

## ORIGINAL ARTICLE

### Inhibition of the Corrosion of Mild Steel in Acidic Medium by Penicillin V Potassium

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#### ABSTRACT

The inhibition of the corrosion of mild steel by penicillin V potassium has been studied. Penicillin V potassium is a good inhibitor for the corrosion of mild steel in H<sub>2</sub>SO<sub>4</sub>. The inhibition efficiency of the inhibitor decreases with increase in temperature and with the period of immersion. However, increase in the concentration of the inhibitor resulted in increase in the inhibition efficiency of penicillin V potassium. Adsorption characteristics of the inhibitor has also been studied and it is found that penicillin V potassium inhibit the corrosion of mild steel by being adsorbed on the surface of mild steel by a physical adsorption mechanism. The adsorption of the inhibitor was also found to be spontaneous and consistent with the assumptions of Langmuir and Frumkin adsorption isotherms.

**Key word:** Corrosion of mild steel, inhibition, penicillin V potassium.

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#### Introduction

Most corrosion inhibitors protect the corrosion of metals when they are adsorbed on the surface of the metal (Abdallah, 2004a,b, 2002; Agrawal *et al.*, 2003; Eddy and Odoemelam, 2008a,b,c; Eddy and Ekop, 2008a; Eddy *et al.*, 2008a,b). The adsorption and inhibitive properties of some corrosion inhibitors containing hetero atoms in their long carbon chain/aromatic structure have also been studied (Abiola *et al.*, 2007,2004; Ashassi-Sorkhabi *et al.*, 2006). Studies have also been conducted on the adsorptive and inhibitive properties of some natural products (Ashassi\_Sorkhabi and Ghalebsaz-Jeddi, 2005). In most of these studies, these properties are found to be strongly influenced by the chemical structure of the compound, the corrosive medium, temperature, concentration of the inhibitor, period of contact, etc.(Arora *et al.*, 2007; Babi-Samordzia *et al.*, 2005). Adsorption characteristics of an inhibitor can be studied by the used of adsorption isotherms and the application of the theory of thermodynamics (Eddy and Odoemelam, 2008a).

Recently, studies on the use of drugs as corrosion inhibitors have been intensified (Awad, 2006; Bendahou *et al.*, 2006; Bouyanzer and Hammouti, 2004). According to Eddy and Odoemelam (2008b), the used of drugs for the inhibition of the corrosion of metals has some advantages over the use of some organic/inorganic inhibitors because of their eco-environmental nature. Therefore, the present study is aimed at investigating inhibitive and adsorptive properties of penicillin V potassium for the corrosion of mild steel in acidic medium. Penicillin V potassium is an antibiotic that is used for the treatment of some infection. This compound can be synthesized from plant and it does not contain heavy metals or other toxic substance.

#### Experimental

Materials used for the study were mild steel sheets of composition (wt %) Mn (0.6), P(0.36), C(0.15) and Si(0.03) and dimension, 5x4x0.11cm. Each coupon was degreased by washing in ethanol, dried in acetone and preserved in a desiccator. The inhibitor was supplied by VERBATA pharmaceutical store, Ikot Ekpene, Nigeria.

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All reagents used for the study were analar grade. Double distilled water was used for the preparation of all solutions. Concentrations of  $H_2SO_4$  used for the study were 1.0, 1.5, 2.0 and 2.5M while the concentrations of the inhibitor were  $3 \times 10^{-4}$ ,  $6 \times 10^{-4}$ ,  $9 \times 10^{-4}$ ,  $12 \times 10^{-4}$  and  $15 \times 10^{-4}$ M. These were respectively dissolved in 2.5M  $H_2SO_4$ .

