

## Research Article

# Pomegranate (*Punica granatum*) Peel Extract as a Green Corrosion Inhibitor for Mild Steel in Hydrochloric Acid Solution

Habib Ashassi-Sorkhabi, Shoja Mirzaee, Taghi Rostamikia, and Robabeh Bagheri

Electrochemistry Research Lab, Physical Chemistry Department, Faculty of Chemistry, University of Tabriz, Tabriz 5166616471, Iran

Correspondence should be addressed to Habib Ashassi-Sorkhabi; [habib.ashassi@gmail.com](mailto:habib.ashassi@gmail.com)

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The inhibition effect of pomegranate peel extract (PPE) on the corrosion of mild steel in hydrochloric acid (HCl) solution was investigated. The polarization, mass loss, and electrochemical impedance techniques were used to evaluate the corrosion inhibition performance of the pomegranate peel extract. The results revealed that PPE acts as a corrosion inhibitor in HCl solution. The inhibition efficiency increased with the increase of extract concentration. The inhibition action was attributed to the adsorption of the chemical compounds present in the extract solution, on mild steel surface.

## 1. Introduction

Corrosion inhibitors are widely used in industry to reduce the corrosion rate of metals and alloys in contact with aggressive environments. Most of the corrosion inhibitors are synthetic chemicals and are expensive and very hazardous to the environment. Therefore, finding cheap and environmentally safe inhibitors is needed [1–3]. There are some reports on the inhibition effects of nontoxic compounds on the corrosion of metals [4–9]. The inhibition effect of amino acids on steel [1] and aluminum [10] corrosion and *Prunus cerasus* juice [11] on steel corrosion in acidic media has been reported. On the other hand, rare earth metals have been proposed as corrosion inhibitors [12–15]. The inhibition effects of some nontoxic organic compounds have been also reported for steel corrosion [16, 17] but they are quite expensive.

Pomegranate (*Punica granatum* L.) is one of the important fruits grown in Turkey, Iran, the USA, the Middle East, and Mediterranean and Arabic countries. It is originated from southeast Asia [18]. The edible part of the fruit contains considerable amount of acids, sugars, vitamins, polysaccharides, polyphenols, and important minerals [19, 20]. It has been reported that pomegranate juice has potent antiatherogenic effects on the health of humans and atherosclerotic effects on mice that may be attributable to the juice's antioxidative properties [21]. The peel part of pomegranate contains

hydroxyl, carbonyl, and aromatic groups with considerable amount of punicalagin [22], punicalin [23], granatin A [24], granatin B [25], maleic acid [26], ursolic acid [27], gallic acid [28], and antioxidant materials [29]. These substances with effective constitutive chemical groups in their structure could show corrosion inhibition performance.

The aim of this study is to investigate the inhibition effect of PPE as a cheap, raw, and nontoxic inhibitor for steel corrosion in hydrochloric acid. Electrochemical measurements and mass loss methods were employed to evaluate and investigate the inhibition efficiency of pomegranate peel extract (PPE).

## 2. Experimental Details

**2.1. Materials and Alloy Samples.** The metal substrate used in this work was mild steel, which had the chemical composition as shown in Table 1. The pomegranate peel powder was purchased from a local market in Tabriz, Iran, and the HCl 37% (Merck, Germany) was used to prepare the aggressive solution (1 M HCl). The concentrations of PPE, employed as inhibitor, were 0.0625, 0.125, 0.25, 0.5, and 1% v/v in 1 M HCl.

All solutions were prepared using the double-distilled water. In the weight loss method for corrosion evaluation, the steel samples with dimensions of 2 cm × 1 cm × 0.01 cm were immersed in the testing solutions for 24 h at 25°C.

TABLE I: Chemical composition of mild steel specimens obtained from quantometric method.

