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Maltodextrin and Chitosan Polymers as Inhibitors for the Corrosion of Carbon Steel in 1.0 M Hydrochloric Acid

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The inhibition impacts of two water-soluble polymers viz., maltodextrin and chitosan on the dissolution of carbon steel in 1.0 M HCl solution were investigated by three dissimilar techniques. The investigation results indicated that the inhibition efficiencies of the examined polymers increased with their concentrations and reduced by raising temperature. The results obtained from polarization measurements proved that the investigated polymers act as mixed type inhibitors. The acquired high inhibition efficiencies of the studied polymers may be owing to powerful adsorption of the polymer molecules on the C-steel surface resulting in the construction of protective layers. Adsorption of the tested polymers on the steel surface was set to accord with Freundlich adsorption isotherm. The inhibition efficiency of chitosan was set to be higher than maltodextrin because of its high molecular mass that increases the surface area of steel covered by the polymer. The acquired thermodynamic parameters for adsorption indicated that the adsorption process is spontaneous and endothermic, and the type of adsorption is physical. The acquired outcomes from the dissimilar measurements were in a good agreement.

Keyword: Carbon steel; Polymers; Corrosion Inhibitors; Kinetic parameters; Adsorption

1. INTRODUCTION

Corrosion of carbon steel (C-steel) in hydrochloric acid media and trying to inhibit it using organic compounds is an important topic that has multiple applications in many industries that serve the national economy. Scientists are trying to discover new compounds that are inexpensive, nontoxic, environment friendly and have high effectiveness in inhibiting corrosion [1]. In recent years, scientists have intensified their efforts to reduce the risk of carbon steel corrosion by using many synthetic organic compounds [2-8], surface active agent (ionic, nonionic, cationic, Gemini) molecules [8-14], natural plant extracts [15-20] and pharmaceutical drug [21-24]. The inhibition capacity of these compounds ascribed to their adsorption characteristics. There are several factors that affect the efficiency of these compounds as corrosion inhibitors, including the type of inhibitor, whether it is an organic or inorganic compound or a mixture, the type of solution used, the temperature, the hydrogen ion concentration, the presence of electro donating or repelling groups, the presence of the active centers and other factors [25].

Some scientists have used some polymers as inhibitors because their functional groups form complexes with metal ions. These complexes coverage a high surface area and isolate the metals from the aggressive attack solutions [26-28]. The main objective of this study was to investigate the inhibiting affinity of two significant naturally-occurring water-soluble polymers, namely, maltodextrin (I) and chitosan (II), on the corrosion of carbon steel in 1.0 M HCl solution using various techniques; weight loss, (WL) galvanostatic polarization (GAP), potentiodynamic anodic polarization (PAP) and electrochemical impedance spectroscopy (EIS) measurements.

2. EXPERIMENTAL

2.1. Materials

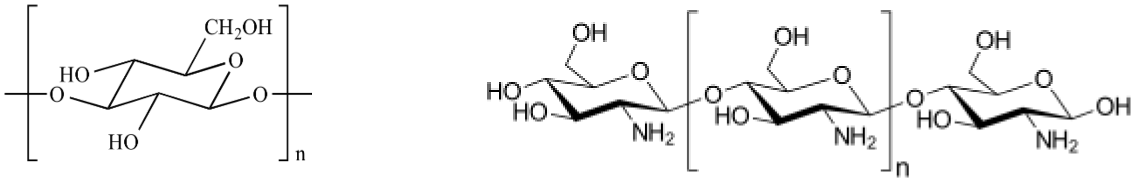
Carbon steel used in the corrosion measurements for this study is produced by Saudi SABIC. It is in the form of a cylindrical rod used in chemical or electrochemical measurements. Its chemical composition is inserted in Table 1. All chemicals were obtained from Sigma Chemicals. The stock solutions of HCl and the two polymer compounds **(**maltodextrin and chitosan**)** were prepared with bidistilled water and the required concentrations were acquired by appropriate dilution.

Table 1. Compositions (wt. %) of the investigated carbon steel specimen.

Element	C	Mn	S	P	Si	Al	Fe
Weight (%)	0.110	0.450	0.050	0.040	0.250	0.039	balance

2.2. Inhibitors

Two polymer molecules were used as received from Sigma-Aldrich. Their structures are illustrated in Figure 1.



Maltodextrin (I) **Chitosan (II)**

Figure 1. Chemical names and structures of the polymers used.