#### Gasometric method

Hydrogen evolution measurements were carried out at 303 and 333K as described in literature (Eddy and Ebenso, 2008; Eddy and Ekop, 2008). From the volume of hydrogen evolved per minutes, inhibition efficiency (h), and degree of surface coverage (q) were calculated using Equation 1 and 2 respectively.

$$\%I = \left\{ 1 - \frac{V'_{Ht}}{V^0_{Ht}} \right\} \times 100 \quad (1)$$

$$\theta = \%I/100 \quad (2)$$

where  $V'_{Ht}$  is the volume of hydrogen evolved at time t for inhibited solution and  $V^0_{Ht}$  is the volume of hydrogen evolved at time t for uninhibited solution.

#### Thermometric method

This was also carried out as reported elsewhere (Eddy and Ebenso, 2008). From the rise in temperature of the system per minutes, the reaction number (RN) and inhibition efficiency were calculated using equation 3 and 4 respectively:

$$RN (\text{°C minutes}) = \frac{T_m - T_j}{t} \quad (3)$$

$$\%I = \frac{RN_b - RN_w}{RN_b} \times 100 \quad (4)$$

### Results and discussion

Fig. 1 shows the variation of the volume of hydrogen gas evolved with time during the corrosion of mild steel in various concentrations of  $H_2SO_4$ . The Figure revealed that the rate of evolution of hydrogen increases as the concentration of the acid increases indicating that the rate of corrosion of mild steel increases with increase in the concentration of  $H_2SO_4$ . Values of corrosion rate of mild steel in various concentrations of  $H_2SO_4$  are recorded in Table 1. The results obtained also indicate that the rate of corrosion of mild steel increases with increase in the concentration of  $H_2SO_4$ .

Figs. 2 and 3 show the variation of the volume of hydrogen gas evolved with time for the corrosion of mild steel in 2.5M  $H_2SO_4$  containing various concentrations of penicillin V potassium at 303 and 333K respectively. These Figures revealed that the rate of corrosion of mild steel increase as the temperature increases but decreases as the concentration of penicillin V potassium increases indicating that penicillin G inhibited the corrosion of mild steel in  $H_2SO_4$ .

Table 1 shows values of corrosion rate of mild steel in  $H_2SO_4$  containing various concentrations of penicillin V potassium. Values of inhibition efficiency of penicillin V potassium are also recorded in the Table. From the results, it was seen that the rate of corrosion of mild steel in  $H_2SO_4$  decreases as the concentration of penicillin V potassium increases but increases with increase in temperature. These findings confirmed that penicillin V potassium inhibited the corrosion of mild steel and that the inhibition efficiency of penicillin V potassium for mild steel corrosion decreases as the concentration of penicillin V potassium increases but decreases as the temperature increases. Fig. 4 shows the variation of inhibition efficiency of penicillin V potassium with concentration at 303 and 333K. The Figure clearly shown that the inhibition efficiency of penicillin V potassium for mild steel corrosion decreases with increase in temperature but increases as the concentration of penicillin V potassium increases supporting the mechanism of physical adsorption. For a physical adsorption mechanism, the inhibition efficiency of the inhibitor is expected to decrease with increase in temperature as observed in this work (Ebenso, 2004, 2003; Ebenso *et al.*, 2005).

The Arrhenius equation (Equation 5) was used to investigate the effect of temperature (T) on the rate of corrosion (CR) of mild steel in the presence and absence of penicillin V potassium (Emregul *et al.*, 2004a,b; 2005a,b).

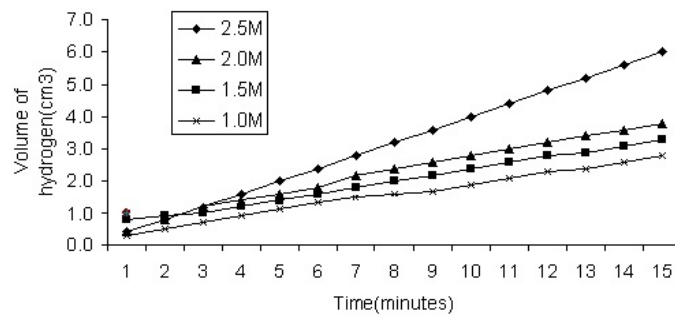
$$CR = A \exp(-E_a/RT) \quad (5)$$

**Table 1:** Values of corrosion rate(cm<sup>3</sup>/minute) and reaction number (°C/minute) for the corrosion of mild steel in H<sub>2</sub>SO<sub>4</sub>

