



## Original Research Article

### Kinetic Studies of Corrosion of Mild Steel in Acidic Medium using *Grewa venusta* Leaves Extract as Inhibitor

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#### ABSTRACT

*Corrosion leads to huge loss of properties especially in the industries. The use of corrosion inhibitor is one way of preventing corrosion from occurring. The aim of this work was to investigate the kinetic and adsorption study of corrosion of mild steel in acidic medium using *Grewa venusta* leaves extract as an inhibitor. The experimental data from the work of Suleiman *et al.* were fitted into the developed kinetics corrosion process models with the aid of an in-built Software Excel version 7. The data were also fitted to the adsorption of corrosion process model to determine the isotherm model, which best fits the experimental data. The kinetics rate model had a negative fractional order (-0.5958) with a rate constant of 135.6394 h/m. The kinetic rate model validated the experimental data up to 94 percent confidence level. The first-order activity rate model developed fitted the experimental data up to 90 percent confidence level. The first-order activity rate constant obtained in the absence of inhibitor was 7.7459 h while it varied between 1.615 and 2.028 h in the presence of an inhibitor. Kinetic – Thermodynamic model with a constant of 0.7691 best fitted the experimental data. It is clear that the developed kinetics rate model and the first order activity model can be used to navigate the design of corrosion facilities of mild steel in 0.5 M tetraoxosulphate (VI) acid ( $H_2SO_4$ ) using *Grewa venusta* leaves extract as an inhibitor.*

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## 1. INTRODUCTION

Mild steel is one of the most widely used engineering materials as a result of its low cost, availability, good mechanical strength. Easy fabricability, formability and weldability (Tripathi *et al.*, 2012; Suleiman *et al.*, 2013; Odoni *et al.*, 2017; Akintola *et al.*, 2019; Edoziuno *et al.*, 2020). Corrosion is the gradual destruction

of materials usually metals, by chemical reaction with its environment which means electrochemical oxidation of metals in reaction with an oxidation such as oxygen (Salami et al., 2012). Corrosion leads to loss of engineering materials. One way of mitigating corrosion is through the use of inhibitors which are chemicals that reduce the rates of corrosion without significant reactions with the materials (Eddy *et al.*, 2009; Loto *et al.*, 2017). In most cases, inhibitors contain heteroatom such as oxygen, nitrogen, sulphur and multiple bonds in their structure through which they are adsorbed on the surface of the materials (Yahazri et al., 2011; Singh et al., 2012).

Many researchers have worked on corrosion inhibitors (Umoru *et al.*, 2006; Oguzie, 2008; Olusegun and James, 2010; Salami *et al.*, 2012; Suleiman *et al.*, 2013; Karthikaiselvi and Subhashini 2014; Salah, 2016; Omoruwou *et al.*, 2017; Omoruwou *et al.*, 2017; Salami and Umar, 2018; Odejebi and Akinbulumo, 2019; Edoziuno *et al.*, 2020). Olusegun and James (2010) investigated the effects of aloe vera extract on corrosion and kinetics of the process of zinc corrosion in HCl solution. The investigation revealed that inhibition efficiency increased with increasing concentrations of the extract but decreased with increasing temperature. Salami et al. (2012) researched on corrosion inhibition of mild steel in sulphuric acid using *Musa sapientum* peels extract as inhibitor. The research showed that the concentration of *Musa sapientum* peels was directly proportional to inhibition efficiency but inversely proportional to the rates of corrosion. Suleiman *et al.* (2013) developed a mathematical model for the prediction of corrosion rate behavior for mild steel in 0.5 M sulphuric acid using *Grewa venusta* leaves extract as inhibitor. The work revealed a good agreement between the predicted and experimental corrosion rate. However, the kinetics and adsorption studies of the corrosion rate were neglected. Karthikaiselvi and Subhashini (2014) studied adsorption properties and inhibition of mild steel corrosion in HCl medium by water soluble composite. The study showed that the adsorption obeyed Langmuir and El – Awady isotherms. Salah (2016) modeled corrosion kinetics of mild steel in hydrochloric acid in the presence and absence of a drug inhibitor. The model revealed that the corrosion rate profile was of second order kinetics with respect to corrosion activity. Omoruwou *et al.*, (2017) worked on statistical analysis of corrosion inhibition of water hyacinth on mild steel in an acidic medium. The obtained statistical model predicted the experimental data up to 96.95 % confidence level. Salami *et al.*, (2018) studied the thermodynamics of corrosion inhibition of *Hunteria umbellata* seed husk extracts on mild steel. The study showed that the activation energies on the presence of inhibitor ranged between 55.49 and 76.87 kJ/mol for hydrochloric acid extract and varied between 83.78 and 100.11 kJ/mol for sulphuric acid extract in sulphuric acid medium.

