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Mitigation of corrosion of carbon steel in acid medium using some antipyrine derivatives

ABSTRACT

Adsorption and inhibition efficiency of some antipyrine derivatives on the C-steel (CS) in 1M HCl were estimated using three electrochemical techniques (electrochemical impedance spectroscopy (EIS), electrochemical frequency modulation (EFM) and potentiodynamic polarization). Results of polarization showed that these derivatives act as mixed type inhibitors. These derivatives were adsorbed on CS surface following Langmuir adsorption model. EIS results showed that, there was a decrease in double layer capacitance (C_{dl}) and an increase in charge transfer resistance (R_{ct}). Three different techniques gave concordant results.

***Keywords**: Corrosion inhibition, carbon steel, HCl, antipyrine derivatives.*

1. INTRODUCTION

Corrosion is a fundamental process playing an important role in economics and safety, particularly for metals. The use of inhibitors is one of the most practical methods for protection against corrosion, especially in acidic media [1]. Most well-known acid inhibitor are organic compounds containing nitrogen, sulfur, and oxygen atoms. Among them, organic inhibitors have many advantages such as high inhibition efficiency, low price, low toxicity, and easy production [2-5]. Organic heterocyclic compounds have been used for the corrosion inhibition of iron [6-12], copper [13], aluminum [14-16], and other metals [17,18] in different corroding media. The adsorption of the surfactant heterocyclic compounds on the metal surface can markedly change the corrosion-resisting property of the metal [19,20] and so the study of the relations between the adsorption and corrosion inhibition is of great importance. Heterocyclic compounds have shown a high inhibition efficiency for iron in both HCl [21-24] and H_2SO_4 [25] solutions. The effect of two pyrazole-type organic compounds, namely ethyl 5,50-dimethyl-10H-1,30- bipyrazole-3 carboxylate (P1) and 3,5,50-trimethyl-10H-1,30-bipyrazole (P2) on the corrosion behavior of steel in 1MHCl solution was investigated [26] at 308 K by weight loss measurements, potentiodynamic polarization and impedance spectroscopy (EIS) methods. The inhibition of corrosion of steel in molar hydrochloric acid solution by two bipyrazolic compounds is studied [27] by weight loss and electrochemical polarization measurements. The two methods give consistent results. Some antipyrine compounds have been studied as corrosion inhibitors before [28] we also used some derivatives of it to inhibit corrosion of CS in 1M HCl. The inhibition effect of vanillin (4-hydroxy-3-methoxy-benzaldehyde) and protocatechualdehyde (3,4-dihydroxy-benzaldehyde) in hydrochloric acid medium on steel with known composition has been investigated [29].

The objective of the present work is to investigate the inhibiting action of some antipyrine derivatives in 1M HCl at 25^{o}C using different electrochemical techniques.

2. EXPERIMENTAL METHOD

2.1. Materials and methods

The working electrode was made from CS rod. The rod was mounted into a glass tube and fixed by araldite leaving a circle surface geometry of 1 cm diameter to contact the test solution. Prior to each experiment, the working electrode was polished with a different grades of emery paper up to 1200 grit, rinsed with acetone and finally with doubly distilled water. The auxiliary electrode was platinum wire, while reference electrode was a saturated calomel electrode (SCE). These electrodes were connected to conventional electrolytic cell of capacity 100 ml. The experiments were conducted in 1M HCl solution and with different

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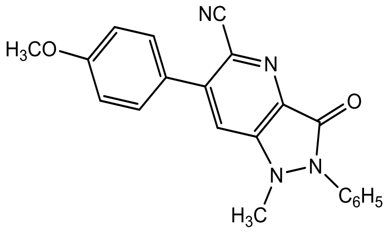
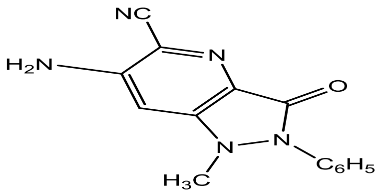
concentration ($1x10^{-6}$, 3 $x10^{-6}$, $6x10^{-6}$, $9x10^{-6}$, $15x10^{-6}$, $18x10^{-6}$ M) of organic compounds. All solutions were freshly prepared using analytical grade reagents and doubly distilled water. All experiments were performed at required temperature 25 ±1°C. The structures, names and molecular weights of the investigated organic compounds are shown below [30] :