2.3. Weight-loss (WL) measurements

WL measurements were carried out in a temperature-controlled system. The cylindrical carbon steel samples with surface areas of about 12.78 cm^2. Before any measurements, the carbon steel rods were polished with various grades of sanding paper from 200 to 1200, then washed with distilled water and lately washed with acetone. The procedure of the WL method as mentioned previously [29]. The corrosion rate (CR) was computed in mils penetration per year (mpy) using the following equation [30]:

$$\mathrm{CR} = \frac{KW}{Atd} \qquad (1)$$

where, K is a constant, W is the WL in grams, A is the surface area in cm^2, t is time in hour and d is the density.

The inhibition efficiencies (% IE) and the degrees of surface coverage (θ) of the polymer molecules were calculated as follows [30]:

$$\%\ \mathrm{IE} = \theta \times 100 = \left[1 - \frac{CR_{inh}}{CR}\right] \mathrm{x}\ 100 \qquad (2)$$

where, CR and $\mathrm{CR_{inh}}$ are the corrosion rates in the free 1.0 M HCl and with the addition of the polymer compounds, respectively.

2.4. Electrochemical measurements

The galvanostatic polarization measurements (GPM) and potentiodynamic polarization measurements (PDP) were done using PGSTAT30 potentiostat / galvanostat in a triple cell with platinum electrode (CE), reference electrode (RE) and working electrode (WE), carbon steel. All measurements were made at a constant temperature in a temperature-controled system. The values of %IE were calculated for the tested polymers from the following equation [30]:

$$\%\ \mathrm{IE} = 1 - \left[1 - \frac{I\ \mathrm{corr(inh)}}{I_{corr}}\right] \mathrm{x}\ 100 \qquad (3)$$

where, I_{corr} and $I_{\mathrm{corr(inh)}}$ are the corrosion current densities in the free 1.0 M HCl and with addition of the polymer compounds, respectively. PDP measurements were carried out at a scan rate of 1.0 $\mathrm{mVsec^{-1}}$. The values I_{corr} was determined by extrapolation of the slopes of cathodic and anodic Tafel lines (β_c, β_a), of the polarization curves with the corrosion potentials (E_{corr}).

EIS measurements were conveyed out in a frequency range of 100 kHz to 0.1 Hz with an amplitude of 4.0 mV peak-to-peak using AC signals at OCP. The %IE values were computed from the charge transfer resistance (R_{ct}) data using the following equation [30]:

$$\%\ \mathrm{IE} = \left[1 - \frac{R_{ct}}{R_{ct(inh)}}\right] \mathrm{x}\ 100 \qquad (4)$$

where, R_{ct} and $R_{\mathrm{ct(inh)}}$ in the free 1.0 M HCl and with the addition of the polymer compounds, respectively.

3. RESULTS AND DISCUSSION

3.1. WL Measurements

3.1.1. Effect of Polymers Concentrations

Figure 2 presents the relationship between WL and immersion time for C-steel in the free 1.0 M HCl solution and in the presence of some concentrations of chitosan polymer, ranging from 100 to 500 ppm. Like curves are acquired in the presence of maltodextrin, but do not appear here. It is evident that WL decreases with increasing chitosan concentration. This donates that the examined polymer molecules act as inhibitors by reducing the rate of steel corrosion of in the aggressive medium, 1.0 M HCl solution. Examining Figure 2, it can be observe that the plots in this figure were linear, which indicates that there is no insoluble film formation on the iron surface throughout corrosion, and the polymer molecules are absorbed on the iron surface. The process of suppression of corrosion is carried out either by blocking the interactive sites or by modifying both the cathodic and partial process mechanisms.

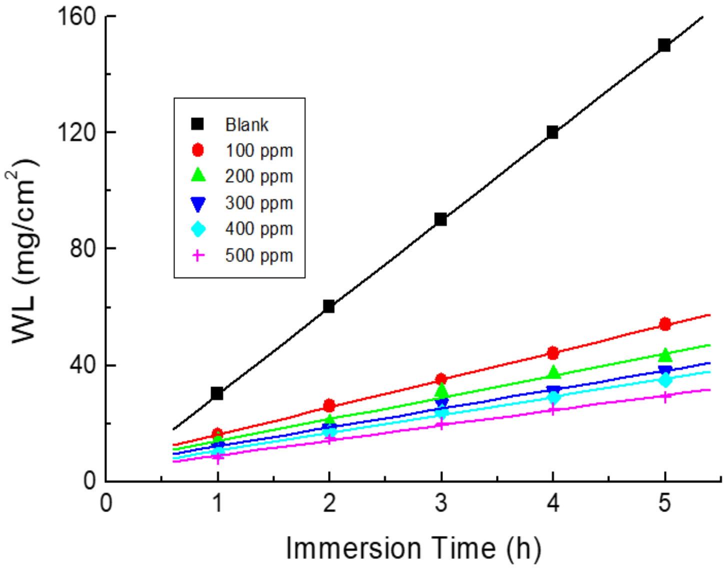


Figure 2. Weight loss (WL) versus immersion time for the corrosion of C-steel in 1.0 M HCl solution (blank) and with various concentrations of chitosan at 298 K.

