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Evaluation of Tectona Grandis Leaf Extract As A Green Corrosion Inhibitor for Aluminium in 1M NaOH

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Abstract

A kind of tropical hardwood tree called the Tectona grandis tree that is indigenous to India. The name "teak tree" is another common name for this kind of tree. It is a member of the plant family that is referred to as Lamiaceae. The primary purpose of this research is to investigate whether or not the plant extract has a corrosion-inhibiting quality. In this study, an aqueous extract of Tectona grandis leaves, commonly known as TGLE, was employed as a corrosion inhibitor for aluminium (Al) in a solution of sodium hydroxide with a concentration of 1M. The sodium hydroxide solution was prepared by diluting 1M sodium hydroxide with 1M sodium hydroxide. Electrochemical methods, such as Potentiodynamic polarisation (PDP) and Electrochemical Impedance Spectroscopy (EIS), in addition to chemical methods, were used in this study to evaluate the inhibitory effects of this medication (weight loss). A promising inhibitory efficiency (IE) of 80% was achieved with TGLE when it was administered at an optimal dosage of 1.2 g/L; this was found to be consistent across all of the techniques that were used. The results of the trials were combined into an appropriate adsorption isotherm, which allowed for additional study into the inhibitor's method of action to be carried out. In order to investigate the possible influence that temperature may have on the effectiveness of the inhibitor, a temperature range of 303 to 333 degrees Kelvin was investigated. An inquiry of the surface was carried out using microscopic metallurgical research, and the results of this study provided further support for the findings.

Keywords: Corrosion, Aluminium, Green inhibitor, NaOH, *Tectona grandis*, Teak

INTRODUCTION

Aluminum (Al) and the alloys that it forms have a number of distinguishing characteristics, such as high levels of electrical and thermal conductivity, low densities (only 2.7 grammes per cubic centimetre), and a lack of ferromagnetic properties, that allow them to be useful in a wide variety of applications. These applications include construction, automobiles, power generation, and home appliances. Because of where it falls in the electromotive force series, aluminium is an example of a metal that is thermodynamically reactive. Most of the time, aluminium is able to resist corrosion because it forms a thin barrier oxide coating on its surface that is just one nanometer thick. This film can be easily repaired under most environmental circumstances. Because this film is not affected by surroundings with a pH between 4 and 9, aluminium is considered to be passive in these settings. Aluminum corrodes in extremely acidic or alkaline solutions owing to the dissolving of the oxide coating, producing Al^{3+} in strongly acidic solutions and AlO^{2-} in strongly alkaline solutions [1]. This occurs beyond of

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aluminum's passive range. The severe corrosion of aluminium may result in enormous losses of natural resources and wasteful investment, making it a problem that affects the whole world. Corrosion inhibitors are a tried-and-true method that has gained widespread acceptance for preventing metal corrosion. When introduced to a corrosive medium, compounds known as corrosion inhibitors slow or stop the deterioration of metals in the medium. Dihydroxy benzenes [2], thiourea derivatives [3], high molecular weight polymers [4], and aromatic acid derivatives [5] are some examples of organic and inorganic additives that have been documented as being used as corrosion inhibitors. Because the majority of them are both costly and harmful to the environment, people are looking for alternatives that are more environmentally friendly.

The plant extracts are a rich source of phytochemicals such as tannins, saponins, polyphenols, and flavonoids, as well as other phytochemicals. These phytochemicals include hetero atoms such as N, O, and S, which give sites for adsorption onto the surface of metals. The effect could be brought on by an electrostatic interaction between the metal and the inhibitor molecules, a dipole-type interaction between lone pair electron pairs in the inhibitor and the metal, p electrons interacting with the metal, or a combination of all of these things [6]. There have been instances of plant extracts being used as corrosion inhibitors for aluminium in conditions that are both acidic and alkaline. The presence of Abrusprecatorius seed extract in 4N NaOH had an inhibitory impact on aluminium. In their investigation of the effect of Vernonia amygdalina on Al-Si alloy in 0.5N NaOH solution, the researchers were successful in achieving an IE of 87% [7]. Priya et al. 2015, investigated the effect that an aqueous garlic extract had on the rate of aluminium corrosion caused by sodium hydroxide in the presence and absence of Zn^{2+} ions [8]. It has been observed that an extract of Sansevieria trifasciata is effective against the corrosion of aluminium in both acidic and alkaline environments [9].

