Some natural aqueous extracts of plants as green inhibitor for carbon steel corrosion in 0.5 M sulfuric acid

M. Abdallah a,b, Hatem M. Altass b, B. A. AL Jahdaly b and M. M. Salem c

Chemistry Department, Faculty of Science, Benha University, Benha, Egypt; bChemistry Department, Faculty of Applied Science, Umm Al-Qura University, Makkah, Saudi Arabia; cChemistry Department, College of Science, Majmaah University, Al Majmaah, Saudi Arabia

ABSTRACT

The inhibiting impact of natural aqueous extracts of some plants such as curcumin, parsley and cassia bark extracts for the corrosion of carbon steel (C-steel) in 0.5 M H₂SO₄ solution was inspected utilizing some techniques such as galvanostatic and potentiodynamic anodic polarization and weight loss measurements. Outcomes indicated that the percentage inhibition efficiency increases with increasing the concentration of the extract due to its horizontal adsorption on the C-steel surface. The process of adsorption is followed by the Temkin isotherm. These natural extracts acted as pitting corrosion inhibitors by shifting the pitting potential to more noble values. The sequence of inhibition efficiency of the natural extracts decreases in the following order: cassia bark extract > parsley extract > curcumin extract. This arrangement is related to the molecular size of the major components of the three natural extracts used.

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1. Introduction

Sulfuric acid is used in many industrial applications, for example pickling and chemical cleaning of steel. Unluckily, the acidic solution caused the corrosion damage. The addition of the inhibitors is one of the effective methods used to protect the steel from the corrosive acid attack. Most of the inhibitors used are inorganic or organic compounds containing heteroatoms (1–16). These compounds are more efficient for the protection of steel from corrosion damage by its adsorption on the steel surface. Unfortunately, most of the synthetic compounds are toxic and cause damage to public health and the environment. Therefore, most researchers now tend to use some natural extracts for some plants to inhibit the corrosion attack of some metals and alloys in aqueous solutions (17–26).

In the previous work, aqueous extract of the leaves of henna (Lawsonia) was studied as an inhibitor for the corrosion of carbon steel (C-steel) in acidic solutions (27). The inhibiting action of the extract is discussed in view of adsorption of the complex formed between metal cations and Lawsonia molecules on the steel surface. Also, Guar gum (28) and some natural oils such as rosemary (29), parsley, lettuce and radish oils (30) are used as corrosion inhibitors for C-steel in aqueous solutions.

The purpose of this manuscript is to find an environmental-friendly, nontoxic, inexpensive and harmless to human health, efficient inhibitor created from the aqueous extract of some plants such as curcumin, parsley and cassia bark extracts to inhibit the corrosion of carbon steel in 0.5 M H₂SO₄ solution. The study is
conducted by some techniques such as galvanostatic polarization, potentiodynamic anodic polarization at scanning rates of 1 and 50 mV/s and weight loss measurements.

2. Experimental methods

2.1. Electrochemical measurements

The experiments were performed with C-steel of type L-52 used in Egyptian petroleum pipelines and has the following chemical composition (wt-%): 0.14 C, 0.6 Mn, 0.05 S, 0.04 P and the rest is Fe.

A cylindrical rod entrenched in Araldite with exposed surface area of 1 cm² was used for the electrochemical measurements such as galvanostatic and potentiodynamic polarization measurements. The exposed area was refined with different emery papers starting from coarser to finer, followed by degreasing with acetone and finally washed with distilled water twice, just before insertion in the electrolytic cell. The experiments were performed at the 23 ± 1°C using an air thermostat. The cell used in the electrochemical measurements contains three electrodes, C-steel as the working electrode, saturated calomel reference electrode (SCE) and a platinum foil auxiliary electrode.

The galvanostatic and potentiodynamic polarization at scan rates of 50 and 1 mV/s experiments were carried out using a PS remote potentiostat with PS6 software for calculation of some corrosion parameters.

2.2. Weight loss measurements

The weight loss measurements were accomplished in large test tubes suspended in a thermostated water bath. Each tube was open to air. In each experiment, 50 ml of the test solution was used. The test species were cut into 1.0 × 2.0 × 0.3 cm elements. Treatment of carbon steel coupons such as those mentioned in the electrochemical measurements. The cleaned C-steel coupons were weighed before and after immersion in 50 ml of the test solution for a period up to 8 h. The average weight loss for each identical experiments was taken and expressed in mg cm⁻².

2.3. Natural extracts

The natural extracts were obtained from a natural product company, Cairo, Egypt.