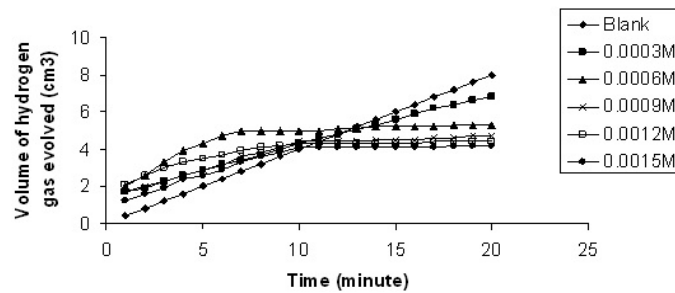
Con. of H <sub>2</sub> SO <sub>4</sub> (M)	CR(303K)	RN(303K)
1.0	0.175	0.0167
1.5	0.180	0.0200
2.0	0.220	0.0267
2.5	0.380	0.0500

**Table 2:** Values of corrosion rate (CR) and reaction number (RN) for the corrosion of mild steel in H<sub>2</sub>SO<sub>4</sub> containing various concentrations of penicillin V potassium

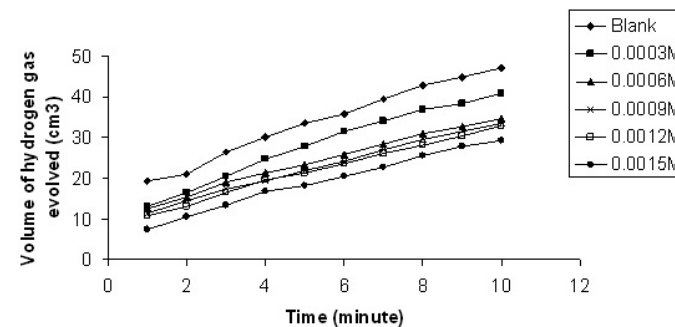
Con. of penicillin V potassium(M)	CR (333K)	CR (303K)	RN (303K)	Gasometric		Thermometric
				%I(303K)	%I(333K)	%I (303K)
3 x 10 <sup>-4</sup>	0.2625	2.3063	0.0370	30.00	16.89	26.00
6 x 10 <sup>-4</sup>	0.2000	2.3063	0.0313	46.67	16.89	37.32
9 x 10 <sup>-4</sup>	0.1813	2.2250	0.0255	51.67	19.82	49.08
12 x 10 <sup>-4</sup>	0.1750	2.0875	0.0255	53.33	24.70	49.08
15 x 10 <sup>-4</sup>	0.1375	2.0063	0.0238	63.33	27.70	52.33



**Fig. 1:** Variation of hydrogen gas evolved with tim for the corrosion inhibition of mild steel at various concentrations of methocarbamol at 303K.



**Fig. 2:** Variation of the volume of hydrogen gas evolved with tim for the inhibition for the corrosion of mild steel by various concentrations of penicillin V potassium at 303K.



**Fig. 3:** Variation of the volume of hydrogen gas evolved with tim for the inhibition for the corrosion of mild steel by various concentrations of penicillin V potassium at 333K.

where E<sub>a</sub> is the activation energy and R is the gas constant. Taking the logarithm of both sides of Equation 5, Equation 6 is obtained,

$$\log CR = \log A - E_a / 2.303RT \tag{6}$$

Assuming that the corrosion rates of mild steel at 303K ( $T_1$ ) and 333K ( $T_2$ ) are  $CR_1$  and  $CR_2$ , then Equation 6 is transformed to Equation 7,

$$\log(CR_2/CR_1) = E_a/2.303R(1/T_1 - 1/T_2) \tag{7}$$

Values of  $E_a$  calculated from Equation 7 were recorded in Table 3. These values ranged from 60.8445 to 75.0473KJ/mol. The values are greater than the value (-55.667J/mol) obtained from the blank indicating that the penicillin V potassium retarded the corrosion of mild steel in  $H_2SO_4$ . The values are also consistent with data expected for the mechanism of physical adsorption ( $E_a < 80KJ/mol$ ).