Element	C	Si	Mn	S	Mo	Ni	Al	Co
w/w %	0.0008	2.080	0.260	0.027	0.005	0.018	0.16	0.353
Element	Nb	Ti	V	W	Pb	Sn	Mg	Zr
w/w %	0.003	0.002	0.007	0.002	0.014	0.001	0.001	0.002
Element	Ca	Sb	Ta	B	Zn	N	Cu	Ce
w/w %	0.002	0.004	0.002	0.000	0.001	0.001	0.009	0.001
Element	As	Cr	Fe	As	Cr			
w/w %	0.002	0.034	97.148	0.002	0.034			

2.2. *Pomegranate Peel Extraction Procedure Using Methanol.* Sifted pomegranate peel powder by a 60-mesh sieve was employed for preparation of solution. Then, the solution slowly was stirred and dried at 30°C for about 4 hours and filtered to obtain a homogenous solution. Moreover, methanol was extracted from the mixture by rotary distillation method [30]. Finally, in this procedure, 13.8 g of extract was obtained from 100 g pomegranate peel powder and was stored in a glass bottle for future uses.

2.3. *Electrochemical Measurements.* Electrochemical experiments were carried out using an Autolab Potentiostat-Galvanostat (PGSTAT 30). A conventional three-electrode system applied for electrochemical studies. The working electrode was a mild steel sheet, mounted in polyester, so that the exposed area was one cm<sup>2</sup>. The test surfaces of the specimens were polished with emery paper number 400 to 1200 grade, then cleaned by acetone, washed with double-distilled water, and finally dried at room temperature before immersion in the test solutions. A saturated calomel electrode and a platinum sheet (approximately one cm<sup>2</sup> surface area) were used as reference and counter electrodes, respectively.

In the case of polarization measurements, the scan rate was one mV/s. The immersion time to achieve an equilibrium potential before each electrochemical measurement was 40 min. The impedance measurements were carried out in the frequency range of 10 kHz to 10 mHz at the open circuit potential, by applying 10 mV sine wave ac voltage. In addition, the impedance data was analyzed using ZView (II) software to determine the parameters of the proposed equivalent electrical circuit models.

Meanwhile, the constant phase element (CPE) and the charge transfer resistance ( $R_{ct}$ ) were calculated from Nyquist plots as described elsewhere [30]. It should be noted that all experiments were performed under the atmospheric ambient. Furthermore, the solutions temperature was controlled using a Memmert thermostat (Germany) in all experiments.

### 3. Results and Discussion

3.1. *Electrochemical Tests.* Tafel polarization technique was used to evaluate the corrosion inhibition efficiency of PPE. Figure 1 shows Tafel curves for potentiodynamic behavior of mild steel in hydrochloric acid containing the different concentration of extract inhibitor.

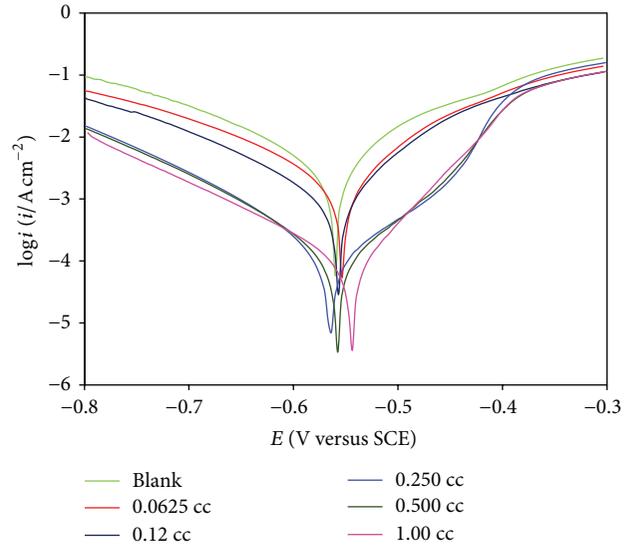


FIGURE 1: The representative Tafel polarization plots for mild steel corrosion in 1M HCl solution in the absence and presence of different concentration of PPE inhibitor.

Also, electrochemical parameters including corrosion potential,  $E_{corr}$ , polarization resistance,  $R_p$ , cathodic Tafel slop,  $b_c$ , anodic Tafel slop,  $b_a$ , corrosion current density,  $I_{corr}$ , and inhibition efficiency percent, IE%, derived from the Tafel curves are given in Table 2.

The following equation was employed to calculate inhibition efficiency (IE) from polarization measurements [2, 7]:

$$IE\% = \left(1 - \frac{i}{i_o}\right) \times 100, \quad (1)$$

where  $i$  and  $i_o$  are the corrosion current densities obtained by extrapolation of the cathodic and anodic Tafel lines, in inhibited and uninhibited solutions, respectively.

