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# *Ocimum basilicum* Extract as a Potential Green Inhibitor for Corrosion of Mild Steel in 0.5M $H_2SO_4$ Solution

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The objective of the study is to determine the effect of extract of *Ocimum basilicum* on corrosion of mild steel in aqueous 0.5M $H_2SO_4$. The systematic study was done by using weight loss method, potentiodynamics polarization technique and electrochemical impedance spectroscopy. The corrosion inhibition is found to occur on mild steel in presence of the extract and the inhibition efficiency of the extract increases with increase in concentration. Polarization measurement indicates that *Ocimum basilicum* acts as a mixed-type inhibitor and the inhibition efficiency decreases with rise in temperature. The corrosion inhibition on mild steel is found to occur due to the adsorption of inhibitor molecules on metal surface, which obeys Langmuir adsorption isotherm. The increasing values of activation energies ($E_a$) in presence of the extract indicate the retardation in rate of corrosion on metal surface. Scanning electron microscopic study confirmed the inhibition of corrosion on metal surface.

**Key Words: Mild steel, Acidic corrosion, Electrochemical impedance spectroscopy, SEM, Adsorption.**

## INTRODUCTION

The study of corrosion inhibition using inhibitor in acidic media is one of the challenges in the current research due to its potential applications in industries such as acid pickling, industrial cleaning, acid descaling, oil-well acid in oil recovery and petrochemical processes[1-5]. The ability of a compound to serve as inhibitor is dependent on its ability to form a compact barrier film and/or nature of adsorption on metal surface. The majority of well-known inhibitors are organic compounds containing heteroatoms, such as O, N, S and multiple bonds[6]. Although many synthetic compounds show good anticorrosive properties, most of them are highly toxic to both human beings and environments[7]. The known hazardous effect of most synthetic organic inhibitors and restrictive environmental regulations have now made researchers to focus on the need to develop cheap, non-toxic and environmental friendly inhibitors like natural products. The natural product extracts are viewed as an incredibly rich source of naturally synthesized chemical compounds that can be extracted by simple procedures with low cost and are biodegradable in nature[8]. This area of research is of much importance because in addition to being environmentally friendly and ecologically acceptable, plant products are inexpensive, readily available and renewable source of materials[9]. The use of these natural products such as extracted compounds from the leaves, flowers, seeds and roots as corrosion inhibitors have been widely reported by several authors[10-14].

The aim of the present work is to find a naturally occurring, cheap and environmentally safe substance that could be used for inhibiting corrosion of mild steel in acidic medium. Therefore, in this present work, *Ocimum basilicum* is chosen for the study and an attempt has been made to ascertain their corrosion inhibition properties. The aqueous extract of its leaves in 0.5M sulphuric acid was tested by using weight loss, potentiodynamic polarization and electrochemical impedance techniques. Scanning electron microscopic (SEM) was also used to study the surface morphologies.

*Ocimum basilicum* was found to use worldwide for its gustative qualities and especially in Mediterranean Cuisine and had long been recognized in folk medicine as a medicinal plant having various medicinal uses such as cardiotonic, abdominal pain reliever and antidiarrhoeal agent. Leaf extracts has antiinflamatory and antioxidant effect. Aqueous extract of *Ocimum basilicum* was also found recently that it decreases plasma total cholestoral, LDL-cholestoral *etc*. and its major chemical constituents are polyphenol (tannins) and flavonoids[15]. However, literature search reveals that no study had been done on the inhibitive effects of *Ocimum basilicum* extract on acidic corrosion of mild steel.

## EXPERIMENTAL

**Preparation of extract of *Ocimum basilicum*:** Double distilled water and analytical reagents-grade $H_2SO_4$ (E Merk, India, AR Grade) were used for preparing solutions. The leaves of *Ocimum basilicum* was dried for 6 h in an oven at 70 ºC and ground into powder and 10 g of the powder of *Ocimum basilicum* was refluxed in 100 mL double distilled water for 1 h. The extract of the plant was prepared by evaporating the filtrate. The required concentrations of solution were prepared by using the residues in aqueous solution of 0.5M $H_2SO_4$.

