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► **To cite this version:**

M.R. Laamari, J. Benzakour, F. Berrekhis, A. Derja, Didier Villemin. Adsorption and corrosion inhibition of carbon steel in hydrochloric acid medium by hexamethylenediamine tetra(methylene phosphonic acid). *Arabian Journal of Chemistry*, 2016, 9, pp.S245 - S251. 10.1016/j.arabjc.2011.03.018 . hal-01829419

HAL Id: hal-01829419

<https://hal.science/hal-01829419>

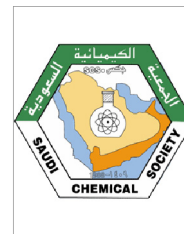
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ORIGINAL ARTICLE

Adsorption and corrosion inhibition of carbon steel in hydrochloric acid medium by hexamethylenediamine tetra(methylene phosphonic acid)



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Received 13 January 2011; accepted 27 March 2011

Available online 1 April 2011

KEYWORDS

Carbon steel;
Thermodynamic parameters;
Phosphonic acid;
Acid inhibition

Abstract The inhibitive effect of the hexamethylenediamine tetra(methylene phosphonic acid) (HMDTMPA) on the corrosion of carbon steel in 1.0 M HCl solution has been investigated by weight loss measurement, potentiodynamic polarization and electrochemical impedance spectroscopy (EIS) techniques. The presence of (HMDTMPA) reduces remarkably the corrosion rate of carbon steel in acidic solution. The effect of temperature on the corrosion behavior of carbon steel was studied in the range of 298–328 K. Results clearly reveal that the (HMDTMPA) behaves as a mixed type corrosion inhibitor with the highest inhibition at 4×10^{-3} M. The adsorption of HMDTMPA on the carbon steel surface obeys to the Langmuir's adsorption isotherm. Surface analysis via scanning electron microscope (SEM) shows a significant improvement on the surface morphology of the carbon steel plate.

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1. Introduction

Acid solutions are widely used in industries such as pickling, cleaning and descaling which lead to corrosive attack. The commonly used acids are hydrochloric acid, sulfuric acid and nitric acid. Since acids are aggressive, inhibitors are usually used to minimize the corrosive attack on metallic materials. A wide variety of compounds are used as corrosion inhibitors for metals in acids media (Schmitt, 1984; Mernari et al., 1998; Onuchukwu, 1988; Ashassi-Sorkhabi and Nabavi-Amri, 2000; Ebenso, 2002; Gomma, 1998). Their inhibitive

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action is related to several factors including the structure and charge distribution of the molecule, the number and types of adsorption sites, and the nature of interaction between the molecule and the metal surface. Corrosion inhibition occurs via adsorption of the organic molecule on the metal surface following some known adsorption isotherms with the polar groups acting as the reactive centers in the molecules. The resulting adsorption film acts as a barrier that isolates the metal from the corrosive media.

Recent works involve the study of corrosion inhibition of Aramco iron and carbon steel in different media using phosphonic acid and monofluorophosphate compounds (Laamari et al., 2011, 2010b, 2001, 2004; Amar et al., 2008).

The aim of this study is to evaluate the inhibitive effect of hexamethylenediamine tetra(methylene phosphonic acid) (HMDTMPA) as a corrosion inhibitor of carbon steel in 1 M HCl solution. The assessment of the corrosion behavior was studied using weight loss, potentiodynamic polarization measurement, electrochemical impedance spectroscopy (EIS) and the scanning electron microscope (SEM). Thermodynamic data were also obtained from adsorption isotherms and Arrhenius plots.

2. Experimental

2.1. Electrochemical cell

Electrochemical experiments were performed using a conventional three electrode cell assembly. The working electrode is a carbon steel rotating disk with surface area of 1 cm². It is abraded with different emery papers up to 1200 grade, washed thoroughly with double-distilled water, degreased with AR grade ethanol, acetone and drying at room temperature.