In the current study, the inhibitory effect of Tectona grandis leaves extract as a green inhibitor on Al corrosion in 1M NaOH was investigated using weight loss (WL) and electrochemical techniques such as potentiodynamic polarisation (PDP) and electrochemical impedance spectroscopy. Weight loss (WL) was used to measure the amount of corrosion and PDP was used to measure the rate of corrosion (EIS).

EXPERIMENTAL METHOD

Materials and Test Solutions

The WL tests were carried out on aluminium coupons with dimensions of 5 × 1 × 0.2 cm deep. A variety of types of water-resistant abrasive paper were used to polish the coupons before each experiment, after which they were washed and dried with acetone. The same coupon that was used for electrochemical measurements also served as the working electrode. The exposed area on this coupon was 1 cm^2. For each of the tests that were performed, a coupon that had just been cleaned and polished was utilised. To prepare the 1M NaOH test solution, 40 g of NaOH were dissolved in 100 millilitres of double-distilled water.

Preparation of *Tectona Grandis* Leaves Extract (TGLE)

Fresh *Tectona grandis* leaves were procured from the Pondicherry University campus in Puducherry. The leaves were rinsed with distilled water to remove dirt prior to getting dried in a hot air oven at 50°C for 6 hours until being finely powdered. This powder (10 g) was refluxed in 100mL distilled water for 2 hours, then filtered with Whatman paper (No.1). Filtrate was evaporated in a water bath to acquire the dried inhibitor compound. To prepare the TGLE solution, 30, 60, 90, and 120 mg of the TGLE were dissolved in 100 mL NaOH to obtain 300, 600, 900, and 1200 ppm (0.3, 0.6, 0.9, 1.2 g/L) concentrations respectively [10].

Procedures

WL Measurements

WL measurements were made as per prior report. Al coupons were immersed in TGLE solutions (0.3–1.2 g/L concentrations) and a blank set-up without TGLE for 2 hours. The coupons were removed, rinsed with water, dried with acetone, and reweighed to get final weight. Difference between initial and final weights provided experimental WL.

The IE%, corrosion rate (CR) and surface coverage (θ) were computed using the formulae [11, 12],

$$\text{IE\%} = ((W_B - W_I)/\, W_B) \times 100 \tag{1}$$

$$\text{CR, mmpy} = \frac{8.76 \times 10^4 \times W}{DAT} \tag{2}$$

$$\text{Surface coverage } (\theta) = (W_B - W_I)/W_B \tag{3}$$

where, W_B is the WL without TGLE, W_I is the WL with TGLE, W is the WL (g), A is the area (cm^2), T is the Time (hrs), and D is the density of Al (2.70 gm/cm^3).

The influence of temperature on inhibition action was probed by operating the WL study in the temperature range 303–333K.

Electrochemical Measurements

After determining a consistent open circuit potential (Eoc), scans were carried out using a three-electrode configuration in conjunction with a biologic electrochemical workstation. Aluminum coupons with an exposure area of one centimetre square were used as the working electrode. Platinum and a conventional calomel electrode were used, respectively, as the counter and reference electrodes. Every experiment was carried out at 303 degrees Kelvin. The EIS were recorded throughout a frequency range of 100 kHz to 1 Hz, with an amplitude of 10mV for the AC signal. The spectra were analysed using EC lab software to determine the impedance values, and they were matched with a model of an electrical circuit equivalent (Figure 1). The IE% was computed using the formula [13, 14],

$$\text{IE } (\%) = \frac{\mathrm{R_{ct}} - \mathrm{R^*_{ct}}}{\mathrm{R_{ct}}} \times 100 \tag{4}$$

where, R^*_{ct} is the charge transfer resistance without TGLE, and R_{ct} is the charge transfer resistance with TGLE.

where R_1 is solution resistance (R_S), C_1 is the double layer capacitance (C_{dl}), and R_2 is the charge transfer resistance (R_{ct}).