**Weight loss method:** Mild steel coupons having per cent composition of C (0.18), Si (0.19), Mn (0.51), P (0.044), S (0.057), Cr (0.14), Ni (0.09), Mo (0.02), Cu (0.06), V (less than 0.01) and remaining Fe (chemical analysis: % by weight by equipment, IS:228 and ICP-OES) were used. The size of the coupon is 1 cm × 4 cm × 1 cm. The specimens were polished successively using the emery papers of 150, 180, 320, 400, 600 and 1000 grade. The polished surface were degreased with acetone and washed with distilled water before the experiment. Weight loss of mild steel coupons immersed in 100 mL of the electrolyte with and without the extract was determined after 4 h at 298 K. The percentage inhibition efficiency (I %) was calculated from the following equation[16]:

$$I\,(\%) = \frac{(W_o - W_i)}{W_o} \times 100 \tag{1}$$

where $W_o$ and $W_i$ are weight losses of mild steel in absence and presence of the extract.

**Electrochemical measurements:** An electrochemical cell assembly of three electrodes was used for potentiodynamic polarization and electrochemical impedance measurements, in which mild steel, calomel electrode and platinum wire were used as the working electrode, reference and counter electrode, respectively. The working electrode was coated thoroughly with epoxy resin keeping surface area of 1 $cm^2$ for the study. The surface of the mild steel was abraded into uniform surface with the help of grinding machine by using 150, 320, 400, 600 grade emery papers and finally polished by 1000 grade emery papers. The polished surface were degreased with acetone and washed with distilled water before the experiment. The measurements were done by using computer controlled electrochemical workstation of CHI 760c model. Before each polarization and electrochemical impedance spectroscopy (EIS) measurement, the working electrode was introduced into the test solution and kept for 4 h to attain the open circuit potential (OCP). Polarization measurements were made under thermostatic conditions at 298, 308, 318 and 328 K and the measurements were carried out in the range of potential from -1.2 to 2.0 V with scan rate of 0.01 (V/s). The percentage inhibition efficiency (I %) from the polarization measurement was calculated using the following equation[17]:

$$I\,(\%) = \frac{(i^o_{corr} - i^i_{corr})}{i^o_{corr}} \times 100 \tag{2}$$

where $i^o_{corr}$ and $i^i_{corr}$ are the corrosion current density values without and with inhibitor, respectively.

Electrochemical impedance measurement was carried out at 298 K and the measurement of the response of the electrochemical system to a.c. excitation with a frequency ranging from 10,0000 to 0.1 Hz and peak to peak a.c. amplitude of 0.005 V was done. The percentage inhibition efficiency (I %) from the electrochemical impedance measurement was calculated using the following equation[18]:

$$I\,(\%) = \frac{[R_{ct(i)} - R_{ct(a)}]}{R_{ct(i)}} \times 100 \tag{3}$$

where $R_{ct(i)}$ and $R_{ct(a)}$ are the values of charge transfer resistance in presence and absence of the inhibitor, respectively.

**Surface analysis:** The test coupons of the size 1 cm × 1 cm were exposed in 100 mL of 0.5M $H_2SO_4$ solutions in absence and presence of 1 and 3 g of the plant extracts for 5 h at 298 K and then washed with distilled water. After drying the specimens, they were examined by scanning electron microscope (SEM) model Leo 435 VP with an Oxford Inca energy dispersion spectrometer system.

## RESULTS AND DISCUSSION

**Weight loss method:** The percentage of inhibition efficiency (I %) at different concentrations of *Ocimum basilicum* extract at 298 K are summarized in the Table-1. It is clear that inhibition efficiency of the extract on corrosion of mild steel increases with increase in concentration. It increases up to 97.9 % when the concentration of extract increases up to 3 g/L.

