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TECHNICAL ARTICLE—PEER-REVIEWED

Corrosion Inhibitive Capacity of Vanillin-Based Schiff Base for Steel in 1 M HCl

G. Banuprakash · B. M. Prasanna · B. M. Santhosh · A. M. Guruprasad · R. S. Malladi

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Abstract The inhibitive action of novel vanillin-based Schiff base for corrosion of steel in 1 M hydrochloric acid solution was evaluated by experimentally at the temperature range of 301–331 K. The computed inhibition efficiency of the inhibitor increases with the increasing its concentration as on 10 to 50 ppm. At the same time, inhibition efficiency decreases with the increasing temperature as on 301 to 331 K. Therefore, the increasing inhibition efficiency was attributed to the adsorption of inhibitor molecules onto the steel surfaces, and decreasing inhibition efficiency is because of desorption of inhibitor molecules from steel surfaces to the solution. The electrochemical polarization measurements proposed that the examined vanillin-based Schiff base as an inhibitor in nature, which reduces the corrosion of steel, decreases both anodic and cathodic reactions. In this study, the adsorption of the inhibitor complies with the Freundlich adsorption isotherm and depending on that, they chose compound as a corrosion inhibitor for steel in 1 M HCl through the mixture of physisorption and chemisorption. At long last SEM pictures show the visual thought of the development of the defensive layer onto the steel surfaces, which hinders the corrosion.

Keywords Vanillin · Schiff base · Electrochemical · Inhibitor

G. Banuprakash (✉)
Department of Chemistry, S. J. B. Institute of Technology, Bangalore, Viswesaraya Technological University, Belagavi 560 060, India
e-mail: banuprakashgas@gmail.com

B. M. Prasanna
Department of Chemistry, Jain Institute of Technology, Davanagere, Visveswaraya Technological University, Belagavi 577 003, India

B. M. Santhosh
Department of Chemistry, G. M. Institute of Technology, Davanagere, Viswesaraya Technological University, Belagavi 577006, India

A. M. Guruprasad
Department of Chemistry, Mangalore Institute of Technology and Engineering, Moodabidre, Viswesaraya Technological University, Belagavi 574 225, India

R. S. Malladi
Department of Chemistry, BLDEA's V.P. Dr. P. G. Halakatti College of Engineering and Technology, Vijayapur, Visvesaraya Technological University, Belagavi, Karnataka 586103, India

Introduction

Continuously the metals are considered as imperative in our day-to-day life because of their boss mechanical and thermal stability. Consequently, metal is utilized in different mechanical and construction applications [1]. Be that as it may, shockingly metal prompts loss in its weight because of a characteristic procedure of corrosion in destructive media [2]. In this manner, the corrosion of metals prompts economic loss for a nation and effects on health issues of industry workers. So, it is necessary to control the corrosion of metal by satisfactory, practically acceptable, eco-friendly, and cost-effective methods for all intents and purposes.

The corrosion inhibitors are commonly heterocyclic organic compounds, which are having electron-rich species such as N, S, O, and π electrons in heterocyclic rings existing in the molecule [3–5]. Through these, the inhibitor molecule adsorbed onto the electron-deficient steel surfaces via the adsorption process. Earlier a variety of sorts of organic compounds such as drug intermediates and herbal agricultural extracts have been suggested as appropriate corrosion inhibitors for the metal in 1 M HCl [6]. Already a wide variety of corrosion inhibitors have been developed for quite a several metals and their alloys in aggressively corrosive media [7]. But current corrosion inhibitors are economically expensive and toxic, and they behave as corrosion inhibitors solely in room temperature. The Schiff base ligands additionally act as proper corrosion inhibitors due to the presence of electron-rich N, S, O, C=N bonds, and non-bonding π-electrons. Through these, inhibitor molecule types covalent bonds with metallic ions at the surfaces and produce protecting barrier at the metallic and solution interface, it reduces the rate of corrosion decreases [8]. Our present investigation concentrates on the corrosion inhibitive ability of vanillin-based Schiff base for the metal in 1 M HCl media at a temperature range of 301–331 K. The investigation includes the electrochemical measurements (i.e., experimental) and scanning electron microscopic measurements (i.e., surface morphology).

