mg  $L^{-1}$ . In literature (Ferreira et al., 2004), it is reported that only when the open circuit potential (OCP) displacement is at least 85 mV in relation to the one measured for the blank solution, can a compound be recognized as an anodic or cathodic inhibitor. From Fig. 1 it can be seen that the displacement

was at most 40 mV with respect to  $E_{\text{corr}(0)}$  (the open circuit potential of blank solution). Therefore, *P. longum* seed extract might act as a mixed-type inhibitor.

# 3.1.2. Electrochemical impedance spectroscopy (EIS) measurements

Nyquist plots of aluminium in 1 M NaOH solution in the absence and presence of different concentrations of P. longum seed extract are given in Fig. 2, where it can be observed that the diameter of the semicircle increases with increasing P. longum extract concentration. This increase in capacitive semicircles suggests that the inhibition action of these inhibitors is due to their adsorption on the metal surface without altering the corrosion mechanism. Further, the figure manifested two depressed capacitive semicircles, typical of Randles element, at higher and lower frequency regions, separated by an inductive loop at low frequencies. Inductive loops can be explained by the occurrence of adsorbed intermediate on the surface. Therefore, adsorbed intermediate species such as  $Al_{ads}^{+} \mbox{ and } Al_{(ads)}^{3+} \mbox{ might be involved in Al dissolution process}$ (Macdonald, 1990). The capacitive semicircle at higher frequencies is attributed to the redox Al-Al<sup>+</sup> reaction since it was assumed to be the rate determining step in the charge transfer process (Abdel-Gaber et al., 2008; Lee and Kim, 2001). Therefore, the resistance value obtained from intercepts of the first capacitive semicircle with real axis corresponds to the Al-Al<sup>+</sup> charge transfer resistance. On the other hand, the second capacitive semicircle could be attributed to the fast complementary redox Al<sup>+</sup>-Al<sup>3+</sup> reaction. The curve manifested that addition of extract to alkaline NaOH solution leads

 Table 2
 Corrosion parameters for aluminium in aqueous solution of 1 M NaOH in absence and presence of different concentrations of *Piper longum* from weight loss measurements at 308 K for 1 h.

Inhibitor concentration (mg $L^{-1}$ )	Weight loss (mg cm <sup><math>-2</math></sup> h <sup><math>-1</math></sup> )	$IE_{w}$ (%)	$CR (mm y^{-1})$
1 M NaOH	15.1	-	56
50	5.1	66	19
100	3.7	75	14
200	1.4	91	5
300	1.2	92	4
400	0.8	94	3

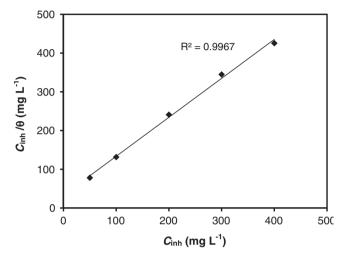


Figure 3 Langmuir adsorption isotherm plot for corrosion of aluminium in 1 M NaOH containing different concentration of *P. longum* seed extract at 308 K.

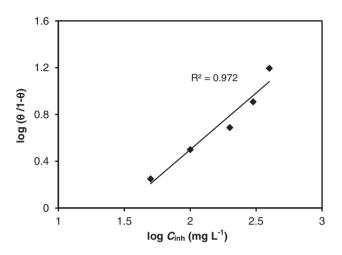
to increase in the size of the capacitive semicircles, indication for increasing the resistances and decreasing corrosion rate.

#### 3.2. Weight loss measurements and adsorption isotherm

The values of inhibition efficiency  $(IE_w\%)$  obtained from weight loss measurements for different concentrations of *P. longum* seed extract in 1 M NaOH are given in Table 2. The inhibition efficiencies,  $IE_w\%$ , were calculated by the following equation:

$$IE_w\% = \frac{W_0 - w}{W_0} \times 100$$
(2)

 $W_0$  and W are the corrosion rates in the absence and presence of the extract, respectively. The obtained results (Table 2) suggested that the inhibition efficiency increases with increasing concentration of extract. As the concentration reached 400 mg L<sup>-1</sup>, the inhibition efficiency of extract obtained a high value of 95%, which represented excellent inhibitive property. Adsorption of *P. longum* seed extract can be explained on the basis that adsorption of the inhibitor was mainly via hetero atoms (viz., N) present in different constituents of extract in addition to the availability of  $\pi$  electrons in the aromatic system (Khaled, 2008). The phytoconstituents of *P. longum* fruits include several pieridine alkaloids such as piperine, piplartine, piperlongumine, piperlonguminine, pipernonaline and piperundecalidine, etc., a few hitherto unidentified steroids



**Figure 4** El-Awady and Ahmed (El-Awady and Ahmed, 1985) model plot for corrosion of aluminium in 1 M NaOH containing different concentration of *P. longum* seed extract at 308 K.