It is possible to infer that the inhibition efficiency improved with increasing the concentration of PPE inhibitor. This may be attributed to the improvement in PPE adsorption on metal surface, which leads to its corrosion protection performance. Furthermore, it is clear that, in the presence of PPE inhibitor, while the  $E_{corr}$  values shifted to more positive, there is no considerable variation with different concentration of inhibitor.

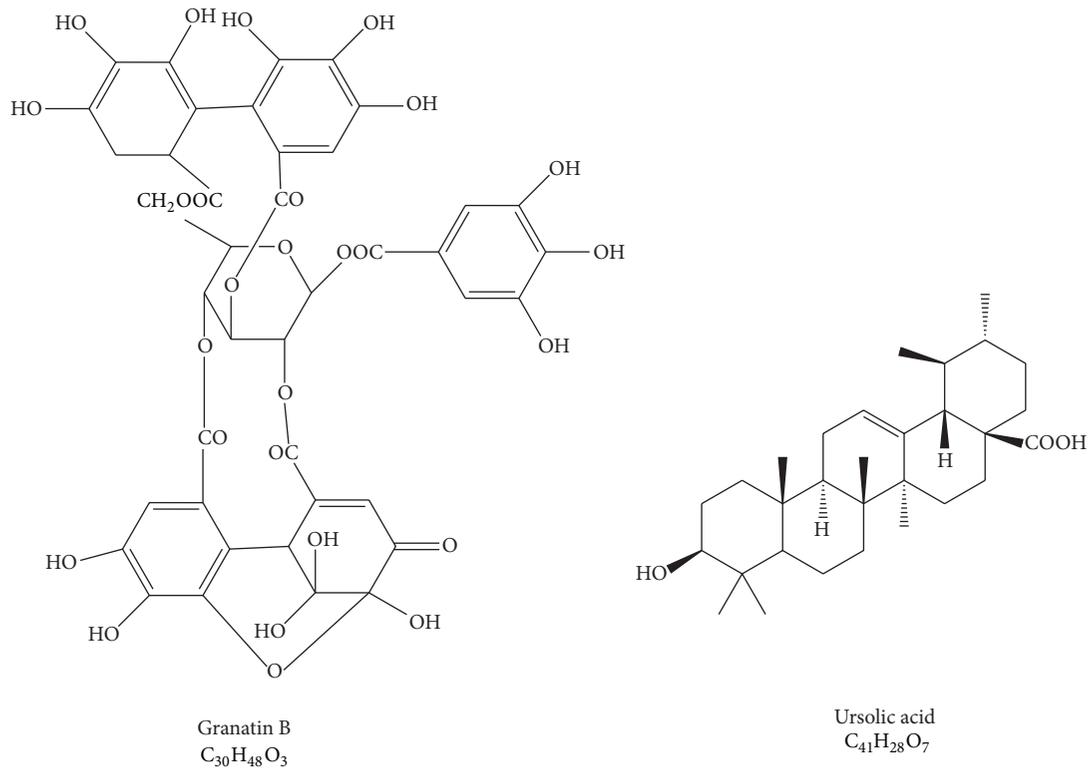


FIGURE 2: The molecular structures of granatin B and ursolic acid.

TABLE 2: The corrosion parameters obtained from polarization plots of mild steel in blank and inhibited HCl solution.

Inhibitor concentration (v/v %)	$E_{\text{corr}}$ (mV)	$R_p$ ( $\Omega \text{ cm}^2$ )	$b_a$ (mV/dec)	$b_c$ (mV/dec)	$I_{\text{corr}}$ ( $\mu\text{A cm}^{-2}$ )	IE%
0.000	-0.548	6.052	0.0103	0.0103	4.1440	—
0.625	-0.550	18.70	0.0790	0.0127	1.1300	72
0.125	-0.550	42.240	0.0760	0.0120	0.047	88
0.250	-0.553	131.40	0.0710	0.0107	0.0414	97
0.500	-0.573	400.05	0.0880	0.0126	0.0056	98
1.000	-0.553	400.80	0.0730	0.0102	0.0038	99