The computed values of CR, %IE and θ are included in Table 2. With an increase in the concentration of maltodextrin and chitosan, CR is reduced and the values of % IE and θ increase, which proves the inhibitory impacts of the two examined polymer compounds. Also, the values of %IE of chitosan at all concentrations studied were found to be more than those of maltodextrin. This point will be interpreted later in the mechanism of inhibition.

Table 2. Corrosion parameters acquired from the corrosion of C-steel in 1.0 M HCl solution and in the presence of maltodextrin and chitosan at 298 K.

Inhibitors	**Inh. Conc. (ppm)**	**10^5 CR mg cm^{-2} min^{-1}**	**% IE**	**ϴ**
--	**0**	**0.580**	**-**	**-**
Maltodextrin	**100**	0.129	77.76	0.777
	200	0.118	79.65	0.796
	300	0.115	80.17	0.802
	400	0.113	80.60	0.806
	500	0.108	81.38	0.814
Chitosan	**100**	0.121	79.13	0.791
	200	0.078	86.55	0.865
	300	0.075	87.10	0.871
	400	0.071	87.79	0.878
	500	0.066	88.62	0.886

3.1.2. Temperature Effect

The influence of temperature rise on WL of C-steel was studied in a free 1.0 M HCl solution and in the presence of a 500 ppm of the polymers by WL measurements at different temperatures from 298 K to 328 K. The values of CR and %IE are inserted in Table 3. From the data listed in Table 3, it is evident that, the CR values increase with increasing temperature and, therefore, the values of %IE increase. This explained by the desorption of the film formed at the surface of C-steel at an elevated temperature. This showed that the adsorption of maltodextrin and chitosan compounds was physical.

Table 3. Impact of rising temperature on the corrosion parameters for C-steel in 1.0 M HCl solution and in the presence of a 500 ppm of maltodextrin and chitosan.

Medium	**T K**	**CR mg cm^{-2} h^{-1}**	**% IE**
1.0 M HCl	**298**	0.580	--
	308	0.842	--
	318	1.020	--
	328	1.141	--
1.0 M HCl + 500 ppm of Maltodextrin	**298**	0.108	81.38
	308	0.198	77.24
	318	0.286	71.96
	328	0.374	68.07
1.0 M HCl + 500 ppm of Chitosan	**298**	0.066	88.62
	308	0.168	80.69
	318	0.242	76.27
	328	0.316	73.15

The activation thermodynamic parameters such as the activation energy (E_a^*) enthalpy of activation (ΔH^*) and entropy of activation (ΔS^*) were determined from the Arrhenius and transition state equations [31,32]:

$$\ln CR = \ln A - \frac{E_a^*}{RT} \qquad (5)$$

$$\ln\left(\frac{CR}{T}\right) = \left(\ln\frac{R}{Nh} + \frac{\Delta S^*}{R}\right) - \frac{\Delta H^*}{R}\frac{1}{T} \qquad (6)$$

where, A, R, T, N and h are Arrhenius constant, universal gas constant, temperature Avogadro's number and Planck's constant, respectively.

From the slopes of the linear relationship between log (R_{corr}) with (1/T) (Fig. 3) we can compute the values of E_a^* which were found to be 18.19 KJ mol^{-1} for free 1.0 M HCl solution and equal to 26.81 and 27.72 KJ mol^{-1} in the presence of a 500 ppm of maltodextrin and chitosan, respectively. Evidently, the E_a^* values increase in the presence of polymer compounds indicating their adsorption on the C-steel surface by creating a barrier for mass and charge transfer.

The enthalpy and entropy of activation for C-steel corrosion in free 1.0 M HCl and in the presence of polymer molecules have been calculated from the slope [-ΔH^*/2.303 R] and the intercept [log (R/Nh -ΔS^*/2.303R)] of the relationship between of (log R_{corr}/T), with (1/T) as shown in Fig . 4. The ΔH^* values were found to be 16.14 KJ mol^{-1} in free 1.0 M HCl and equal to 24.98 and 28.72 KJ mol^{-1} in the presence of a 500 ppm of maltodextrin and chitosan, respectively. The positive signs of ΔH^* denote that the adsorption the two polymer compounds onto the C-steel surface is an endothermic process. The calculated values of ΔS^* are equal to -163.12 J mol^{-1} K^{-1} in the free 1.0 M HCl solution and equal to -53.36 and -261.81 J mol^{-1} K^{-1} in presence of a 500 ppm of maltodextrin and chitosan, respectively. The values of ΔS^* are negative suggesting that the construction of the activation complex is the rate-determining step which represents an association rather than dissociation [33].

