EFFECT OF ADENINE, GUANINE AND HYPOXANTHINE ON THE CORROSION OF MILD STEEL IN H_3PO_4

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Abstract

The corrosion inhibition potentials of adenine (AD), guanine (GU) and hypoxanthine (HYP) on the corrosion of mild steel in 0.1 M H_3PO_4 was investigated at 303 and 333 K using weight loss and potentiodynamic polarization techniques. The results obtained, indicated that AD, GU and HYP inhibited the corrosion of mild steel in phosphoric acid. Results obtained from potentiodynamic polarization studies showed that AD, GU and HYP acted as mixed corrosion inhibitors by retarding both the anodic and cathodic half reactions of the corrosion process. Iodide ions, I^- increased inhibition efficiencies of the purines. Adsorption of AD, GU and HYP on the mild steel surface was spontaneous and aligned with Langmuir isotherm model.

Keywords: *Adsorption, Corrosion inhibition, Potentiodynamic polarization.*

INTRODUCTION:

Phosphoric acid is a major chemical product most of which is produced from phosphate rock by the wet process. Phosphoric acid (H_3PO_4) has many important uses especially in the production of fertilizers and surface treatment of steel such as chemical and electrolytic polishing or etching, chemical coloring, removal of oxide film, phosphating, passivating, and surface cleaning (Sivaraju and Kannan, 2010; Oguzie *et al.*, 2012; Zarrouk *et al.*, 2013). Consequently, the steel is prone to corrosion with its attendant serious economic, health, safety, technological, and cultural consequences to our society (Gunter, 2009).

In order to reduce this menace due to corrosion of steel in industrial installations and equipment, several steps have been adopted for protecting mild steel against corrosion one of which is the use of corrosion inhibitors. Corrosion inhibitors are organic compounds that contain nitrogen, oxygen, sulphur, phosphorus, and multiple bonds or aromatic rings in their structures (Sinko, 2001; Bothi and Sethuraman, 2008; Shukla *et al.*, 2009; Jia-Jun *et al.*, 2010). The molecular electronic structures and electron densities around these functional groups are the key structural features that determine the effectiveness of inhibition (Bothi and Sethuraman, 2008; Umoren *et al.*, 2009).

Several organic inhibitors have been investigated including triazoles, benzotriazoles, organic dyes, amino acids, schiff bases and imidazoles (Gasparac *et al.*, 2000; Abdeli *et al.*, 2009; Hosseini *et al.*, 2009; Jia-Jun *et al.*, 2010; Matheswaran and Ramasamy, 2010; Li *et al.*, 2011).

The present study sets to investigate the potentials of adenine (AD), guanine (GU) and hypoxanthine (HYP) (Fig (1)), as inhibitors for the corrosion of mild steel in

0.1 M H_3PO_4 solutions using gravimetric, and potentiodynamic techniques.

MATERIALS AND METHODS

Weight Loss Measurements

Mild steel coupons of dimension 5.0 cm × 4.0 cm x 0.15 cm were cut and wet-polished with silicon carbide abrasive paper (from grade #400 to #1000), rinsed with distilled water, dried in acetone and warm air, and weighed before immersion in the test solutions. Tests were conducted under total immersion conditions in 250 mL of the aerated and unstirred test solutions. The mild steel coupons were retrieved from the test solutions at 24 h progressively for 120 h at 303 and 333 K respectively. The retrieved coupons were appropriately cleaned, dried, and reweighed. The weight loss was taken to be the difference between the weight of the coupons at a given time and its initial weight. All tests were run in duplicates and the data showed good reproducibility. Average values for each experiment was used in subsequent calculations (Oguzie *et al.,* 2010, 2012, 2012b; Momoh- Yahaya *et al.,* 2013).

From the initial and final weights of mild steel, the weight loss (g h^{-1}), corrosion rate ($gh^{-1}cm^2$), inhibition efficiency and the degree of surface coverage (θ) were calculated using equations 1 to 3 respectively (Wabanne and Okafor, 2001).

$$CR\left(gh^{-1}cm^{-2}\right)=\frac{\Delta W}{At} \qquad 1$$

$$IE_{\exp}=\left(1-\frac{W_1}{W_2}\right)x100 \qquad 2$$

$$\theta=1-\frac{W_1}{W_2} \qquad 3$$

where W_1 and W_2 are the weight losses (g) for mild steel in the presence and absence of the purine compound respectively, θ is the degree of surface coverage of the inhibitor, A is the area of the metal coupon (in cm^2), t is the period of immersion (in hours) and $\Delta W = W_2 - W_1$ is the weight loss of mild steel after time, t.

POTENTIODYNAMIC POLARIZATION MEASUREMENTS

Metal samples for electrochemical experiments were machined into test electrodes of dimension 2 × 2 cm^2 and sealed with epoxy resin in such a way that only one square surface area (1 cm^2) was left uncovered. The exposed surface was cleaned using the procedure described above. Electrochemical tests were conducted in a Model K0047 corrosion cell using a VERSASTAT 400 complete DC voltammetry and corrosion system, with V3 Studio software. A graphite rod was used as a counter electrode and a saturated calomel electrode (SCE) as a reference electrode. The latter was connected via a Luggin capillary. Measurements were performed in aerated and unstirred solutions at the end of 1 h of immersion at 30°C. Impedance measurements were made at corrosion potentials (E_{corr}) over a frequency range of 100 kHz– 10 mHz, with a signal amplitude perturbation of 5 mV. Potentiodynamic polarization studies were carried out in the potential range ±250 mV versus corrosion potential at a scan rate of 0.33 mV/s. Each test was run in duplicate (Oguzie *et al.,* 2010, 2012; Momoh- Yahaya *et al.,* 2013).