The main components in the three natural extracts are given in Table 1.

3. Results and discussion

3.1. Galvanostatic polarization

Figure 1 clarifies the impact of different concentrations of the cassia bark extract on the galvanostatic anodic and cathodic polarization curves of C-steel electrode in 0.5 M H₂SO₄ solutions. Analogous curves were also obtained for the parsley extract and curcumin extract, but it is not shown here. Some corrosion parameters were anodic (bₐ) and cathodic (bₜ) Tafel constants, corrosion potential (E₉رت), corrosion current density (I₉رت), surface coverage (θ) and the percentage inhibition efficiency (%IE). The corrosion parameters were calculated from the intercept of the anodic and cathodic Tafel lines and are present in Table 2.

![Figure 1](image-url)
The percentage inhibition efficiency (%IE) and surface coverage (θ) were calculated using equations (1) and (2):

$$\%IE = \left( \frac{l_{\text{uninh}} - l_{\text{inh}}}{l_{\text{uninh}}} \right) \times 100,$$

$$\theta = \left( \frac{l_{\text{uninh}} - l_{\text{inh}}}{l_{\text{uninh}}} \right).$$

where $l_{\text{uninh}}$ and $l_{\text{inh}}$ are the corrosion current densities in uninhibited and inhibited solution, respectively.

It is evident that from Figure 1 and Table 2, as the concentrations of the extract of curcumin, parsley and cassia bark increase, the polarization curves shift toward more negative potential and lower current density values. The values of $b_a$ and $b_c$ are diverse a bit, indicating the inhibition impact of these extracts by its adsorption on the C-steel due to blocking adsorption mechanism (31). Also, these natural extracts are categorized as a mixed type inhibitor. The values of $E_{\text{corr}}$ are slightly shifted toward negative direction, the values of $i_{\text{corr}}$ are lowered and the values of $\theta$ and consequently %IE increase. These results confirm the inhibitory effect of these extracts. The efficiency of inhibition of natural extracts is reduced in the following order:

cassia bark extract > parsley extract > curcumin extract.

### 3.2. Potentiodynamic anodic polarization

The potentiodynamic anodic polarization curves of C-steel electrode in 0.5 M H$_2$SO$_4$ containing different concentrations of cassia bark extract at a scanning rate of 50 mV/s is presented in Figure 2. Analogous curves were also obtained for the parsley extract and curcumin extract, but are not shown here.

It is obvious from this figure that there is only one anodic peak (A) followed by passive region before O$_2$ evolution.

Peak (A) was explicated due to the active dissolution of Fe into Fe$^{+2}$ ion according to a previous mechanism (32). At certain potential, the current drops to low values, indicating the onset of passivity. The electrode is considered now to be covered by a passivating oxide film of mainly ferric oxide ($\gamma$Fe$_2$O$_3$). This oxide can be created in the film covering the electrode surface by the oxidation of Fe$^{+2+}$ ions according to Abdallah and Megahed (33):

$$2\text{Fe} + 2 + 3\text{H}_2\text{O} \rightarrow \gamma\text{Fe}_2\text{O}_3 + 6\text{H}^+ + 2e^-.$$ 

Table 2. Corrosion parameters obtained from galvanostatic polarization measurements of C-steel in 0.5 M H$_2$SO$_4$ solution containing different concentrations of natural extracts of curcumin, parsley and cassia bark.

| Comp.          | Cons, ppm | $b_a$, mV dec$^{-1}$ | $b_c$, mV dec$^{-1}$ | $E_{\text{corr}}$, mV (SCE) | $i_{\text{corr}}$, mA cm$^{-2}$ | %IE  |
|----------------|-----------|----------------------|----------------------|-----------------------------|---------------------------------|------|
| 0.5 M H$_2$SO$_4$ | 0.5 M     | 69.5                 | 91                   | 478                         | 0.712                           | –    |
| Curcumin extract | 100       | 153                  | 152                  | 522                         | 0.389                           | 45.36|
|                | 200       | 156                  | 170                  | 541                         | 0.340                           | 52.25|
|                | 300       | 173                  | 180                  | 544                         | 0.242                           | 66.01|
|                | 400       | 180                  | 188                  | 543                         | 0.165                           | 76.82|
|                | 500       | 193                  | 200                  | 551                         | 0.135                           | 81.04|
| Parsley extract | 100       | 143                  | 141                  | 533                         | 0.372                           | 47.75|
|                | 200       | 164                  | 176                  | 537                         | 0.295                           | 58.56|
|                | 300       | 168                  | 181                  | 541                         | 0.212                           | 70.22|
|                | 400       | 177                  | 188                  | 543                         | 0.144                           | 79.77|
|                | 500       | 190                  | 196                  | 548                         | 0.116                           | 83.70|
| Cassia bark extract | 100   | 141                  | 165                  | 511                         | 0.366                           | 48.59|
|                | 200       | 148                  | 157                  | 522                         | 0.298                           | 58.14|
|                | 300       | 157                  | 178                  | 537                         | 0.186                           | 72.51|
|                | 400       | 165                  | 183                  | 538                         | 0.142                           | 80.05|
|                | 500       | 175                  | 193                  | 541                         | 0.095                           | 86.66|