Experimental

Steel

The strip of metal with an uncovered region 1 cm^2 was used for this investigation procured commercially. And the strips have been descaled, cleaned, and washed to take away the preliminary rust dust particles over the metallic surfaces. The easy dry and rust-free metal strips have been automatically polished by way of SIC emery papers with the lowest grade quantity of 80 up to very best grade wide variety of 2000 to get smooth surfaces. Then, they are stored in a desiccator to keep away from moisture attack on steel, and it was used as a working electrode for the actual corrosion experiments.

Inhibitor

The newly synthesized vanillin-based Schiff base was examined as a corrosion inhibitor for the metal in 1 M HCl. The IUPAC nomenclature of the chosen inhibitor 2-{[(E)-(4-hydroxy-3-methoxyphenyl) methylidene] amino}-3-(1H-indol-2-yl) propanoic acid is with the molecular formulation of $C_{19}H_{18}N_2O_4$ with the molecular weight of 338.35. Firstly, inhibitor was used to be dissolved in 1 cm^3 of ethanol and then miscible with 1 M HCl as an inhibited sample. The inhibitor was once dissolved in the concentration of 10–50 ppm in 1 M HCl and used as an inhibitor solution for all the corrosion experiments. The molecular structure of vanillin-based Schiff base is proven as shown in Fig. 1.

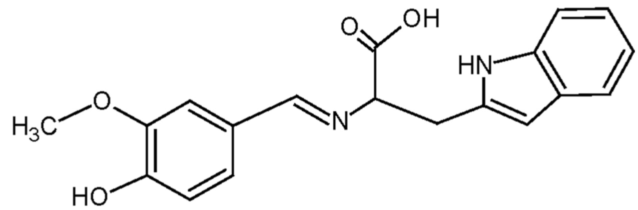

Fig. 1 The molecular structure of vanillin-based Schiff base

Electrochemical Measurements

The electrochemical trials were completed in an electrochemical workstation CHE608D which is associated with a three-electrode system. The three-electrode system comprises a steel strip as a working terminal, immersed calomel cathode as a source of the reference electrode and a platinum counter terminal as a counter electrode.

The inhibition efficiency of vanillin-based Schiff base for steel in 1 M HCl at temperature scope of 301–331 K. For the potentiodynamic polarization estimations, the potential–current bends were plotted at a scan rate of 0.01 V s-1 within the given possible range. The electrochemical impedance spectroscopic investigations were led by an AC signal with an amplitude of 5 mV at open-circuit potential at the frequency region of 100–0.1 kHz. The acquired EIS information was fit into an electrical circuit with the assistance of Z-Simp 3.21 expert programming.

Adsorption Isotherm

The corrosion inhibition effectiveness of vanillin-based Schiff base is credited because of the adsorption of inhibitor molecules onto the surfaces of metal from the solution. The method of adsorption was concentrated by the assistance of registered thermodynamic boundaries fitting into a proper adsorption isotherm model. The examined novel Schiff base in this examination is good for the Freundlich adsorption isotherm, and figured boundaries are talked about.

Scanning Electron Microscopic Measurement (SEM)

The polished strips of steel were immersed within the absence and presence of 50 ppm of compound vanillin-based Schiff base in 1 M HCl over an amount of 6 h. Then, the strips were taken out, washed with triple water and dried at temperature. The surface morphology of each of the strips was obtained by scanning microscope (SEM) VEGA3 TESCAN SEM instrument at an associate fast beam of 20 kV.

