PLANTS AS A SOURCE OF GREEN CORROSION INHIBITORS: THE CASE OF GUM EXUDATES FROM *ACACIA SPECIES (A. DREPANOLOBIUM AND A. SENEGAL)*

J Buchweishaija
Chemistry Department, College of Natural and Applied Science,
P.O. Box 35061, University of Dar es Salaam, Dar es Salaam, Tanzania

ABSTRACT

The inhibitive performance of gum exudates from Acacia drepanolobium *and Acacia senegal from Tanzania, towards the corrosion of mild steel in fresh water has been investigated. The experimental methods include potentiodynamic polarization and electrochemical impedance spectroscopy (EIS) studies. The results indicate that gum exudates (*Acacia drepanolobium *and* Acacia senegal*) exhibit good inhibition characteristics to corrosion on mild steel under fresh water medium and the inhibition efficiency of up to 90.7% and 99.7% respectively was attained at 30 ^{o}C. Furthermore, the studies have shown that the inhibition performance remained above 90% on both* Acacia *exudates independent of the raise in temperature. Polarization measurements revealed that the investigated inhibitors are mixed type for mild steel corrosion in fresh water with significant reduction of anodic current densities (this sentence is not clear). The results obtained in this work show that these gum exudates of* Acacia senegal *and* Acacia drepanolobium *can serve as effective green inhibitors for the corrosion of mild steel in fresh water systems.*

INTRODUCTION

Corrosion is a major destructive process affecting the performance of metallic materials in applications in many construction sectors. Corrosion is a naturally occurring phenomenon commonly defined as deterioration of metal surfaces caused by the reaction with the surrounding environmental conditions. The consequences of corrosion are quite many and are considered a serious problem in industry, constructions and civil services such as electricity, water and sewage systems. To prevent or minimize internal corrosion in these systems, inhibitors are used especially in flow and closed systems, such as fresh water distribution systems. A corrosion inhibitor is a substance which when added in small concentration to an environment, effectively reduces the corrosion rate of a metal exposed to it. Large numbers of organic compounds have been studied and are still being studied to assess their corrosion inhibition potential. However, most of these substances are not only expensive but also poses health and environmental hazards (Raja and Sethuraman 2008) prompting the search for their replacement.

Plants have been recognized as sources of naturally occurring compounds that are generally referred to as 'green' compounds, some with rather complex molecular structures and having a variety of physical, chemical and biological properties. A number of these compounds are enjoying use in traditional applications such as pharmaceuticals and biofuels (Farooqi *et al.* 1997, Mukherjee *et al.* 1997, Nkunya 2002). Furthermore, there has been a growing trend in the use of natural products as corrosion inhibitors for metals in various corrosive media (Orubite and Oforka 2004). In fact, plants have become important as environmentally acceptable, readily available and renewable source of a wide range of chemicals. Due to bio-degradability, eco-friendliness, low cost and easy availability, the extracts of some common plants based chemicals and their by-products have been tried as inhibitors for metals under different environments (Abdel-Gaber *et al.* 2006, Ebenso and Ekpe 1996, Ebenso *et al*. 2004, Ekpe *et al.* 1994, Kliskic *et al.* 2000).

The use of the gum exudate from *Acacia seyal* var *seyal* as corrosion inhibitor for mild steel in fresh water has been reported (Buchweishaija and Mhinzi 2008). In the continued effort to contribute to the search for green corrosion inhibitors to replace the toxic fossil origin organic inhibitors, the present study reports on the corrosion inhibition and adsorption behaviour of the gum exudates from *Acacia drepanolobium* and *Acacia senegal* on mild steel in fresh water at the temperature range of 30 to 70 °C using potentiodynamic polarization and electrochemical impedance spectroscopy methods.

EXPERIMENTAL

The tests were carried out in a 100 mL three electrodes thermostatically controlled cell made of pyrex glass with a Rotating Disk Electrode (RDE) assembly which served as working electrode (Mutasingwa 2004). The cell was also equipped with counter electrode, reference electrode and a thermometer. The experimental set up has been described in detail elsewhere (Philip *et al.* 2001). Two electrochemical techniques were employed to observe the corrosion processes for both inhibited and uninhibited media. These were electrochemical impedance spectroscopy and potentiodynamic polarization techniques.

Materials preparation

Materials used for the study were cylindrical mild steel machined from a parent metallic pipe used for domestic water piping and plumbing in Dar es Salaam, Tanzania. The composition and preparation of mild steel samples were as previously reported by Buchweishaija and Mhinzi, 2008. The specimens were mounted in Teflon tube with an opening exposing a surface area of 0.07 cm^2. Prior to carrying out the corrosion tests, the metal specimens were mechanically ground successively with 400 to 4000 grit silicon carbide papers followed by polishing with diamond paste (9 to 1.0 μm) to achieve mirror finish surfaces. The specimens were then ultrasonically degreased in absolute ethanol, washed thoroughly with distilled water, dried with a jet of hot air followed with acetone and immediately stored in moisture-free desiccators before carrying out corrosion studies.