NO	Structures and names	Chemical Formula and Molecular weight
1	 6-(4-methoxyphenyl)-1-methyl-3-oxo-2-phenyl-2,3-dihydro-1*H*-pyrazolo[4,3-*b*]pyridine-5-carbonitrile	$C_{21}H_{16}N_4O_2$ 356.38
2	 6-amino-1-methyl-3-oxo-2-phenyl-2,3-dihydro-1*H*-pyrazolo[4,3-*b*]pyridine-5-carbonitrile	$C_{14}H_{11}N_5O$ 265.28

Tafel polarization curves were obtained by changing the electrode potential automatically from -500 to +500 mV at open circuit potential with a scan rate of 1 mVs^{-1}. Stern-Geary method [31] used for the determination of corrosion current is performed by extrapolation of anodic and cathodic Tafel lines to a point which gives log i_{corr} and the corresponding corrosion potential (E_{corr}) for inhibitor free acid and for each concentration of inhibitor. Then i_{corr} was used for calculation of inhibition efficiency and surface coverage (θ) as below:

$$IE\ \% = \theta \times 100 = [1- (i_{corr(inh)} / i_{corr(free)}] \times 100 \qquad (1)$$

Where $i_{corr(free)}$ and $i_{corr(inh)}$ are the corrosion current densities in the absence and presence of inhibitor, respectively.

Impedance measurements were carried out in frequency range from 100kHz to 10mHz with amplitude of 5 mV peak-to-peak using ac signals at open circuit potential. The experimental impedance were analyzed and interpreted on the basis of the equivalent circuit. The main parameters deduced from the analysis of Nyquist diagram are the resistance of charge transfer R_{ct} (diameter of high frequency loop) and the capacity of double layer C_{dl} which is defined as:

$$C_{dl} = 1/ (2 \pi f max\ R_{ct}) \qquad (2)$$

Where f_{max} is the maximum frequency. The inhibition efficiencies and the surface coverage (θ) obtained from the impedance measurements were defined by the following relation:

$$IE\ \% = \theta \times 100 = [1- (R^{\circ}_{ct}/R_{ct})] \times 100 \qquad (3)$$

Where R^{o}_{ct} and R_{ct} are the charge transfer resistance in the absence and presence of inhibitor, respectively.

The electrode potential was allowed to stabilize 30 min before starting the measurements. All the experiments were conducted at 25±1°C. Measurements were performed using Gamry (PCI 300/4) Instrument Potentiostat/Galvanostat/ZRA. This includes a Gamry framework system based on the ESA 400. Gamry applications include DC105 for corrosion measurements and EIS300 for electrochemical impedance spectroscopy along with a computer for collecting data. Echem Analyst 5.58 software was used for plotting, graphing, and fitting data.

3. RESULTS AND DISCUSSION

3.1. Polarization method

Figures 1&2 depict the Potentiodynamic polarization curves of CS immersed in HCl solution contains different concentrations of inhibitors 1 & 2 respectively. The corrosion parameters are presented in Table 1. In the presence of inhibitors, the corrosion potentials are shifted to both cathodic and anodic sides. The largest shift evidenced by the two inhibitors system is 34 mV and 31 mV. Therefore, it is ensured that the system functions as mixed type inhibitors. Simultaneously, in the presence of inhibitors, the corrosion current decreases (Table 1). The decrease in the corrosion rate indicates that the adsorption of the inhibitors on the metal surface block the active sites and inhibit corrosion and reduce the corrosion rate with the protective film formation on the metal surface.