RESULTS AND DISCUSSION

Weight Loss Measurements

Corrosion Rate of Mild Steel as a Function of Time

The variation of corrosion rate of mild steel in 0.1 M H_3PO_4 in the absence and presence of 0.002, 0.006 and 0.010 M of AD, GU and HYP were studied at 303 and 333K respectively. The result Figs (2a-2c) reveals that though the corrosion rate of mild steel in 0.1 M H_3PO_4 increased with time, corrosion rate of mild steel decreased with increase in the concentration of the purines. This suggests that as the concentration of the purines increased, there was an increase in the number of adsorption of the purine molecules on the surface of the mild steel which constituted

a barrier for mass transfer and prevented further corrosion (Oguzie *et al.,* 2012b; Olasehinde *et al.,* 2012).

Corrosion Rate of Mild Steel as a Function of Concentration of AD, GU and HYP

Figs (3a- 3c) show the variation of corrosion rate of mild steel with different concentrations of AD, GU and HYP. From the plots, it is evident that corrosion rate of mild steel in H_3PO_4 decreased in the presence of AD, GU and HYP indicating that the purines retarded the corrosion of mild steel in the acid medium. It can also be deduced from the plots that although the weight loss of mild steel increased over time, the corrosion rates of mild steel in the presence of the inhibitors decreased with increasing inhibitor concentrations.

IE% as a Function of Concentration of AD, GU and HYP

The inhibition efficiency of mild steel exposed to various concentrations of AD, GU and HYP in 0.1 M H_3PO_4 is shown in Figs (4a, 4b and 4c). The inhibition efficiencies of the purines was found to increase with increasing concentration as a result of increase in the fraction of the mild steel surface covered by the adsorbed purine molecules. The maximum IE% of 53.49 %, 68.88% and 45.38%, at 303 K were obtained for AD, GU and HYP respectively, signifying that GU functioned as a better inhibitor for the corrosion of mild steel in H_3PO_4..

Potentiodynamic Polarisation Measurements

Typical potentiodynamic polarization curves for mild steel in 0.1 M H_3PO_4 in the absence and presence of 0.010 M AD, GU and HYP are shown in Fig (5) while the electrochemical parameters derived from the polarization curves and inhibition efficiency values are summarized on Table 1.

The values of the corrosion current density in the absence ($i_{corr,bl}$) and presence of inhibitor ($i_{corr,inh}$) were used to estimate the inhibition efficiency from polarization data (IE%) as follows:

$$IE_i\% = \left(1 - \frac{i_{corr,inh}}{i_{corr,bl}}\right) \times 100 \qquad 4$$

($i_{corr,bl}$) and ($i_{corr,inh}$) are the corrosion current densities in the absence and presence of AD, GU and HYP.

The displayed data show that the addition of the purines decreased the corrosion current density in the phosphoric acid media. The shapes of the polarization curves are obviously related to the activity of the reducing species present at the interface. Addition of the purines is seen to affect both the anodic and cathodic half reactions, shifting the corrosion potential (E_{corr}) slightly toward more positive (anodic) values and reducing the anodic and cathodic current densities and the corresponding corrosion current density (i_{corr}). This indicates that the purines not only inhibited the corrosion of mild steel in H_3PO_4 but also functioned as mixed-type inhibitors Fig (5).

Effect of 5 mM KI

Figs (6 and 7) show the effect of KI on the corrosion inhibitive properties of AD, GU and HYP.

It is generally accepted that the presence of halide ions in acidic media synergistically increases the inhibition efficiency of some organic compounds. It is thought that the halide ions are able to improve adsorption of the organic cations by forming intermediate bridges between the positively charged metal surface and the positive end of the organic inhibitor. Corrosion inhibition synergism thus results from increased surface coverage arising from ion-pair interactions between the organic cations and the anions (Oguzie *et al.,* 2007; Umoren *et al.,* 2010).

From Figs (6 and 7), it can be observed that the presence of iodide ions increased the inhibition efficiencies of AD, GU and HYP against the corrosion of mild steel in 0.1 M phosphoric acid.

Thermodynamics and Adsorption Considerations

From Fig (3a), it is evident that corrosion rates are higher for all the systems at 333 K. This is due to increased rate of mild steel dissolution and partial desorption of the inhibitors from the metal surface at increased temperature (Oguzie *et al.*, 2012b; Olasehinde *et al.,* 2012). Hence, inhibition efficiencies of the purines decreased at 333 K (Figure 4), a trend that is normally attributed to physical rather than chemical adsorption of the inhibitor molecule on the corroding metal surface.

Arrhenius equation was used in calculating the activation energies (E_a),

$$log(CR_2/CR_1) = {E_a}/{2.303R} \qquad 5$$

where CR_1 and CR_2 are the corrosion rates at temperatures T_1 and T_2, respectively.

An estimate of the heats of adsorption (Q_{ads}) was obtained from the trend of surface coverage with temperature as follows:

$$Q_{ads} = 2.303R\left\{log\left[\frac{\theta_1}{1-\theta_2}\right] - log\left[\frac{\theta_1}{1-\theta_1}\right]\right\} \qquad 6$$

where θ_1 and θ_2 are values of the degree of surface coverage at temperatures T_1 and T_2, respectively and R is the gas constant.