The corrosive medium was chlorinated fresh water collected from Upper Ruvu water work, Coast region, Tanzania. The corrosion inhibitors used were *Acacia* gum exudates collected from Singida in Tanzania namely *A. drepanolobium and A. senegal.* The performances of the gum exudates from *Acacia* species as corrosion inhibitors were monitored in the concentration range from 0 to 1000 ppm (v/v) at 30 °C. For each experiment, fresh water solutions as well as freshly polished metallic samples were used. The study presented in this work was carried out at a single rotation speed of 3000 rpm that corresponds to a peripheral velocity of 7.1 m/s, as explained elsewhere (Mutasingwa 2004). The effect of temperature on the performance of these gum exudates was also studied at 50 and 70 °C.

Electrochemical impedance spectroscopy measurements

It was carried out by a computer controlled AUTOLAB Frequency Response Analyzer (FRA) in the frequency range of 10 kHz to 10 mHz at a sweeping rate of 10 points per decade, logarithmic division at the open circuit potential. A sine wave with 10 mV amplitude was used to perturb the system. Measurements were taken at various time intervals ranging from 2 to 12 hours at open circuit potential. The results were analysed by fitting the observed data to a simple equivalent circuit (Randles type of circuit) using a Complex Nonlinear Least Squares fitting program EQUIVCRT to estimate equivalent circuit parameters and corrosion current densities were calculated using Stern Geary relation (Philip *et al.* 2001).

Potentiodynamic polarization measurements

Potentiodynamic polarization measurements were performed using a computer-controlled potentiostat, AUTOLAB PGSTAT20. Cathodic and anodic polarization curves were each recorded on separate electrode by sweeping at a rate of 1 mV/s over a range of 100 mV from the open circuit potential. Platinum sheet was used as a counter electrode and a saturated Ag/AgCl as a reference electrode. All polarization curves were taken at the end of the exposure time to avoid alteration of the surface. From the polarization curves, corrosion current densities were obtained by Tafel extrapolation.

Corrosion rate and percentage inhibition evaluation

The corrosion rate, R_{corr} (mm/y) was evaluated from the corrosion current densities using the Faraday's law from the relation:

$$R_{Corr} = \frac{1.6 \times 10^{-3} \times i_{corr} \times M}{\rho} \qquad (1)$$

where i_{corr} is the current density in μA/cm^2, M is the weight of metal or alloy in g/mol and ρ is the density of metal in g/cm^3. The percent of the inhibition efficiency (IE)% of the *Acacia* gum exudates against corrosion was evaluated from the relation:

$$\mathrm{IE}(\%) = \frac{R_{corr,O} - R_{corr,I}}{R_{corr,O}} \times 100 \qquad (2)$$

Where $R_{\mathrm{corr,O}}$ and $R_{\mathrm{corr,I}}$ are the corrosion rates in the absence and presence of the inhibitor, respectively.

RESULTS AND DISCUSSION

Polarization measurements

Polarization behaviour of mild steel in aerated fresh water in the absence and presence of varied concentrations of inhibitors, *A. drepanolobium* and *A. senegal* gum exudates is shown in Figure 1. The data obtained from polarization curves by Tafel extrapolations are presented in Table 1. These include corrosion potential ($E_{\mathrm{corr.}}$), corrosion current density ($i_{\mathrm{corr.}}$), cathodic Tafel slope, anodic Tafel slope, calculated corrosion rates in millimetre per year (mm/y) and percentage inhibition efficiency (IE%) at varied inhibitor concentrations.

It is seen from Figure 1 that the increase in *Acacia* gum exudates concentration lowers the current densities and shift the corrosion potential anodically. In contrast to *Acacia drepanolobium* gum inhibitor (Figure 1(a)), a significant reduction of current densities as well as anodic shift of open circuit potential was observed when *Acacia senegal* gum concentration is increased (Figure 1(b)). However, only a slight reduction in current density was registered on cathodic and anodic polarization curves when the inhibitor concentration was increased above 400 ppm. These inhibitors might therefore be characterized as anodic inhibitors. However, this is somewhat a misleading description since both inhibitors suppress both anodic and cathodic processes with a considerable reduction of anodic current densities. Based on the shift of open circuit potential and the parallel shift in both cathodic and anodic branches of the polarization curves towards lower current densities, these inhibitors can be considered as inhibitors of the mixed type with a predominant anodic effect (Kaesche and Hackerman 1958). Similar observations have been reported by Buchweishaija and Mhinzi, 2008.