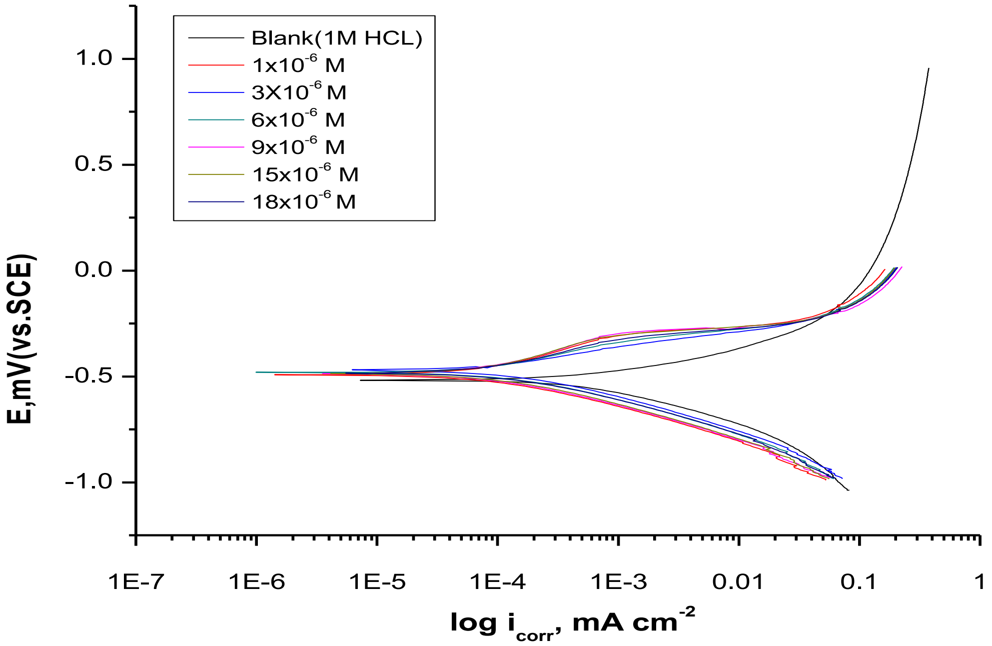


Figure 1. Potentiodynamic polarization curves for the dissolution of CS in 1M HCl in the absence and presence of different concentrations of compound (1) at 25° C
Slika 1. Potentiodinamičke polarizacione krive za CS u 1 M HCl u odsustvu i prisustvu različitih koncentracija jedinjenja (1) na 25°C

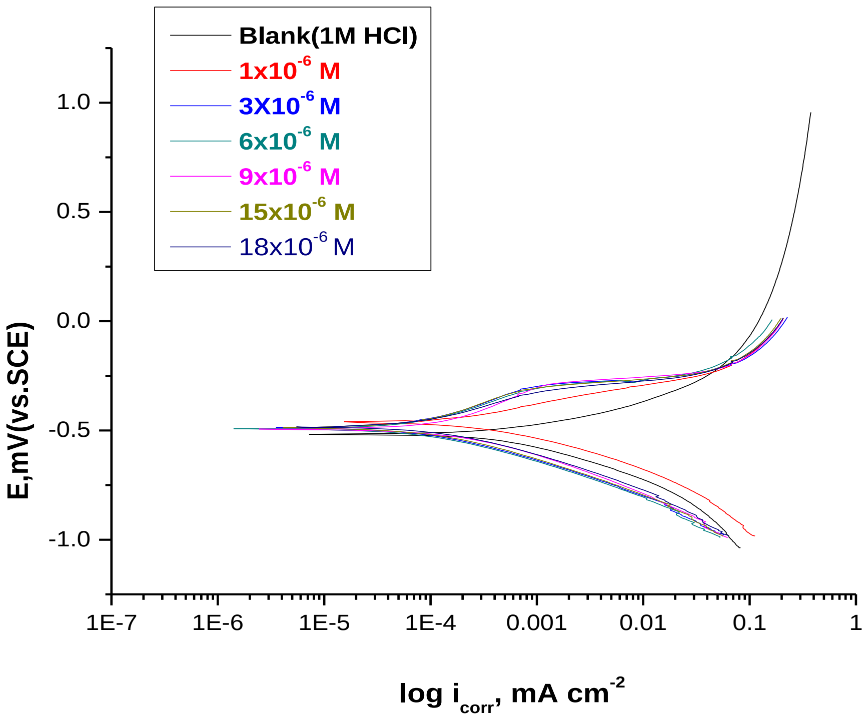


Figure 2. Potentiodynamic polarization curves for the dissolution of CS in 1M HCl in absence and presence of different concentrations of compound (2) at 25° C
Slika 2. Potentiodinamičke polarizacione krive za CS u 1 M HCl u odsustvu i prisustvu različitih koncentracija jedinjenja (2) na 25°C