Odejebi and Akinbulumo (2019) carried out modeling and optimization of the inhibition efficiency of *Euphorbia heterophylla* extracts-based corrosion inhibitor of mild steel corrosion in HCl media using a response surface methodology. The developed model predicted the experimental data for corrosion rate and inhibition efficiency up to 98.80 and 93.10 % respectively. Edoziuno et al. (2020) worked on optimization and development of predictive models for corrosion inhibition of mild steel in sulphuric acid by methyl – 5 – benzoyl – 2 – benzimidazole carbamate (mebendazole). The optimum inhibition efficiency obtained in the work was 88.40 % with desirability value of 0.914.

It evident from myriad of available literature that the kinetics and adsorption studies of corrosion of mild steel in acidic medium using *Grewa venusta* leaves extract as inhibitor has not been carried out. Therefore, the aim of this work is to carry out the kinetics and adsorption studies of corrosion of mild steel in acidic medium using *Grewa venusta* leaves extract as inhibitor. This work will help chemists, chemical engineers and corrosion engineers in the design of the process facilities which justifies the work.

## 2. METHODOLOGY

The kinetics of corrosion of mild steel in acidic medium using *Grewa venusta* leaves extract as inhibitor was represented by Equation (1) which was linearised to obtain Equation (2).

$$r = kc^n \quad (1)$$

$$\ln r = \ln k + n \ln c \quad (2)$$

where  $r$  is the corrosion rate of mild steel,  $k$  the rate constant for the process, whose unit depends on the order of reaction,  $n$  and  $c$  the inhibitor concentration.

The rate of change in activity for the first order reaction shown in Equation (3) was integrated using the condition that at  $t = 0$ ,  $a = 1$  to obtain Equation (4). Equation (4) was put in Equation (5) (Levenspiel, 1999) and upon linearising, yielded Equation (6).

$$-\frac{da}{dt} = k_1 a \quad (3)$$

$$a = \exp(-k_1 t) \quad (4)$$

$$a = r/r_0 \quad (5)$$

$$\ln r = \ln r_0 - k_1 t \quad (6)$$

where  $a$  is the activity as a function of time ( $t$ ),  $r$  is the corrosion rate at time  $t \neq 0$ ,  $r_0$  is the corrosion rate at time  $t = 0$  and  $k_1$  is the first order activity rate constant.

The rate of change in activity for the second order reaction presented in Equation (7) was also integrated using the stated conditions to arrive at Equation (8) which was then put in Equation (5) and linearised to obtain Equation (9).

$$-\frac{da}{dt} = k_2 a^2 \quad (7)$$

$$a = 1/(1 + k_2 t) \quad (8)$$

$$\frac{1}{r} = \frac{1}{r_0} + \left(\frac{k_2}{r_0}\right)t \quad (9)$$

where  $k_2$  is the second order activity rate constant.