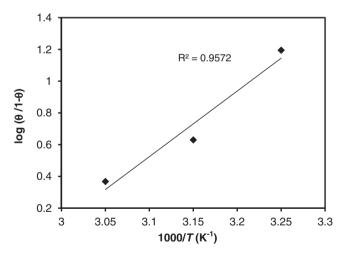


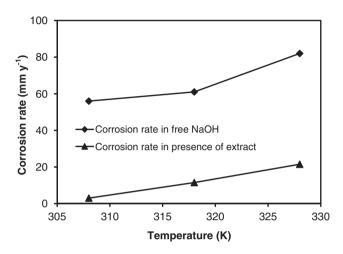
Figure 5 Plot of log  $(\theta/1 - \theta)$  versus 1/T for corrosion of aluminium in 1 M NaOH solution containing 400 mg L<sup>-1</sup> *P. longum* seed extract.

and some reducing sugars and their glycosides (Desai et al., 1989; Hamrapurkar et al., 2011).

The establishment of isotherms that describe the adsorptive behaviour of a corrosion inhibitor is an important part of its study, as they can provide important clues to the nature of

**Table 3** The values of adsorption constant ( $K_{ads}$ ), free energy of adsorption ( $\Delta G_{ads}^{\circ}$ ), number of active sites (1/y) and heat of adsorption (Q) for adsorption of compounds present in P. longum seed extract on aluminium in 1 M NaOH in the presence of 400 mg L<sup>-1</sup> concentrations of the extract.

Langmuir isotherm		Kinetic m	Kinetic model		
$K_{\rm ads} \ ({\rm mg} \ {\rm L}^{-1})^{-1}$	$\Delta G_{\rm ads}^{\circ} \; ({\rm kJ} \; {\rm mg}^{-1})$	1/y	$K_{\rm ads} \ ({\rm mg} \ {\rm L}^{-1})^{-1}$	$\Delta G_{\rm ads}^{\circ} ~({\rm kJ}~{\rm mg}^{-1})$	
0.076	-3.696	4.27	0.081	-3.862	-79.2



**Figure 6** Effect of temperature on dissolution of aluminium in 1 M NaOH.

the metal-inhibitor interaction. The values of surface coverage  $(\theta)$  are defined as  $IE_w\%/100$  and obtained from weight loss measurements at 308 K. Several adsorption isotherms were assessed to fit  $\theta$  values, but the best fit was found to obey Langmuir adsorption isotherm (Langmuir, 1917) which may be expressed by:

$$\frac{C_{\rm inh}}{\theta} = \frac{1}{K_{\rm ads}} + C_{\rm inh} \tag{3}$$

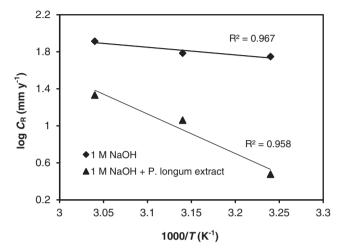
where  $C_{\rm inh}$  is inhibitor concentration and b is equilibrium constant of adsorption. It is well known that the standard adsorption free energy ( $\Delta G_{\rm ads}^{\circ}$ ) is related to equilibrium constant of adsorption ( $K_{\rm ads}$ ) and can be calculated by the following equation (Khamis, 1990):

$$K_{\rm ads} = \left(\frac{1}{55.5}\right) \exp\left(\frac{-\Delta G_{\rm ads}^{\circ}}{RT}\right) \tag{4}$$