Table 1. Corrosion parameters of CS immersed in HCl solution in the absence and presence of various concentrations of investigated inhibitors obtained by polarization method

Tabela 1. Korozioni parametri CS potopljeni u rastvoru HCl u odsustvu i prisustvu različitih koncentracija ispitivanih inhibitora dobijenih polarizacijonom metodom

% IE	Θ	C.R mpy	β_c mV dec^{-1}	β_a mV dec^{-1}	i_{corr} μA cm^{-2}	$-E_{corr}$ mV vs SCE	[inh] M	Comp.
-----	----	226.0	158	113	495.0	517	Blank	1
77.5	0.775	50.8	165	128	111.0	492	1×10^{-6}	
78.5	0.785	48.4	147	115	106.0	471	3×10^{-6}	
79.3	0.793	46.8	141	133	102.0	481	6×10^{-6}	
81.0	0.810	42.5	149	141	93.10	487	9×10^{-6}	
81.5	0.815	41.9	131	102	91.60	487	15×10^{-6}	
82.4	0.824	39.7	147	149	87.00	484	18×10^{-6}	
56.4	0.564	98.8	108	221	216.0	462	1×10^{-6}	2
59.4	0.594	90.8	137	238	199.0	483	3×10^{-6}	
66.1	0.661	76.8	125	235	168.0	493	6×10^{-6}	
69.3	0.693	69.4	143	231	152.0	494	9×10^{-6}	
69.5	0.695	68.9	131	223	151.0	498	15×10^{-6}	
70.5	0.705	66.8	139	251	146	458	18×10^{-6}	

3.2. Adsorption isotherm

Adsorption isotherms are very important in determining the mechanism of organ electrochemical reaction. The most frequently used isotherms are Langmuir, Frumkin, Hill de-Boer, Parsons, Temkin, Flory-Huggin and Freundlich. Basic information on the interaction between the inhibitor and the CS surface can be provided by the adsorption isotherm and in general, inhibitors can function either by physical (electrostatic) adsorption or chemisorptions with the metal. To obtain more information about the interaction between the inhibitors and CS surface, different adsorption isotherms were used. The fractional surface coverage Θ at different concentrations was determined from the potentiodynamic polarization measurements data using the Eq.1. The Langmuir isotherm is presented in the Eq.:

$$C/\Theta = (1/K) + C \quad (4)$$

Where K is the adsorption equilibrium constant, C is the concentration of the inhibitor. The adsorption equilibrium constant K is related to the free energy of adsorption ΔG^o_{ads} as:

$$K = 1/55.5 \exp(-\Delta G^o_{ads}/RT) \quad (5)$$

55.5 mol dm^{-3} is the molar concentration of water, R is the gas constant, T is thermodynamic temperature in K.

The linear relationship obtained on plotting C/Θ as function of C, with slope of unity was shown in Figure 3. The thermodynamic parameters K and ΔG^o_{ads} for the adsorption of the studied inhibitors at 25°C on CS are obtained by Langmuir adsorption isotherm and are given in Table 2.The negative values of ΔG^o_{ads} for the addition of inhibitors indicate that the process of adsorption of studied inhibitors is spontaneous in nature. The free energy of adsorption of the studied inhibitors 1 & 2 on CS at 25°C was found to be 38.18 and 35.27 kJ mol^{-1} respectively. The calculated adsorption ΔG^o_{ads} values for the two inhibitors (Table 2) shows that the adsorption is mixed one i.e. physisorption and chemisorptions. Since the values of ΔG^o_{ads} are less than 40 kJ mol^{-1} and larger than 20 kJ mol^{-1}.

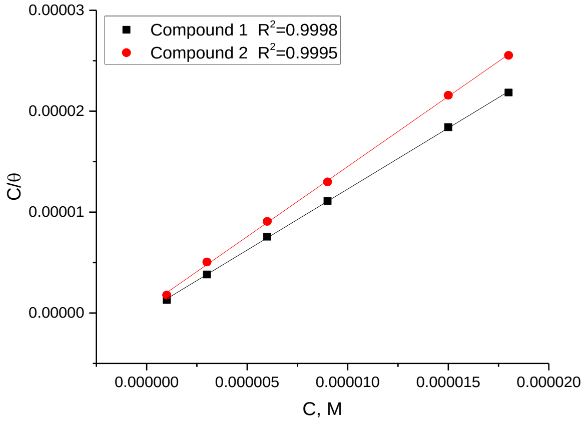


Figure 3. Langmuir adsorption isotherm of studied inhibitors on CS in 1 M HCl at 25°C