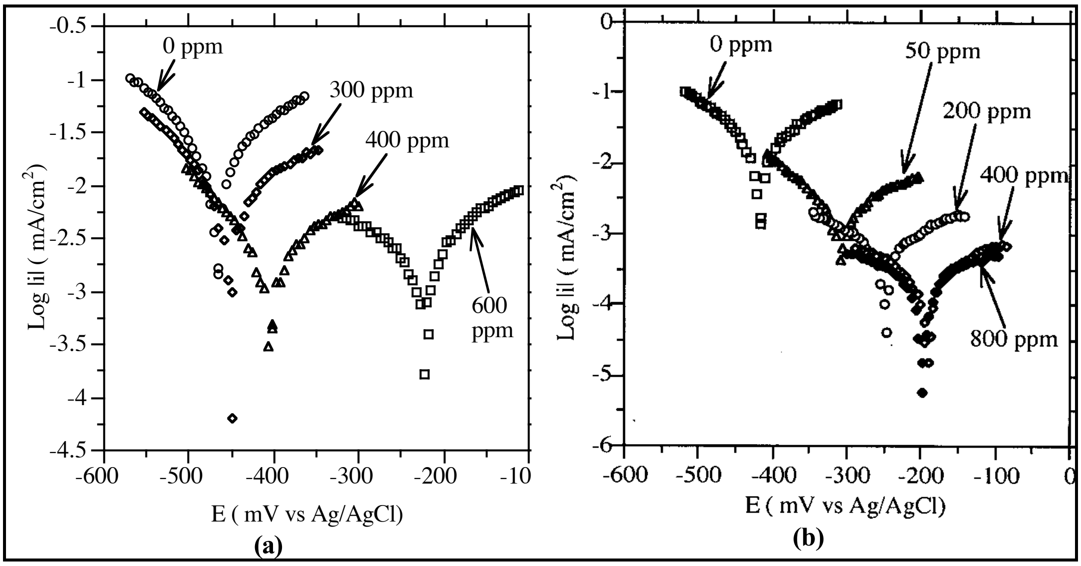


Figure 1: Polarization curves for mild steel in the inhibited fresh water medium at various concentrations of *Acacia* gum exudates at 30 °C for: (a) *Acacia drepanolobium*, (b) *Acacia senegal*

Table 1: **Electrochemical kinetic parameters from polarization curves for mild steel in fresh water without and with various concentrations of *Acacia drepanolobium* gum and *Acacia senegal* gum at 30 °C.**

Gum Species	Conc. (ppm)	E_{corr} (mV)	β_a (mV/dec)	β_c (mV/dec)	$i_{corr.}$ (μA/cm^2)	$R_{corr.}$×10^2/ (mm/y)	IE (%)
Acacia drepanolobium	0	-415	48	-50	6.30	7.17	-
	300	-453	39	-33	1.90	2.16	69.9
	400	-404	41	-47	0.70	0.80	88.8
	600	-224	38	-40	0.60	0.68	90.5
Acacia senegal	50	-306	68	-60	1.02	1.16	83.8
	100	-267	49	-51	0.36	0.41	94.3
	200	-246	34	-52	0.20	0.23	96.8
	400	-203	41	-49	0.07	0.08	98.9
	800	-197	36	-40	0.05	0.06	99.2

