

Turmeric extract as a biocompatible inhibitor of mild steel corrosion in 3.5%NaCl solution

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Abstract

Corrosion inhibitory impacts of turmeric with concentrations of 200-800 ppm on the electrochemical behavior of mild steel were studied in the medium of NaCl 3.5% using several techniques such as electrochemical impedance spectroscopy (EIS) and polarization. Scanning electron microscopy (SEM) was used for analyzing the surface of mild steel after 24 hours of immersion in the electrolyte solution, with and without turmeric. On the one hand, the results of polarization and EIS demonstrated that for the steel sample immersed in a salt solution containing 800 ppm of turmeric extract, the corrosion current density was decreased and the corrosion potential was shifted to positive values. On the other the electric capacitance of the double-layer was decreased and the charge transfer resistance and inhibition efficiency were increased, confirming an improved corrosion resistance as compared to the other sample concentrations. SEM results for the mentioned inhibitor with 800 ppm concentration showed a more even and continuous film formed on the surface of the mild steel and no corrosion products were observed.

Keywords: Mild steel; corrosion protection; turmeric; green inhibitor.

Introduction

Corrosion is one of the most fundamental problems in the application of metal artifacts in this sense, various methods have been employed to reduce its complications [1-4]. Corrosion which is defined as a factor for the gradual destruction of metal properties is a result of electrochemical or chemical reactions

with the environment [1-4]. The use of organic inhibitors for the reduction of surface corrosion in metals exposed to corrosive environments has been extensively studied. However, most inhibitory materials are toxic or harmful for the environment and their existence in the waste can cause environmental issues [5]. Organic inhibitors are typically extracted from

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heterogeneous compounds containing oxygen, nitrogen, and sulfur that have active absorption sites for metal surfaces. One of the major issues concerning organic inhibitors is their high cost, toxicity, and lack of environmental compatibility [6-10].

The use of green inhibitors considering the abundance of natural resources is cost-effective as these compounds are non-toxic and biocompatible [5,11-13]. Plants are massive resources of natural compounds that can be extracted and utilized as natural corrosion inhibitors. Based upon the main mechanism, natural corrosion inhibitors are generally classified into two groups: either they lead to the reduction of corrosion by blocking the active sites accessible, or by adsorption and film formation [5,11-13].

Many natural herbal compounds such as aloe vera [14,15], nettle [16,17], ginger [18], cinnamon [19], black and red pepper [20,21], henna [22,23], Glycyrrhizaglabra extract [24], pelargonium [25], have been used as eco-friendly inhibitors for protection of steel structures in different corrosive environments. One of the rarely investigated green inhibitors is turmeric extract which can be utilized as a preferable alternative for organic chemical inhibitors such as azole derivatives and the others [6]. Turmeric has several applications including anti-bacterial [26,27], anti-virus [28], and also anti-inflammatory effects [29-31]. The main reason in which turmeric is known as biocompatible materials is that these type of materials do not produce a toxic or immunological response when exposed to the body or bodily fluids and are free of heavy metals in their structure.

About Shahba et al. [32] investigated the corrosion behavior of

mild steel in the presence of turmeric extract with the concentration range of 50-300 ppm in an environment containing 1M HCl by polarization and EIS. The results of polarization revealed that with the increase of turmeric extract concentration to 300ppm the corrosion current density decreased up to 3 folds as compared to the control sample. Moreover, the corrosion potential shifted to positive values which were $44.4 \mu\text{A}/\text{cm}^2$ and 524 mV , respectively; also the inhibition efficiency reached 63%. On the other hand, the results of EIS confirmed that with the increase of inhibitor concentration up to 300 ppm, the charge transfer resistance and the electric capacitance of the double layer were decreased and were $261 \Omega.\text{cm}^2$ and $0.638 \text{ F}.\text{cm}^2$, respectively. The increase of the potential and charge transfer resistance and also the reduction of corrosion current density and electric capacitance of the double layer demonstrated the adsorption of turmeric molecules on mild steel and formation of a protecting film against corrosive factors and an increase of corrosion resistance.