A saturated calomel electrode (SCE) was used as the reference electrode. All the measured potentials presented in this paper are referred to this electrode. The counter electrode was a platinum plate with a surface area of 2 cm².

The aggressive solutions were made of AR grade 37% HCl. Appropriate concentration of acid is prepared using double distilled water. The inhibitor is added to freshly prepared 1.0 M HCl in the concentration range of 10⁻⁴–4 × 10⁻³ M.

The corrosion inhibitor used in this work is hexamethylenediamine tetra(methylene phosphonic acid) (HMDTMPA). The organic compound is synthesized by the micro-wave technique. The obtained product is purified and characterized by ¹H, ¹³C, ³¹P NMR spectroscopy and IR techniques. The molecular structure is shown in Fig. 1.

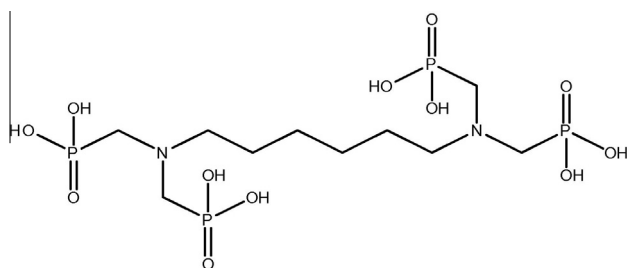


Figure 1 Structure of hexamethylenediamine tetra(methylene phosphonic acid) (HMDTMPA).

2.2. Methods

2.2.1. Gravimetric measurements

Gravimetric measurements were carried out in a double walled glass cell equipped with a thermostat-cooling condenser. The solution volume was 100 mL of 1.0 M HCl with and without addition of different concentrations of inhibitor. The carbon steel specimens used have a rectangular form (2 × 2 × 0.05 cm). The immersion time for the weight loss was 24 h at 298 K and 6 h at the other temperatures. After the corrosion test, the specimens of carbon steel were carefully washed in double-distilled water, dried and then weighed. The rinse removed loose segments of the film of the corroded samples. Triplicate experiments were performed in each case and the mean value of the weight loss is reported. Weight loss allowed us to calculate the mean corrosion rate as expressed in mg cm⁻² h⁻¹. The inhibition efficiency (IE%) was determined by using the following equation:

$$IE\% = \frac{CR_0 - CR}{CR_0} \times 100 \quad (1)$$

where, CR and CR₀ are the corrosion rates of carbon steel with and without the inhibitor, respectively.

2.2.2. Electrochemical measurements

Two electrochemical techniques, namely DC-Tafel slope and AC-electrochemical impedance spectroscopy (EIS), were used to study the corrosion behavior. All experiments were performed in one-compartment cell with three electrodes connected to a Voltalab 10 (Radiometer PGZ 100) system controlled by the Volta master 4 corrosion analysis software model.

Polarization curves were obtained by changing the electrode potential automatically from -800 to 0 mV versus open circuit potential (*E*_{ocp}) at a scan rate of 1 mVs⁻¹. The inhibition efficiency is calculated by the following equation:

$$IE\% = \frac{i_{corr}^0 - i_{corr}}{i_{corr}^0} \times 100 \quad (2)$$

Where *i*_{corr}⁰ and *i*_{corr} are the corrosion current density values without and with inhibitor, respectively. EIS measurements were carried out under potentiostatic conditions in the frequency range 100–0.1 Hz, with an amplitude of 10 mV peak-to-peak, using AC signal at *E*_{ocp}. All experiments were performed after immersion for 60 min in 1.0 M HCl with and without addition of inhibitor.

2.2.3. Surface morphology

For morphological study, surface features (0.9 × 0.8 × 0.2 cm) of carbon steel were examined after exposure to 1 M HCl solution after 1 day with and without inhibitor. JEOL JSM-5500 scanning electron microscope was used for this investigation.