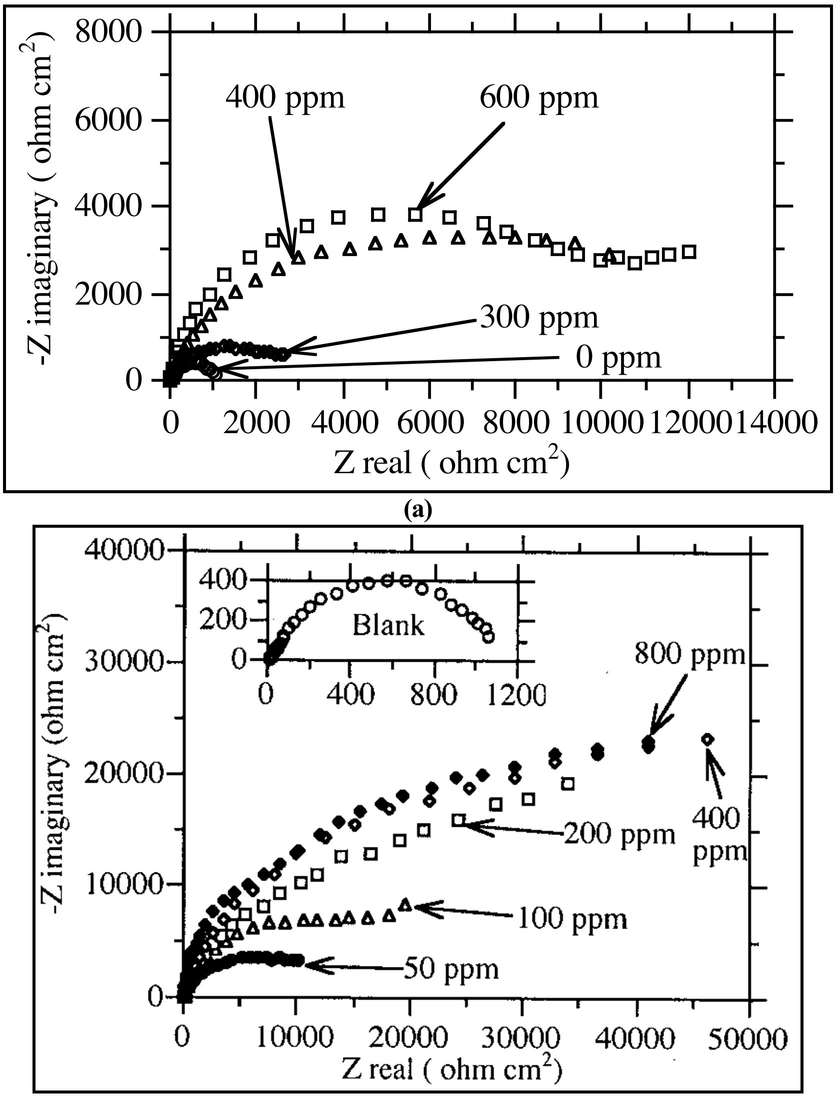
It is also seen that the degree of inhibition for a given concentration of inhibitor is considerably greater with *Acacia senegal* gum than with *Acacia drepanolobium* gum inhibitor. The latter requires 600 ppm to obtain 90.9% inhibition, while the former requires only 100 ppm to attain similar percentage inhibition. This implies that *Acacia drepanolobium* gum, to some degree has poor adsorption properties and thus a considerably higher concentration is required to attain an acceptable protection. It is also an indication that there are differences in terms of the active constituents present in these *Acacia* gum exudates that are active to the metal surface.

Electrochemical impedance measurements

The inhibition processes of the gum exudates were also studied by electrochemical impedance spectroscopy technique. Impedance measurements in the Nyquist format for mild steel in fresh water in the absence and presence of various concentrations of both *Acacia drepanolobium* and *Acacia senegal* gum exudates at 30 ^{o}C are presented in Figure 2. It is obvious from these plots that the impedance response of mild steel has significantly changed after addition of the gum exudates. It is quite clear that the shapes of the impedance plots for inhibited mild steels are not substantially different from those of uninhibited electrodes. The presence of the inhibitor just increases the impedance without changing other aspects of the behaviour. This capacitive loop is associated with the charge transfer at the metal/solution interface and consequently to corrosion rate (Silverman 1993). It means that the faradaic process which take place on the surface uncovered by the inhibitor film decreases with increasing inhibitor concentration. Thus, the inhibitors do not alter the electrochemical reactions responsible for corrosion but inhibit corrosion primarily through adsorption of inhibitor molecules on the metal surface (Khaled 2008). This increase in size of semicircle as the inhibitor concentration increases demonstrates the corrosion inhibition properties of these *Acacia* gum exudates. The size is relatively larger on *Acacia senegal* gum as compared to *Acacia drepanolobium* gum under the same concentration. This suggests that more inhibitor is adsorbed and more protective film is formed on the metal surface when *A. senegal* gum is applied giving rise to a slower corrosion process. This was also confirmed by the visual aspect of the electrode surface at the end of the test.

On the other hand, the size of the capacitive arc increases with increase in the inhibitor concentrations up to 400 ppm and remained almost unchanged with further increase of the concentration up to 600 ppm. This suggests that, 400 ppm could be considered as the optimum inhibitor concentration, forming the optimum protective film on the substrate surface against the charge transfer.

As can further be observed from these Figures, the impedance spectra obtained are depressed semicircles. According to Juttner 1990, this kind of behaviour is attributed to metal surface roughness. Therefore, a constant phase element (CPE) is used instead of the double layer capacitance (C_{dl}). The impedance spectra for different Nyquist plots were analysed by fitting the experimental data to a simple equivalent circuit model (a Randles type circuit with only one capacitive loop) using EQUIVCRT fitting program as described elsewhere (Philip *et al.* 2001). The C_{dl} were calculated using the CPE, solution resistance (R_s), charge transfer resistance and a dimensionless parameter, n, based on the given relation as described elsewhere (Brug *et al.* 1984). The calculated capacitances together with corresponding charge transfer resistances at different concentrations and calculated corrosion rates and inhibitor efficiencies are summarised in Table 2.