The calculated values of E_a and Q_{ads} are presented on Table 3. From Table 3, calculated values of E_a ranged from 33.00 to 34.52, 33.40 to 36.68 and 31.69 to 32.72 kJ mol^{-1} respectively for AD, GU and HYP. These values are larger than the value for the blank which is 26.97 kJ mol^{-1}, confirming a physical or coulombic type of adsorption where the decrease in inhibition efficiency with rise in temperature is observed. Negative values of Q_{ads} presented on Table 3 signify that the adsorption of the purines onto the metal surface is an exothermic process (Popova *et al.*, 2003; Eddy *et al.*, 2010; Olasehinde *et al.*, 2012; Momoh-Yahaya *et al.*, 2014).

Adsorption isotherms provide information about the interaction among adsorbed molecules themselves as well as their interactions with the metal surface.

The surface coverage data obtained from IE% values of weight loss studies gave best fit with the adsorptions model of Langmuir (equation 7) taking into cognizance that the plots gave linear slopes with regression coefficients, R^2, values greater than 0.8 (Saratha, 2009).

$$\frac{C}{\theta} = \frac{1}{K_{ads}} + C \qquad 7$$

The divergence of the slope from unity and the non-zero intercepts on the y-axes (Figure 8) is attributable to interactions between adsorbate molecules on the metal surface as well as changes in the adsorption heat with increasing surface coverage (Oguzie, 2008; Yan *et al.*, 2008; Olasehinde *et al.*, 2012; Momoh- Yahaya *et al*., 2013).

Thermodynamic parameters associated with the adsorption process were deduced using the relationship between the adsorption equilibrium constant (k_{ads}) and the standard free energy of adsorption, $\Delta G_{ads}°$ (equation 8):

$$logK_{ads} = -1.744 - \frac{\Delta G^{\circ}_{ads}}{2.303RT} \qquad 8$$

Negative ΔG_{ads} values indicate spontaneity of the adsorption process (Eddy *et al.*, 2010; Momoh-Yahaya *et al.*, 2014).

Generally, ΔG_{ads} values with magnitude much less than 40 kJ mol^{-1} have typically been correlated with the electrostatic interactions between organic molecules and charged metal surface (physisorption), whilst those of magnitude in the order of 40 kJ mol^{-1} and above are associated with charge sharing or transfer from the organic molecules to the metal surface (chemisorption) (Eddy *et al.*, 2010; Olasehinde *et al.*, 2012). Calculated values of ΔG_{ads} were negative and ranged from -10.67 to – 19.64 kJ mol^{-1} indicating that the adsorption of the purines is spontaneous and that the mechanism of adsorption is physisorption.

Conclusions

1. AD, GU and HYP inhibited the corrosion of mild steel in 0.1 M H_3PO_4.
2. Inhibition efficiency values increased with increase in concentrations of AD, GU and HYP but decreased with rise in temperature.It was however observed that inhibition efficiencies decreased with inmersion time.
3. Inhibition efficiencies of AD, GU and HYP were enhanced in the presence of KI.
4. Polarization measurements showed that the adsorbed purines inhibited the corrosion process via mixed inhibition mechanism, affecting both the anodic dissolution reaction of the mild steel and the cathodic hydrogen evolution reaction.
5. From the values of activation energies (E_a), heats of adsorption (Q_{ads}) and standard free energies of adsorption (ΔG_{ads}) obtained from the study, it is evident that AD and GU adsorbed on the mild steel surface through physical interactions, and that the adsorption process is exothermic and spontaneous. Adsorption characteristics of the purines gave best fit with Langmuir isotherm.

Acknowledgements

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Table 1: Polarization data for mild steel in 0.1 M H_3PO_4 in the absence and presence of 0.01M adenine (AD), guanine (GU), hypoxanthine (HYP) at 303 K.

System	E_{corr} (mV vs SCE)	i_{corr} (μA cm^{-2})	IE%
H_3PO_4 (Blank)	-568.31	229.27	
AD	-550	124.38	45.75
GU	-560.1	73.80	67.81
HYP	-528.38	134.53	41.32

Table 2: Weight loss and polarisation results on the effect of 5 mM potassium iodide (KI) on inhibition efficiency for mild steel corrosion in 0.1 M H_3PO_4 in the absence and presence of 0.01 M adenine (AD), guanine (GU) and hypoxanthine (HYP) at 303 K.

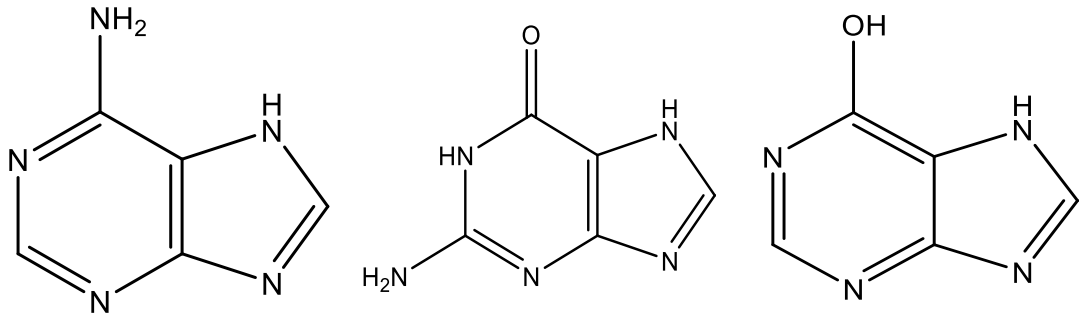
SYSTEM	INHIBITION EFFICIENCY (IE%)	
	WEIGHT LOSS	POLARISATION
KI	50.13	48.21
AD	53.49	45.75
AD + KI	57.32	56.70
GU	68.88	67.81
GU + KI	78.01	74.05
HYP	45.38	41.32
HYP + KI	60.38	63.50

Table 3: Calculated values of activation energies (E_a) and heats of adsorption (Q_{ads}) for the corrosion of mild steel in 0.1 M H_3PO_4 in the absence and presence of various concentrations of adenine (AD), guanine (GU) and hypoxanthine (HYP).