3. Results and discussion

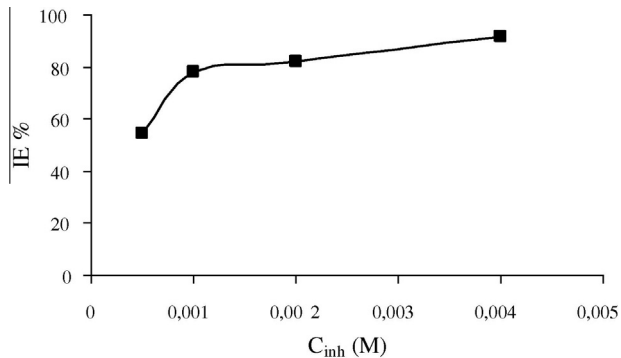
3.1. Weight loss measurements

3.1.1. Effect of inhibitor concentration

Table 1 collects the corrosion rates and Fig. 2 shows the variation of inhibition efficiencies evaluated from weight loss measurements for different inhibitor concentrations in 1.0 M HCl.

Table 1 Corrosion parameters obtained from weight loss measurements for carbon steel in 1 M HCl containing various concentrations of HMDTMPA at 298 K.

C (M)	CR (mg cm ⁻² h ⁻¹)	IE%
Blank	0.51	–
0.0005	0.23	54.58
0.001	0.16	77.97
0.002	0.11	81.87
0.004	0.05	91.61

**Figure 2** Variation of inhibition efficiency (a%) with the concentration of HMDTMPA for carbon steel in 1 M HCl.

The corrosion rate decreases with HMDTMPA concentration and in turn the inhibition efficiency (IE%) increases to attain 91%. The increasing of IE% might be due to the result of increased adsorption and coverage of inhibitor on the carbon steel surface with increasing inhibitor concentration (Zhao and Mu, 1999). In order to gain better understanding of thermodynamic properties of carbon steel corrosion processes in the presence of HMDTMPA, a detailed study on corrosion behavior of carbon steel was carried out at a temperature range 298–328 K using the weight loss technique.

3.1.2. Effect of temperature

Generally, the metallic corrosion in acidic media is accompanied by the evolution of H₂ gas and rises with temperature that usually accelerates the corrosion reactions, involving higher dissolution rate of the metal. Further, the value of inhibition efficiency indicates the adsorption ability of inhibitor molecules on the metal surface; the higher inhibition efficiency results in the higher adsorption.

The effect of temperature on the adsorption behavior of HMDTMPA was investigated using weight loss measurements in the temperature range 298–328 K with and without inhibitor at different concentrations in 1.0 M HCl. The values of inhibition efficiency obtained are given in Table 2 and Fig. 3.

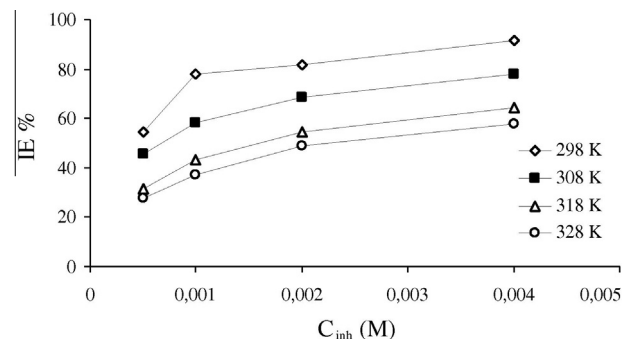
It is clear that inhibition efficiency increased as far as inhibitor concentration increases. The maximum value of inhibition efficiency (IE%) obtained for 4×10^{-3} M HMDTMPA is 91% at 298 K. The inhibition efficiencies decrease slightly with increasing temperature, indicating that higher temperature dissolution of carbon steel predominates on adsorption of HMDTMPA at the metal surface. The temperature effect on the inhibited acid-metal reaction is very complex according to Bentiss et al. (2005); because many changes occur on the metal surface such as rapid etching, desorption of inhibitor and the inhibitor itself may undergo decomposition.