Slika 3. Langmirova adsorpciona izoterma ispitivanih inhibitora za CS u 1 M HCl na 25°C

Table 2. Thermodynamic adsorption parameters for the adsorption of studied inhibitors on CS in 1 M HCl at25°C

Tabela 2. Termodinamički adsorpcijski parametri za adsorbciju ispitivanih inhibitora na CS u 1 M HCl na 25°C

Langmuir Isotherm			Inhibitor
$-\Delta G^o_{ads.}$ kJ mol^{-1}	K x 10^6 M^{-1}	Slope	
38.18	5.12	1.2	1
35.27	1.58	1.3	2

3.3. Electrochemical frequency modulation (EFM) method

The EFM technique is used to calculate the anodic and cathodic Tafel slopes as well as the corrosion current densities for the investigated compound. Figures (4,5) show the EFM inter-modulation spectra (spectra of current response as a function of frequency) of C-steel alloy in 1M HCl. The calculated electrochemical parameters (i_{corr}, β_c, β_a, CF-2, CF-3 and %IE) are given in Table (3). The values of causality factors obtained in absence and presence of investigated compounds. It shown from the Table that the corrosion current density decreases with increasing the concentration of the investigated compounds with respect to blank and hence the inhibition efficiency increases and indicate that the investigated compounds inhibit the acid corrosion of the alloy through adsorption [32].

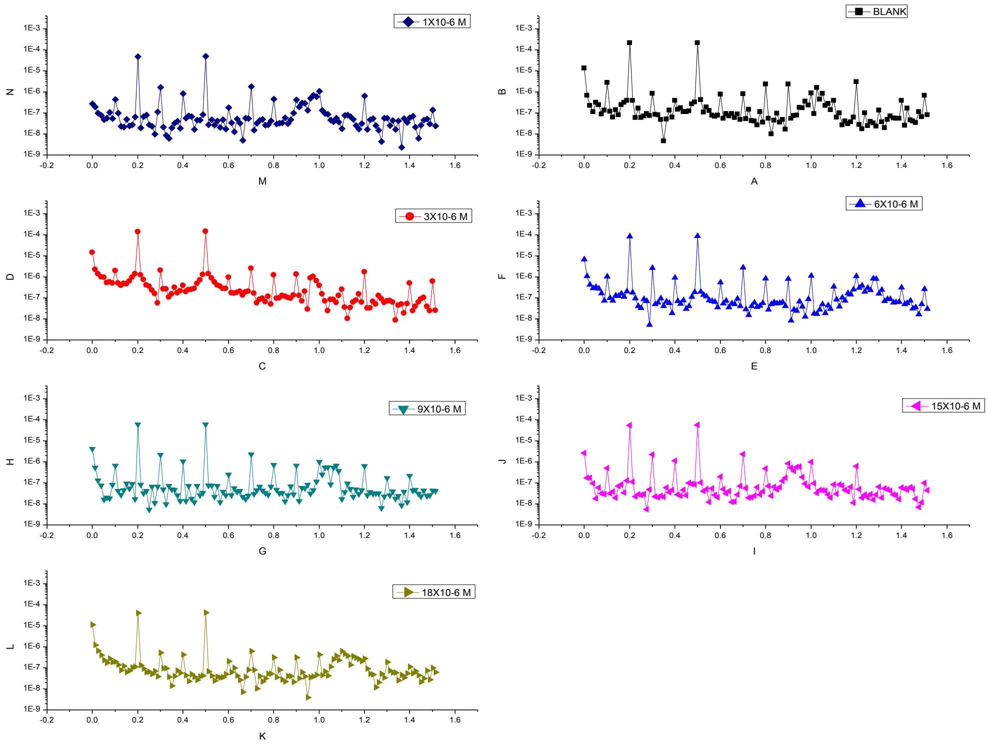


Figure 4. EFM intermodulation spectra of CS in 1M HCl for inhibitor (1) at 25°C
Slika 4. EFM intermodulacijski spektri CS u 1M HCl u prisustvu inhibitora (1) na 25°C