Al Fakih et al. [33] evaluated the inhibitory effects of turmeric on the corrosion behavior of mild steel in 1 M HCl and a concentration range of 2.5-10 g/L using the weight loss method. The results of weight loss showed that with the increase of turmeric concentration to 10 g/L, the corrosion rate was remarkably decreased compared to the control sample and corrosion efficiency was increased and were 2.23 mm/year and 91.83%, respectively.

Further, the importance of mild steel corrosion in a neutral saline medium is related to its industrial applications in heat exchangers and recirculating water systems [34]. Also,

it is worth noting that mild steel corrosion causes huge economic losses, energy losses, and safety issues [34]. Therefore, it is significant and necessary to study the corrosion inhibition in neutral saline medium [34]. In addition, the price of turmeric is much lower against chemical inhibitors. Between \$ 1 and \$ 2. While the price of chemical inhibitors is higher.

In the present study, the use of turmeric extract as a natural inhibitor for postponing of the electrochemical reactions and corrosion was investigated by means of EIS, polarization and SEM methods. So far, most of the studies have evaluated the anti-corrosion influences of turmeric extract in acidic media, however, reports based on the use of turmeric in corrosive environment containing NaCl 3.5% by the mentioned techniques have not yet been published on the world's renowned journals or as national or international patents, which accounts for the novelty of this investigation.

The main objective of this study is founding of optimum amount of turmeric extract for corrosion inhibiting of steel structure in 3.5% NaCl media that can be a perspective for future development as active protective system in surface coating. It can cause to extend the life time of metal structure if it capsulated with nano and micro carriers to enable its controlled release.

Experimental

Materials

The materials used in this research are turmeric powder (molecular weight: 368.38 g/mol, molecular formula: C₂₁H₂₀O₆, purity: 98%) purchased from Sigma Aldrich Company, electrolyte solution consisting of sodium chloride and distilled water was acquired from Merck Company(Germany), and mild steel which was used as a material for evaluation of corrosion behavior was purchased from Isfahan's Mobarakeh Steel Company. The elements present in the mild steel samples are presented in Table 1.

Table 1. Elemental composition of the mild steel samples

<i>Element</i>	C	Si	Mn	Cr	Mo	Co	Cu	Nb	Fe
<i>Wt(%)</i>	0.190	0.288	1.390	0.026	0.018	0.388	0.297	0.334	97.06

Preparation of turmeric extract and mild steel surface

For the preparation of the salt solution containing turmeric extract, different turmeric concentrations were dispersed in 1 liter of NaCl 3.5% for 24 h under magnetic stirring at 600 rpm and afterward, filtration was performed using centrifugation and filter paper. For electrochemical impedance spectroscopy and polarization tests, mild steel samples were portioned to 1 cm² fragments. The fragments were

prepared with sandpapers no. 400 and 800 and finally degreased by ethanol and were immersed in the aforementioned solution.

Instrument

The polarization tests were performed using a Corrtest CS350 (China) potentiostat/galvanostat and a common three-electrode system that included a saturated calomel reference electrode for controlling the applied potential in uncoated samples, a Pt auxiliary

electrode for establishing the current, and a working electrode of an uncoated mild steel sample. These tests were carried out in the potential range of ± 200 mV with a scan rate of 0.5 V/s. The corrosion current density and other information were obtained via the Tafel extrapolation method. Additionally, these tests were performed at room temperature and were repeated three times to enhance the accuracy of testing of similar samples.

EIS was performed using a Corrtest CS350 potentiostat/galvanostat in conjunction with a common three-electrode system that included a reference electrode, an auxiliary electrode, and a steel sample (1-cm² area) as the working electrode. Sample preparation for EIS was similar to that for the polarization tests. The EIS experiments were conducted at open-circuit potential (OCP) using ± 10 mV perturbation in a frequency range from 10 kHz to 10 MHz after the samples had been immersed for 1, 4 and 24 h.

Frequency response analyzer software was used for data analysis.

The morphology of the surface of the steel samples after immersion in extracted solutions was analyzed by scanning electron microscopy (SEM) on a LEO1455-VP (Japan) scanning electron microscope. The samples were covered with a thin layer of gold *via* a physical vapor deposition (PVD) technique through the sputtering method.

Results and discussion

Electrochemical measurement (solution phase study)

Corrosion inhibitory effects of sodium chloride 3.5% solution, with and without various concentrations of turmeric, was assessed by electrochemical methods (EIS and polarization). The results of EIS after 1, 4, and 24 h of immersion are showed as Nyquist and Bode plots in Figures 1 and 2.