Table 3. Corrosion parameters obtained from potentiodynamic anodic polarization measurements of C-steel in 0.5 M H$_2$SO$_4$ containing different concentrations of natural extracts of curcumin, parsley and cassia bark at a scan rate 50 mV/s.

| Comp.          | Conc, ppm | $i_{\text{p}}$, mA cm$^{-2}$ | $E_{\text{corr}}$, V (SCE) | %IE  |
|----------------|-----------|-------------------------------|-----------------------------|------|
| 0.5 M H$_2$SO$_4$ | 0.5 M     | 4.99                          | 0.76                         | –    |
| Curcumin extract | 100       | 2.74                          | 0.74                         | 45.09|
|                | 200       | 2.46                          | 0.69                         | 50.70|
|                | 300       | 1.62                          | 0.65                         | 67.53|
|                | 400       | 1.13                          | 0.60                         | 77.35|
|                | 500       | 0.88                          | 0.54                         | 82.36|
| Parsley extract | 100       | 2.69                          | 0.72                         | 46.09|
|                | 200       | 2.35                          | 0.74                         | 52.90|
|                | 300       | 1.54                          | 0.68                         | 69.14|
|                | 400       | 1.08                          | 0.62                         | 78.35|
|                | 500       | 0.77                          | 0.56                         | 84.57|
| Cassia bark extract | 100   | 2.65                          | 0.66                         | 46.89|
|                | 200       | 2.30                          | 0.63                         | 53.91|
|                | 300       | 1.49                          | 0.49                         | 70.14|
|                | 400       | 1.01                          | 0.38                         | 79.75|
|                | 500       | 0.62                          | 0.35                         | 87.57|
As the concentration of the extract increases, the passive film increases as shown in Figure 2. As the potential become positive, the current rises again due to the evolution of oxygen according to

$$2\text{H}_2\text{O} \rightarrow \text{O}_2 + 4\text{H}^+ + 4\text{e}.$$  \hspace{1cm} (4)

The values of peak current density ($i_p$) and the peak potential ($E_p$) are calculated and listed in Table 3.

The percentage inhibition efficiency ($\%\text{IE}$) was calculated from the following equation (34):

$$\%\text{IE} = \left( \frac{i_p(\text{uninh}) - i_p(\text{inh})}{i_p(\text{uninh})} \right) \times 100,$$  \hspace{1cm} (5)

where $i_p(\text{inh})$ and $i_p(\text{uninh})$ are the peak current densities in the presence and absence of inhibitors.

Inspection of the curves of Figure 2 and Table 3 indicates that, as the concentration of natural extracts increases, the values of $E_p$ are shifted to more positive values and $i_p$ to lower values and the values of $\%\text{IE}$ increases. This indicates an increased resistance to the active dissolution C-steel. The values of $\%\text{IE}$ of the natural extracts decrease in the following order:

cassia bark extract > parsley extract > curcumin extract.

### 3.3. Natural extract as pitting corrosion inhibitors

Figure 3 represents the effect of the addition of different concentrations of the natural cassia bark extract on the potentiodynamic anodic polarization curves of C-steel electrode in 0.5 M H$_2$SO$_4$ containing 0.5 M NaCl as pitting corrosion agent at a scanning rate of 1 mV/s. Analogous curves were also obtained for the parsley extract and curcumin extract, but are not shown here. It was found that the pitting potential of the C-steel electrode shifts to more positive (noble) values with increasing the concentration of these natural extracts. This indicates an increased resistance to pitting attack (35, 36).