Table 3. Electrochemical parameters for CS determined from potentiodynamic polarization curves in1 M HCl solution without and with the addition of different concentrations of studied inhibitors at 25°C

Tabela 3. Elektrohemijski parametri za CS određeni iz potentiodinamičkih polarizacionih krivih u 1 M HCl bez i sa dodatkom različitih koncentracija ispitivanih inhibitora na 25°C

% IE	Θ	C.R mpy	CF-3	CF-2	β_c mVdec^{-1}	β_a mVdec^{-1}	i_{corr} µA cm^{-2}	[inh] M	Inhibitor
----	---	154.3	3.5	1.3	105.6	103	337.8	0	Blank
30.8	0.308	107.8	5	2	115	103	235.8	1 x10^{-6}	1
57.9	0.579	64.9	2.6	2.3	124	100	142.1	3 x10^{-6}	
68.6	0.686	48.5	4.6	2.1	128	99	106.2	6 x10^{-6}	
71.8	0.718	43.4	4	2.1	130	98	95.06	9 x10^{-6}	
73.0	0.730	41.7	1.8	1.3	152	135	91.27	15 x10^{-6}	
73.2	0.732	41.3	3	1.7	135	105	90.41	18 x10^{-6}	
27.4	0.274	112.2	3.5	2.3	120	94	245.4	1 x10^{-6}	2
45.0	0.450	84.8	2.4	2.3	139	103	185.7	3 x10^{-6}	
48.7	0.487	79.2	2.7	2.3	147	95	173.3	6 x10^{-6}	
52.9	0.529	72.6	2.5	2.5	136	94	158.8	9 x10^{-6}	
56.0	0.560	67.8	2.6	2.3	146	99	148.5	15 x10^{-6}	
59.4	0.594	62.7	1.9	2.2	160	110	137.2	18 x10^{-6}	