Inhibitor	Concentration (mol dm^{-3})	Activation Energy, E_a(kJ mol^{-1})	Heat of Adsorption, Q_{ads} (kJ mol^{-1})
	Blank	26.97	
AD	0.002	34.21	-22.29
	0.004	33.00	-16.11
	0.006	34.30	-16.86
	0.008	34.52	-16.82
	0.010	34.15	-15.26
GU	0.002	36.29	-23.76
	0.004	35.86	-18.40
	0.006	36.68	-18.95
	0.008	35.93	-16.04
	0.010	33.40	-17.97
HYP	0.002	31.69	-16.49
	0.004	32.72	-17.86
	0.006	31.80	-13.32
	0.008	31.94	-13.20
	0.010	32.06	-12.62

Table 4: Calculated thermodynamic parameters from Langmuir adsorption isotherms.

Temperature	Inhibitor	Intercept	Slope	k_{ads}	R^2	ΔG_{ads} (kJ mol^{-1})
303 K	AD	0.0018	1.694	555.56	0.9956	-19.16
	GU	0.0017	1.309	588.24	0.9925	-19.64
	HYP	0.0019	2.034	526.32	0.9972	-19.03
333 K	AD	0.0043	2.082	232.56	0.9971	-11.92
	GU	0.0050	1.251	200.00	0.9269	-10.67
	HYP	0.0034	2.455	294.12	0.8238	-13.87



(a) Adenine (b) Guanine (c)Hypoxanthine

Fig (1): Chemical structures of (a) Adenine (b) Guanine and (c) Hypoxanthine.

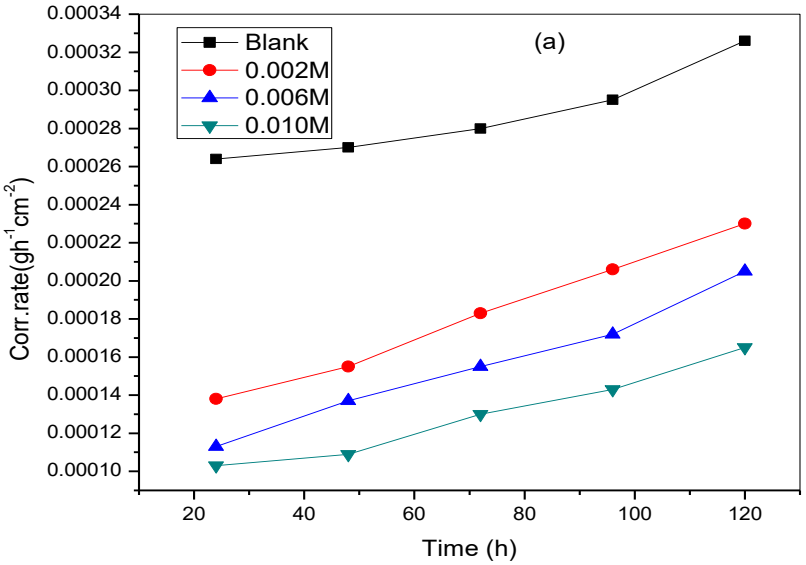


Fig (2a): Variation of corrosion rate ($gh^{-1}cm^{-2}$) of mild steel with time (h) in 0.1 M H_3PO_4 in the absence and presence of different concentrations of Adenine (AD) at 303 K.

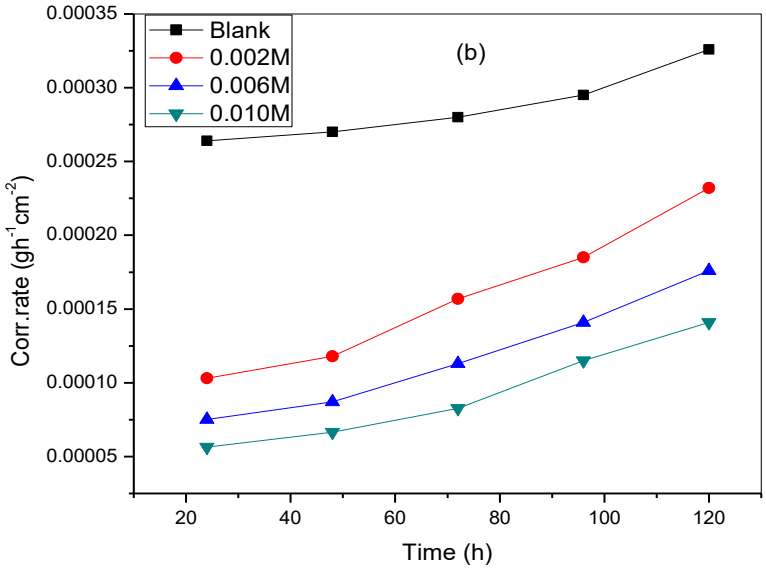


Fig (2b): Variation of corrosion rate ($gh^{-1}cm^{-2}$) of mild steel with time (h) in 0.1 M H_3PO_4 in the absence and presence of different concentrations of Guanine (GU) at 303 K.