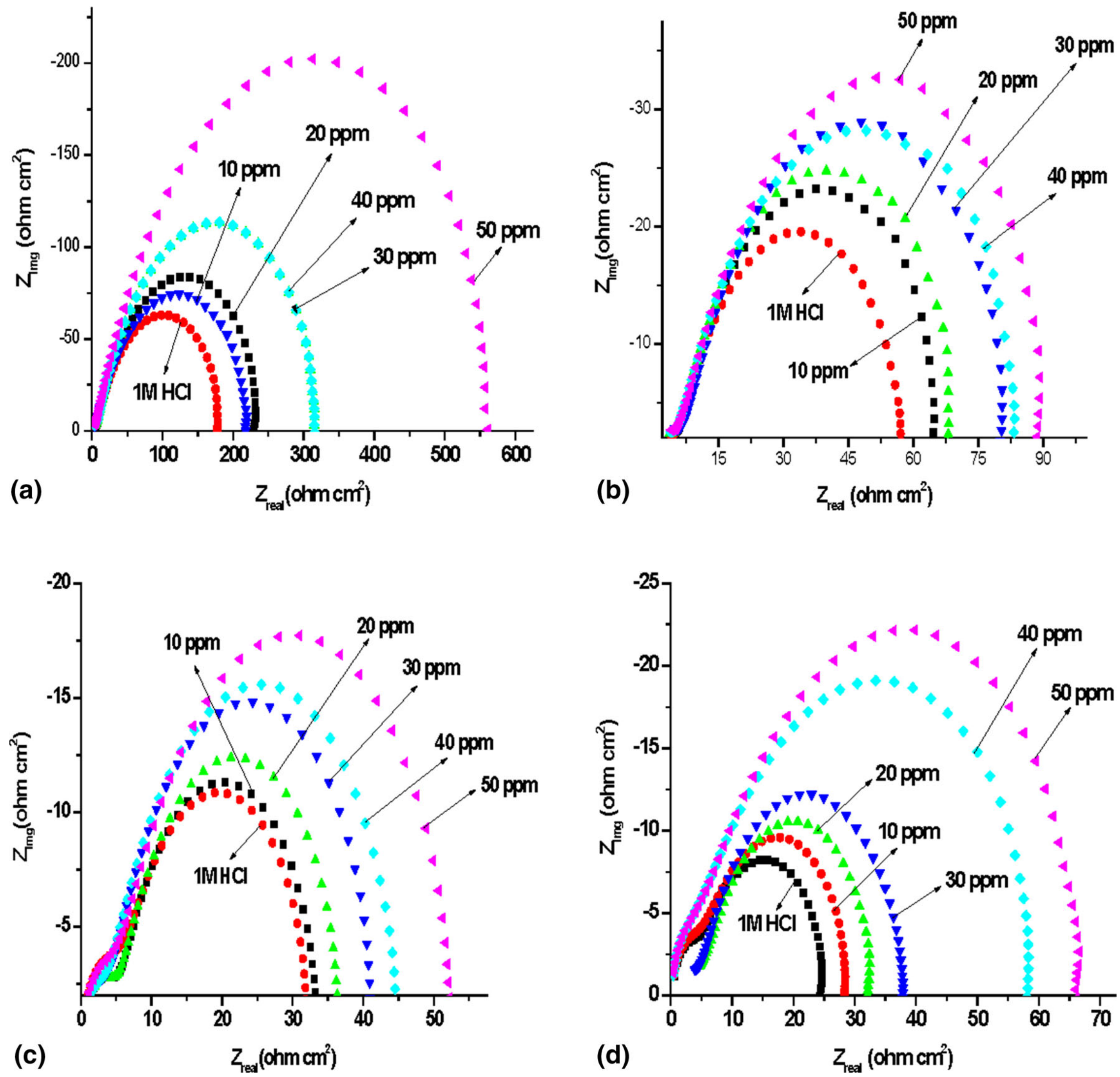

Fig. 2 EIS spectrum for steel without and with the vanillin-based Schiff base as inhibitor in 1 M HCl at temperature of (**a**) 301 K (**b**) 311 K (**c**) 321 K (**d**) 331 K