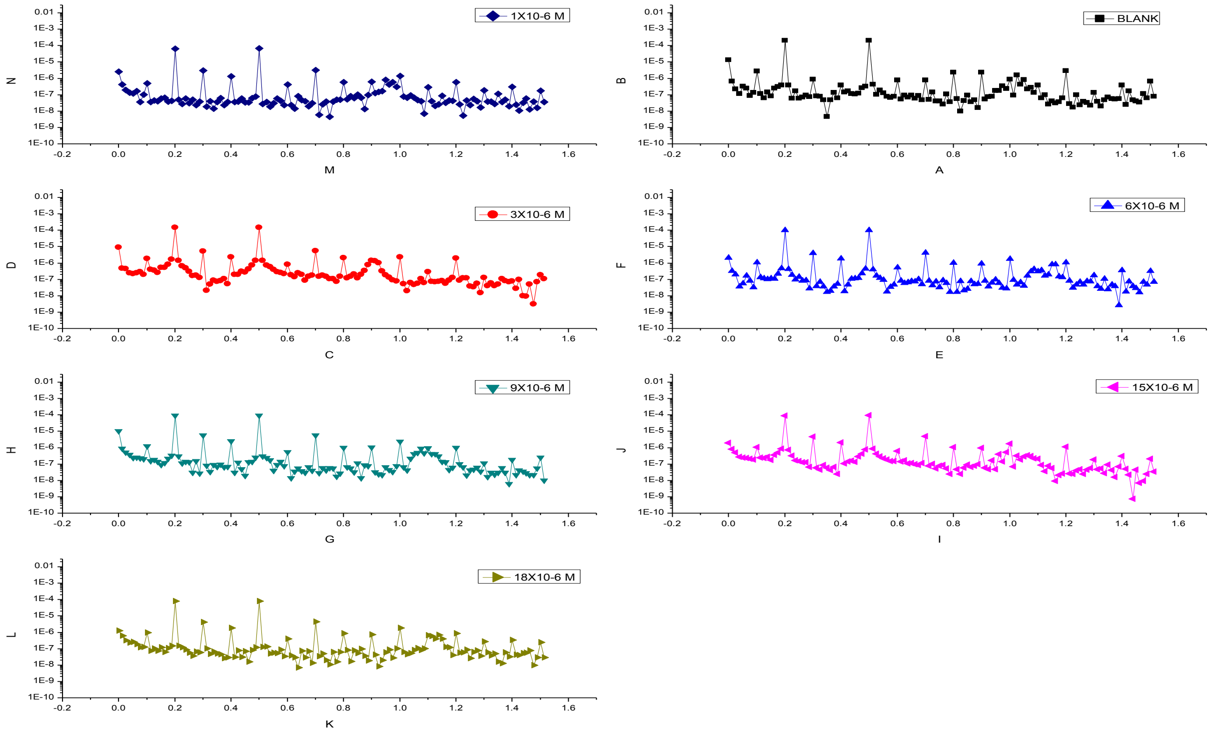


Figure 5. EFM intermodulation spectra of CS in 1M HCl for inhibitor (2) at 25°C
Slika 5. EFM intermodulacijski spektri CS u 1M HCl u prisustvu inhibitora (2) na 25°C

3.4. Electrochemical impedance spectroscopy (EIS) method

The effect of inhibitor concentration on the impedance behavior of CS in 1M HCl solution at 25 ºC is presented in Figures (6 a, b & 7 a, b) as Nyquist and Bode plots. The curves show a similar type of Nyquist plots for CS in the presence of various concentrations of investigated inhibitors. The existence of single semi-circle showed the single charge transfer process during dissolution which is unaffected by the presence of inhibitor molecules. Deviations from perfect circular shape are often referred to the frequency dispersion of interfacial impedance which arises due to surface roughness, impurities, dislocations, grain boundaries, adsorption of inhibitors, and formation of porous layers and in homogenates of the electrode surface [33].

Table 4: Electrochemical kinetics parameters obtained by EIS technique for the investigated inhibitors in 1M HCl at 25°C

Tabela 4. Elektrohemijski kinetički parametri dobijeni EIS tehnikom za ispitivane inhibitore u 1M HCl na 25°C

Inhibitor	[inh] M	R_{ct}, Ω cm^2	R_s, Ω cm^2	C_{dl}, μFcm^{-2}	Θ	% IE
Blank	1 M HCl	63.13	1.45	6.17	-----	------
1	1 x10^{-6}	88.6	1.55	3.34	0.760	28.7
	3 x10^{-6}	152.2	1.42	3.41	0.801	58.5
	6x10^{-6}	206.2	1.34	3.93	0.862	69.4
	9 x10^{-6}	226.1	1.31	3.20	0.892	72.1
	15 x10^{-6}	254.5	1.44	3.88	0.911	75.2
	18 x10^{-6}	295.8	1.18	3.24	0.921	78.6
2	1 x10^{-6}	81.8	1.39	9.74	0.694	22.9
	3 x10^{-6}	121.3	1.52	5.92	0.696	47.9
	6 x10^{-6}	130.4	1.28	5.25	0.714	51.6
	9 x10^{-6}	131.9	1.27	4.37	0.755	52.1
	15 x10^{-6}	155.1	1.35	4.60	0.781	59.3
	18 x10^{-6}	183.6	1.14	3.82	0.812	66.0