The experimental data for optimum exposure of 3 h obtained from the work of Suleiman et al. (2013) shown in Table 1 were fitted to the kinetics of corrosion process model of Equation (2) with the aid of an in-built Software in Excel version 7.0 to arrive at the rate model parameters. A graph of  $\ln r$  against  $\ln c$  was plotted from which slope ( $n$ ) was determined and  $k$  was evaluated from the intercept. The experimental data in Table 1 were also applied to models of Equations (6) and (9) using the in-built Software Excel version 7.0 to obtain the first- and second-order activity rate parameters. A graph of  $\ln r$  against  $t$  was plotted from which the ( $k_1$ ) was obtained using the slope. A plot of  $(1/r)$  against  $t$  was also plotted from where  $k_2$  was evaluated using the intercept.

The data in Table 1 were also fitted in the adsorption of corrosion process models of linear isotherm, 2-p Freundlich isotherm, Langmuir isotherm and kinetic-thermodynamic model presented in Equations (10), (11), (12) and (13) respectively, with the help of the in-built Software Excel version 7.0 to determine the isotherm model which best fitted the experiment data.

$$\theta = k_H c \quad (10)$$

$$\ln \theta = \ln k_F + (1/N) \ln c \quad (11)$$

$$\frac{c}{\theta} = \frac{1}{K_L} + c \quad (12)$$

$$\ln\left(\frac{\theta}{1-\theta}\right) = \ln K_T + y \ln c \quad (13)$$

where  $\theta$  is the fraction covered by the adsorbed molecules ( $0 < \theta < 1$ ), known as surface coverage,  $k_H$  is the linear adsorption equilibrium constant,  $K_F$  is a Freundlich adsorption equilibrium constant,  $N$  is the Freundlich isotherm constant,  $K_L$  is the Langmuir adsorption equilibrium constant,  $K_T$  is the kinetic-thermodynamic adsorption constant and  $y$  is the kinetic-thermodynamic isotherm constant.

Table 1: Numerical values of corrosion rates and surface coverage at difference inhibitor concentration and exposure time at temperature 30 °C (Suleiman et al., 2013)

Inhibitor concentration (v/v)	Exposure time (h)	Corrosion rate (m/h)	Surface coverage
0	2.25	4542.206	-
	3.0	4375.31	-
	3.50	3832.459	-
0.02	2.25	2043.158	0.5502
	3.0	1423.068	0.6748
	3.50	928.469	0.7577
0.04	2.25	1498.55	0.6701
	3.0	872.251	0.8006
	3.50	728.194	0.81
0.06	2.25	1271.045	0.7201
	3.0	756.302	0.8271
	3.50	599.947	0.8435
0.08	2.25	1048.81	0.7691
	3.0	607.853	0.8611
	3.50	580.622	0.8485

### 3. RESULTS AND DISCUSSION

Table 2 presents the experimental and predicted values for corrosion rate as well as percentage difference between the experimental and predicted values at optimum exposure time of 3 h. The numerical values of corrosion rates at different exposure time and inhibitor concentration are shown in Table 3 while Tables 4 and 5 depict the parameters for the second and first order activity rate. In Table 2, as the experimental rates decreased, the predicted rates also followed the same trend. This showed that the experimental rates were directly proportional to the predicted rates. However, there was no definite trend in the relationship between the rates and the percent error. The percentage error between the experimental and predicted rates varied between - 354 and 89.9166 percent as shown in Table 3. This indicated that the experimental rates were not in good agreement with the predicted rates considering second order activity rate. Table 4 showed a very good agreement between the experimental and predicted rates which implied that the first order activity rate can be used to navigate the corrosion system. The adsorption isotherm parameters by linear method are shown in Table 5. The Langmuir isotherm showed the best fit for the experimental data in term of  $R^2$  value. The experimental and predicted values as well as the percentage difference for adsorption isotherms are depicted in Table 6. The kinetic - thermodynamic adsorption model showed the best fit for the experimental data in term of Percentage error between the experimental and the predicted data.