Results and Discussion

EIS

The corrosion inhibition capability of novel vanillin-based Schiff base was investigated by electrochemical impedance measurement at the temperature variations of 301–331 K. Nyquist's plot carries with it a collection of semicircles with high-frequency (HF) depressed semicircles (Nyquist plots). Figure 2 shows the Nyquist's plot, and also the EIS data work into an appropriate electrical equivalent circuit is shown in Fig. 3. The corrosion parameters like polarization resistance (RP) and double-layer capacitance (C_{dl}) were computed. The inhibition potency (ηZ) of vanillin-based Schiff base is calculated by the subsequent equation as [9],

$$\eta_Z = \frac{R_p - R_p^0}{R_p} \times 100$$

where R_p and R_P^0 are polarization resistance values without and with the addition of vanillin-based Schiff base as an inhibitor in 1 M HCl solution. The diameter of the semi-circle is delineated as R_p value, and therefore, the double-layer capacitances were measured by the subsequent equation as,

$$C_{dl} = \left(QR_{ct}^{1-n}\right)^{1/n}$$

where Q is that the constant phase element (CPE) and n is the CPE exponent which provides details concerning the degree of surface inhomogeneity.

All calculated corrosion parameters by EIS measure are reported in Table 1. The results found by the EIS measurement showed the values of η_Z are increased with increasing the concentration of the inhibitor. The increasing inhibition efficiency was attributed to the surface adsorption of the inhibitor molecules onto the metal surfaces, which forms a protecting layer that ends up in the decreasing corrosion rate. During this investigation, we tend to accomplish the maximum inhibition efficiency of the inhibitor as around 70% for an optimized concentration of 50 ppm of inhibitor at 301 K temperature. After that, the inhibition efficiency step by step decreases with the increasing temperatures. The lesser inhibition result of around 38% for 50 ppm of inhibitor was achieved at 331 K. As a result of the increasing temperature, the adsorbed layer over the metal surfaces diminishes. Therefore, natural action takes place over the surface assimilation of the substance from resolution to metal surfaces. Thus, the substance is not simpler to scale back the corrosion of metal at high temperatures. The issue that explains the increasing or decreasing of inhibition efficiency is the double-layer capacitance (C_{dl}). The decreasing values of C_{dl} urged that increasing the inhibition result of substance because of either decrease in dielectric constant or increase within the thickness of the electrical double layer suggesting that the novel vanillin-based Schiff base adsorbed on the metal/solution interface. In step with this investigation by the EIS technique, the studied inhibitor acts as a good inhibitor for steel in 1 M HCl.

Potentiodynamic Polarization Measurements

The corrosion kinetic parameters are examined by Tafel polarization for steel in the absence and presence of novel vanillin-based Schiff base in 1 M HCl at the raised temperature range of 301–311 K. In these estimations, Tafel's plots were plotted as potential against log i by changing potential by ± 0.2 V to the open-circuit potential (E_{corr}) with a scan rate of 0.01 mV/s as shown in Fig. 4. The corrosion parameters, viz. corrosion current density (i_{corr}), corrosion rate (v), cathodic Tafel slope (β_c), anodic Tafel slope (β_a) and inhibition efficiency (η_p), are reported in Table 2. The following equation condition is utilized to calculate the inhibition efficiency (η_p) of the studied inhibitor for steel in 1 M HCl,

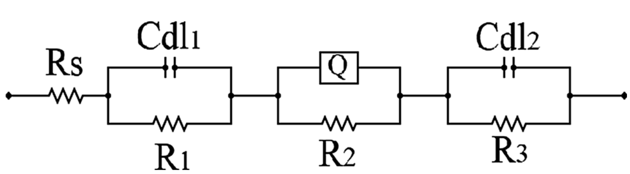

Fig. 3 Electrical equivalent circuit

$$\eta_p = \frac{i^0_{Corr} - i_{Corr}}{i^0_{Corr}} \times 100$$

where i^0_{Corr} and i_{corr} are corrosion current density in the absence and presence of vanillin-based Schiff base in 1 M HCl, respectively.