Fig. 3 represents the plot of  $(C_{inh}/\theta)$  against  $C_{inh}$  for investigated *P. longum* seed extract. Also, it is found that the kinetic-thermodynamic model of El-Awady and Ahmed (El-Awady and Ahmed, 1985), i.e.,

$$\log\left(\frac{\theta}{1-\theta}\right) = \log k' + y \log C_{inh} \tag{5}$$

is valid to operate the present adsorption data.  $K_{ads} = K'^{(1/y)}_{ads}, K'_{ads}$  is constant, and 1/y is the number of the surface active sites occupied by one inhibitor molecule and  $C_{inh}$  is the bulk concentration of the inhibitor. Plotting  $\log(\theta/1 - \theta)$  against  $\log C_{inh}$  for the extract is given in Fig. 4, where straight line relationships were obtained suggesting the validity of this model for the present case. A plot of  $\log(\theta/1 - \theta)$  versus 1/T at the extract concentration of 400 mg L<sup>-1</sup> (Fig. 5) gives straight lines according to the following equation:



**Figure 7** Arrhenius plots of  $\log C_R$  versus 1/T for aluminium in 1 M NaOH in the absence and presence of 400 mg L<sup>-1</sup> *P. longum* seed extract.

$$\log\left(\frac{\theta}{1-\theta}\right) = \log A + \log C_{\rm inh} - \left(\frac{Q}{2.303RT}\right) \tag{6}$$

The Q value was obtained from the slope of this line. The values of  $\beta$  and  $\Delta G_{ads}^{\circ}$  calculated by Langmuir isotherm and 1/y,  $\beta$ , and  $\Delta G_{ads}^{\circ}$  calculated by the kinetic model, and the values of Q are given in Table 3. The negative values of  $\Delta G_{ads}^{\circ}$  suggest that the adsorption of inhibitor molecules onto steel surface is a spontaneous process. The magnitude of heat of adsorption reaches the magnitude of heat of chemical reaction, which is the result of the transference of electron from donating atoms in the phytoconstituent molecules present in the extract to the d-orbital of the iron atom. The negative values of Q show that the process of adsorption is exothermic. It is noted that the value of 1/y is more than unity. This suggests that the studied plant extract will form monolayer on the aluminium/solution interface.

#### 3.3. Effect of temperature

The effect of temperature on the rate of dissolution of aluminium in 1 M NaOH containing 400 mg L<sup>-1</sup> of the *P. longum* seed extract was tested by weight loss method over a temperature range from 308 to 328 K. The effect of increasing temperature on the corrosion rate values is given in Fig. 6. The results revealed that on increasing temperature there is an increase of corrosion rate for both in the absence and presence of extract. The increase in corrosion rate in the absence of extract is higher at all temperatures studied, suggests more aggressiveness of free alkaline solution. The increase in corrosion rate with increase in temperature may be probably due to decreasing strength of adsorption (shifting the adsorption–desorption equilibrium toward desorption) and roughening of the electrode surface which results from enhanced corrosion.

The activation energy of the corrosion process was calculated using the Arrhenius equation (Putilova et al., 1960):

$$C_R = \lambda \exp\left(\frac{-E_a}{RT}\right) \tag{7}$$

where  $C_R$  is the corrosion rate,  $\lambda$  is the pre-exponential factor,  $E_a$  is the apparent activation energy, R is the universal gas constant, and T is the absolute temperature. The apparent activation energy was calculated by linear regression between  $\ln C_R$  and 1/T (Fig. 7). The value of  $E_a$  can be obtained from the slope of the straight line which was found to be 15.9 kJ mg<sup>-1</sup> and 81.8 kJ mg<sup>-1</sup> in the absence and presence of 400 mg L<sup>-1</sup> *P. longum* seed extract, respectively. The higher value of  $E_a$  in the presence of extract than its absence indicates a strong inhibitive action of the extract by increasing the energy barrier for the corrosion process.

#### 4. Conclusion

*P. longum* seed extract was found to inhibit the corrosion of aluminium in 1 M NaOH solution and inhibition efficiency increases with increasing extract concentration. At the highest extract concentration of 400 mg  $L^{-1}$ , the inhibition efficiency is increased markedly and reached 94%. Potentiodynamic polarization curves proved that the *P. longum* seed extract was a mixed-type inhibitor. EIS plots indicated that the charge transfer resistances increase with increasing concentration of the extract. The adsorption model obeys the Langmuir adsorption isotherms model. The *P. longum* seed extract inhibits the corrosion of aluminium in alkali media by adsorption mechanism.

#### Acknowledgment

We gratefully acknowledge the financial support of the Council of Science and Industrial Research (CSIR), New Delhi, India.

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