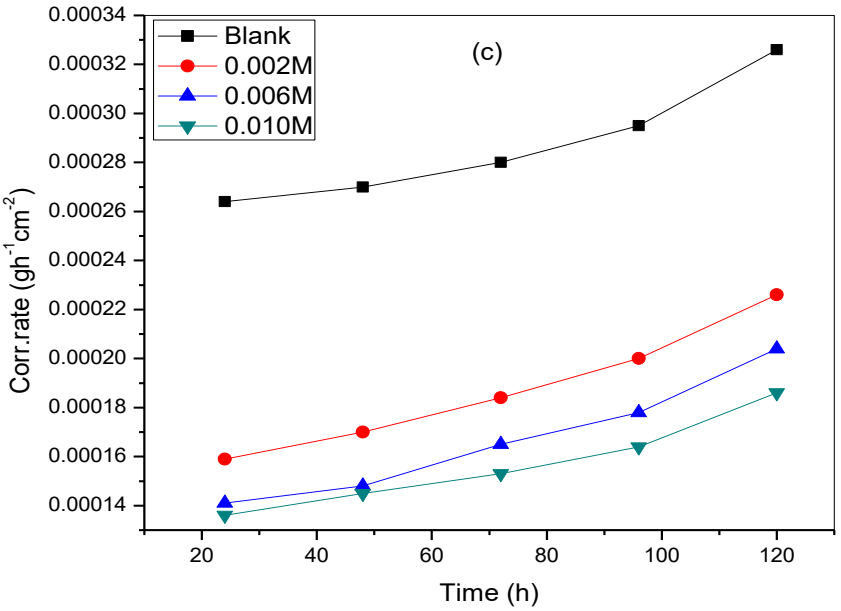


Fig (2c): Variation of corrosion rate (gh^{-1}cm^{-2}) of mild steel with time (h) in 0.1 M H_3PO_4 in the absence and presence of different concentrations of Hypoxanthine (HYP) at 303 K.

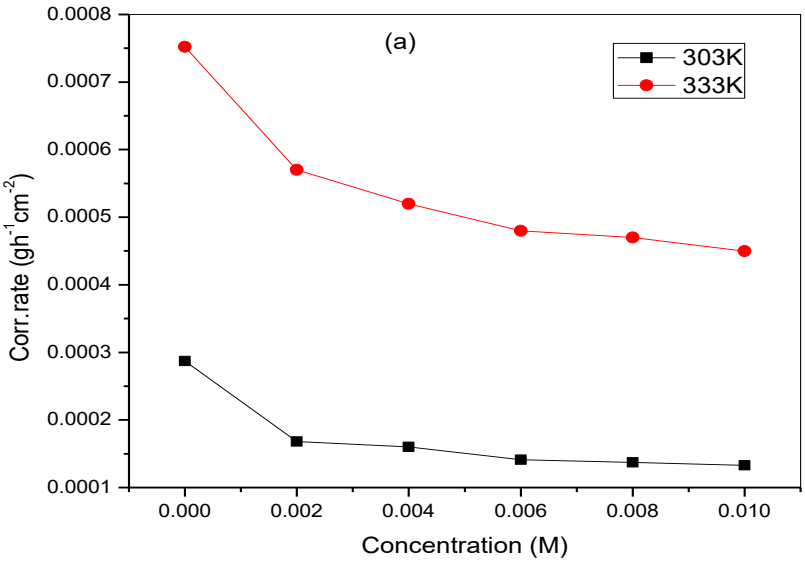


Fig (3a): Variation of corrosion rate (gh^{-1}cm^{-2}) with different concentrations (M) of adenine (AD) for corrosion of mild steel in 0.1 M H_3PO_4 at 303 and 333 K.

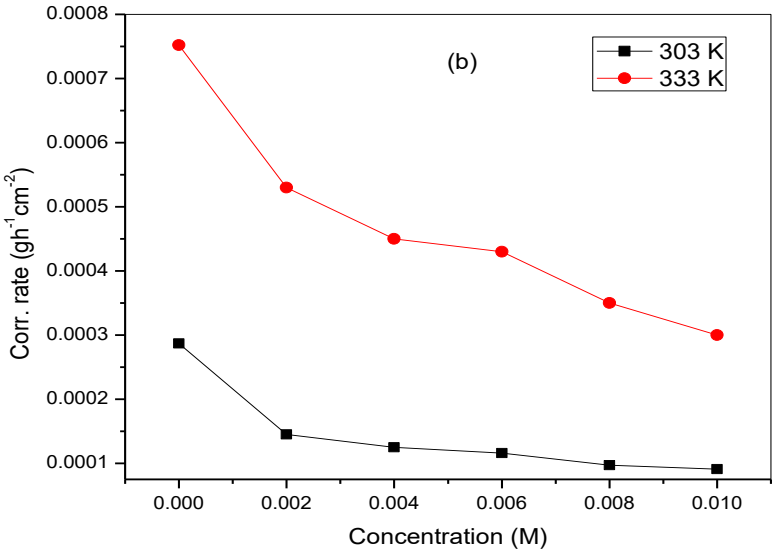


Fig (3b): Variation of corrosion rate ($gh^{-1}cm^{-2}$) with different concentrations (M) of guanine (GU) for corrosion of mild steel in 0.1 M H_3PO_4 at 303 and 333 K.