TABLE-1
CORROSION PARAMETERS FOR MILD STEEL IN 0.5M $H_2SO_4$ SOLUTION IN ABSENCE AND PRESENCE OF DIFFERENT *Ocimum basilicum* EXTRACT CONCENTRATIONS

| Temperature (K) | Solution | Concentration (g/L) | I (%) |
|---|---|---|---|
| 298 | 0.5M $H_2SO_4$ | 0.0 | – |
| | | 1.0 | 83.4 |
| | | 2.0 | 89.8 |
| | | 3.0 | 97.9 |

**Potentiodynamic polarization measurement:** Potentiodynamic polarization curves for mild steel in 0.5M $H_2SO_4$ solutions in absence and presence of various concentrations of *Ocimum basilicum* extract at 298 K are shown in Fig. 1. The extrapolation of Tafel straight line allows the calculation of the corrosion current density ($i_{corr}$). The values of icorr, the corrosion potential ($E_{corr}$), cathodic and anodic Tafel slopes ($\beta_c$ and $\beta_a$) and the percentage of inhibition efficiency (I %) are given in the Table-2.

At a given temperature, the addition of the extract of *Ocimum basilicum* to the acid solution increases both the anodic and cathodic overpotentials and decreases the corrosion current density ($i_{corr}$). The change in cathodic and anodic Tafel slopes ($\beta_c$ and $\beta_a$) shown in the Table-2 indicates that adsorption of *Ocimum basilicum* extract modify the mechanism of the anodic dissolution as well as cathodic hydrogen evolution. From Fig. 1, it is clear that both cathodic and anodic reactions are inhibited and the inhibition increases as the inhibitor concentration increases in acid media. From Table-2, it is clear that there is no definite trend in the shift of $E_{corr}$ values, in presence of various concentration of *Ocimum*

TABLE-2
ELECTROCHEMICAL PARAMETERS FOR MILD STEEL CORROSION IN 0.5M $H_2SO_4$ SOLUTION IN ABSENCE AND PRESENCE OF DIFFERENT *Ocimum basilicum* EXTRACT CONCENTRATIONS

| Temp. (K) | Solution | Conc. (g/L) | $-E_{corr}$ (mV *vs.* SCE) | $\beta_c$ (mV/dec) | $\beta_a$ (mV/dec) | $i_{corr}$ (mA/cm$^2$) | I (%) | θ |
|---|---|---|---|---|---|---|---|---|
| 298 | 0.5M $H_2SO_4$ | 0.0 | 475 | 54 | 61 | 8.11 | – | – |
| | | 1.0 | 512 | 66 | 49 | 1.62 | 80.0 | 0.80 |
| | | 2.0 | 495 | 71 | 52 | 1.30 | 83.9 | 0.839 |
| | | 3.0 | 496 | 78 | 46 | 0.33 | 95.9 | 0.959 |
| 308 | 0.5M $H_2SO_4$ | 0.0 | 475 | 53 | 59 | 14.99 | – | – |
| | | 1.0 | 488 | 62 | 62 | 5.20 | 65.3 | 0.653 |
| | | 2.0 | 498 | 63 | 63 | 4.71 | 68.6 | 0.686 |
| | | 3.0 | 512 | 65 | 49 | 2.11 | 85.9 | 0.859 |
| 318 | 0.5M $H_2SO_4$ | 0.0 | 481 | 48 | 51 | 16.39 | – | – |
| | | 1.0 | 478 | 60 | 61 | 7.47 | 54.4 | 0.544 |
| | | 2.0 | 501 | 56 | 51 | 6.72 | 58.9 | 0.589 |
| | | 3.0 | 497 | 51 | 50 | 4.65 | 71.6 | 0.716 |
| 328 | 0.5M $H_2SO_4$ | 0.0 | 500 | 49 | 50 | 19.98 | – | – |
| | | 1.0 | 469 | 53 | 57 | 12.60 | 36.9 | 0.369 |
| | | 2.0 | 479 | 55 | 66 | 10.84 | 45.7 | 0.457 |
| | | 3 .0 | 483 | 57 | 58 | 8.43 | 57.8 | 0.578 |