The final results acquired from Tafel polarization measurements are given in Table 2; the corrosion current density decreases [10] with the increasing concentrations of vanillin-based Schiff base as an inhibitor in 1 M HCl. This is attributed to the adsorption of inhibitor molecules onto the surfaces of metallic from the bulk of the solution. The values of corrosion potential (Ecorr) for inhibited solution vary concerning that of the uninhibited solution is less than ± 85 mV, the inhibitor acts as a mixed type in nature [11]. The inhibitor retards both the anodic (i.e., metal dissolution) and cathodic (i.e., Hydrogen liberation) reactions, which results in the corrosion. In our study, we discovered the variant of Ecorr of the inhibited solution for the uninhibited solution is around or less than 30 mV, and it was advised that vanillin-based Schiff base acts as a mixed type in nature. Therefore, the small change in

Table 1 EIS parameters for steel without and with the vanillin-based Schiff base in 1 M HCl

Temperature (K)	Inhibitor conc^n (ppm)	R_p (Ω cm^2)	C_{dl} (μF cm^{-2})	η_z	Surface coverage θ
301	Blank	175.2	575	…	…
	10	219.38	571	20.13	0.201
	20	225.07	523	22.15	0.221
	30	265.89	496	34.10	0.341
	40	317.22	523	44.77	0.447
	50	574.74	580	69.51	0.695
311	Blank	23.52	459	…	…
	10	27.67	274	14.45	0.144
	20	30.63	635	23.21	0.232
	30	35.02	689	32.83	0.328
	40	57.11	456	58.81	0.588
	50	65.41	514	64.04	0.640
321	Blank	31.13	329	…	…
	10	32.57	357	4.42	0.422
	20	35.20	600	11.56	0.115
	30	39.81	322	21.80	0.218
	40	43.84	646	28.99	0.289
	50	51.29	637	39.30	0.393
331	Blank	54.49	592	…	…
	10	63.19	594	13.7	0.137
	20	66.19	610	17.67	0.176
	30	77.57	682	29.75	0.297
	40	81.47	224	33.11	0.331
	50	86.87	554	37.27	0.372