Table 2: Experimental and predicted values for corrosion rate as well as percentage difference between the experimental and predicted values

$r_{\text{expt}}$ (m/h)	$r_{\text{pred}}$ (m/h)	% error
1423.008	1395.186	1.96
872.251	923.163	5.84
756.302	725.042	4.13
607.853	610.836	0.49

Table 3: Parameters for second order activity rate

$c$ (v/v)	$t$ (h)	$r_{expt}$ (m/h)	$r_{pred}$ (m/h)	% error	$R^2$
0	2.25	4542.206	587.0284	87.0761	0.8168
	3.0	4375.31	449.9315	89.7166	
	3.50	3832.459	389.316	89.8416	
0.02	2.25	2043.158	957.825	53.1204	0.9279
	3.0	1423.068	631.8412	55.5982	
	3.50	928.469	514.9935	44.5330	
0.04	2.25	1498.55	1209.5925	19.2825	0.9929
	3.0	872.251	767.4992	12.0094	
	3.50	728.194	617.1296	15.2520	
0.06	2.25	1271.045	811.9163	36.1221	1.000
	3.0	756.302	524.1259	30.6989	
	3.50	599.947	423.9453	29.3362	
0.08	2.25	1048.81	4771.946	- 354.987	0.9026
	3.0	607.853	2375.9884	- 290.882	
	3.50	580.622	1780.129	-206.59	

Table 4: Parameters for first order activity rate

$c$ (v/v)	$t$ (h)	$r_{expt}$ (m/h)	$r_{pred}$ (m/h)	% error	$R^2$
0	2.25	4542.206	4620.347	- 1.7203	0.8282
	3.0	4375.31	4193.957	- 4.1449	
	3.50	3832.459	3931.79	- 2.5918	
0.02	2.25	2043.158	2104.22	- 2.9886	0.9738
	3.0	1423.068	1322.528	7.0611	
	3.50	928.469	970.319	- 4.5153	
0.04	2.25	1498.55	1456.546	2.8028	0.9727
	3.0	872.251	936.6409	- 7.3820	
	3.50	728.194	697.8121	4.1722	
0.06	2.25	1271.045	1248.315	1.7883	0.9895
	3.0	756.302	791.3181	- 4.6299	
	3.50	599.947	583.941	2.6679	
0.08	2.25	1048.81	980.218	6.5310	0.8895
	3.0	607.853	673.2387	- 10.7568	
	3.50	580.622	524.083	9.7377	

Table 5: Adsorption isotherm parameters by linearisation

Isotherm	Parameters	Value
Langmuir	$K_L$	1.3558
	$R^2$	0.9978
	$N$	5.7405
Freundlich	$k_F$	1.3558
	$R^2$	0.9529
	$y$	0.7691
Kinetic – Thermodynamic model	$K$	43.567
	$R^2$	0.9816
	$k_H$	1.3555
Linear	$R^2$	0.9529

Table 6: Experimental and predicted values as well as percentage difference for adsorption isotherms

$c(v/v)$	$\theta_{expt}$	Langmuir		Freundlich		Kinetic-thermodynamic		Linear	
		$\theta_{pred}$	% error	$\theta_{pred}$	% error	$\theta_{pred}$	% error	$\theta_{pred}$	% error
0.02	0.6748	0.6857	- 1.62	0.6857	- 1.62	0.6826	- 1.15	0.0271	95.98
0.04	0.8006	0.7737	3.359	0.7737	3.359	0.7835	1.872	0.0542	93.23
0.06	0.8271	0.8303	0.833	0.8303	- 0.39	0.8335	- 0.77	0.0813	90.17
0.08	0.8611	0.8730	0.862	0.8730	- 1.38	0.8627	- 0.10	0.1084	87.41

Figure 1 shows a graph of corrosion rate against inhibitor concentrations at optimum exposure of 3 h. The relationship between corrosion and inhibitor concentrations is inversely proportional that is as inhibitor concentrations increased, the corrosion rate declined. This revealed that the inhibitor molecules adsorbed on the surface of the metal inhibited corrosion rate. Figures 2 and 3 present the variation of corrosion rate against time at different inhibitor concentrations for second and first order activity rate respectively. In Figure 2, as the values of inhibitor concentrations increased, the values of inverse of corrosion rates also increased at constant exposure time. Moreover, as the exposure time increased, the values of inverse of corrosion rate also increased at constant inhibition concentrations while Figure 3 revealed that as the exposure time increased, the logarithm of the corrosion rate declined.