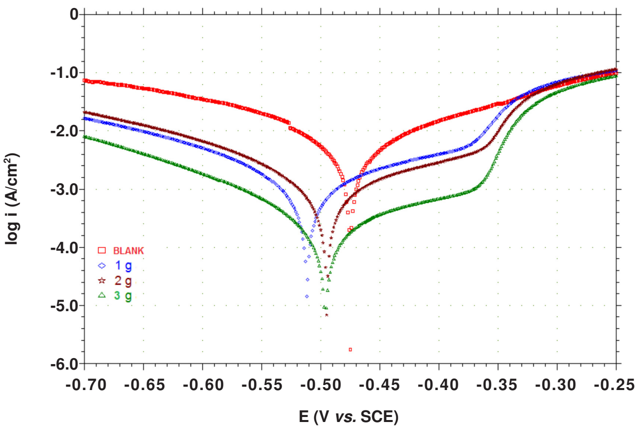


Fig. 1. Potentiodynamic polarization curves for mild steel in 0.5M $H_2SO_4$ solution in absence and presence of different concentrations of *Ocimum basilicum* extract at 298 K

*basilicum* extract in 0.5M $H_2SO_4$ solutions. This result indicates that *Ocimum basilicum* extract can be classified as mixed type of inhibitor in 0.5M $H_2SO_4$ solutions[19].

**Effect of temperature:** The effect of temperature on the inhibition efficiency is summarized in the Table-2. It shows that an increase in temperature decreases the inhibition efficiency. This can be explained on the fact that an increase in temperature usually accelerates corrosive processes, particularly in media in which $H_2$ gas evolution accompanies corrosion, giving rise to higher dissolution rates of the metal. The activation energies ($E_a$) for the corrosion process in absence and presence of inhibitor are evaluated from Arrhenius equation[20]:

$$k = Ae^{(-Ea/RT)} \tag{4}$$

where A is the pre-exponential factor, T is absolute temperature, R the gas constant and k is the rate constant of metal dissolution reaction which is directly related to corrosion current density. Therefore, the equation can be rewritten as[21]:

$$i_{corr} = Ae^{(-Ea/RT)} \tag{5}$$

where $i_{corr}$ is the corrosion current density. The activation energies of corrosion reaction in presence and absence of the inhibitor are determined by plotting log $i_{corr}$ against 1/T as shown in Fig. 2. The values of the activation energies are given in Table-3.

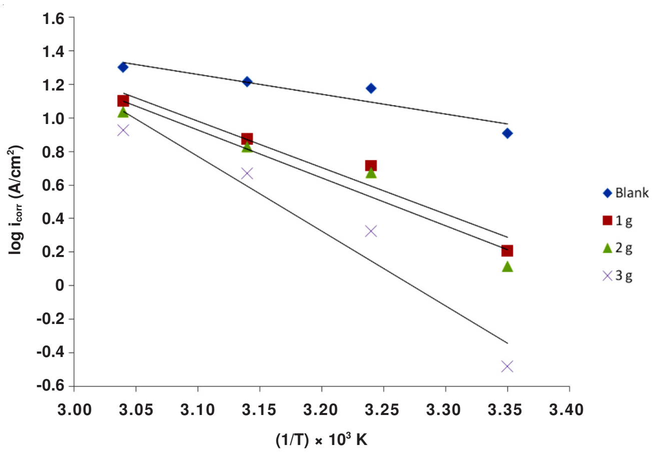


Fig. 2. Arrhenius plots of log $i_{corr}$ *versus* 1/T for mild steel corrosion in 0.5M $H_2SO_4$ solution in absence and presence of different concentrations of *Ocimum basilicum* extract

TABLE-3
CALCULATED VALUES OF ACTIVATION ENERGIES ($E_a$) FOR VARIOUS CONCENTRATIONS OF *Ocimum basilicum* EXTRACT DURING MILD STEEL CORROSION IN 0.5M $H_2SO_4$ SOLUTION

| Solution | Conc. (g/L) | $E_a$ (kJ/mol) | $R^2$ |
|---|---|---|---|
| 0.5 M $H_2SO_4$ | 0.0 | 22.72 | 0.876 |
| | 1.0 | 52.90 | 0.944 |
| | 2.0 | 54.59 | 0.924 |
| | 3.0 | 85.30 | 0.938 |

The values of activation energies ($E_a$) increased in presence of the extract at all the studied concentration in 0.5M $H_2SO_4$, which suggested that the adsorbed organic matters create a physical barrier to charge and mass transfer leading to reduction in corrosion rate[21].