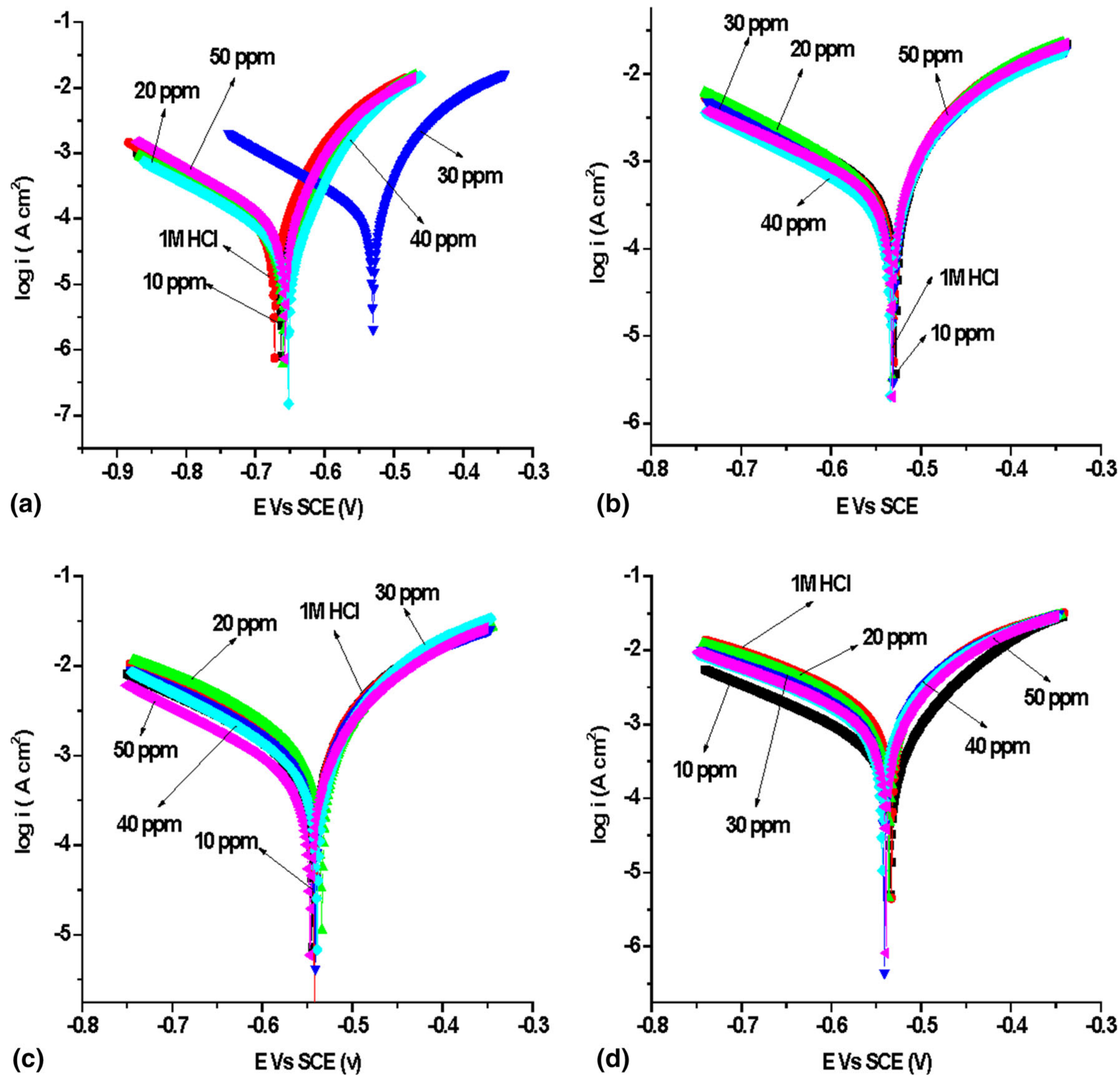

Fig. 4 Tafel's plots for steel without and with the vanillin-based Schiff base as inhibitor in 1 M HCl at temperature of (**a**) 301 K (**b**) 311 K (**c**) 321 K (**d**) 331 K

slopes of anodic and cathodic curves for the inhibited solution compared to uninhibited solution advised that the strong barrier for proton discharge results in lesser hydrogen evolution and the inhibitor changed into first adsorbed onto the metallic surfaces which blocking the corrosion active sites on metallic surfaces without affecting the anodic reaction [12]. The corrosion rate was found to be lower with increasing inhibitor concentrations; we found around 72% at 50 ppm concentration of inhibitor at 301 K. This shows the revolutionary adsorption of inhibitor at the metallic surfaces with an increase in concentration [13]. The inhibition efficiency decreases with the increasing temperature of 301–331 K; that is attributed to that the desorption of inhibitor molecules takes area at an excessive rate from the metallic surfaces in preference to the adsorption process. The results suggested that the polarization measurements had been nearly much like that of the effects discovered from EIS results. The investigated vanillin-based Schiff base acts as an effective corrosion inhibitor for steel in 1 M HCl.

Thermodynamics

The inhibitive capability of inhibitor is attributed to the adsorption of molecules from the bulk of the solution onto the metallic surfaces. The mode of adsorption is investigated through an appropriate adsorption isotherm model. The data found from EIS measurement were fit into a unique adsorption isotherm model. If the regression coefficient (R^2) had been near to unity taken into consideration as that is the maximum suitable adsorption isotherm version for the adsorption method. The vanillin-based Schiff

Table 2 Corrosion parameters by polarization measurement for steel without and with the vanillin-based Schiff Base in 1 M HCl