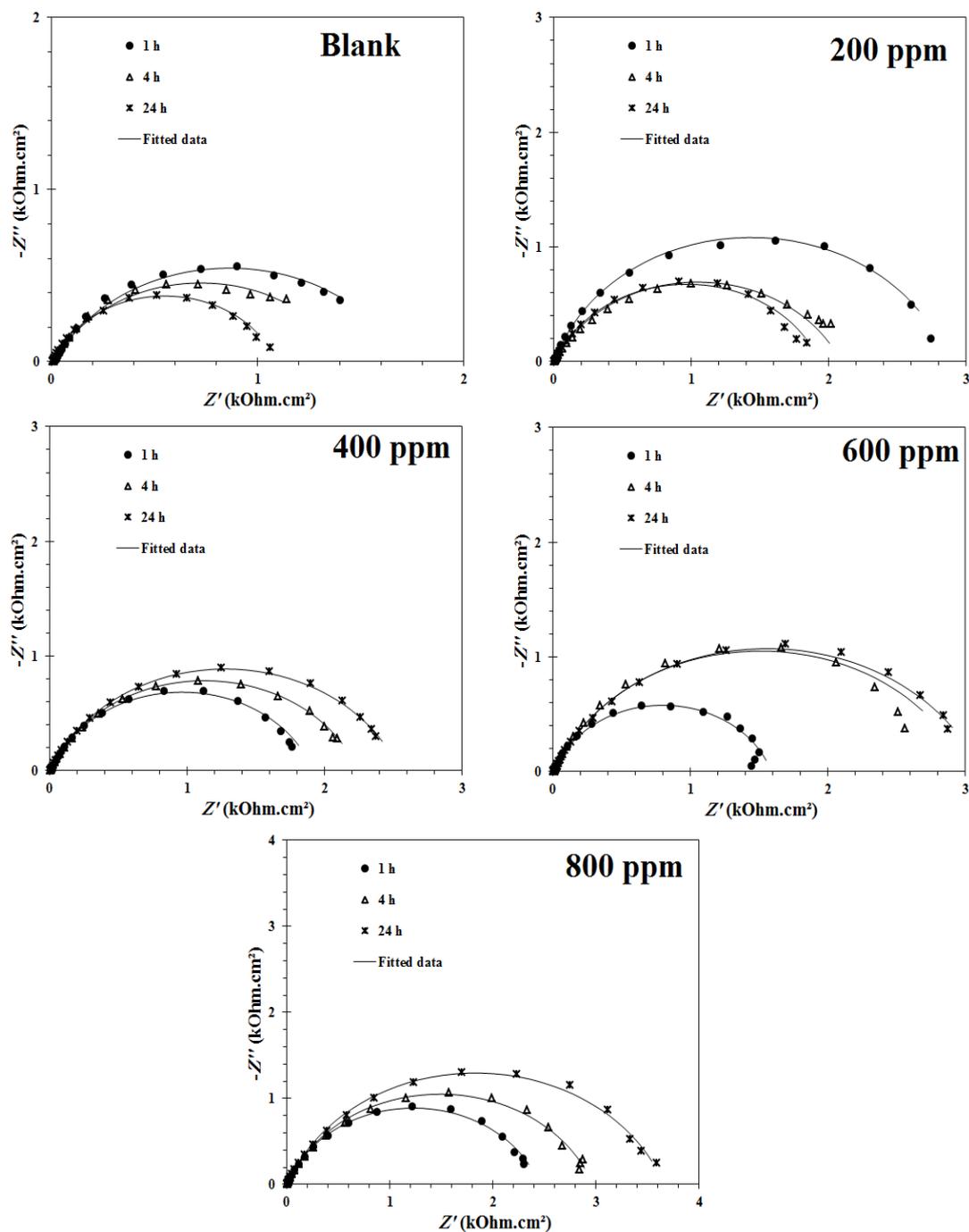


Figure 1. Nyquist plots for all samples immersed in sodium chloride 3.5% solution, with and without various turmeric, after 1, 4, and 24 h