**Adsorption isotherms:** Two main types of the interaction often describe adsorption of organic inhibitors on a corroding metal surface *viz.*, chemical adsorption and physical adsorption. It has been suggested that physisorbed molecules are attached to the metal at local cathodes and essentially retard metal dissolution by stifling the cathodic reaction whereas chemisorbed molecules protect anodic areas and reduce the inherent reactivity of the metal at the sites where they are attached. The more efficient inhibitors appear to protect anodic areas preferentially by chemisorption.

Basic information on the interaction between the inhibitor and mild steel surface can be provided by the adsorption isotherms. For this purpose, the values of surface coverage ($\theta$) at different concentrations of *Ocimum basilicum* extract in acid media in the temperature range from 298-328 K have been calculated to explain the best isotherm to determine the adsorption process. The value of the surface coverage ($\theta$) was calculated using the relationship[22]:

$$\theta = \frac{[I\,\%]}{100} \quad (6)$$

Attempts were made to fit these $\theta$ values to various isotherms including Langmuir, Temkin, Frumkin, El-Awady, Freundlich and Flory-Huggins, *etc*.

The best fit was obtained with Langmuir isotherm as suggested by the plot between C/$\theta$ and C (Fig. 3) and the linear correlation coefficient of the fitted data was close to 1, indicating that the adsorption of the inhibitor molecules obey the Langmuir adsorption isotherm[23,24]:

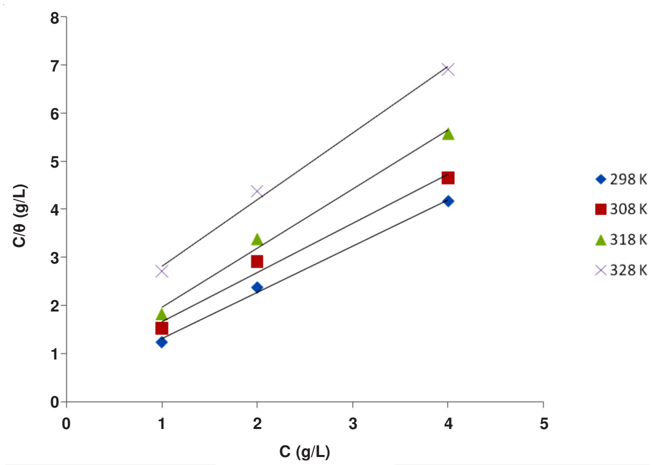


Fig. 3. Langmuir adsorption isotherm for adsorption of *Ocimum basilicum* extract on the surface of mild steel in 0.5M $H_2SO_4$

$$\left[\frac{C}{\theta}\right] = C + \left[\frac{1}{K_{ads}}\right] \quad (7)$$

where C is the inhibitor concentration and $K_{ads}$ is the equilibrium constant for adsorption/desorption process of the inhibitor molecules on the metal surface. $K_{ads}$ values were calculated from the intercept of the plot for adsorption process. The adsorption equilibrium constant, $K_{ads}$, is related to the standard free energy ($\Delta G^o_{ads}$) by the following equation[25]:

$$K_{ads} = \left[\frac{1}{55}\right]\exp\left[\frac{-\Delta G^o_{ads}}{RT}\right] \quad (8)$$