Temperature (K)	Inhibitor conc^n (ppm)	Corrosion potential E_{corr} (V)	Corrosion current density i_{corr} (A cm^{-2})	Corrosion rate v_{corr} (mpy)	Cathodic Tafel slope β_c (mV/decade)	Anodic Tafel slope β_a (mV/decade)	Inhibition efficiency η_p (%)
301	Blank	− 0.672	0.0636	28.60	6.43	11.91	…
	10	− 0.663	0.0478	12.44	6.35	12.43	24.84
	20	− 0.660	0.0384	9.36	6.36	13.49	39.62
	30	− 0.630	0.0328	7.52	6.06	10.31	48.42
	40	− 0.652	0.0264	5.63	6.52	14.63	58.49
	50	− 0.657	0.018	5.16	6.28	11.93	71.69
311	Blank	− 0.547	0.1464	28.63	5.61	8.36	…
	10	− 0.542	0.0825	59.37	5.70	7.02	43.64
	20	− 0.534	0.0741	53.31	5.97	7.60	49.38
	30	− 0.541	0.0652	46.91	5.91	7.39	55.46
	40	− 0.534	0.0560	40.27	6.32	7.99	61.74
	50	− 0.539	0.0507	36.52	6.08	8.43	65.36
321	Blank	− 0.530	0.1418	26.57	5.59	8.07	…
	10	− 0.528	0.1359	26.15	5.52	8.07	41.60
	20	− 0.532	0.1262	24.67	5.98	8.48	11.00
	30	− 0.534	0.0926	19.77	5.67	8.09	34.69
	40	− 0.531	0.1011	18.12	6.13	8.25	28.70
	50	− 0.535	0.0751	14.68	5.30	8.82	47.03
331	Blank	− 0.533	0.1011	72.78	5.78	7.14	…
	10	− 0.534	0.0949	18.57	5.77	10.27	61.32
	20	− 0.536	0.0844	60.76	5.76	7.50	16.51
	30	− 0.541	0.0762	54.87	5.70	7.49	24.62
	40	− 0.543	0.0676	48.64	5.56	7.67	33.13
	50	− 0.539	0.0620	44.64	5.48	8.24	38.67

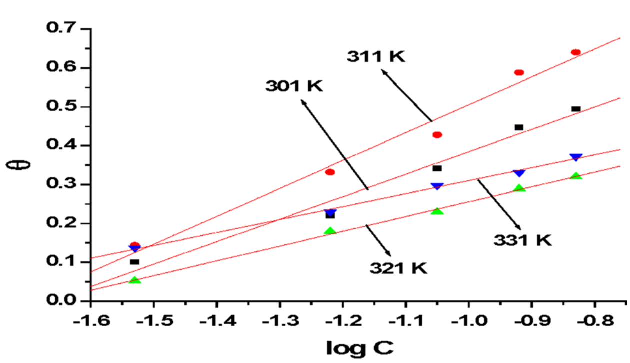

Fig. 5 Freundlich adsorption isotherm model

base acts as a corrosion inhibitor for steel through adsorption method, which obeys Freundlich adsorption isotherm as shown in Fig. 5. Therefore, the Freundlich adsorption isotherm plot includes the log of the concentration of inhibitor (C) in millimoles against surface coverage (θ) of the metal, which results in a set of straight lines with slope and intercept. According to the Freundlich adsorption isotherm,

Table 3 Adsorption isotherm and thermodynamic parameters

Temperature (K)	K_{ads} (M^{-1})	ΔG^0_{ads} (kJ/mol)
301	1572	− 30.55
311	1557	− 31.28
321	1039	− 27.43
331	819	− 27.72

$$\log \theta = \log K + n \log C.$$

The Freundlich adsorption isotherm model was used to calculate the adsorption parameters. The values of the equilibrium constant of adsorption (K_{ads}) are calculated by the intercept values of straight lines found on plot, and the standard free energy change of adsorption ΔG^0_{ads} was calculated by the following expressions as

$$\Delta G_{ads} = -\ln(K_{ads} \times 55.5) \times R \times T.$$

The computed values of K_{ads} and ΔG^0_{ads} are reported in Table 3.