In order to obtain more details on the corrosion process, activation kinetic parameters such activation energy (E_a); enthalpy (ΔH°) and entropy (ΔS°) are obtained from the effect of temperature using Arrhenius law (Eq. (3)) and the alternative formulation of Arrhenius equation (Eq. (4)) (Behpour et al., 2009):

$$\log(\text{CR}) = \log A - \frac{E_a}{2.303RT} \quad (3)$$

$$\text{CR} = \left(\frac{RT}{Nh}\right) \exp\left(\frac{\Delta S^\circ}{R}\right) \exp\left(-\frac{\Delta H^\circ}{RT}\right) \quad (4)$$

where CR is the corrosion rate, R is the universal gas constant, T is the absolute temperature, A is the pre-exponential factor, h is Planck's constant (6.626176×10^{-34} Js) and N is Avoga-

**Figure 3** Variation of inhibition efficiency (IE%) with concentration of HMDTMPA for carbon steel in 1 M HCl at different temperatures.**Table 2** Corrosion parameters obtained from weight loss for carbon steel in 1 M HCl containing various concentrations of HMDTMPA at different temperatures.

C (M)	CR (mg cm ⁻² h ⁻¹)				IE%			
	298	308	318	328	298	308	318	328
Blank	0.51	1.05	1.80	3.35	–	–	–	–
0.0005	0.23	0.57	1.23	2.42	54.58	45.71	31.66	27.76
0.001	0.16	0.44	1.02	2.10	77.97	58.09	43.33	37.31
0.002	0.11	0.33	0.82	1.91	81.87	68.57	54.44	48.95
0.004	0.05	0.17	0.54	1.42	91.61	78.09	64.44	57.61

dro's number ($6.02252 \times 10^{23} \text{ mol}^{-1}$). The plot of $\log CR$ against $1/T$ for carbon steel corrosion in 1.0 M HCl without and with different concentrations of HMDTMPA is presented in Fig. 4. All parameters are given in Table 3. The results show that the addition of HMDTMPA to the acid solution increases the value of E_a . Previous studies (Aljourani et al., 2009; Elachouri et al., 1996; Ferreira et al., 2004) showed that compared with the activation energy in the absence of inhibitor, higher values for E_a were found in the presence of inhibitor. This explains that the energy barrier of corrosion reaction increases with the concentration of inhibitor. In addition, the value of activation energy which is around 40–80 kJ mol⁻¹ can suggest that it obeys the physical adsorption mechanism (physisorption) (Orubite and Oforka, 2004). Physisorption is often related with this phenomenon, where an adsorptive film with electrostatic character is formed on the carbon surface (Popova et al., 2003).

The thermodynamic parameters (ΔS° and ΔH°) were calculated from the linear regression of transition state (Fig. 5) using Eq. (4).

Examination of the kinetic values shows that the increase of inhibitor concentration leads to increases of all parameters of corrosion process (Table 3). The positive value of the enthalpy (ΔH°) means that the process is endothermic and it needs more