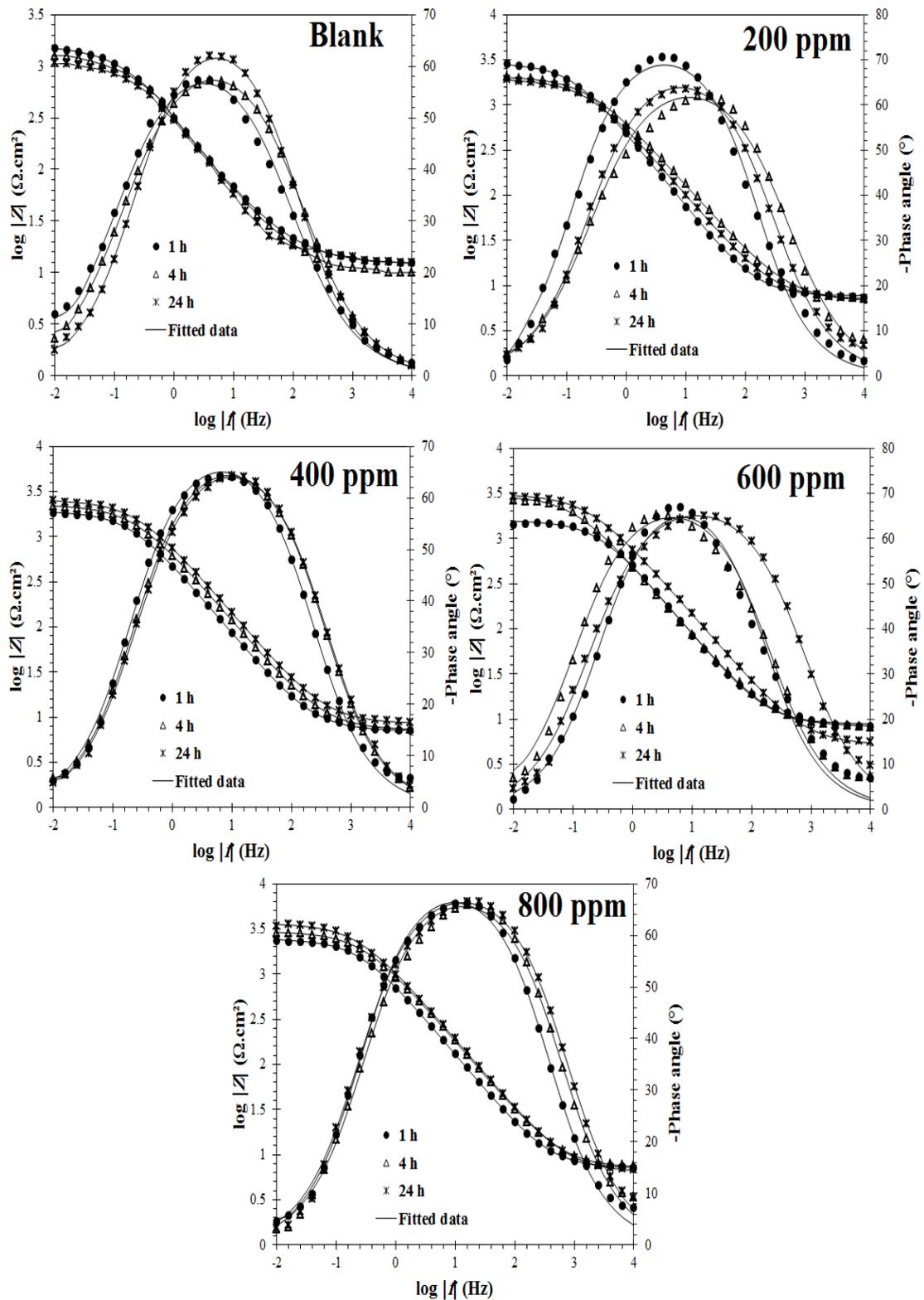


Figure 2. Bode plots for all samples immersed in sodium chloride 3.5% solution, with and without various turmeric, after 1, 4, and 24 h