The plotting of log $K_{ads}$ against 1/T (Fig. 4) gives the value of the standard free energy ($\Delta G^o_{ads}$) which is equal to -34.62 kJ/mol. The negative values of $\Delta G^o_{ads}$ ensure the spontaneity of the adsorption process and the stability of the adsorbed layer on the mild steel surface. It is well known that values of $\Delta G^o_{ads}$ of the order of -20 kJ/mol or lower are consistent with the electrostatic interaction between organic molecules and metal surface, indicating a physisorption and the value around -40 kJ /mol or higher involve charge sharing to transfer from the organic molecules to the metal surface to form a co-ordinate type of bond, indicating a chemisorption[26]. The calculated $\Delta G^o_{ads}$ value indicates that the adsorption mechanism of the investigated extract on mild steel in 0.5M $H_2SO_4$ solution is a typical of chemisorption. Since the major constituents of the extract of *Ocimum basilicum* are polyphenol (tannins) and flavanoids which contains oxygens atoms and $\pi$-bonds in the rings, it may be possible that these organic molecules adsorbed on metal surface and inhibited the corrosion.

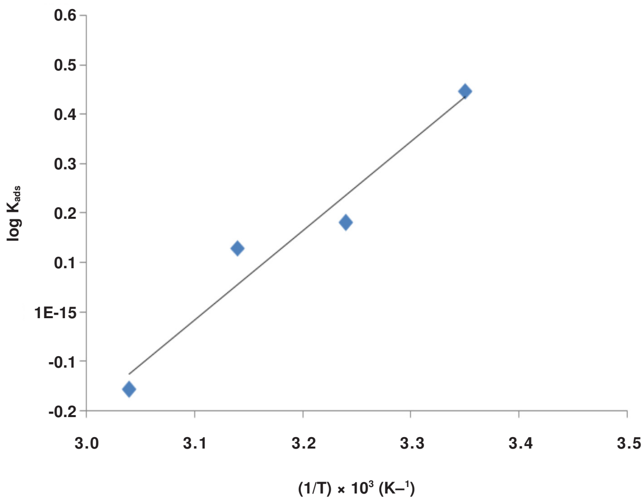


Fig. 4. Plots of log $K_{ads}$ against 1/T for mild steel in absence and presence of different concentrations of *Ocimum basilicum* extract in 0.5M $H_2SO_4$

By using the transition state equation[27]:

$$\log\left(\frac{i_{corr}}{T}\right) = \log\left(\frac{R}{Nh}\right) + \frac{\Delta S^o_{ads}}{2.303R} - \frac{\Delta H^o_{ads}}{2.303RT} \quad (9)$$

where N is the Avogadro's number and h is the plank's constant. Hence, a plot of log ($i_{corr}$/T) against 1/T gives a straight line as shown in Fig. 5 and the standard enthalpy change $\Delta H^o_{ads}$ is evaluated from the slope and the mean value of standard enthalpy change ($\Delta H^o_{ads}$) is found to be -62.49 kJ/mol. The standard adsorption entropy ($\Delta S^o_{ads}$) is calculated by using the following equation[28]:

$$\Delta G^o_{ads} = \Delta H^o_{ads} - T\Delta S^o_{ads} \quad (10)$$

The value of $\Delta S^o_{ads}$ is found to be 93.55 J $mol^{-1}$ $K^{-1}$ in presence of *Ocimum basilicum* in 0.5M $H_2SO_4$ solution which is large and positive, meaning that an increase in disordering takes place in going from reactants to the metal-adsorbed species reaction complex.

**Electrochemical impedance spectroscopy measurements:** Electrochemical impedance spectroscopy technique was applied to investigate the electrode/electrolyte interface and corrosion processes that occur on mild steel surface in presence and absence of *Ocimum basilicum* extract. To ensure complete characterization of the interface and surface processes, EIS measurements were made at open circuit potential in a wide