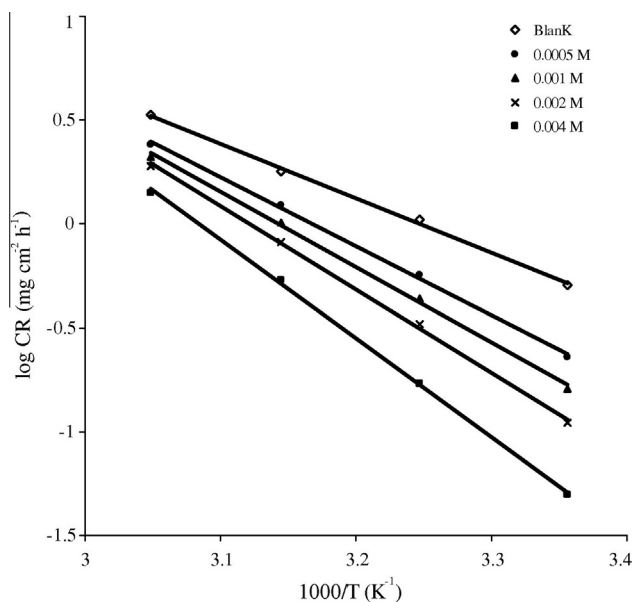


Figure 4 Arrhenius plots for carbon steel corrosion rates (CR) in 1 M HCl with and without different concentrations of HMDTMPA.

Table 3 Corrosion kinetic parameters for carbon steel in 1 M HCl with and without different concentrations of HMDTMPA.

C (M)	E_a (kJ mol ⁻¹)	R	ΔH° (kJ mol ⁻¹)	ΔS° (J mol ⁻¹)
Blank	50.33	0.998	47.73	-90.40
0.0005	63.75	0.998	61.16	-51.84
0.001	69.76	0.997	67.16	-34.62
0.002	77.12	0.998	74.53	-13.07
0.004	91.13	0.999	88.53	27.19

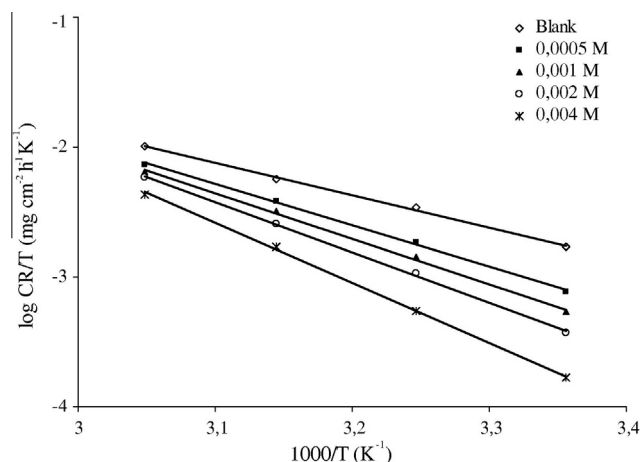


Figure 5 Transition-state plots for carbon steel corrosion rates (CR) in 1 M HCl with and without different concentrations of HMDTMPA acid.

energy to achieve the activated state or equilibrium (Bouklah et al., 2006a,b; Orubite and Oforka, 2004; Wahyuningrum et al., 2008). In addition, the entropy ΔS° , in the absence of inhibitor is negative and tends to become positive at higher concentration. The increase of ΔS° is generally interpreted by the occurrence of a disorder which takes place on going from reactants to the activated complex (Elachouri et al., 1996).

3.2. Adsorption isotherm

In order to get a better understanding of the electrochemical process on the metal surface, adsorption characteristics are also studied for HMDTMPA. This process is closely related to the adsorption of the inhibitor molecules (Hackerman and Sudbury, 1950; Hackerman, 1962; Cheng et al., 2007) and adsorption is known to depend on the chemical structure (Ateya et al., 1984; Akiyama and Nobe, 1970). Adsorption isotherms are very important in determining the mechanism of organic electrochemical reactions. Several adsorption isotherms can be used to assess the adsorption behavior of the inhibitors. The most frequently used are Langmuir, Temkin and Frumkin. In the hydrochloric acid solution, the organic compound follows the Langmuir adsorption isotherm (Trachli et al., 2002; Li and Mu, 2005; Benabdellah et al., 2011).