Recordings of Tafel polarisation curves were made throughout a potential range of minus 250 mV from open circuit potential (Eoc). The results were collected using a scan rate of 25 mVs^{-1} while the electrode potential was monitored in the range of -1.850 V to -1.350 V. In order to calculate the corrosion current density, linear anodic and cathodic Tafel curves were extrapolated to the corrosion potential (E_{corr}) using EC lab software (I_{corr}). The following formula was used to get the IE%:

$$\mathrm{IE}(\%) = (\mathrm{I^*_{corr}} - \mathrm{I_{corr}}) * 100)/\mathrm{I^*_{corr}} \tag{5}$$

where, $\mathrm{I^*_{corr}}$ and $\mathrm{I_{corr}}$are the corrosion current densities without and with TGLE, respectively.

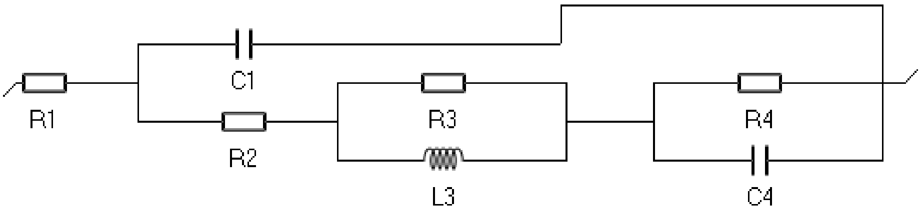

Figure 1. Implemented equivalent circuit to fit EIS data.

Surface Analysis

Al coupons measuring 5×1×0.2 cm were submerged in a blank solution consisting of 1M NaOH as well as the optimal TGLE concentration at which maximum IE% was measured. After that, the coupons were washed and dried, and metallurgical microscopy was used to observe any changes that occurred on the surface.

RESULTS AND DISCUSSION

WL Measurements

The WL data for aluminium in 1M sodium hydroxide are shown in Table 1 together with and without varying amounts of TGLE.

The CR is very high in the absence of TGLE, providing evidence of its inhibitory function. Increasing the concentration of TGLE from 0.3 to 1.2 g/L has the effect of reducing CR while simultaneously increasing IE. At a concentration of 1.2 g/L of TGLE, the greatest inhibitory effectiveness (IE) was measured, which indicated that this was the ideal concentration for corrosion inhibition. Bashire et al. 2019, found that raising the concentration of TGLE led to a stronger adsorption of TGLE molecules on the surface of the aluminium, which led to an increase in the surface coverage and a better separation of the aluminium from the sodium hydroxide [15].

Effect of Temperature

Due to the desorption of TGLE molecules at higher temperatures, the IE decreases as the temperature increases (Table 2). The IE decreased as the temperature increased demonstrates that TGLE molecules interact with the surface of Al through physisorption.

Activation energy (E_a), enthalpy of activation (ΔH), and entropy of activation (ΔS) were derived for the blank solution and optimal TGLE concentration (1.2 g/L) using the Arrhenius law and Eyring transition state equation:

$$\text{Arrhenius equation: } k = Ae^{-Ea/RT} \tag{6}$$

where, k is the CR, E_a is the energy of activation for corrosion, A is the Arrhenius frequency factor, R is the universal gas constant, and T is the absolute temperature.

Eyring transition state equation:

$$CR = \frac{RT}{Nh}\exp\left(\frac{\Delta S}{R}\right)\exp\left(\frac{\Delta H}{RT}\right) \tag{7}$$

Table 1. Corrosion data obtained from WL measurements for Al in 1M NaOH

[TGLE] (g/L)	WL (g/cm^3)	CR (mmpy)	IE (%)	Surface coverage (θ)
Blank	0.2625	823.73	-	-
0.3	0.1038	325.79	60.5	0.605
0.6	0.0799	250.90	69.5	0.695
0.9	0.0576	180.80	78.1	0.781
1.2	0.0524	164.39	80.0	0.800

Table 2. CR and IE of Al in 1M NaOH without and with optimum TGLE concentration at varied temperature computed by WL measurements

[TGLE] (g/L)	Temperature (K)			
	303	*313*	*323*	*333*
	IE (%)			
0.3	59.91	58.05	50.53	46.00
0.6	69.76	63.42	60.09	53.67
0.9	77.47	75.84	69.93	68.27
1.2	80.04	77.86	75.41	71.69

where, N is the Avogadro's number (6.023×10^{23}), and h is the Planck's constant (6.626×10^{-34}Js).