Values of the heat of adsorption of penicillin V potassium on mild steel surface were calculated using Equation 8 (Eddy and Ebenso, 2008).

$$Q_{ads} = 2.303R[\log(\theta_2/1-\theta_2) - \log(\theta_1/1-\theta_1)] \times (T_2T_1)/(T_2 - T_1) \tag{8}$$

where R is the gas constant,  $q_1$  and  $q_2$  are the degree of surface coverage at temperatures,  $T_1$  and  $T_2$  respectively. Values of  $Q_{ads}$  calculated from Equation 8 ranged from -15.6509 to -31.5849KJ/mol indicating that the adsorption of penicillin V potassium on mild steel surface is exothermic.

Data obtained for degree of surface coverage were used to fit curves for different adsorption isotherms including Langmuir, Frumkin, Freundlich, Bockris-Swinkcl, Temkin and Florry - Huggins adsorption isotherms. The results revealed that the isotherms that best described the adsorption characteristics of penicillin V potassium on mild steel surface are Langmuir and Frumkin adsorption isotherms.

Starting from Langmuir adsorption isotherm, the degree of surface coverage ( $\theta$ ) and the concentration of the inhibitor in the bulk electrolyte are related according to Equation 9 (Eddy and Odoemelum, 2008a,b; Oguzie, 2007,2006a,b).

$$C/\theta = 1/k+C \tag{9}$$

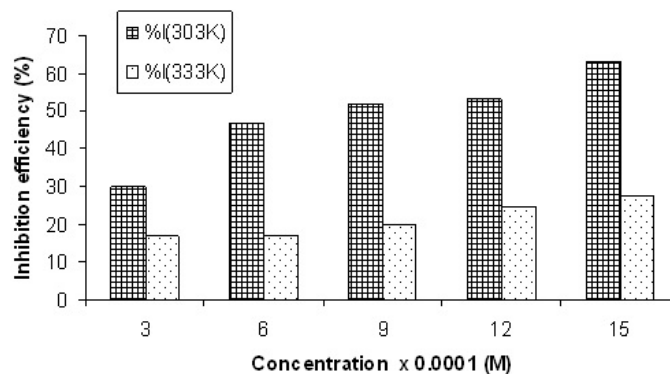
where k is the equilibrium constant of adsorption of penicillin V potassium on mild steel surface. Taking the logarithm of both sides of Equation 9, Equation 10 is obtained (Sheatty *et al.*, 2006; Shockry *et al.*, 2006; Rajendran *et al.*, 2005)

$$\log(C/\theta) = \log C - \log K \tag{10}$$

From Equation 10, a plot of  $\log(C/\theta)$  is expected to produce a straight line provided the assumptions of Langmuir isotherm are valid. Fig. 5 shows Langmuir isotherm for the adsorption of penicillin V potassium on the surface of mild steel.

**Table 3:** Some thermodynamic parameters for the adsorption of penicillin V potassium on mild steel surface

Concentration. of penicillin V potassium (M)	$E_a$ (KJ/mol)	$Q_{ads}$ (KJ/mol)
$3 \times 10^{-4}$	60.8445	-15.6509
$6 \times 10^{-4}$	68.4582	-30.6254
$9 \times 10^{-4}$	70.2095	-30.7169
$12 \times 10^{-4}$	69.4060	-26.1792
$15 \times 10^{-4}$	75.0473	-31.5849