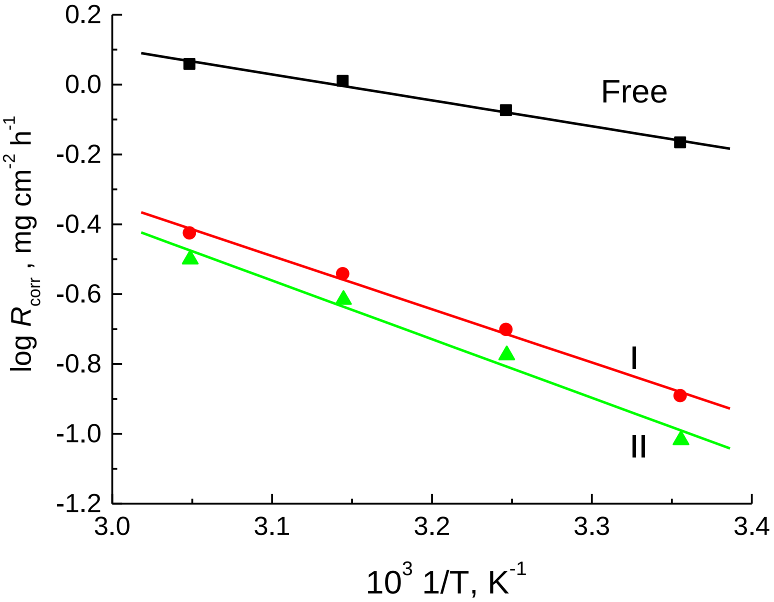


Figure 3. Arrhenius plots for the corrosion of C-steel in 1.0 M HCl solution and in the presence of a 500 ppm of: I) maltodextrin, II) chitosan.

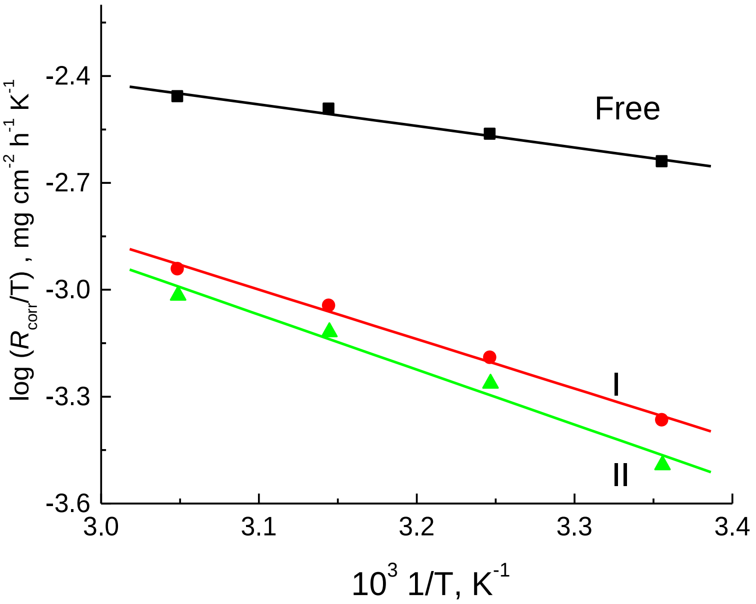


Figure 4. Transition state plots for the corrosion of C-steel in 1.0 M HCl solution and in the presence of 500 ppm of: I) maltodextrin, II) chitosan.

3.1.3. Galvanostatic Polarization Measurements (GPM)

Figure 5 displays the GPM for C-steel in a free 1.0 M HCl solution and in the presence of some concentrations of chitosan ranging from 100 to 500 ppm. Similar curves were obtained in the presence of maltodextrin but did not inserted here. Some corrosion parameters such as corrosión potential, E_{corr}, corrosión current density, I_{corr}, β_a, β_c and %IE are computed and recorded in Table 4.