(a)

(b)

Figure 2: Impedance plots in the Nyquist format for mild steel in the inhibited fresh water medium at various concentrations of *Acacia* gum exudates at 30 °C for: (a) *Acacia drepanolobium*, (b) *Acacia senegal* and blank solution (0 ppp *Acacia senegal*) is also shown as an insert.

Table 2: Electrochemical kinetic parameters from impedance results (Nyquist format) for mild steel in fresh water without and with various concentrations of *Acacia drepanolobium* and *Acacia senegal* gum exudates at 30 °C.

Gum Species	$C_{INH.}$ (ppm)	E_{OCP} (mV vs Ag/AgCl)	R_{CT} (Ωcm^2)	C_{DL} ($\mu F\ cm^2$)	$i_{corr.}$ ($\mu A/cm^2$)	$R_{corr.}\times 10^2$/ (mm/y)	IE (%)
Acacia drepanolobium	0	-450	1,091	1,075	10.19	11.60	-
	300	-450	2,297	306	3.53	4.02	65.3
	400	-406	8,215	133	1.21	1.38	88.1
	600	-221	9,469	45	0.93	1.06	90.9
Acacia senegal	50	-311	9,565	137	1.51	1.72	85.2
	100	-295	17,235	105	0.66	0.75	95.5
	200	-250	22,364	51	0.42	0.48	95.9
	400	-204	37,986	46	0.27	0.31	97.3
	800	-200	38,105	44	0.23	0.26	97.8

It is seen from Table 2 that, as inhibitor concentration increases, the R_{CT} values increases while the double-layer capacitance, C_{dl}, decreases. This indicates that, gum exudates from *Acacia drepanolobium* and *Acacia senegal* are corrosion inhibitive in nature. The decrease in C_{dl} results from a decrease in local dielectric constant and/or an increase in the thickness of the double layer, suggesting that exudates molecules have been adsorbed at the interface (Lagrenee *et al.* 2002). Buchweishaija and Mhinzi 2008, Philip *et al.* 2001 reported similar behaviour of the gum exudates from *Acacia seyal* var *seyal* when subjected under similar conditions. These adsorbed molecules from gum exudate reduce the active sites on the metal surface undergoing corrosion. By comparing the values of inhibition efficiencies obtained from impedance method with that obtained from polarization method, it is observed that there is good agreement between them.

Effect of Temperature on the corrosion inhibition of mild steel

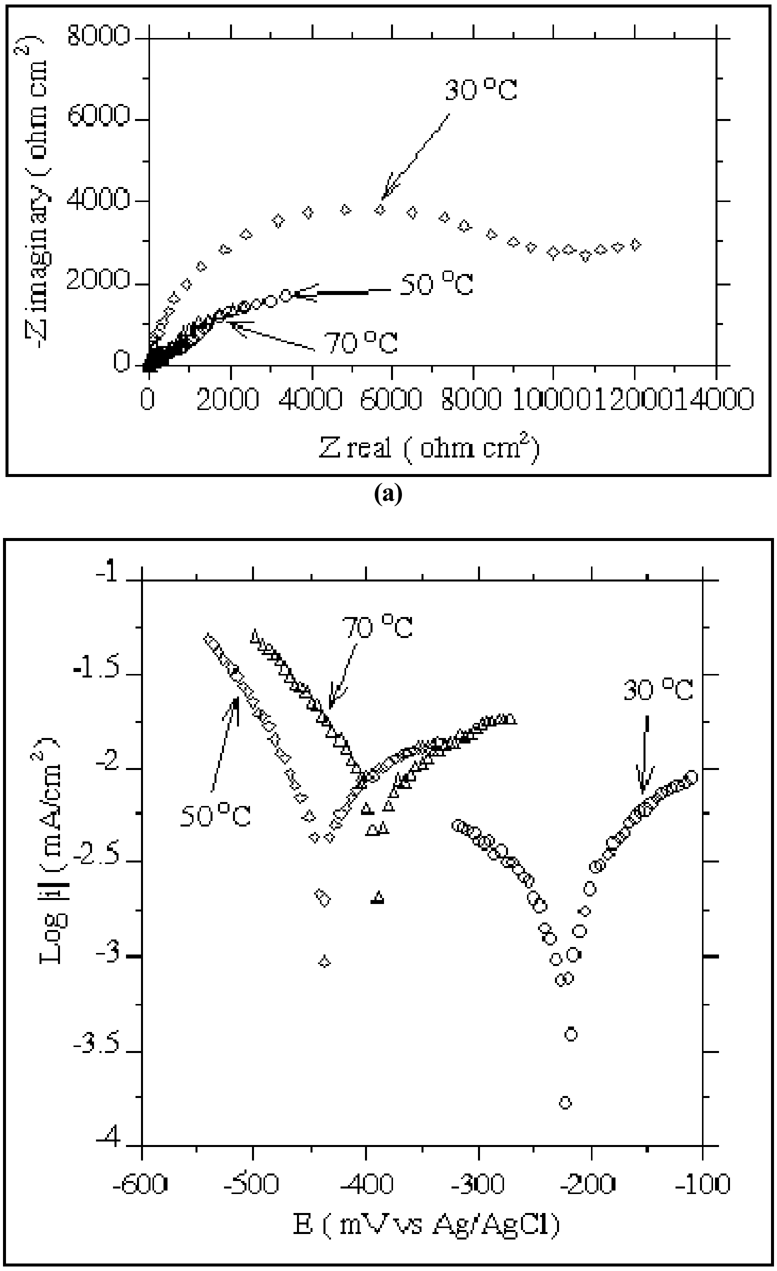
It is well known that temperature has a profound effect not only on corrosion rates, but also influences the inhibitor requirements because it affects inhibitor stability and solubility (Donham 1990, Mutasingwa 2004). According to Schaschl 1973, some organic inhibitors have lower and others have higher solubility at elevated temperatures. Inhibitor solubility in the medium is necessary for the inhibitor to reach the metal surface which may result in a positive effect of the inhibitor performance. On the other hand, temperature can affect the adsorption of the inhibitor to the metal surface. It has been reported that, if the adsorption is due to chemisorption an increase in temperature can result in an increase in the inhibition efficiency because of the activation energy of the chemical adsorption process. However, there will be no effect by temperature if physical adsorption takes place (Trabanelli 1989).