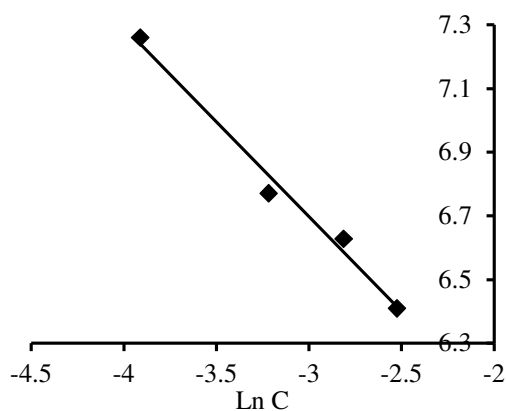


Figure 1: Variation of corrosion rate against inhibitor concentrations at optimum exposure time of 3 h

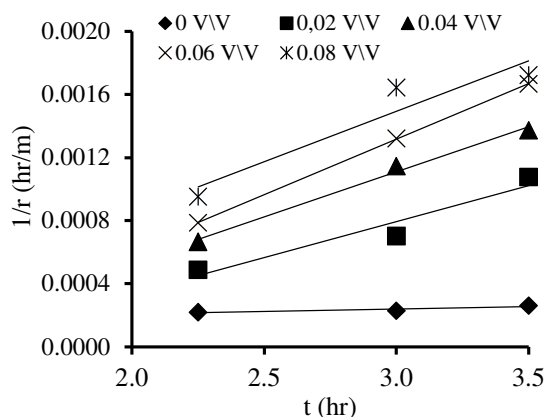


Figure 2: Variation of corrosion rate against time at different inhibitor concentrations for second-order activity rate

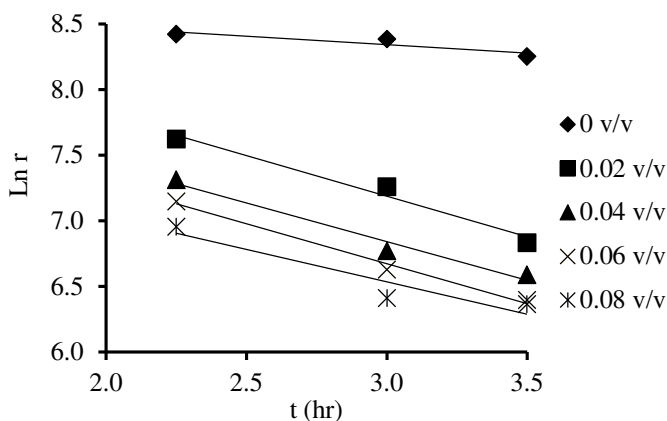


Figure 3: Variation of corrosion rate against time at different inhibitor concentrations for first-order activity rate

Figures 4–7 depict the Langmuir, Freundlich, Kinetic-thermodynamic model and linear adsorption isotherm plots respectively.