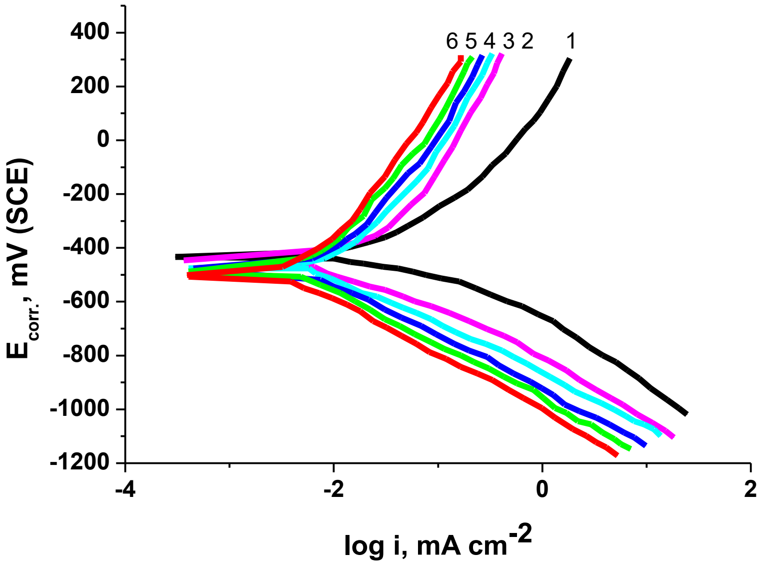


Figure 5. GPM for C-steel in 1.0 M HCl solution and in the presence of different concentrations (ppm) of chitosan: 1) 00.0 2) 100 3) 200 4) 300 5) 400 6) 500.

Examining Table 4, it is evident that, the increased concentration of the two polymers resulted in slightly shifting the values of $E_{corr.}$ towards negative direction and the I_{corr} values were reduced at all studied concentrations, suggesting the inhibitory action of these polymeric compounds. The values of Tafel slopes (β_a & β_c) were changed slightly in the presence of the polymers, indicating that maltodextrin and chitosan polymers are considered as mixed type inhibitors [34]. That is, these compounds influenced both cathodic hydrogen evolution and anodic steel corrosion reactions. The order of the inhibition efficiency of the studied polymeric compounds is chitosan > maltodextrin.

Table 4. Effect of increasing concentrations (ppm) of the examined polymers on the corrosion parameters obtained from GPM of C-steel in 1.0 M HCl solution at 298 K.

Inhibitors	Inh. Conc. (ppm)	$-E_{corr}$ (mV (SCE))	β_a (mV/decade)	$-\beta_c$ (mV/decade)	I_{corr} (μA/cm^2)	%IE
--	**0**	434	78	113	0.781	--
Maltodextrin	**100**	440	80	118	0.342	80.78
	200	454	81	120	0.291	83.65
	300	487	88	130	0.258	85.50
	400	492	98	133	0.223	87.47
	500	498	102	134	0.183	89.72
Chitosan	**100**	492	79	112	0.284	84.04
	200	498	80	119	0.218	87.75
	300	511	87	124	0.180	89.88
	400	537	94	129	0.162	90.89
	500	539	110	133	0.132	92.58

3.1.4. Polymer compounds as pitting corrosion inhibitors

The two selected polymer compounds, chitosan and maltodextrin, have been examined as inhibitors for pitting corrosion of C-steel. Figure 6 displays the PDP curves of C-steel in the free 1.0 M HCl solution and in the presence of some concentrations of chitosan (ranging from 100 to 500 ppm) at a scan rate 1.0 mVs^{-1}. Similar curves in the presence of maltodextrin are obtained, but not appeared here. Through this figure there are no anodic peaks suggesting a good stable oxide film on the surface of C-steel. As the potential increase, the current remains constant until a certain potential, then the current increase rapidly due to breakdown of the passive film formed on the C-steel surface and the initiation of pitting attack. This potential is defined as pitting corrosion potential (E_{pit}) [35-38]. As the concentration of polymer compounds increases the values of E_{pit} is transferred to more positive potentials donating the resistance of pitting attack.