Figure 4 represents the relationship between pitting potential and the logarithm of the molar concentration of the added compounds. Straight lines were obtained and the following conclusions can be drawn:

The increase of inhibitor concentration causes the shift of the pitting potential into more positive values in accordance with the following equation (28, 37):

$$E_{\text{pitt}} = a_2 + b_2 \log C_{\text{inh}},$$  \hspace{1cm} (6)

where $a_2$ and $b_2$ are constants which depend on both the composition of additives and the nature of the electrode.

Inhibition afforded at the same concentrations of the natural extracts decreases in the following order:

cassia bark extract > parsley extract > curcumin extract.

### 3.4. Weight loss measurements

The effect of increasing concentrations of the natural cassia bark extract on the weight loss of C-steel electrode in 0.5 M H$_2$SO$_4$ solution is presented in Figure 5. The same curves were obtained for the parsley extract and curcumin extract, but are not shown. As shown from these figures, it is obvious that by increasing the concentration of the natural extract, the weight loss of C-steel is decreased. This means that the presence of these extracts retards the corrosion of C-steel in 0.5 M H$_2$SO$_4$ solution.
The linear relationship obtained in Figure 5 indicates the absence of insoluble surface film during corrosion. In the absence of any surface films, the inhibitors are first adsorbed onto the metal surface and thereafter affect corrosion.

The corrosion rate values \( k \) \( (\text{mg cm}^{-2} \text{min}^{-1}) \) were calculated from equation (7):

\[
\text{Corrosion rate } (k) = \frac{\text{loss in weight (mg cm}^{-2})}{\text{time (min)}}. \quad (7)
\]

The inhibition efficiency (%IE) and the surface coverage (\( \theta \)) of the natural extract were calculated from the following equations:

\[
\%\text{IE} = 1 - \frac{K_{\text{inh}}}{K_{\text{uninh}}} \times 100, \quad (8)
\]

where, \( K_{\text{inh}} \) and \( K_{\text{uninh}} \) are the corrosion rate in devoid of and containing the natural extracts, respectively.

The values of \( k \), \( \theta \) and %IE for various concentrations of three natural extracts are given in Table 4. The obtained data from Table 4 reveal that the inhibition efficiency of these natural extracts is arranged in the following order:

\[
\text{cassia bark extract} > \text{parsley extract} > \text{curcumin extract}.
\]

### 3.5. Adsorption isotherms and explanation of inhibition

The inhibition of general and localized pitting corrosion of C-steel in 0.5 M \( \text{H}_2\text{SO}_4 \) solution by some natural extracts, e.g. curcumin extract, parsley extract and cassia bark extract, were inspected. The primary step of the inhibitory action of these aqueous extract toward the corrosion of C-steel in 0.5 M \( \text{H}_2\text{SO}_4 \) solution is usually by the adsorption of these extracts on the steel surface. The adsorption of the natural extracts on the steel surface is considered as an alternative adsorption process between the natural extract compounds in the aqueous solution \( (\text{Ext}_{\text{aq}}) \) and the water molecules adsorbed on the C-steel surface \( (\text{H}_2\text{O})_{\text{ads}} \)

\[
\text{Ext}_{\text{aq}} + x(\text{H}_2\text{O})_{\text{ads}} \rightarrow \text{Ext}_{\text{ads}} + x\text{H}_2\text{O}_{\text{aq}},
\]

where \( x \) is the ratio of the number of water molecules substituted by one molecule of extract adsorbate. The adsorption process depends on the chemical structure of the

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**Table 4. Variation of degree of surface coverage (\( \theta \)), corrosion rate (\( k \)) and percentage inhibition efficiency (%IE) of C-steel in the presence of different concentrations of curcumin, parsley and cassia bark extracts.**

| Comp.          | Conc, ppm | \( k \), mg cm\(^{-2}\) min\(^{-1}\) | \( \theta \) | %IE  |
|----------------|-----------|---------------------------------|-------------|------|
| 0.5 M \( \text{H}_2\text{SO}_4 \) | 0.5 M \( \text{H}_2\text{SO}_4 \) | 7.35 | – | – |
| Curcumin extract | 100 | 4.12 | 0.439 | 43.94 |
| | 200 | 3.45 | 0.531 | 53.06 |
| | 300 | 2.46 | 0.665 | 66.53 |
| | 400 | 1.75 | 0.762 | 76.19 |
| | 500 | 1.27 | 0.827 | 82.72 |
| Parsley extract | 100 | 3.85 | 0.476 | 47.62 |
| | 200 | 3.38 | 0.540 | 54.01 |
| | 300 | 2.25 | 0.694 | 69.38 |
| | 400 | 1.58 | 0.785 | 78.50 |
| | 500 | 1.12 | 0.848 | 84.76 |
| Cassia bark extract | 100 | 3.73 | 0.492 | 49.25 |
| | 200 | 3.12 | 0.575 | 57.55 |
| | 300 | 2.08 | 0.717 | 71.70 |
| | 400 | 1.42 | 0.807 | 80.68 |
| | 500 | 0.88 | 0.880 | 88.02 |
extract and the presence of the active group in it, the number of adsorption active centers in the molecule and their charge density, molecular size, mode of adsorption, the nature of the metal surface used, temperate, type of the corrosive acidic solution, and the potential of the metal–solution interface.