According to this isotherm, θ is related to C_{inh} by:

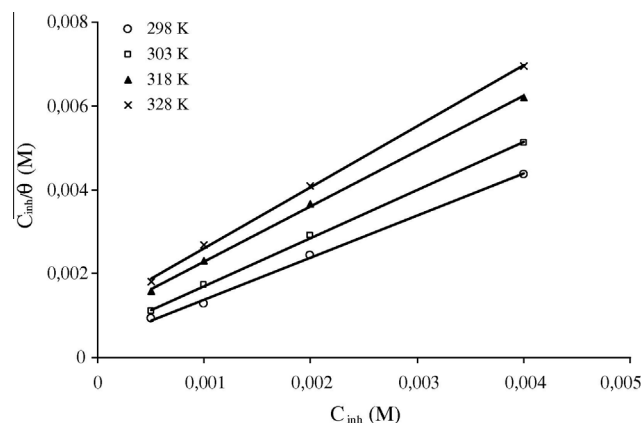
$$\frac{C_{\text{inh}}}{\theta} = \frac{1}{K_{\text{ads}}} + C_{\text{inh}} \quad (5)$$

Where θ is the surface coverage, C_{inh} is the molar concentration of inhibitor and K_{ads} is the equilibrium constant of the adsorption process.

From the values of surface coverage, the linear regressions between $\frac{C_{\text{inh}}}{\theta}$ and C_{inh} are calculated and the parameters are listed in Table 4. Fig. 6 shows the relationship between $\frac{C_{\text{inh}}}{\theta}$ and C_{inh} at various temperatures. These results show that the linear regression coefficients (R) are almost close to 1.000, indicating that the adsorption of inhibitor onto steel surface agrees to the Langmuir adsorption isotherm.

Table 4 Thermodynamic parameters for the adsorption of HMDTMPA on the carbon steel in 1 M HCl at different temperatures.

Temperature (K)	R^2	K_{ads}	ΔG_{ads}° (kJ mol ⁻¹)
298	0.999	2.50×10^3	-29.36
308	0.999	1.91×10^3	-29.65
318	0.999	1.51×10^3	-29.98
328	0.997	1.21×10^3	-30.33

**Figure 6** Langmuir's isotherm adsorption model of HMDTMPA on the carbon steel surface in 1 M HCl at different temperatures.

The equilibrium constants of the adsorption process (K_{ads}) decrease upon increasing the temperature values (Table 4). It is well known that K_{ads} indicates the adsorption power of the inhibitor onto the carbon steel surface. Clearly, HMDTMPA gives higher values of K_{ads} at lower temperatures, indicating that it was adsorbed strongly onto the carbon steel surface. Thus, the inhibition efficiency decreased slightly with the increase in temperature as the result of the improvement of desorption of HMDTMPA from the metal surface.

The standard adsorption free energy (ΔG_{ads}°) is obtained according to the following equation

$$\Delta G_{ads}^{\circ} = -2.303RT \log(55.5K_{ads}) \quad (6)$$

where R is the universal gas constant, T the thermodynamic temperature and the value 55.5 is the molar concentration of water in the solution.

The negative values of ΔG_{ads}° point to the spontaneity of the adsorption process and stability of the adsorbed layer on the carbon steel surface (Ali et al., 2005). Generally, values of ΔG_{ads} up to -20 kJ mol⁻¹ are consistent with physisorption while those around -40 kJ mol⁻¹ or higher are associated with chemisorption as a result of sharing or transferring of electrons from organic molecules to the metal surface to form a coordinate type of bond (Obot and Obi-Egbedi, 2008; Bouklah et al., 2006a,b; Ateya et al., 1984). In the present study, the calculated values of ΔG_{ads}° obtained for HMDTMPA range between -29.36 and -30.33 kJ mol⁻¹ (Table 4), indicating that the adsorption mechanism of HMDTMPA on carbon steel in 1.0 M HCl solution at the studied temperatures may obey to a physical adsorption (Ahamad et al., 2010; Trachli et al., 2002; Li and Mu, 2005; Benabdellah et al., 2011).