E_a is obtained from the slope of the plot log (CR) vs (1/T) (Figure 2).

ΔH and ΔS data are derived from the slope and intercept of the plot log(CR/T) vs (1/T), (Figure 3). The activation parameters data are presented in Table 3 and Table 4.

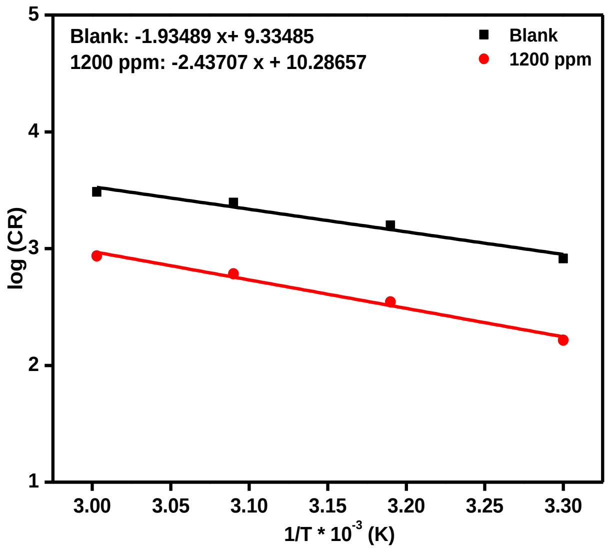

Figure 2. Arrhenius plots for Al in 1M NaOH without and with optimum TGLE concentration.

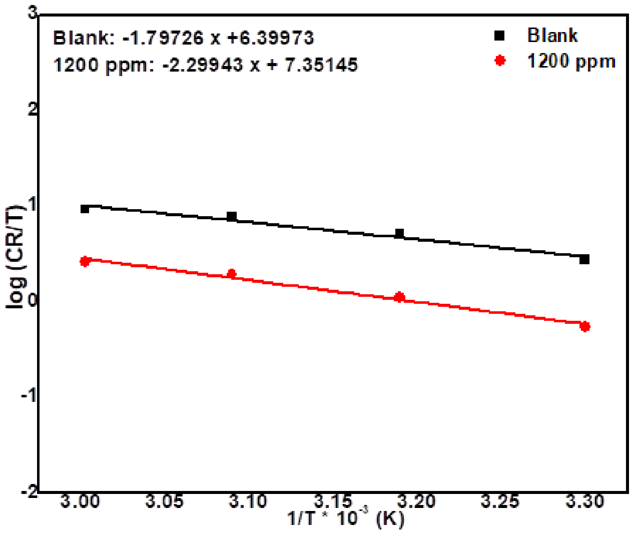

Figure 3. Variation of log(CR/T) vs. (1/T) for Al in 1M NaOH in the without and with optimum TGLE concentration.

Table 3. Computed values of activation energy (E_a) and free energy of adsorption ($\Delta G°_{ads}$) without and with optimum TGLE concentration

System	**E_a (kJ/mol)**			**$-\Delta G°_{ads}$ (kJ/mol)**			
	Temperature (K)			***Temperature (K)***			
	303-313	*313-323*	*323-333*	*303*	*313*	*323*	*333*
Blank	49.31	37.19	20.07	-	-	-	-
1.2 g/L of TGLE	57.15	45.91	33.54	14.46	14.29	14.04	13.84

Table 4. Computed data of entropy of adsorption (ΔS) and enthalpy of adsorption (ΔH) for Al without and with optimum TGLE concentration

Concentration	**ΔS ($JK^{-1}mol^{-1}$)**	**ΔH ($kJ\ mol^{-1}$)**
Blank	-75.04	34.41
1.2 g/L of TGLE	-56.82	44.03

The fact that the Ea values for the blank solution are lower than those for the solution containing 1.2g/L of TGLE is evidence that TGLE has an inhibiting effect. The fact that the values of Ea were greater when the TGLE was present is further evidence that the TGLE layer on the surface of the aluminium was generated via physisorption. The H and S measurements for the blank solution are lower when compared to the results for 1.2 g/L of TGLE. According to Obot and Obi-Egbedi (2010), a negative value for H denotes that the associative step rather than the dissociative step is involved in the activated complex during the rate-determining step. This indicates that the randomness decreases as one moves from the reactants to the activated complex.