It is well known that the pomegranate peel contains the organic acids with phenolic, hydroxyl, and carbonyl groups and antioxidant materials [29, 31]; some of these organic compounds have been used as organic corrosion inhibitors for metals [32]. On the other hand, the adsorption of organic molecules such as phenolic compounds (gallic acid and granatin B, Figure 2) may be due to the presence of an oxygen atom (a heteroatom),  $\pi$  electron of aromatic rings, and electron donating groups. In other words, the heteroatoms such as oxygen are the major adsorption center in organic compounds for its interaction with the metal surface [33]. The adsorption can also occur via electrostatic interaction between a negatively charged surface, which is provided by a specifically adsorbed anion ( $\text{Cl}^-$ ) on iron, and the positive charge of the inhibitor [34]. Furthermore, according to literature [35] organic acid component itself can form passive film on substrate surface. From the presented investigation,

it can be deduced that the inhibitor molecules adsorb on the metal surface by blocking the active corrosion sites [17].

As a result, the presence of PPE caused a potential shift toward more positive values, indicating the anodic reaction is inhibited. We can conclude that decreasing the oxidation rate of substrate in corrosive media is due to the formation of protective layer.

Figures 3 and 4 show the Nyquist diagrams in the absence and presence of different concentrations of PPE inhibitor. One capacitive depressed semicircle is present for the samples in the Nyquist diagrams. Analysis of the impedance spectra was performed by means of Randles circuit, which is the most common equivalent circuit. In addition, Table 3 presents the calculated data obtained by ZView fitting program.  $R_s$  and  $R_{ct}$  are the solution resistance and the charge transfer resistance, respectively, and the CPE is constant phase element for the double-layer capacitance.

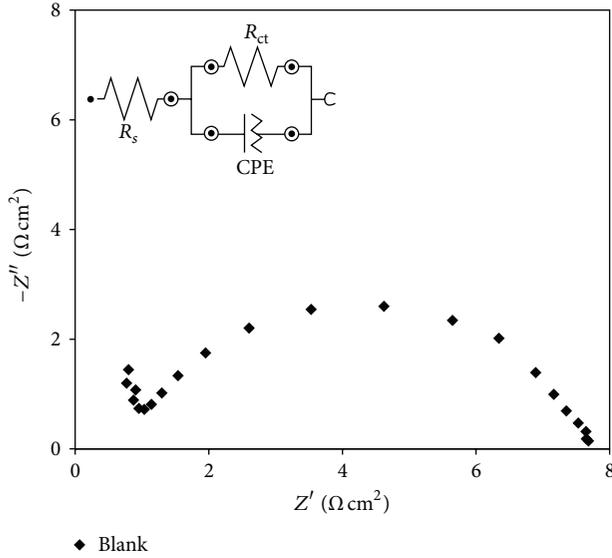


FIGURE 3: The representative Nyquist diagrams for mild steel corrosion in 1 M HCl solution in the absence of inhibitor at 25°C.

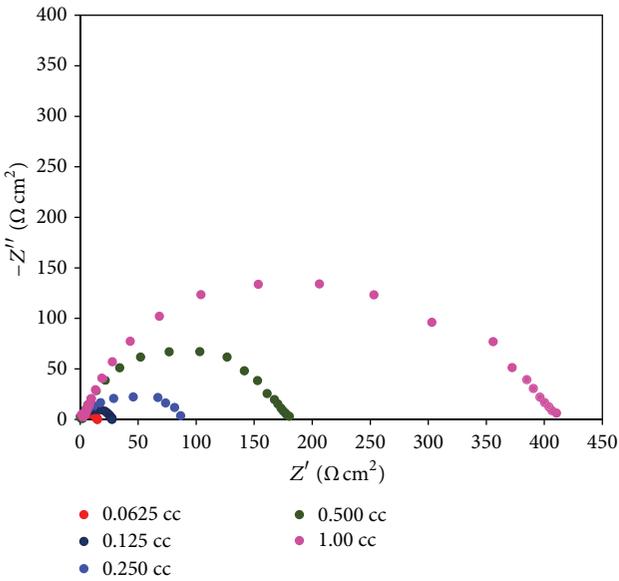


FIGURE 4: The representative Nyquist diagrams for mild steel corrosion in 1 M HCl solution in the presence of different concentration of inhibitor at 25°C.