Figure 1 demonstrates that the semicircle diameter, which is an indicator of charge transfer resistance in the electric double layer, is smaller for the sample without turmeric (blank) in all times confirming the corrosion and destruction in sodium chloride solution [34]. The semicircle diameter of the extracted solution containing different concentrations of turmeric is larger than the blank which reveals the inhibitory effect on mild steel. Furthermore, the semicircle diameter is enlarged for the extracted solutions with 400, 600, and 800 ppm concentrations which shows improvement of corrosion resistance in these systems. The largest diameter after 24 hou belongs to the sample with 800 ppm concentration which demonstrates a better performance compared to the other samples. Additionally, in accordance with the results from Nyquist plot, the highest

impedance are obtained in low frequencies for this sample. In Bode and Nyquist plots only one time constant is observed for all solutions. The equivalent circuits used for fitting the data of the single time constant systems are depicted in figure 3. In these circuits, R_s represents the solution resistance, R_{ct} is the charge transfer resistance, CPE_{dl} is the constant phase element of the double-layer, and R_f and CPE_f respectively represent the resistance and the constant phase element of the inhibitor film. The capacitance of the double layer and capacitance of the inhibitor film are calculated using the Equation 1[34].

$$C = (Y_0(R_{ct})^{1-n})^{\frac{1}{n}} \quad (1)$$

In this equation, Y_0 represents the admittance of the constant phase element and n is the constant related to surface heterogeneity.

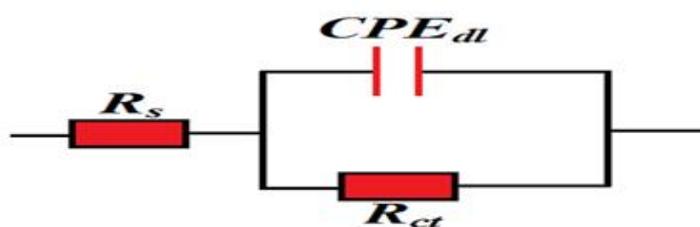


Figure 3. Electrical equivalent circuits used for modeling the impedance curves with single-time-constant

Amounts of electrochemical parameters resulted from the data fitting were calculated for 1, 4, and 24

hours with 3 times repeat and the average results are presented in Table 2.

Table 2. Electrochemical parameters obtained from EIS on mild steel immersed in extracted solutions after 1, 4, and 24 h

Sample	Time (h)	$R_{ct}^a(\Omega.cm^2)$	CPE_{dl}		$C_{dl}(\mu F.cm^{-2})$	η (%)
			$Y_0^b(\mu s^n.\Omega^{-1}.cm^{-2})$	n^c		
Blank (without inhibitor)	1	1602	591.6	0.81	584.2	-
	4	1430	702.1	0.80	739.2	-
	24	1021.5	1218.2	0.80	1049.9	-
200 ppm	1	2855	432.5	0.83	134.4	43.88
	4	2080	346.8	0.75	45.7	31.25
	24	1924	396.3	0.78	75.5	46.90
400 ppm	1	1910	442.4	0.79	95.1	16.12
	4	2236	336.5	0.78	61.5	36.04
	24	2536	280	0.78	50.7	59.72
600 ppm	1	1880	408.3	0.81	108.9	14.78
	4	2999	457	0.78	94.8	52.31
	24	3107	283.3	0.77	40.4	67.12
800 ppm	1	2443	281.9	0.80	59.6	34.42
	4	2960	213.4	0.79	38	51.68
	24	3652	201.9	0.79	34.4	72.02

^aThe standard deviation range for R_{ct} values is between 2.53% and 8.7%.

^bThe standard deviation range for Y_0 values is between 1.3% and 8.2%.

^cThe standard deviation range for n values is between 0.5% and 1.3%.

According to Table 2, the amounts of charge transfer resistance for solutions containing various concentrations of turmeric is raised as compared to the blank sample which reveals the improvement of corrosion inhibitory influence in the presence of turmeric. In Table 2, the results show that as the time passes, the amounts of charge transfer resistance is reduced in the extracted solutions with 200 ppm concentration and blank; whereas they

are increasing for the solutions with 400, 600, and 800 ppm of turmeric. This increase in the charge transfer resistance can upturn the current tendency to cross the capacitor in the circuit and show the corrosion inhibitory role of turmeric. Moreover, the rising manner of samples demonstrates the formation of shielding film on the steel surface. However, the second time constant was not detectable for any of the samples indicating that

time constant for film formation is very close to the corrosion time constant.