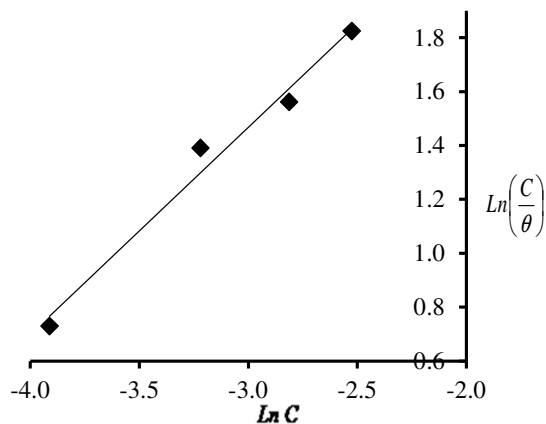


Figure 4: Langmuir adsorption isotherm plot

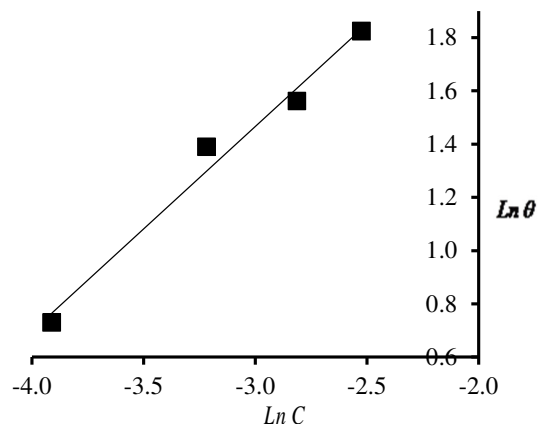


Figure 5: Freundlich adsorption isotherm plot

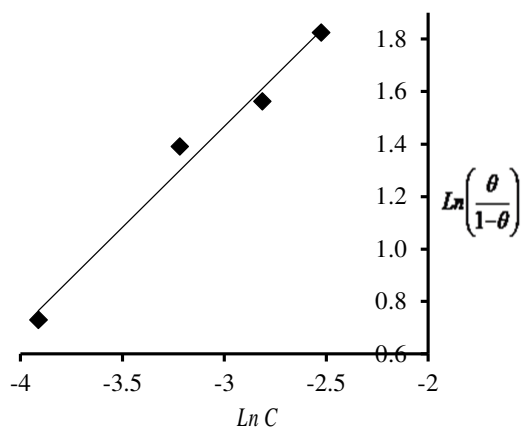


Figure 6: Kinetic thermodynamic model plot

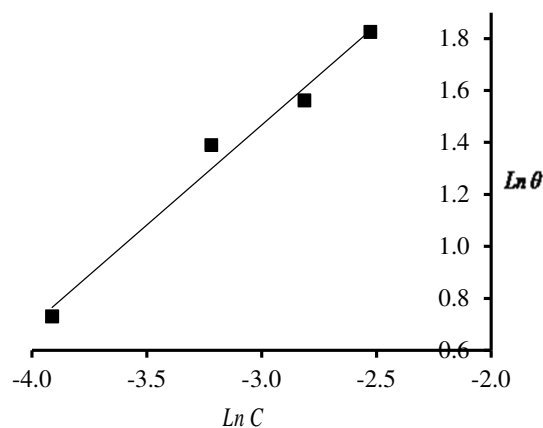


Figure 7: Linear adsorption isotherm plot

The corrosion rate of mild steel in 0.5 M sulphuric acid using *Grewa venusta* leaves extract as inhibitor is presented in Equation (14). The rate is a fractional order with a rate constant of 135.6394 h/m. The obtained rate model of Equation (14) was used to validate the experimental data and the percentage difference between the experimental and predicted values varied between 0.49 and 5.84 as shown in Table 3. This revealed the obtained model of Equation (14) predicted the experimental data up to 94 percent confidence level hence the model can be used to navigate the design of the corrosion facilities of mild steel in 0.5 M sulphuric acid using *Grewa venusta* leaves extract as inhibitor.

$$r = 135.6394c^{-0.5958} \quad (14)$$

The obtained second order activity rate model was used to validate the experimental data. Based on the coefficient of determination ( $R^2$ ) that is the goodness fit, it can be said the second order activity rate model predicted the experimental data as  $R^2$  values ranged between 0.8168 and 1.000. However, based on the percentage difference between the experimental and predicted values as shown in Table 5, there was a poor agreement indicating the second order activity rate model did not fit for the experimental data. This has

shown clearly that using coefficient of determination value alone to determine the fitness and validity of a model may be misleading. Models obtain in any work should be used to predict experimental values to determine validity of the models. The work of Olusegun and James, (2010), Salah (2016, Omoruwou *et al.* (2017), Odejobi and akinbulumo (2019) and Edoziuno *et al.* (2020) have validated their obtained models using coefficient of determination values alone hence there is a need for them to use their obtained model to predict the experimental data in their work.