**Fig. 4:** Variation of the inhibition efficiency of penicillin V potassium with concentration at 303 and 333K.

The expression for Frumkin isotherm is as given by Equation 11(Eddy and Ebenso, 2008),

$$\theta/1-\theta = B.C.e^{2aq} \tag{11}$$

From the logarithm of Equation 11, Equation 12 is obtained:

$$\log[(\theta/(1-\theta))*[C]]=\log K+2a \theta \tag{12}$$

where q is the of surface coverage, C is the concentration of the adsorbate, K is the adsorption-desorption equilibrium constant and a is an interaction parameter. From Equation 12, a plot of  $\log[(\theta/(1-\theta))*[C]]$  versus q should produce a straight line if Frumkin isotherm is applicable. Fig. 6 shows Frumkin isotherm for the adsorption of penicillin V potassium on the surface of mild steel. Values of Frumkin adsorption parameters are recorded in Table 4. From the results, it was seen that values of the adsorption parameters were positive indicating the attractive behaviour of the inhibitor (Eddy and Ebenso, 2008).

The free energy of adsorption of penicillin V potassium is related to the equilibrium constant of adsorption according to Equation 13 (Okafor *et al.*, 2008,2007a,b;Rajappa and Vekateshal, 2002).

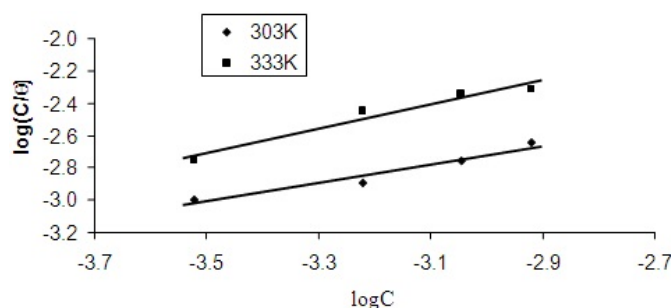
$$\Delta G_{ads} = -2.303RT\log(55.5K) \tag{13}$$

where R is the gas constant and T is the temperature. Values of K obtained from Langmuir and Frumkin adsorption isotherms were used to calculate values of  $\Delta G_{ads}$  according to Equation 13. These values are recorded in Table 4. The results indicated that the adsorption of penicillin V potassium is spontaneous ( $\Delta G_{ads}$  is negative) and suggest the applicability of the mechanism of physical adsorption ( $\Delta G_{ads} < 40\text{KJ/mol}$ ) (Eddy and Odeomalm, 2008a,b; Eddy and Ekop, 2008).

The chemical structure of penicillin V potassium is as shown by Fig. 7. From the structure, it can be stated that the inhibition ability of this compound is largely contributed by the presence of nitrogen and sulphur in their aromatic/cyclic structure. These atoms tend to enhance the electron donation ability of the inhibitor. The compound also contained amino and carbonyl functional groups. We therefore proposed that the mechanism of adsorption of penicillin V potassium (hence its inhibition efficiency) is due to the donation of electron by the molecule of penicillin V potassium to a vacant d-orbital of Fe in mild steel (Fig. 8). The formation of Fe-penicillin V potassium complex thereby stabilized the mild steel and prevent it against further corrosion attack.

**Table 4:** Values of Langmuir, Frumkin and Freundlich adsorption parameters

	Temperature (K)	Slope	logK	R <sup>2</sup>	$\Delta G_{ads}$ (KJ/mol)
Langmuir	303	0.5748	0.9978	0.9554	-15.9085
	333	0.7477	0.8810	0.9557	-16.7389
	Temperature (K)	$\alpha$	logK	R <sup>2</sup>	$\Delta G_{ads}$ (KJ/mol)
Frumkin	303	2.0121	3.7917	2.2141	-22.9650
	333	2.2984	0.9869	0.8818	-25.7762



**Fig. 5:** Langmuir isotherm for the adsorption of penicillin V potassium on mild steel surface.

**Conclusion**

- From the results and findings of the study, the following conclusions were drawn,
- Penicillin V potassium is a good inhibitor for the corrosion of mild steel in H<sub>2</sub>SO<sub>4</sub>.
  - The adsorption of the inhibitor on mild steel surface is exothermic, spontaneous and is consistent with the mechanism of physical adsorption. Langmuir and Frumkin isotherms best described the adsorption characteristics of the inhibitor.