The best inhibitory performance was obtained for solutions with 800 ppm turmeric concentration. After 24 h, the amount of charge transfer resistance for this sample was more compared to the other ones which confirm better corrosion inhibitory properties that can be related to the inhibitor molecules in turmeric structure.

Evaluating the admittance of samples revealed that the lowest amount was obtained in a solution containing 800 ppm turmeric and there was also a similar performance for the electric capacitance of double layer. Reduction of the admittance and electric capacitance of double layer amounts could be related to the formation of shielding film or the

increase in the thickness of the electric double layer. These findings clearly demonstrate a better inhibitory performance for the sample containing 800 ppm turmeric.

The results of EIS obviously indicate that turmeric has corrosion inhibitory properties and the increase of concentration has improved the inhibitory performance of the system. The inhibition efficiency was calculated by Equation 2.

$$\eta_{imp}\% = 100\left(1 - \frac{R_{ct,blank}}{R_{ct}}\right) \quad (2)$$

Polarization methods were used for the investigation of the inhibitory mechanism of turmeric in sodium chloride solution. Polarization curves, after 24 h of immersion of samples in extracted solution, are depicted in Figure 4.

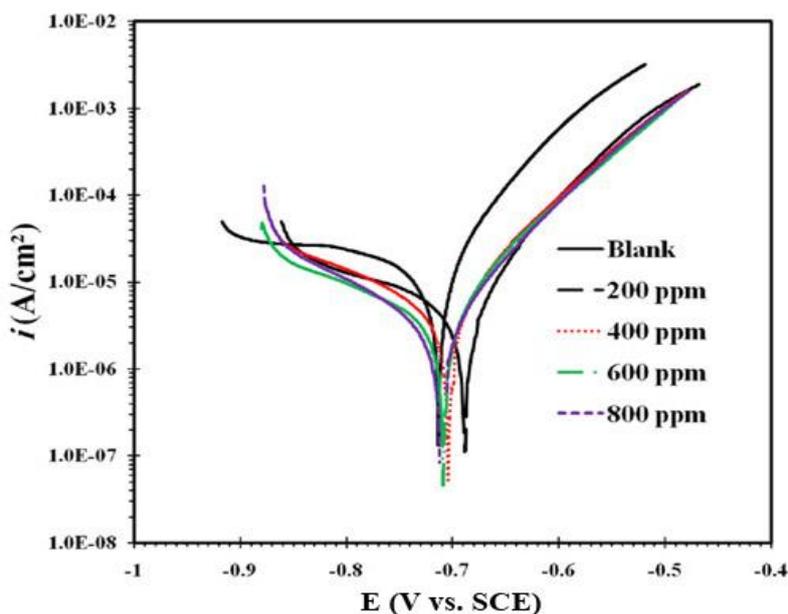


Figure 4. Polarization test for all samples immersed in sodium chloride 3.5% solution, with and without various turmeric, after 24 h

The polarization curves obviously show that the corrosion potential has changed for samples containing turmeric as compared to the blank. For these samples, both anodic and cathodic branches were shifted to lower currents showing mixed corrosion inhibitory

effect (both anodic and cathodic) of turmeric. However, the amounts of potential are shifted to positive values which can be related to the dominant anodic inhibitory mechanism.

Electrochemical corrosion parameters such as corrosion potential

(E_{corr}), corrosion current density (i_{corr}), anodic Tafel slope (b_a), and cathodic Tafel slope (b_c) were obtained after 24

h of immersion using Tafel extrapolation method and the results are presented in Table 3.

Table 3. Electrochemical parameters obtained from Tafel extrapolation polarization teston mild steel immersed in extracted solutions after 24 h

Solution	E_{corr} vs. SCE (mV)	i_{corr} ($\mu\text{A}/\text{cm}^2$)	b_a (V/dec)	$-b_c$ (V/dec)	η (%)
Blank (without inhibitor)	-748	17.4	0.09	0.43	-
200 ppm	-700.1	5.8	0.09	0.3	66.7
400 ppm	-714	5.2	0.09	0.2	70.1
600 ppm	-723.7	4.1	0.09	0.2	76.4
800 ppm	732.2	3.2	0.09	0.2	81.6

As one can observe, the corrosion current density is decreased for the samples immersed in the extracted solution containing 800 ppm turmeric compared to the other samples. Reduction of corrosion current density is the result of the inhibitor sediment layer on the steel surfaces, blockage of the anodic and cathodic active sites, and restriction of the corrosive species attack on the surface.