The  $R_{ct}$  values are calculated from the difference in impedance at lower and higher frequencies, as suggested by Elkadi et al. [36]. To obtain the double-layer capacitance ( $C_{dl}$ ), the frequency at maximum ( $-Z_{imax}$ ) imaginary component of impedance was determined and  $C_{dl}$  values were calculated from the following equation [36]:

$$f(-Z_{imax}) = \frac{1}{2\pi C_{dl} R_{ct}}. \quad (2)$$

It is well known that the corrosion of mild steel is obviously inhibited in the presence of the inhibitor and the  $R_{ct}$  values significantly improved by increasing the PPE inhibitor

TABLE 3: The parameters obtained from EIS measurements of mild steel in blank and inhibited HCl solution.

Inhibitor concentration (v/v %)	$R_s$ ( $\Omega \text{ cm}^2$ )	$R_{ct}$ ( $\Omega \text{ cm}^2$ )	$C_{dl}$ ( $\mu\text{F cm}^{-2}$ )	IE%
0.000	1.1	6	$1.84 \times 10^{-4}$	—
0.625	1.1	20	$7.72 \times 10^{-5}$	69
0.125	1.0	41	$3.48 \times 10^{-5}$	85
0.250	0.1	111	$1.94 \times 10^{-5}$	94
0.500	0.1	348	$1.61 \times 10^{-5}$	98
1.000	0.8	378	$1.24 \times 10^{-5}$	99

TABLE 4: Weight loss data of mild steel in blank and inhibited HCl solution.

Inhibitor concentration (v/v %)	Weight loss (g)	$\eta_w$ %
0.000	0.18	—
0.625	0.07	61
0.125	0.032	82
0.250	0.030	83
0.500	0.014	92
1.000	0.009	95

concentration. The value of  $R_{ct}$  increases from six  $\Omega \text{ cm}^2$  (in the absence of PPE inhibitor) to 378  $\Omega \text{ cm}^2$  (in the presence of PPE inhibitor) and corresponding  $C_{dl}$  value decreases from  $1.836 \times 10^{-4}$  to  $1.24 \times 10^{-5} \mu\text{F cm}^{-2}$ . In other words, as the inhibitor concentration increased, the  $R_{ct}$  values increased, but the  $C_{dl}$  values tended to decrease. This is mainly due to the adsorption of inhibitor on the metal surface [37]. In the case of the electrochemical impedance spectroscopy, the inhibition efficiency was calculated using the charge transfer resistances as follows [36]:

$$\text{IE\%} = \frac{R_{ct(\text{inh})} - R_{ct}}{R_{ct(\text{inh})}} \times 100, \quad (3)$$

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

Comparing the calculated results confirmed that the inhibition efficacy (IE%) of PPE inhibitor was enhanced by increasing its concentration. This phenomenon is consistent with results obtained by polarization method. Therefore, there is relatively good agreement between the polarization resistances, obtained from both electrochemical methods in the presence of high concentration range of PPE.

**3.2. Weight Loss Measurements.** Table 4 shows the weight loss data of mild steel in 1 M HCl in the absence and presence of various concentrations of inhibitors. The corrosion efficiencies ( $\eta_w$ %) were calculated according to [38] by the following equation:

$$\eta_w\% = \frac{W_{\text{corr}} - W_{\text{corr}(\text{inh})}}{W_{\text{corr}}} \times 100, \quad (4)$$

where  $W_{\text{corr}}$  and  $W_{\text{corr}(\text{inh})}$  are the weight loss of mild steel in the absence and presence of the inhibitor, respectively.

The results show that the corrosion efficiency increases with increasing inhibitor concentration.

These observations verify the results obtained from polarization and EIS measurements in the presence of high concentration range of extract.

#### 4. Conclusion

The obtained results from electrochemical and weight loss methods showed that the pomegranate peel extract acts as a nontoxic, cheap, and easily prepared inhibitor for corrosion of mild steel in hydrochloric acid media. Corrosion inhibition action of pomegranate juice increases as its concentration increases in the corrosive solution. Moreover, inhibition behavior of pomegranate juice may be explained by adsorption of constitutive organic compounds in PPE on the metal surface.

#### Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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