Trials were made to insert the \( \theta \) values to some adsorption isotherm such as Temkin, Frumkin, Frundich and Langmuir isotherms. In the present work, the adsorption process obeys the Temkin's adsorption isotherm (38), according to the below equation:

\[
\exp \left( \frac{-2a \theta}{K_{ads}} \right) = C_{inh}, \tag{11}
\]

where \( K \) is the equilibrium constant of the adsorption process, \( a \) is the molecule interaction parameter and \( C \) is the inhibitor concentration in the bulk solution.

Figure 6 clarifies the relation between \( \theta \) and \( \log C \) for C-steel in the presence of natural extracts. Straight lines were obtained indicating that the adsorption of these natural extracts obeys Temkin’s adsorption isotherm. The natural extracts block the reaction sites on the surface of C-steel samples by adsorption and reduce the available area for further corrosion reaction.

The values of %IE obtained by galvanostatic and potentiodynamic anodic polarization and weight loss techniques indicate that the extent of inhibition efficiency of the natural extracts toward the corrosion of C-steel in 0.5 M H\(_2\)SO\(_4\) solution obeys the following order:

cassia bark extract > parsley extract > curcumin extract.

The inhibiting vigor of natural extracts could be interpreted by strong blocking adsorption on the C-steel surface due to the high molecular weight of these natural extracts. We expect horizontal adsorption of its components on the C-steel surface. The adsorbed layer acts as a barrier between the C-steel surface and corrosive H\(_2\)SO\(_4\) solution leading to a decrease in the corrosion rate. This difference in the inhibition efficiencies could be explained on the basis of the molecular size. The three natural extracts had a high molecular size and this led to facilitate the adsorption process and hence increase the surface coverage.

The values of %IE which were evaluated for the three natural extracts used toward the corrosion of carbon steel in 0.5 M H\(_2\)SO\(_4\) solution using different techniques show an agreement and conformity of the experimental results. However, there is a small difference in the values obtained from the different techniques. This observed discrepancy could be attributed to the differences of the experimental condition.

4. Conclusions

1. Curcumin, parsley and cassia bark extracts inhibit corrosion of C-steel in 0.5 M H\(_2\)SO\(_4\) solution.
2. The inhibition efficiency of the three extracts used increases with increasing the concentration of the extract.
3. The polarization curves proved that the natural extracts act as a mixed inhibitor.
4. The inhibition was explained in view of its horizontal adsorption on the surface of the steel.
5. The process of adsorption follows Temkin’s isotherm.
6. The order of the inhibition efficiency depends on the molecular size of the major component of three natural extract used.
7. The natural extracts inhibit the pitting corrosion of C-steel by shifting the pitting potential to more noble values.

Disclosure statement

No potential conflict of interest was reported by the authors.

Notes on contributors

M. Abdallah is a Professor of physical chemistry since 2004. He published more than 130 papers in international journals. Most publications are in the field of corrosion inhibition of some metals and alloys in aqueous solutions. Focusing on plant extract, natural oils, surfactant compounds, polymer compounds, organic and inorganic compounds. He supervisor of 40 Master Degree and 18 Ph.D. Degree.

Hatem M. Altass is an assistant professor of physical chemistry, Dean of Faculty of Applied Science, Umm Al Qura university, Makkah, Saudi Arabia. He published some papers in nanotechnology science and corrosion inhibition of metals and alloys in international journals.

B. A. AL Jahdaly is an assistant professor of physical chemistry, Faculty of Applied Science, Umm Al Qura university, Makkah, Saudi Arabia. She has published some papers in corrosion and electrochemistry fields.

M. M. Salem is an assistant professor of physical chemistry Faculty of Science in Zulfi, Majmaah University, Saudi Arabia. She has published some papers in corrosion inhibition of metals and alloys in different media.

ORCID

M. Abdallah © http://orcid.org/0000-0002-6132-8849

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