The effect of temperature on the corrosion rate of mild steel in fresh water containing the optimal concentration of the inhibitor was studied in the temperature range 30 to 70 °C using electrochemical techniques. Figure 3 shows the impedance and polarization results recorded on mild steel in fresh water in the presence of 600 ppm of *Acacia drepanolobium* gum at 30, 50 and 70 °C. Similarly, Figure 4 shows the impedance and polarization results recorded on mild steel in fresh water in the presence of 400 ppm of *Acacia senegal* gum at 30, 50 and 70 °C. For comparison purposes, the impedance and polarization measurement results for mild steel in fresh water without inhibitor at the same temperatures are also shown in Figure 5. It is seen from Figure 5 that there is a general increase in current density as well as corrosion current density as the temperature is increased from 30 to 70

°C. This is in accordance with the general rule guiding the rate of chemical reactions which states that chemical reaction increases with increasing temperature. Their corresponding kinetic parameters are summarized in Table 3. The displayed data (Table 3) show that increasing the temperature on the inhibited system increases the corrosion current density and consequently the corrosion rate. However, the inhibition power (calculated inhibition efficiencies) of the additives remains almost unchanged as the temperature of the system increases. This shows that *Acacia* gum exudates are not affected by temperature variations. The slight variation of Tafel slopes (β_a, β_c) (Table 3) indicate that the inhibiting action takes place by the simple blocking of the available sites (anodic and cathodic) on the metal surface.

This is in agreement with what was reported on gum exudate, *Acacia seyal* var *seyal* as reported by Buchweishaija and Mhinzi 2008. These results indicates that the inhibitors studied in this work remain quite effective within the temperature range studied (30 to 70 °C). However, the addition of 400 ppm of *Acacia senegal* gum at 30 °C caused the corrosion rate to drop from 0.0717 to 0.0008 mm/y which corresponds to 98.9% efficiency. The same concentration of inhibitor, when injected at 70 °C caused the corrosion rate to drop from 0.4856 to 0.0090 mm/y which corresponds to 98.1% efficiency. Similarly, addition of the optimal concentration of 600 ppm of *Acacia drepanolobium* gum at 30 °C caused the corrosion rate to drop from 0.0717 to 0.0068 mm/y corresponding to 90.5 % efficiency whereas the same concentration when injected at 70 °C caused the corrosion rate to drop from 0.4856 to 0.0501 mm/y corresponding to 89.7%.

It is also noted from Table 3 that, the values of the double layer capacity (C_{dl}) decreased significantly in the presence of the *Acacia* gum exudates. The decrease in C_{dl} values as the results of inhibitor addition regardless of the rise in temperature might have been caused by a complex formation at the metal surface which reduces the thickness of the double layer charges around the metal surface (Quraishi *et al.* 1999). Generally the impedance results (corrosion rate and inhibition efficiency) were in agreement with those obtained with potentiodynamic studies.