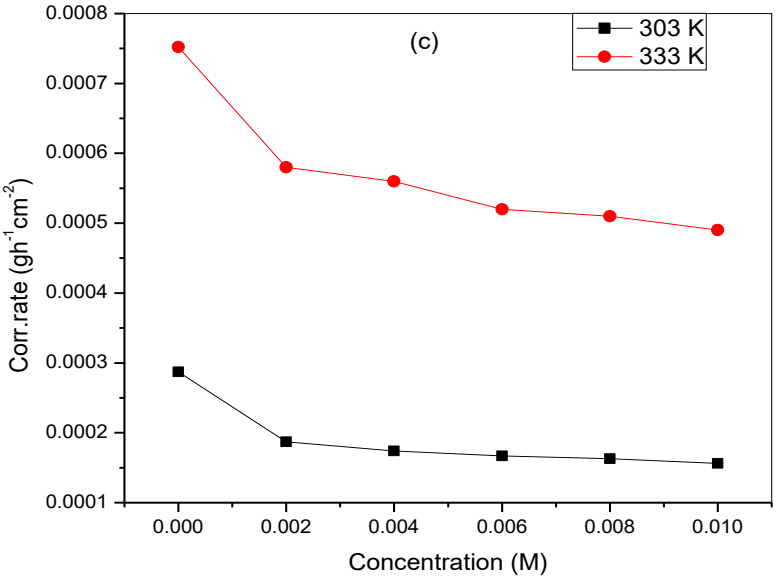


Fig (3c): Variation of corrosion rate ($gh^{-1}cm^{-2}$) with different concentrations (M) hypoxanthine (HYP) for corrosion of mild steel in 0.1 M H_3PO_4 at 303 and 333 K.

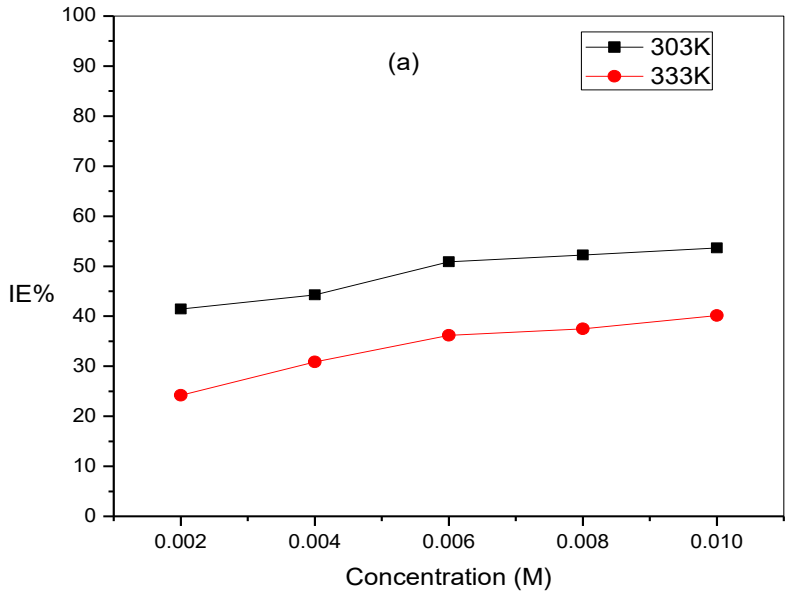


Fig (4a): Variation of inhibition efficiency (%IE) with different concentrations (M) of adenine for corrosion of mild steel in 0.1 M H_3PO_4 at 303 and 333 K.

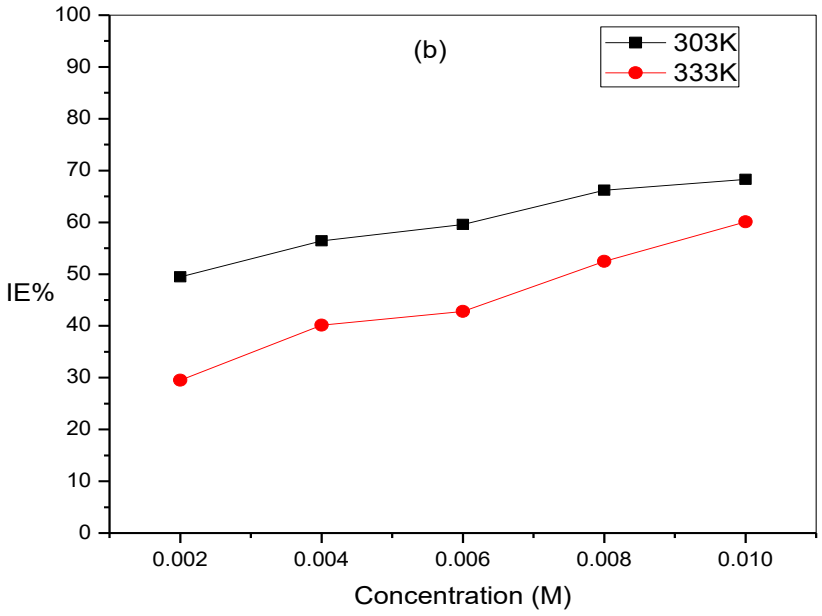


Fig (4b): Variation of inhibition efficiency (%IE) with different concentrations (M) of guanine (GU) for corrosion of mild steel in 0.1 M H_3PO_4 at 303 and 333 K.