It is possible to say that the corrosion-inhibiting activity of TGLE takes place due to its adsorption onto the surface of the aluminium, which then creates a barrier between the aluminium and the sodium hydroxide. Both physisorption and chemisorption are possible forms of adsorption, although physisorption is the more common one.

It is crucial to apply appropriate adsorption isotherms in order to have an understanding of the nature of the interaction that occurs between TGLE and Al. The surface coverage values that were acquired through WL measurements were found to have a good match with the Langmuir adsorption isotherm, and this was discovered after the measurements were carried out.

$$\frac{C}{\theta} = \frac{1}{K_{ads}} + C \qquad (8)$$

where, C is the concentration of the TGLE(g/L), θ is the degree of surface coverage, and K_{ads} is the equilibrium constant for adsorption.

Figure 4 gives the plot C/ θ vs. C, straight lines with regression coefficients (R^2) close to one are obtained. The reciprocal of the intercept gives the adsorption constant (K_{ads}) which can be related to the standard free energy of adsorption ($\Delta G°_{ads}$) by the equation:

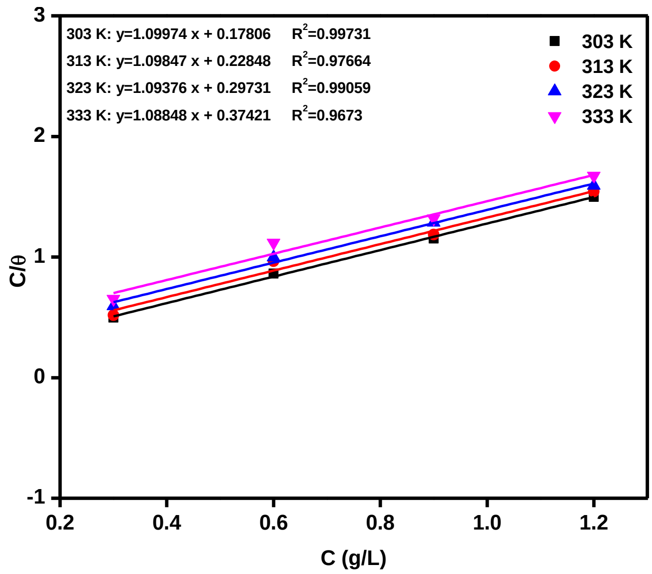

Figure 4. Langmuir adsorption isotherm plots for Al in the blank solution and optimum TGLE concentration.

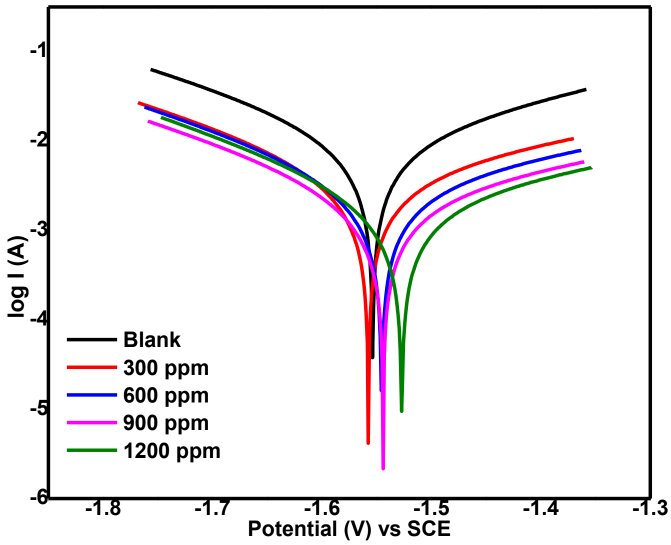

Figure 5. PDP curves for Al in 1M NaOH without and with varied TGLE concentrations.

Table 5. PDP parameters for Al in 1M NaOH without and with varied TGLE concentrations

[TGLE] g/L	**I_{corr} (mA)**	**E_{corr} (mV)**	**β_c (mV)**	**β_a (mV)**	**IE (%)**
Blank	12.02	-1554	273	374	-
0.3	4.75	-1558	270	483	60.5
0.6	3.66	-1545	258	491	69.6
0.9	2.64	-1554	260	479	78.0
1.2	2.40	-1527	244	480	80.0

$$\Delta G^{\circ}_{ads}=-RT\ \ln\ (55.5 * K_{ads}) \tag{9}$$

where, R is the universal gas constant, T is the absolute temperature, and 55.5 is concentration of water in solution (mol L^{-1}).