Lower current density, charge transfer resistance (resulting from EIS), and higher inhibition efficiency (η) in the extracted solution with 800 ppm turmeric proves a better corrosion inhibitory effect in this concentration. The polarization inhibition efficiency (η_p) is calculated according to the Equation 3.

$$\eta_p(\%) = \frac{i_{\text{corr}/\text{blank}} - i_{\text{corr}}}{i_{\text{corr}/\text{blank}}} \quad (3)$$

Based on the studies of Abou Shahba et al. [32] and compare with this project we lead to that, turmeric extract with concentration of 200 ppm in 3.5% NaCl media with 66.7% inhibition efficiency has more performance to turmeric extract with 300 ppm concentration in 1M HCl media with 63.0%. This is the best

reason for excellent performance of turmeric extract in 3.5% NaCl media.

As follows from polarization and EIS measurements, corrosion of mild steel in 3.5% NaCl solution is postponed in the presence of Turmeric extract. The results obviously depicted that the inhibition mechanism involves blocking of mild steel surface by inhibitor molecules *via* adsorption. In general, the phenomenon of adsorption is affected by the nature of metal and chemical structure of inhibitor [32].

Turmeric extract may act as a good corrosion inhibitor with at least one polar unit. These polar units can provide the free electron pairs and p-electrons, also hydroxyl group (-OH), which is an electron donator, moderately activated by virtue of presence of lone pair of electrons on the oxygen (O) atom [32,35-38]. So turmeric extract can absorb into mild steel surface through the transference of free electron pairs and p-electrons to the d-orbital in iron atom, another previous studies show that the phosphate groups play an important role in the complexation process as a whole more effective donor than carboxylate groups. The presence of the aromatic ring increases the power

complexing of these molecules [32,35-38]. Turmeric consists of volatile and non-volatile constituents [39]. The non-volatile constituents are rich with phenolic compounds such as curcumin and other curcumin's derivatives [39]. The corrosion inhibition effect of turmeric is attributed to the adsorption of phenolic constituents onto metal surface [39].

SEM analysis

Analysis of the surface of steel samples after 24 hours of immersion in a salt solution containing turmeric with different concentrations was evaluated with SEM and the results are shown in Figure 5. For the salt solution without the inhibitor (blank), figure 5a, corrosion, and accumulation of the corrosion products were observed on the surface of mild steel. As observed in figures 5b and 5c, for steel samples immersed in a salt solution containing 200 and 400 ppm turmeric extract a film was adsorbed on the mild steel but the surface coverage was not adequate to shield it all and the corrosion products were partially detected in some spots. Figures 5d and 5f reveal that among the samples immersed in salt solution containing 600 and 800 ppm turmeric extract, a more even and continuous film was formed on the surface of the sample containing 800 ppm turmeric; this is a great proof for better inhibitory effect of this concentration that is presented by EIS and polarization tests. That is, a suitable relationship exists between the sediment on the surface and the

inhibition-corrosion performance: the more the sediment on the surface, the better the corrosion-inhibition performance. In general, due to proper adhesion of the shielding film to the metal substrates, the tendency of water molecules to metal is decreased. This improvement of adhesion reduces the penetration of corrosive factors to the interface of the shielding film and steel during the electrochemical tests and leads to better inhibitory effects.

Conclusion

1. The results of EIS demonstrated that, as compared to the other concentrations, after 24 h of immersion, the turmeric extract with 800 ppm concentration in sodium chloride 3.5% with inhibition efficiency of 72% and minimum electrical capacitance of the double layer which is $34.4 \mu\text{F}\cdot\text{cm}^{-2}$, has the best corrosion inhibitory effect.

2. The results of polarization confirmed that for a turmeric extract with 800 ppm concentration both anodic and cathodic branches shifted to lower current densities indicating a mixed corrosive inhibition (both anodic and cathodic) and with the highest inhibition efficiency (81.6%) first-ranked compared to the other rival.

The results of SEM clearly showed that in 800 ppm concentration a thorough continuous film of the inhibitor was adsorbed on mild steel which resulted in blockage of anodic and cathodic areas of metal surface and an increase of the corrosion resistance; this was in accordance with the results of EIS and polarization tests.