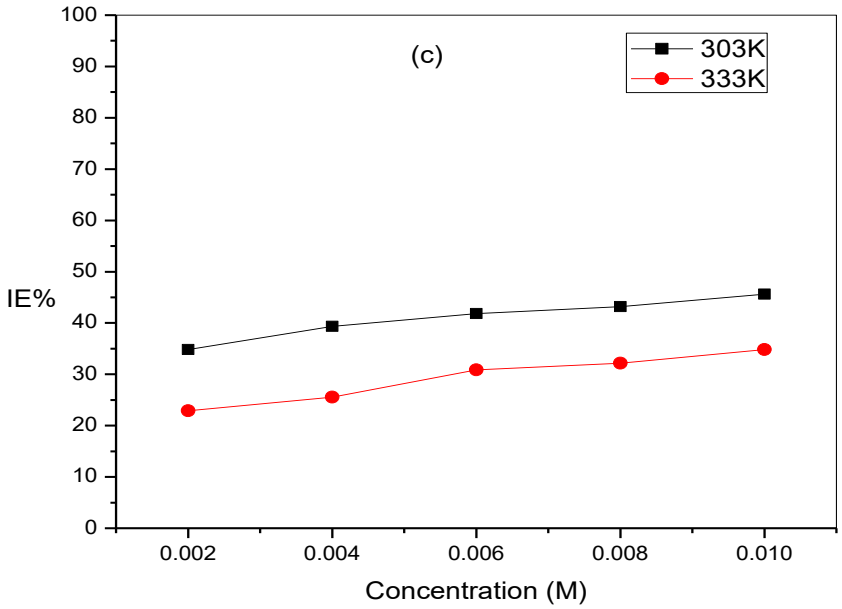


Fig (4c): Variation of inhibition efficiency (%IE) with different concentrations (M) of hypoxanthine (HYP) for corrosion of mild steel in 0.1 M H_3PO_4 at 303 and 333 K.

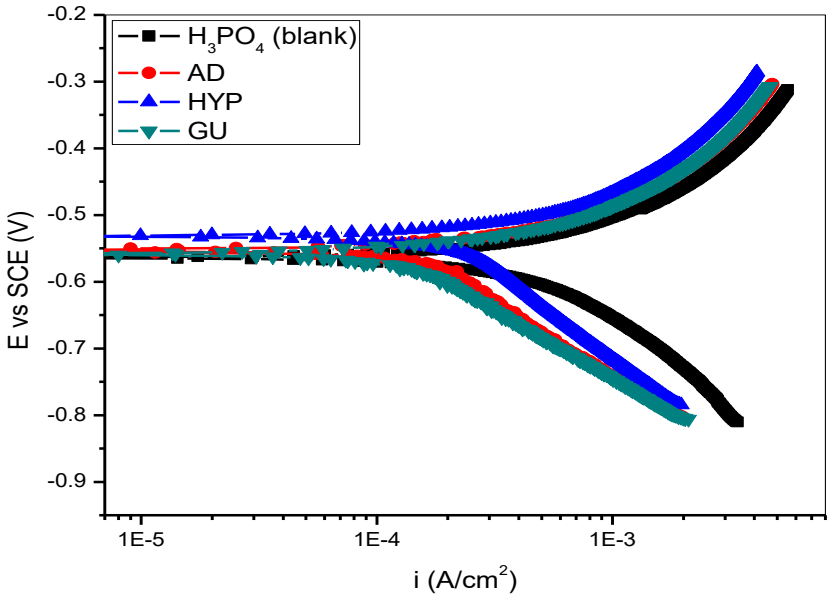


Fig (5): Polarisation curves of mild steel in 0.1 M H_3PO_4 in the absence and presence of 0.01 M adenine (AD), guanine (GU) and hypoxanthine (HYP) at 303 K.

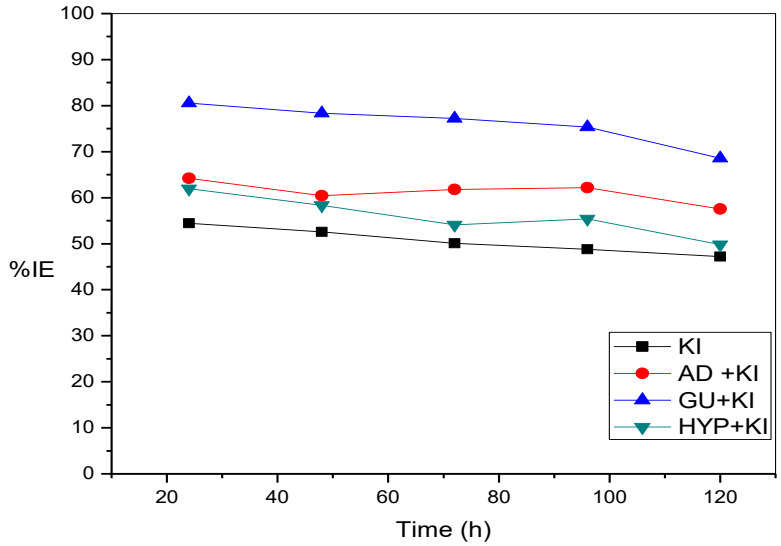


Fig (6): Effect of 5 mM KI on the inhibition efficiency of 0.01 M AD, GU and HYP as a function of time (h) from weight loss measurement at 303 K.