Calculated values of ΔG_{ads} (Table 3) are negative and below -20 kJ/mol, which indicate that TGLE is spontaneously adsorbed on Al surface. ΔG_{ads}upto -20 kJ/mol indicate physisorption involving electrostatic interaction between charged TGLE molecules and the charged Al, while those more negative than -40 kJ/mol indicate chemisorption. This confirms physical adsorption mechanism for TGLE on Al [16–18].

Electrochemical Measurements

Potentiodynamic Polarization Studies

Figure 5 illustrates the PDP curves for Al in the blank solution and varied TGLE concentrations. I_{corr}, E_{corr}, and Tafel slopes (β_a and β_c) are presented in Table 5. In the presence of TGLE, I_{corr} is significantly reduced, suggesting its inhibitive efficacy. A maximum IE of 80% was recorded at 1.2 g/L TGLE concentration. An inhibitor is anodic or cathodic, if its E_{corr}values shift by ±85 from the blank solution. Since there is no significant shift in E_{corr} for TGLE, it acts as a mixed- type inhibitor [19, 20].

Electrochemical Impedance Spectroscopy

The Nyquist plots that were produced for aluminium in 1M sodium hydroxide with and without TGLE are shown in Figure 6. There are three loops that may be seen, and they are a capacitive loop at a higher frequency, an inductive loop at a lower frequency, and another capacitive loop at a lower frequency [21, 22]. The existence of a protective oxide coating on the metal surface and the interfacial interactions at the metal/oxide/electrolyte interface are two primary causes for the high frequency capacitive loop. Both of these factors contribute significantly to the phenomenon. The processes include the creation of Al^+ ions at the interface between the metal and the oxide, followed by the migration of these ions across the contact between the oxide and the electrolyte, where they are oxidised to Al^{3+}. At the contact between the oxide and the electrolyte, OH^- or O^{2-} ions may also be produced. The presentation of these three responses by one loop might be the result of an overlap of processes, or the dominance of one of the processes could be responsible for the suppression of the others. At intermediate frequencies, the inductive loop may be caused by the surface or bulk relaxation of species that are present in the oxide layer. According to Wang et al. (2009), the inductive loop may have been caused by an adsorbed intermediate ([AlOH]ads), while the capacitive loop at low frequency may have been caused by the disintegration of the oxide layer [23].

The results of measuring the impedance of aluminium in 1M NaOH and varying concentrations of TGLE are shown in Table 6. When Cdl was lowered in conjunction with an increase in Rctreflect, an increase in inhibition could be seen at higher TGLE concentrations.

Metallurgical Microscopic Analysis

The surface alterations that took place on the aluminium coupons are shown in Figure 7 when they were submerged in 1M NaOH and TGLE (1.2 g/L). The Al surface that was submerged in TGLE solution had much less corrosion than the surface that was exposed to 1M NaOH. It is implied that TGLE has adsorbed into the surface of the aluminium, hence decreasing the corrosive damage that was caused by the medium [24, 25].

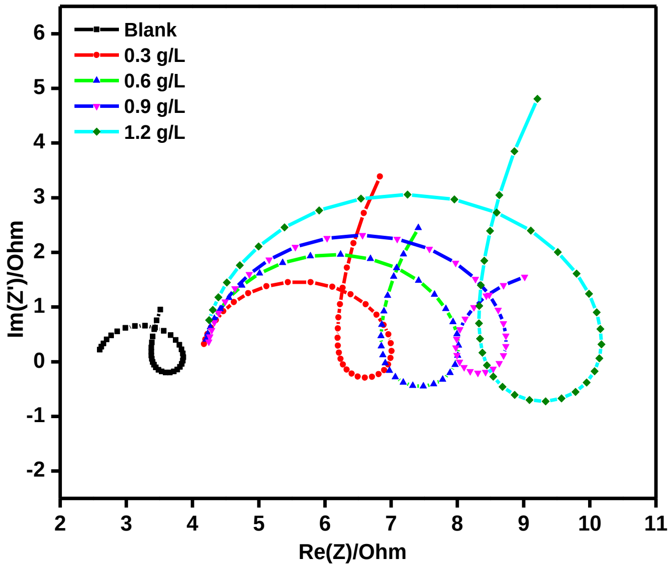

Figure 6. EIS obtained for Al in 1M NaOH in absence and presence of various concentrations of TGLE.