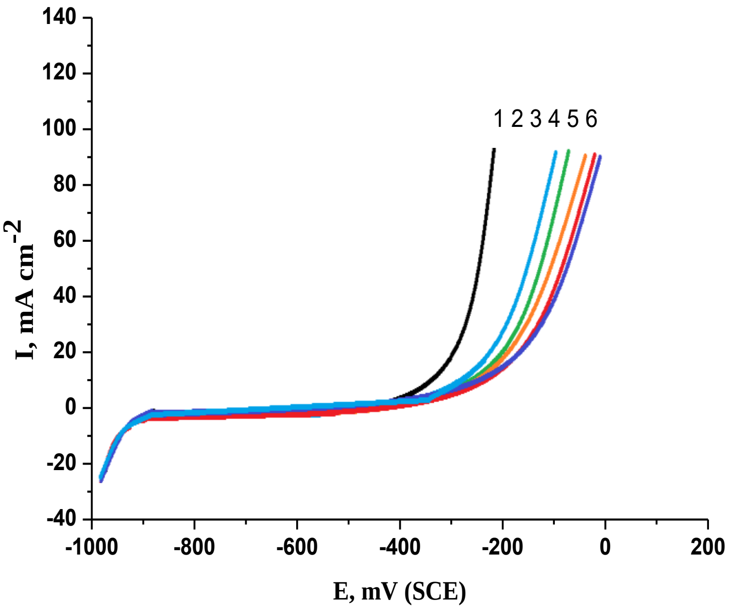


Figure 6. PDAP curves for C-steel in 1.0 M HCl + 0.1 M NaCl solution and in the presence of different concentrations (ppm) of chitosan: 1) 00.0 2) 100 3) 200 4) 300 5) 400 6) 500.

Figure 7 displays the relationship between E_{pit} and the logarithm of the concentrations of polymer compounds. Obviously from this figure the values of E_{pit} is directed to more positive direction as the concentrations of the two polymer compounds increases satisfying the following equation [37,38]:

$$E_{pit} = A + B \log C_{inh} \quad (7)$$

where, A and B are constants depending on the types of the electrode and inhibitor employed.

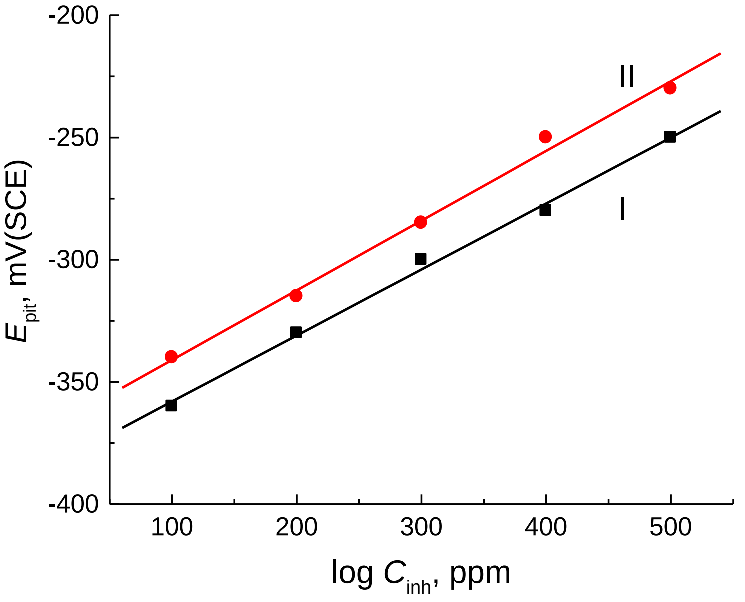


Figure 7. The relationship between E_{pit} and log (conc.) of polymer compounds: I) maltodextrin and II) chitosan for C-steel in 1.0 M HCl + 0.1 M NaCl solution.

3.1.5. EIS Measurements

Nyquist plots of C-steel in 1.0 M HCl solution and in the presence of some concentrations of chitosan compound are shown in Figure 8. Similar curves in the presence of maltodextrin are obtained but not appeared here. It is clear from this figure, that in supreme of these cases the impedance diagram does not show a complete half-circle. This due to the frequency dispersion [39] as a result of roughness and homogeneity of steel surface. The increase in semicircle diameters with the concentration of two polymer compounds indicates an increase in the protective properties of C-steel surface. The impedance diagrams for two polymer compounds have a semicircular appearance; proving that C-steel corrosion is mainly controlled by a charge transfer process [40].

The equivalent circuit proposed by Randles has been used previously as mentioned above [41]. Two parameters were derived from analysis of Nyquist plots, the first parameter being the charge transfer resistance (R_{ct}) obtained from the intercepts of the semicircle with the axis of the real component. The second parameter is the capacity of double layer (C_{dl}) computed from the angular frequency ($\omega = 2\pi f$) at the maximum imaginary component and the R_{ct} according to the following equation:

$$C_{dl} = [1/\omega_{max} R_{ct}] = [1 / 2\pi f_{max} R_{ct}] \qquad (8)$$

where, f is frequency, ω is the angular velocity.