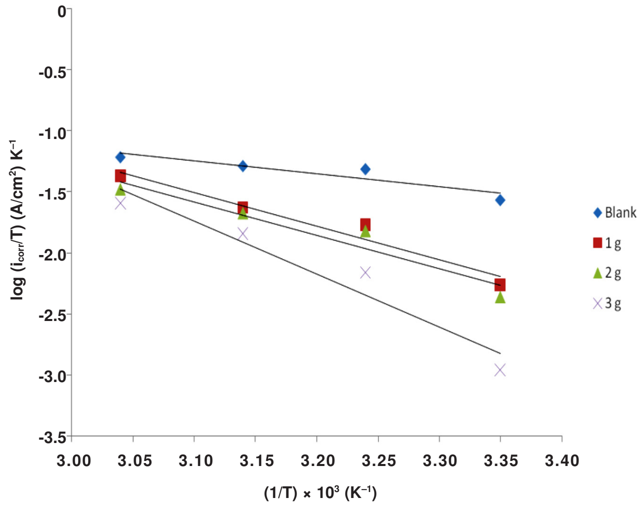


Fig. 5. Transition-state plots of log ($i_{corr}$/T) *versus* 1/T for mild steel corrosion in 0.5M $H_2SO_4$ in absence and presence of various concentrations of *Ocimum basilicum* extract

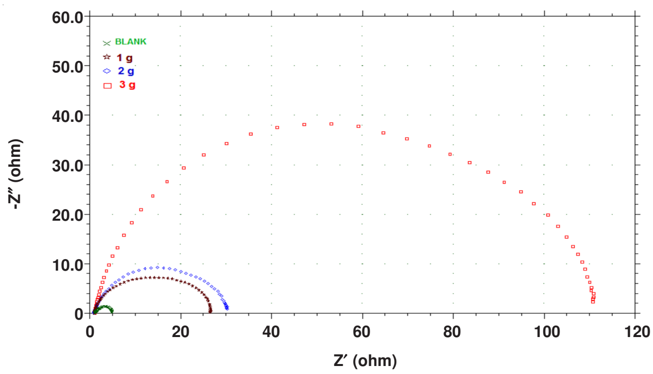


Fig. 6. Nyquist plots (EIS) of mild steel immersed in 0.5M $H_2SO_4$ in absence and presence of different concentrations of *Ocimum basilicum* extract at 298 K

frequency range at 298 K. Fig. 6 shows Nyquist plots for mild steel electrode immersed in 0.5M $H_2SO_4$ solution at 298 K in absence and presence of various concentrations of the extract at the respective open circuit potential. It is cleared from the Fig. 6 that the diameter of the semicircle increases with the increase in inhibitor concentration in the electrolyte, indicating an increase in corrosion resistance of the material.

The value of electrochemical double layer capacitance ($C_{dl}$) was calculated at the frequency, $f_{max}$ using the following equation[29]:

$$C_{dl} = \frac{1}{2\pi f_{max} R_{ct}} \quad (11)$$

where $f_{max}$ is the frequency at which the imaginary component of the impedance is maximal.

The impedance data listed in the Table-4 indicate that the values of both $R_{ct}$ and I % are found to increase by increasing in inhibitor concentration, while the values of $C_{dl}$ are found to decrease. This behaviour can be attributed to a decrease in dielectric constant and/or an increase in the thickness of the electric double layer, suggesting that the inhibitor molecules act by adsorption mechanism at mild steel/acid interface[29].

**Scanning electron microscopy studies:** SEM micrograms of the polished surface of mild steel exposed for 5 h in

TABLE-4
ELECTROCHEMICAL IMPEDANCE PARAMETERS FOR MILD STEEL CORROSION IN 0.5M $H_2SO_4$ SOLUTION IN ABSENCE AND PRESENCE OF DIFFERENT *Ocimum basilicum* EXTRACT CONCENTRATIONS

| Temp. (K) | Solution | Conc. (g/L) | $C_{dl}$ (F cm$^{-2}$) | $R_{ct}$ (Ω cm$^2$) | I (%) |
|---|---|---|---|---|---|
| 298 | 0.5M $H_2SO_4$ | 0.0 | $105 \times 10^{-3}$ | 4.1 | – |
| | | 1.0 | $11.84 \times 10^{-3}$ | 29.88 | 86.3 |
| | | 2.0 | $10.56 \times 10^{-3}$ | 33.49 | 87.8 |
| | | 3.0 | $4.3 \times 10^{-3}$ | 118.58 | 96.8 |

0.5M $H_2SO_4$ solutions in absence and presence of 3 g of *Ocimum basilicum* extract were shown in Fig. 7(a-b). In the comparison of the SEM micrograms, there were a rough surface on mild steel in absence of the extract and a smooth surface with deposited extract in presence of the extract[30,31]. This confirms that the extract inhibited corrosion of mild steel through adsorption of the inhibitor molecules on metal surface.