Table 6. Impedance parameters for Al corrosion in 1M NaOH without and with varied TGLE concentrations

[TGLE] (g/L)	R_s (Ohm)	R_{ct} (Ohm)	C_{dl} (μF)	IE %
Blank	2.56	0.80	7.03	-
0.3	4.17	2.02	2.91	60.6
0.6	4.21	2.62	2.73	69.6
0.9	4.24	3.62	2.60	78.0
1.2	4.25	4.08	2.06	80.5

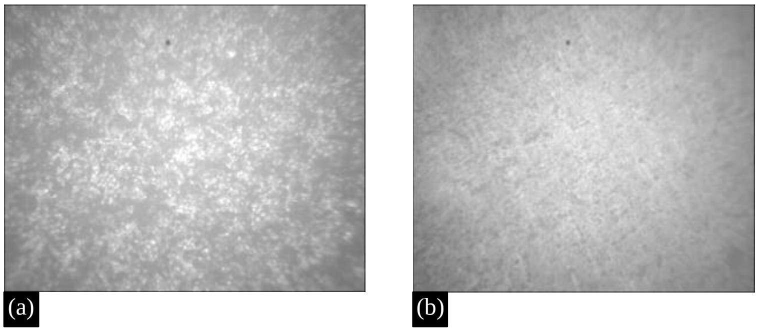

Figure 7. Surface images of (a) Al coupon immersion in 1M NaOH, (b) Al coupon immersion in 1.2 g/L TGLE

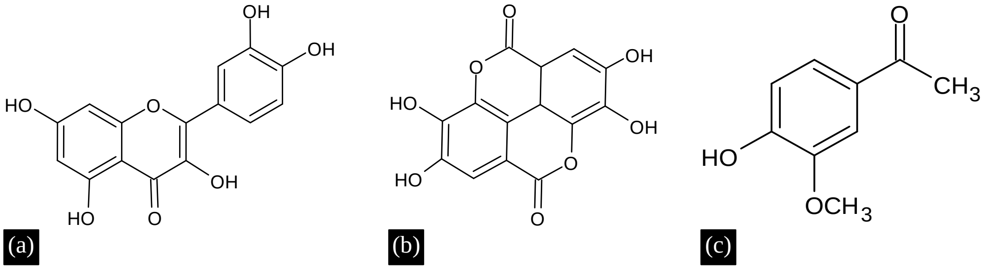

Figure 8. Structure of (a) Quercetin, (b) Ellagic acid (c) Acetovanillone.

Inhibition Mechanism

The phytoconstituents in TGL are responsible for the product's anti-corrosion properties. TGL are abundant in flavonoids like rutin and quercetin as well as phenolic compounds like gallic acid, ellagic acid, acetovanillone, and E-isofuraldehyde [26]. These heteroatoms that interact with the surface of the aluminium make up the bioactive components seen in Figure 8(a-c).

CONCLUSION

It was observed that an IE of 80% could be achieved using TGLE's inhibition of Al corrosion at a concentration of 1.2 g/L, and this result was shown to be consistent across all of the methodologies that were investigated. The PDP investigations demonstrated that TGLE operated as a mixed-type inhibitor since there was no discernable change in the Ecorr values during the course of the investigation. In order to have a better understanding of the dynamics of adsorption, the Langmuir isotherm was modified. On the basis of the fact that $-G^{\circ}_{ads}$ was stated to be less than -20 kJ/mol, physisorption was recommended. The inhibitory effectiveness of TGLE was validated by metallurgical microscopic inspection because an aluminium surface that was dipped in 1M sodium hydroxide had greater corrosion damage than an aluminium surface that was dipped in the TGLE system. The phytoconstituents of TGLE are what make up the heteroatoms, and they are what are responsible for its inhibitory function. As a consequence of this, TGLE functions in alkaline media as a potentially useful green inhibitor for aluminium.

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