The inhibition efficiency, %IE, was determined from the following equation:

$$\%IE = \left(1 - \frac{(R_{ct})_{free}}{(R_{ct})_{add.}}\right) x100 \qquad (9)$$

where, $(R_{ct})_{free}$ and $(R_{ct})_{add}$ are the charge transfer resistance in the free 1.0 M HCl solution and in the presence of the polymer compounds, respectively. Values of R_{ct}, C_{dl} and %IE are inserted in Table 5.

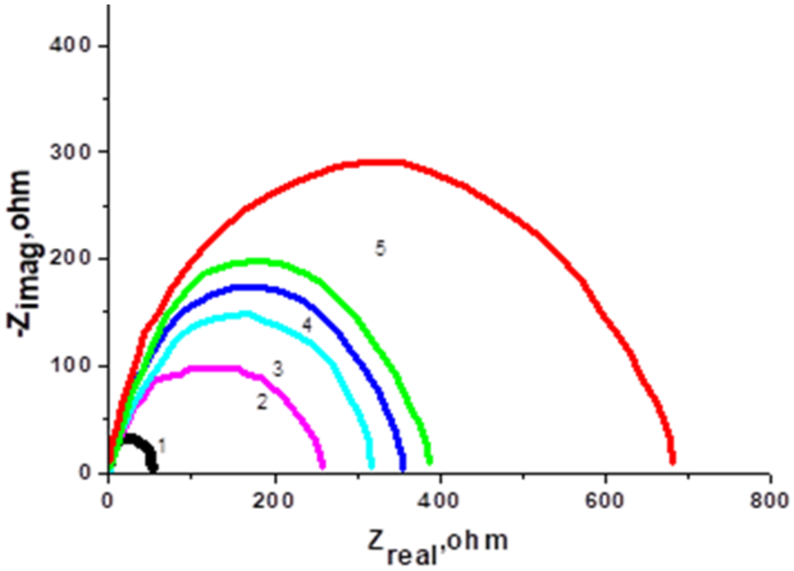


Figure 8. Nyquist plot of C-steel in 1.0 M HCl solution and in the presence of different concentrations (ppm) of chitosan: 1) 00.0 2) 100 3) 200 4) 300 5) 400 6) 500.

It was observed from Table 5 that with increasing the concentrations of the two polymer compounds, the R_{ct} values increase owing to the construction of protective film at the interface between the steel and electrolytic solution. On the other hand, the values of C_{dl} decreased due to the water molecules at the electrode interface are replaced by polymer compounds of lower dielectric constant through adsorption. Also, values of %IE of chitosan were set to more than those of maltodextrin. These results are in a good agreement with the previous measurements used.

Table 5. Electrochemical parameters obtained by the EIS technique for C- steel in 1.0 M HCl solution and containing various concentrations of the examined polymer compounds.

Inhibitors	**Inh. Conc. (ppm)**	**C_{dl}, µF cm^{-2}**	**R_{ct}, Ω cm^{-2}**	**%IE**
--	**0**	**187**	**58**	**--**
Maltodextrin	**100**	151	206	71.84
	200	147	249	76.70
	300	139	271	78.60
	400	132	286	79.72
	500	128	327	82.26
Chitosan	**100**	166	254	77.16
	200	158	311	81.35
	300	151	345	83.20
	400	148	371	84.36
	500	142	678	91.44

3.1.6. Adsorption Isotherm and Inhibition Mechanism

The inhibiting vigor of the two investigated polymeric compounds on the C-steel corrosion in 1.0 M HCl solution based on their adsorption onto the steel surface. The adsorption strength depends on the chemical structure and the molar mass of the studied polymer compounds, the chemical composition of the steel, the type of the aggressive acid, the pH value, the temperature and the electrochemical potential of the steel /electrolyte interface.

In theory, the adsorption process can be considered as replacement process between the polymer compounds in the aqueous phase [$\mathrm{Poly}_{(aq)}$] and water molecules at the carbon steel surface [$H_2O_{(sur)}$] to give the polymer compounds adsorbed on the surface of carbon steel [$\mathrm{Poly}_{(sur)}$] and thus increased inhibition efficiency due to subsequent equation:

$$\mathrm{Poly}_{(aq)} + z\,H_2O_{(sur)} \rightarrow \mathrm{Poly}_{(sur)} + zH_2O_{(aq)} \qquad (10)$$

where, z is the size ratio and simply equals the number of adsorbed water molecules replaced by a single inhibitor molecule. In order to obtain the best isotherm match the results obtained. It was found that the obtained results obeyed Freundlich isotherm which governed by Eq. (11) [42]:

$$\log \theta = \log K_{ads} + n \log C_{inh.} \qquad (11)$$

where, K_{ads} and C_{inh} are the equilibrium constant for the adsorption and concentration of the polymer, respectively.