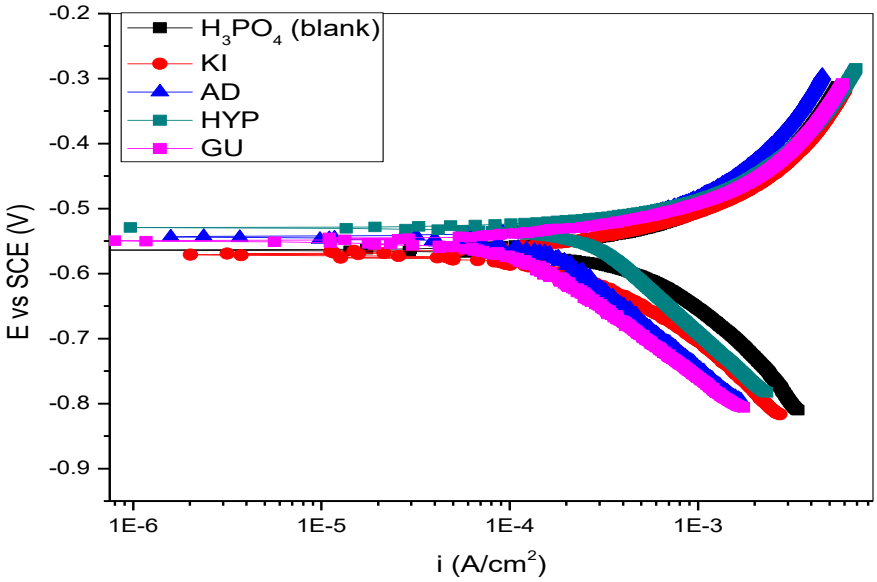


Fig (7): Polarization curves showing the effect of addition of 5 mM potassium iodide (KI) on inhibition efficiency in the absence and presence of 0.01 M adenine (AD), guanine (GU) and hypoxanthine (HYP) during the corrosion of mild steel in 0.1 M H_3PO_4 at 303 K.

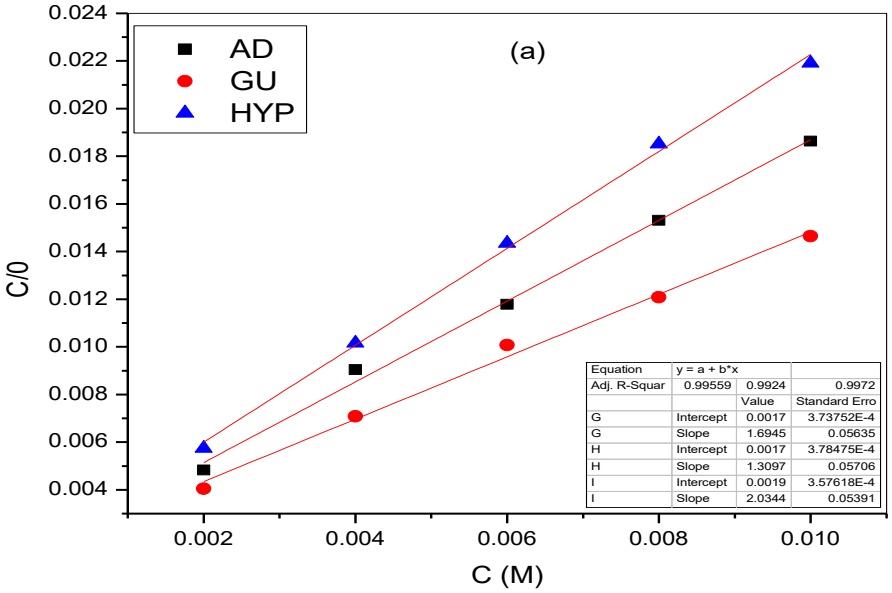


Fig 8(a): Langmuir isotherm for the adsorption of AD, GU and HYP onto mild steel in 0.01 M H_3PO_4 solution at 303 K.

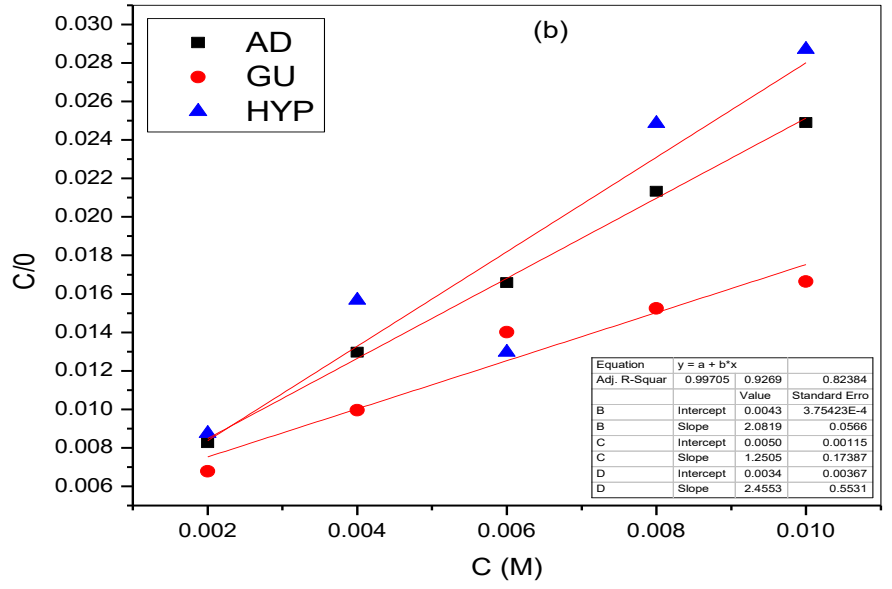


Fig 8(b): Langmuir isotherm for the adsorption of AD, GU and HYP onto mild steel in 0.01 M H_3PO_4 solution at 333 K.

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