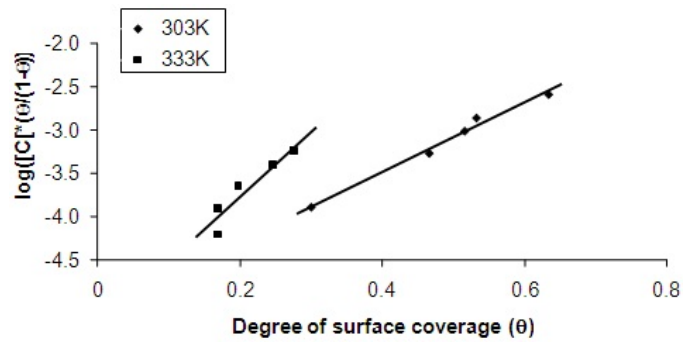


Fig. 6: Frumkin isotherm for the adsorption of penicillin V potassium on mild steel surface.

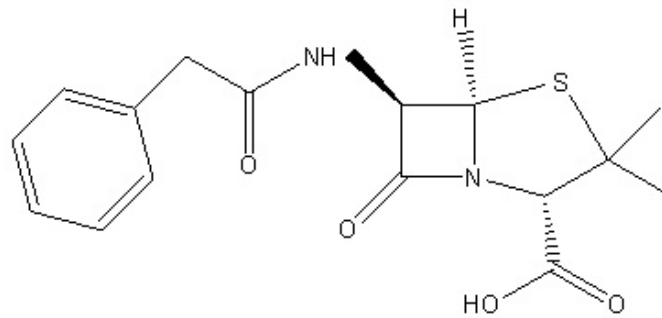


Fig. 7: Chemical structure of penicillin V potassium.

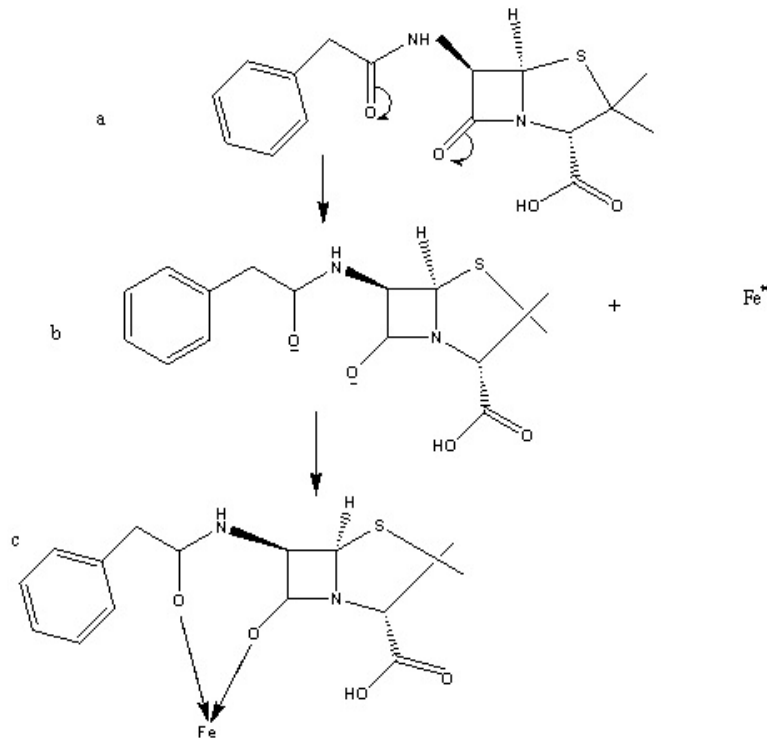


Fig. 8: Mechanism of adsorption of penicillin V potassium on Fe on mild steel surface.

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