Figure 9 displays the relationship of Freundlich isotherm (log θ vs log C_{inh}) of C-steel electrode in 1.0 M HCl solution and containing various concentrations of two polymer compounds. Straight lines were obtained. From the intercept, the K_{ads} values are calculated which relates to the standard free energy of adsorption, ΔG^o_{ads}, according to the following equation:

$$K_{ads} = 1/55.5 \exp(-\Delta G^o_{ads}/RT) \qquad (12)$$

The computed values of ΔG^o_{ads} are equal to -45.23 kJ mol^{-1} and -52.18 kJ mol^{-1} for maltodextrin and chitosan, respectively. The negative value of ΔG^o_{ads} suggests spontaneous adsorption process and strong interaction of the two polymer compounds onto the carbon steel surface.

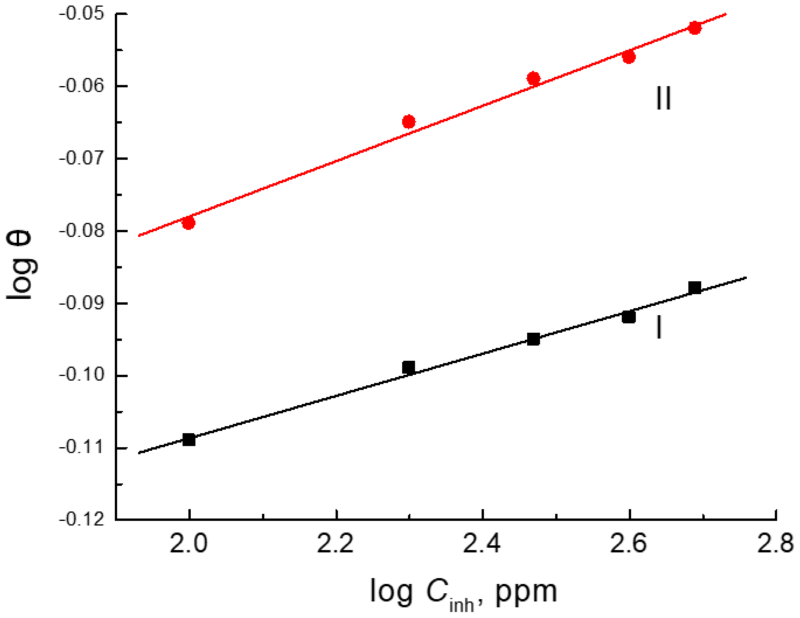


Figure 9. Freundlich isotherms for C-steel in 1.0 M HCl solution and in the presence of 500 ppm of: I) maltodextrin, II) chitosan.

The studied polymer compounds act as a good inhibitor for the carbon steel corrosion in 1.0 M HCl solution. The inhibition ability depends on the concentration, temperature, molar mass and chemical structure of the polymer compounds. The existence of some hetro oxygen atoms, OH group and NH_2 facilitates the adsorption process by forming a coordination bond between the polymer compound and steel by transferring lone pairs of electrons from hetero oxygen atoms to the steel surface. The formed complex is blocked adsorbed onto the steel surface due the formation of more than one active centre. Among all four techniques used the %IE of chitosan is more efficient than maltodextrin. This is due to the possibility of a more complex formation in the chitosan molecule due to the presence of more than an oxygen hetro atoms and the presence of many OH and NH_2 accelerates the adsorption process. Also, the molar mass of chitosan is higher than maltodextrin, which increases the surface area covered by the polymer, which increases the inhibition efficiency. On the other hand, the obtained results in the present research work indicated that inhibitive impact of chitosan as a green corrosion inhibitor for carbon steel in the investigated corrosive medium was set to be higher than that obtained in previous work [43,44].

4. CONCLUSIONS

1. The examined polymer compounds, maltodextrin and chitosan, were found to act as effective inhibitors to reduce the corrosion of C-steel 1.0 M HCl solution.
2. The inhibition efficiency increased with rising concentrations of polymer compounds and lowered with rising temperature.
3. The studied polymers were found to behave as mixed type inhibitors.
4. The inhibiting action of the investigated polymer compounds is due to their spontaneous adsorption on the surface of C-steel.
5. The adsorption of the inhibitors followed Freundlich isotherm.
6. The studied polymers acted as pitting corrosion inhibitors.
7. The inhibition efficiency of chitosan was found to higher than that of maltodextrin.

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