(a)

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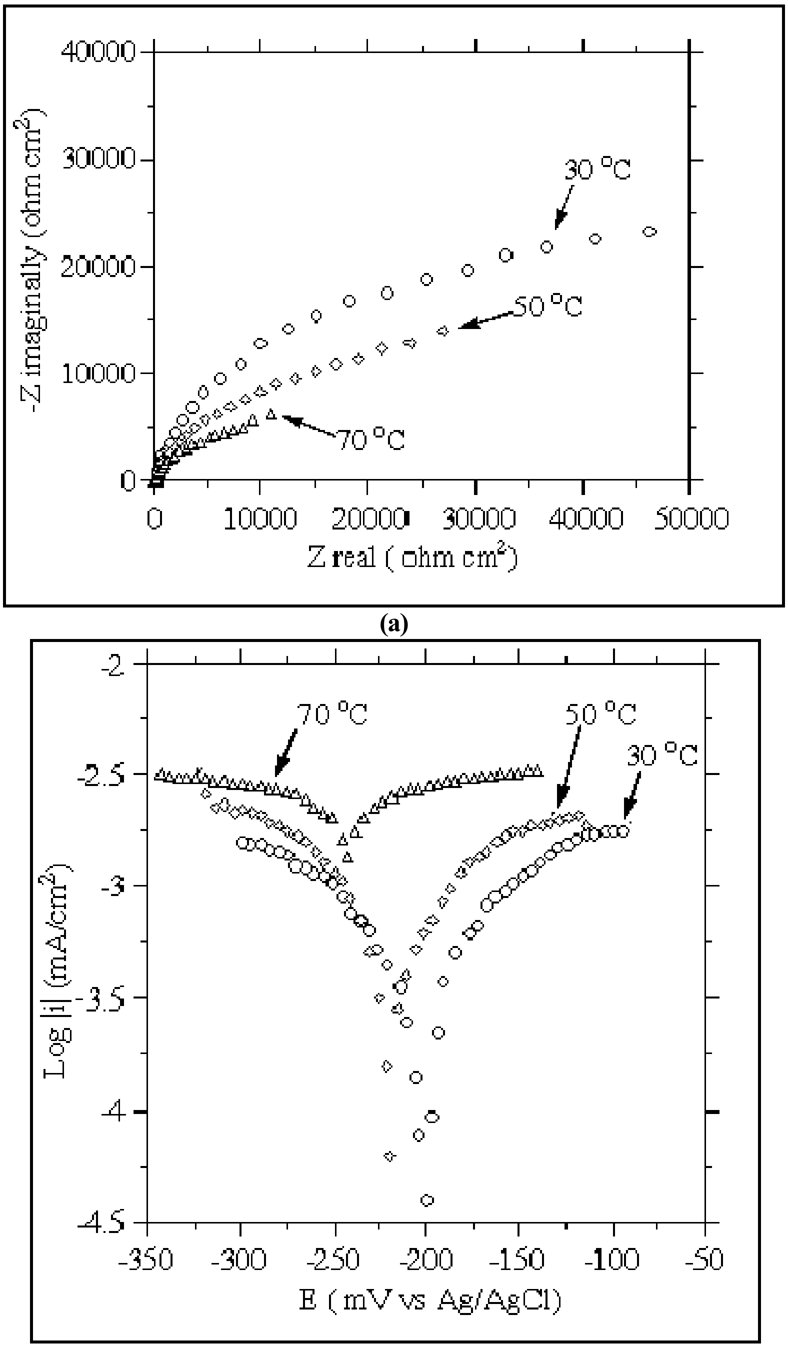
Figure 3: Electrochemical measurements for inhibited mild steel in fresh water with *Acacia drepanolobium* gum exudate at various temperatures: (a) Electrochemical Impedance results (Nyquist format) (b) Potendiodynamic polarization results.



(a)

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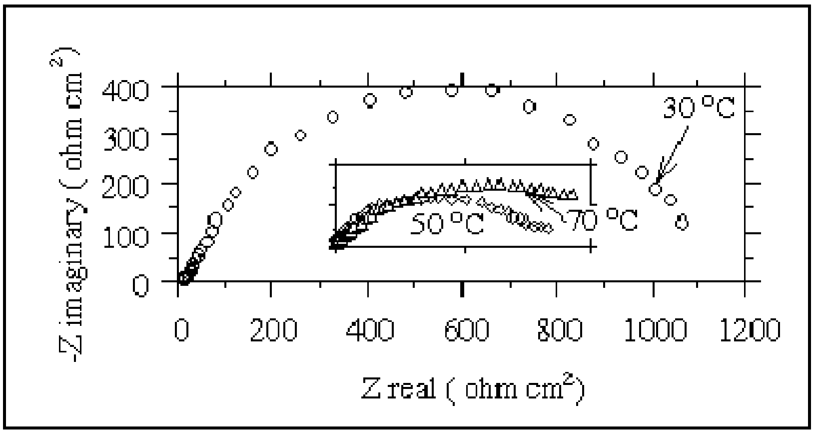
Figure 4: Electrochemical measurements for inhibited mild steel in fresh water with *Acacia senegal* gum exudate at various temperatures: (a) Electrochemical Impedance results (Nyquist format) (b) Potendiodynamic polarization results