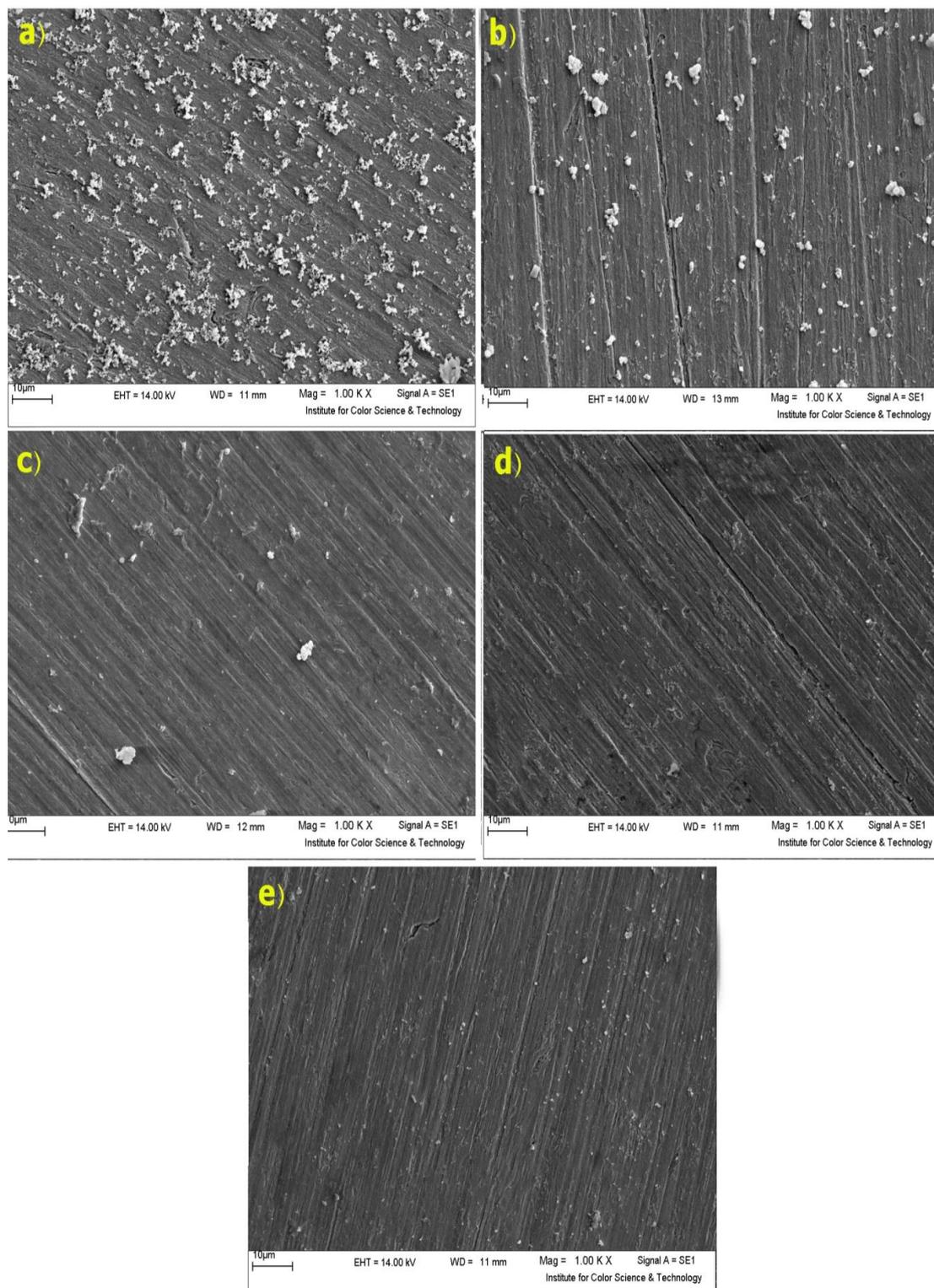


Figure 5. Images of mild steel surface immersed in blank solution (a), extracted solutions containing turmeric with 200 (b), 400 (c), 600 (d), and 800 (e) concentrations, after 24 h

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