3.3. Potentiodynamic polarization studies

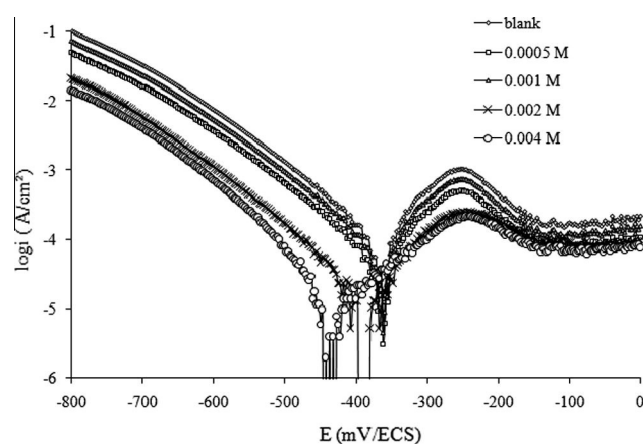
Polarization profiles for carbon steel in 1.0 M HCl in the absence and presence of HMDTMPA acid at various concentrations are shown in Fig. 7. The important corrosion parameters derived from these curves are presented in Table 5.

As shown in Fig. 7 and Table 5, cathodic current–potential curves give rise to parallel Tafel lines indicating that the hydrogen evolution reaction is under activation controlled process. Furthermore, the cathodic current density decreases with the concentration of HMDTMPA accompanied by a slight effect on the anodic portions. These results indicate that HMDTMPA acts as a mixed-type inhibitor. It seems also that the presence of the inhibitor changes slightly the corrosion potential values toward negative value.

From the values, it is clear that the corrosion current (I_{corr}) values decrease from 1400 to 133 $\mu\text{A cm}^{-2}$ with the addition of HMDTMPA. Inhibition efficiency increases upon increasing of inhibitor concentration, which suggests that the number of molecules adsorbed over the carbon steel surface become significant, blocking the active sites on this surface and protecting the metal from corrosion.

3.4. Electrochemical impedance spectroscopy

The Nyquist representations of the impedance behavior of carbon steel in 1.0 M HCl with and without addition of various concentrations of HMDTMPA are given in Fig. 8. The existence of single semi-circle showed the single charge transfer

**Figure 7** Potentiodynamic polarization curves for carbon steel in 1 M HCl containing different concentrations of HMDTMPA.**Table 5** Polarization parameters and the corresponding inhibition efficiency of carbon steel corrosion in 1 M HCl containing different concentrations of HMDTMPA at 298 K.

C (M)	E_{corr} (mV/SCE)	I_{corr} ($\mu\text{A cm}^{-2}$)	B_c (mV dec ⁻¹)	IE%
0	-362	1400	-69	–
5.10^{-4}	-367	615	-61	56.07
10^{-3}	-370	412	-66	70.57
2.10^{-3}	-407	251	-73	82.07
4.10^{-3}	-442	133	-79	90.5

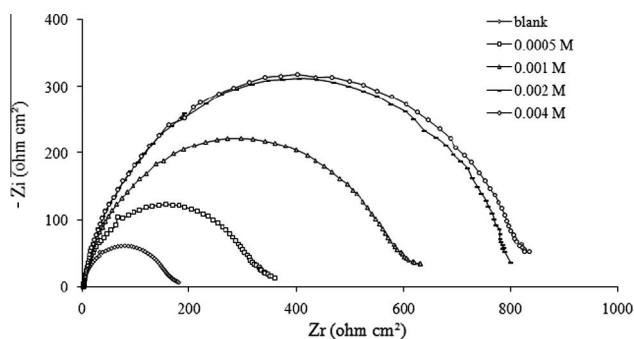


Figure 8 Nyquist plot for HMDTMPA in 1 M HCl with and without different concentrations of HMDTMPA.

Table 6 Electrochemical impedance parameters of carbon steel corrosion in 1 M HCl containing different concentrations of HMDTMPA at 298 K.