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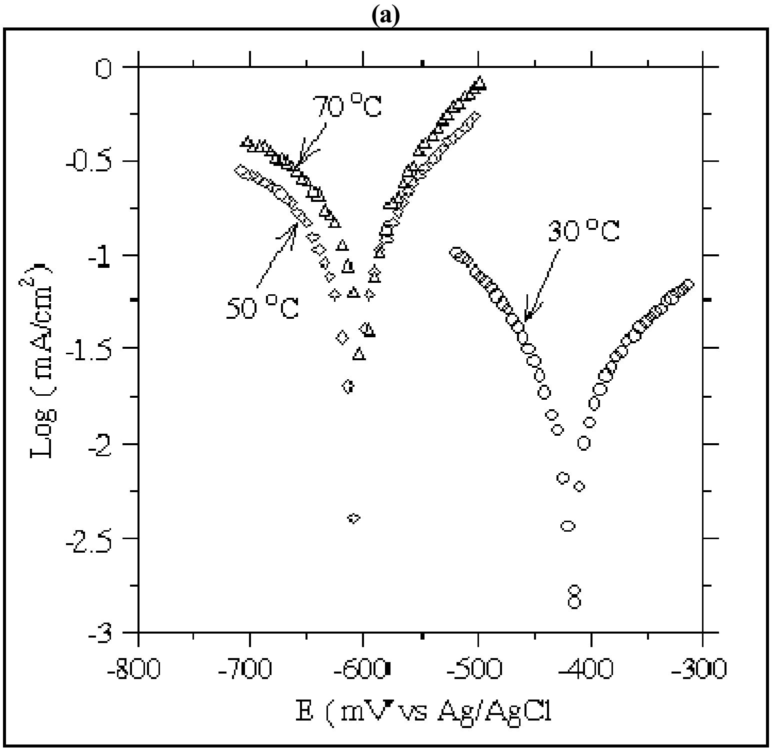

(b)

Figure 5: Electrochemical measurements for uninhibited mild steel in fresh water at various temperatures: (a) Electrochemical Impedance results (Nyquist format) (b) Potendiodynamic polarization results.

Table 3: **Electrochemical kinetic parameters of mild steel in fresh water without and with *Acacia* gum exudates at temperature range 30 – 70 °C:** [FW = Fresh Water, *Ad* = *Acacia drepanolobium*, As = *Acacia senegal*]

(a) Potentiodynamic Polarization results

Medium	**Temp °C**	E_{corr} **(mV)**	β_a **(mV/dec)**	β_c **(mV/dec)**	$i_{corr.}$ **(μA/cm^2)**	$R_{corr.}\times10^2$**/ (mm/y)**	**IE (%)**
FW	30	-415	48	-50	6.30	7.17	
	50	-614	33	-34	16.98	19.33	
	70	-602	46	-41	42.65	48.56	
FW + *Ad*	30	-224	38	-40	0.60	0.68	90.5
	50	-438	50	-42	2.50	2.85	85.2
	70	-393	68	-44	4.40	5.01	89.7
FW + *As*	30	-203	41	-49	0.07	0.08	98.9
	50	-220	49	-35	0.23	0.26	98.7
	70	-243	40	-42	0.79	0.90	98.1

(b) Electrochemical Impedance results.

Medium	**Temp °C**	E_{OCP} **(mV)**	R_{CT} **(Ωcm^2)**	C_{DL} **(μF cm^2)**	$i_{corr.}$ **(μA/cm^2)**	$R_{corr.}\times10^2$**/ (mm/y)**	**IE (%)**
FW	30	-415	1,091	1,075	10.19	11.60	
	50	-600	96	2,235	58.00	66.03	
	70	-604	80	4,998	123.00	140.04	
FW + *Ad*	30	-224	9,469	45	0.93	1.06	90.7
	50	-438	2,479	204	4.18	4.76	92.8
	70	-393	1,725	215	7.03	8.00	94.3
FW + *As*	30	-204	37,986	46	0.27	0.31	99.7
	50	-225	19,306	66	0.48	0.55	99.2
	70	-241	10,169	87	0.91	1.04	99.3

CONCLUSION

The present study shows that gum exudates, from *Acacia drepanolobium* and *Acacia senegal* trees, which are natural products, inhibit the corrosion of mild steel in fresh water with *A. senegal* gum exhibiting better inhibition characteristics compared to *Acacia drepanolobium*. The study also shows that the inhibition performances of the *Acacia* gum exudates are insignificantly affected by temperature rise. Potentiodynamic polarization studies reveals that the gum exudates are mixed type inhibitors of mild steel corrosion in fresh water with significant reduction of anodic current densities.

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