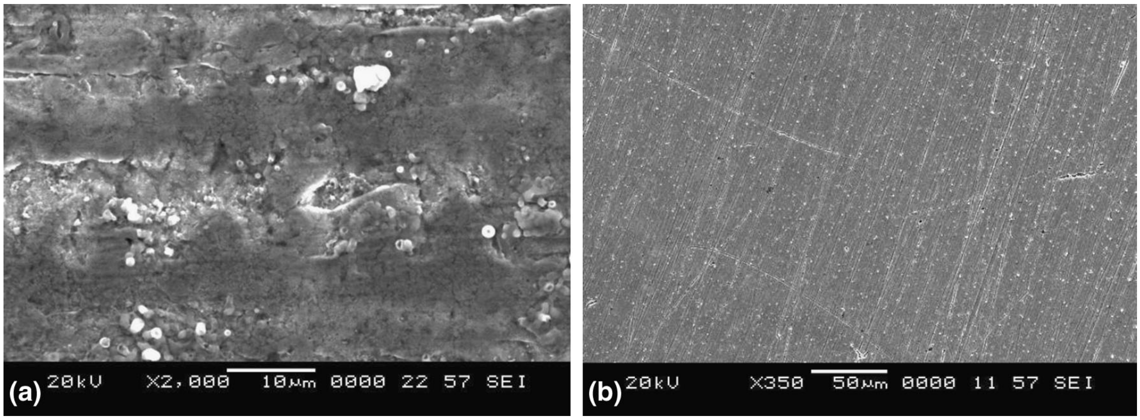

Fig. 6 SEM images of steel immersed in (**a**) uninhibited sample and (**b**) inhibited sample

The computed values of K_{ads} and ΔG^0_{ads} are stated in Table 3. With the information acquired with the aid of using this study (Table 3), the values of K_{ads} describe the adsorption of inhibitor molecules from the solution over the surfaces of steel. The higher the value of K_{ads}, the higher the inhibition effect of the inhibitor for steel.

The results reported as the K_{ads} are 1572 M^{-1} at 301 K; thereafter, the K_{ads} values decrease with increase in temperature of 301–331 K. This suggests that the inhibition effect of vanillin-based Schiff base for metal decreases with the increasing temperature. This is attributed to that the desorption method has a dominant position than the adsorption method. And some other adsorption parameter such as ΔG^0_{ads} additionally explains the inhibitive capability of inhibitor for the metal in 1 M HCl. The bad signal of ΔG^0_{ads} indicates that the inhibitor spontaneously adsorbed onto the metal surfaces. If the value of ΔG^0_{ads} is less than or equal to -20 kJ/mol adsorption is because of the electrostatic interaction (i.e., Physisorption), even as the ones around $-$ 40 kJ/ mol or higher are typically accepted to form a coordinate kind of bond (i.e., chemisorption) [14]. In this cutting-edge investigation, the value of ΔG^0_{ads} is in between $-$ 27.43 and 31.28 kJ/mol. Hence, the adsorption of inhibitor takes area via each physisorption and chemisorption approach. The investigated compound acts as a corrosion inhibitor via each physisorption and chemisorption approach onto the surfaces of metal in 1 M HCl [15].

SEM

The SEM micrographs of steel strips in 1 M HCl solution in the absence and the presence of 50 ppm of vanillin-based Schiff base are defined in Fig. 6. The surface examination of steel in uninhibited solution showed that it has pits and corroded elements as proven in Fig. 6(I). But metal surface was immersed in inhibited sample that has smooth surfaces due to defensive adsorbed layer of the inhibitor molecules over the metal surfaces [16]. Hence, SEM measurements offer a visible concept of the formation of a protective layer in the steel/solution interface, which resists the steel through the attack of corrosion.

Conclusions

The inhibition effect of vanillin-based Schiff base for steel in 1 M HCl was found with maximum inhibition efficiency percentage of 71.69% at optimized concentration of 50 ppm. The inhibition efficiency of studied inhibitor increases with the increasing inhibitor concentration, but decreases with the increasing temperature of 301 K–331 K.

The inhibitive capability of vanillin-based Schiff base for steel in 1 M HCl is facilitated through the adsorption process. Therefore, Freundlich adsorption isotherm model was most likely to fit this investigation. Hence, the values of ΔG^0_{ads} less than $-$ 40 kJ/mol suggest that the inhibitor spontaneously adsorbed onto the metal surfaces through mixture of physisorption and chemisorption method.

The surface morphology of steel in the absence and presence of inhibitor in 1 M HCl was carried out by SEM analysis. The SEM image of steel shows that the inhibitor molecules form a protective film, which reduces the corrosion.

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