C (M)	Re (ohm cm ²)	Cd (μF/cm ²)	Rp	IE%
Blank	0.57	632	159	–
0.0005	0.97	348	325	51.08
0.001	0.16	169	595	73.27
0.002	1.44	110	805	80.24
0.004	2.41	66	1342	88.15

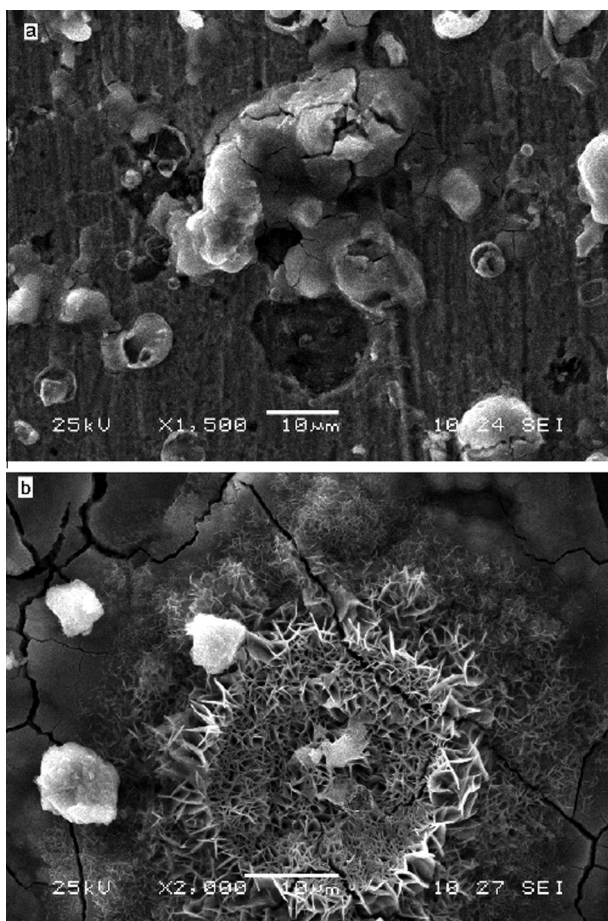


Figure 9 SEM micrographs of samples after immersion in 1 M HCl (a) without (b) with 4×10^{-3} M HMDTMPA.

process during dissolution, which is unaffected by the presence of inhibitor molecules. The charge transfer resistance (R_{ct}) and the interfacial double layer capacitance (C_{dl}) derived from these curves are given in Table 6. It is clear that addition of inhibitor increases the values of R_{ct} and reduces the C_{dl} . The decrease in C_{dl} is attributed to increase in thickness of electronic double layer (Hosseini et al., 2007; Ahamad and Quraishi, 2010). In addition, the increase in R_{ct} value is attributed to the formation of protective film on the metal/solution interface (Bentiss et al., 2000; Murlidharan et al., 1995; Ahamad and Quraishi, 2010). These observations suggest that inhibitor molecules function by adsorption at metal surface thereby causing the decrease in C_{dl} values and increasing of R_{ct} values. The data obtained from EIS technique are in good agreement with those obtained from potentiodynamic polarization and mass loss methods.

3.5. SEM analysis

The SEM photograph Fig. 9a shows that the surface is highly damaged in the absence of the inhibitor while Fig. 9b shows the formation of a film on the metal surface which causes the corrosion inhibition.

4. Conclusion

From these studies, it can be concluded that

- HMDTMPA acid is an efficient inhibitor and the inhibition efficiency increases upon increasing the concentration but decreases upon rising the temperature.
- The inhibition efficiencies obtained by polarization, EIS and weight loss measurements are in good agreement.
- Polarization curves show that the inhibitor is a mixed type one.
- The adsorption of HMDTMPA on carbon steel surface was found to be in accordance with Langmuir adsorption isotherm model. From the thermodynamic point of view, the adsorption process is spontaneous, endothermic and accompanied by a decrease in entropy of the system.
- Surface analysis shows that there are many important improvements on the surface morphology upon addition of the inhibitor.

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