The first order activity rate model fitted the experimental data based on  $R^2$  values as the values ranged between 0.8282 and 0.9738. Furthermore, the difference between the experimental and predicted values using the first order activity rate model varied between  $-10.7568$  and  $9.7377$  percent. This is an indication that the first order activity rate model validated the experimental data up to 90 percent confidence level hence can be used to navigate the design of the corrosion facilities. The first order activity rate constant ( $k_1$ ) according to Salah (2016) is considered as a measure of the speed of decay in the system activity toward corrosion. The  $k_1$  value for the process in the absence of inhibitor was 7.7459 hrs while the  $k_1$  values in the presence of inhibitor ranged between 1.615 and 2.028 hrs. This revealed that in the presence of inhibitor, the speed of decay in the system activity toward corrosion was reduced drastically.

In Figures 4 – 7, based on  $R^2$  values, all the adsorption isotherms considered in this work fitted the experiment data as  $R^2$  values ranged between 0.9529 and 0.9978 as shown in Table 5. The obtained adsorption isotherm models were used to validate the experimental data. The difference between the experimental and predicted values shown in Table 6 indicated that the linear adsorption isotherm did not fit the experimental data as the difference between the experimental and predicted values varied between 87.4068 and 95.9825 percent. Moreover, from Equation (11), the slope of the Langmuir plot in Figure 4 should be unity but in the plot, the slope was found to be 0.8258 which made the Langmuir adsorption isotherm model not to fit the experimental data.

For Freundlich adsorption isotherm model, the difference between the experimental and predicted values ranged between  $-1.6164$  and  $3.359$  percent which means it fitted the experimental data. The value of the Freundlich isotherm constant was 0.1742 which is less than unity. This shows that the ratio of the amount of inhibitor adsorbed onto a given mass of mild steel to the concentration of the inhibitor in the solution was not constant at different concentrations of inhibitor. Moreover, Kinetic – Thermodynamic adsorption model also fitted the experimental data as the difference between the experimental and predicted values ranged between  $-1.15$  and  $1.872$  percent. The value of the Kinetic –Thermodynamic isotherm constant,  $y$ , was 0.7691 which is less than unity. This depicts a given inhibitor molecule will occupy more than one active site and no formation of multilayer of inhibitor on the surface of the mild steel.

Though the Freundlich and kinetic – thermodynamic isotherm model fitted the experimental data, the kinetic – thermodynamic model can be taken to fit the experimental data better than the Freundlich isotherm based on the percentage values between the experimental and predicted data.

#### 4. CONCLUSION

The kinetics and adsorption studies of corrosion of mild steel in acidic medium using *Grewa venusta* leaves extract has been carried out. The kinetic rate model developed has a fractional order of  $-0.5958$  with a rate constant of 135.6394 hr/m. The kinetics rate model validated the experimental data up to 94 percent confidence level which indicates the model can be used to navigate the design of the corrosion facilities of mild steel in 0.5 M sulphuric acid using *Grewa venusta* leaves extract as inhibitor. The first order activity rate model developed also fitted the experimental data up to 90 percent confidence level which means it can also be used to navigate the design of the corrosion facilities. The first order activity rate constant obtained in the absence of inhibitor was 7.7459 hrs while it ranged between 1.615 and 2.028 hrs in the presence of inhibitor. This revealed that the speed of decay in the system activity toward corrosion was declined drastically in the presence of inhibitor. The kinetic-thermodynamic model with isotherm constant of 0.7691 best fitted the experimental data among the isotherms investigated, which means a given inhibitor molecule will occupy more than one active site on the surface of the mild steel.



## 5. CONFLICT OF INTEREST

There is no conflict of interest associated with this work.

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