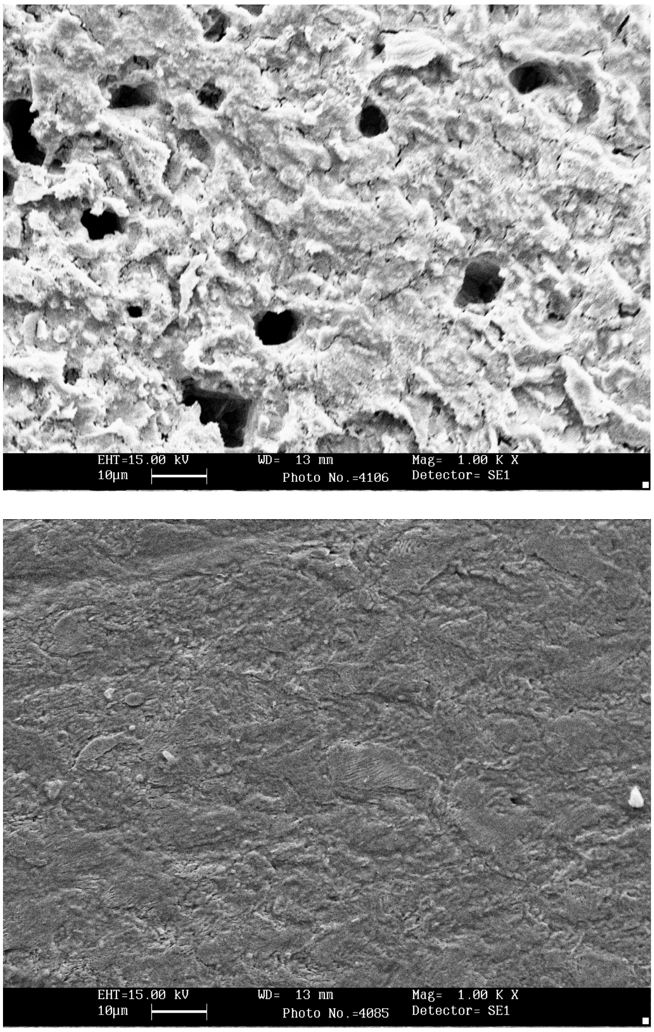


Fig. 7. (a) Scanning electron microgram of polished mild steel (1000 ×) after exposure to 0.5M $H_2SO_4$; (b) Scanning electron microgram of polished mild steel (1000 ×) after exposure to 0.5M $H_2SO_4$ containing 3 g of *Ocimum basilicum* extract

## Conclusion

The inhibition efficiency of *Ocimum basilicum* extract on corrosion of mild steel in 0.5M $H_2SO_4$ solution increases on increasing of concentration of the extract and decrease with rise in temperature. Potentiodynamic polarization measurement show that *Ocimum basilicum* acts as mixed type inhibitor. Adsorption of inhibitor molecules of extract on mild steel surface is found to obey Langmuir adsorption isotherm. The increase in the values of activation energies of corrosion process in presence of the extract indicates that *Ocimum basilicum* extract create a physical barrier to charge and mass transfer leading to reduction in corrosion rate of mild steel in 0.5M $H_2SO_4$ solution. The negative values of $\Delta G^o_{ads}$ and $\Delta H^o_{ads}$ highlight that the inhibition of corrosion of mild steel through adsorption is spontaneous and exothermic. Their values also reveal that the adsorption process is of chemisorptions in nature. EIS measurement reveals that charge transfer resistance increases with increase in concentration of the extract, indicating that the inhibition increases with increase in concentrations. SEM study confirms that corrosion inhibition of mild steel in 0.5M $H_2SO_4$ is due to adsorption of the extract on it.

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