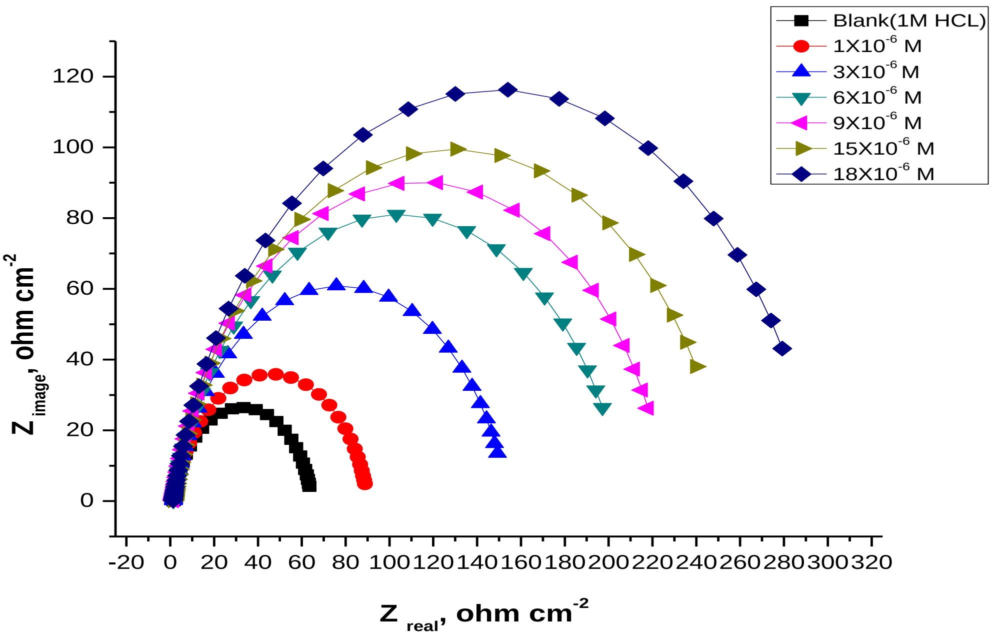


Figure 6a. The Nyquist plots for corrosion of CS in 1 M HCl in the absence and presence of different concentrations of inhibitor (1) at 25°C

Slika 6a. Nyquist-ove krive za koroziju CS u 1 M HCl u odsustvu i prisustvu različitih koncentracija inhibitora (1) na 25°C

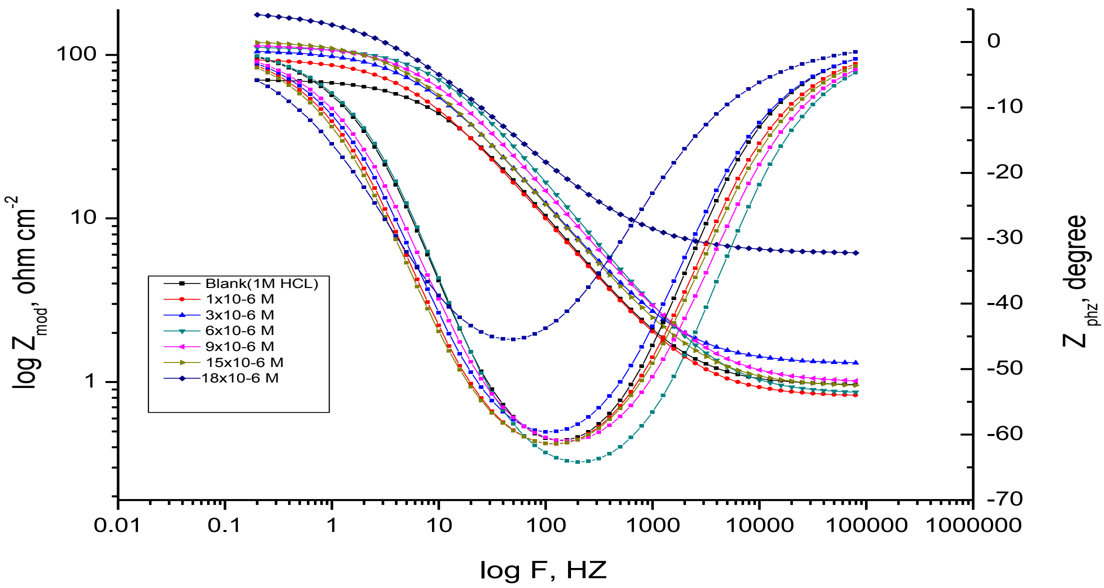


Figure 6b. The Bode plots for corrosion of CS in 1 M HCl in the absence and presence of different concentrations of inhibitor (1) at 25°C

Slika 6b. Bode-ove krive za koroziju CS u 1 M HCl u odsustvu i prisustvu različitih koncentracija inhibitora (1) na 25°C

The AC impedance parameters are given in Table 4. It is obvious from this Table that in presence of inhibitors, the R_{ct} raises from 63.13 ohm cm^2 to 295.8 and 183.6 ohm cm^2 for inhibitors 1 & 2 respectively and C_{dl} decreases from 6.17 μF cm^{-2} to 3.24 and 3,82 μF cm^{-2} for inhibitors 1 & 2 respectively. This decrease in C_{dl} and increase in Rct confirmed that the CS dissolution is reduced due to the adsorption of inhibitors on the metal surface that is indicated by the raise in impedance value

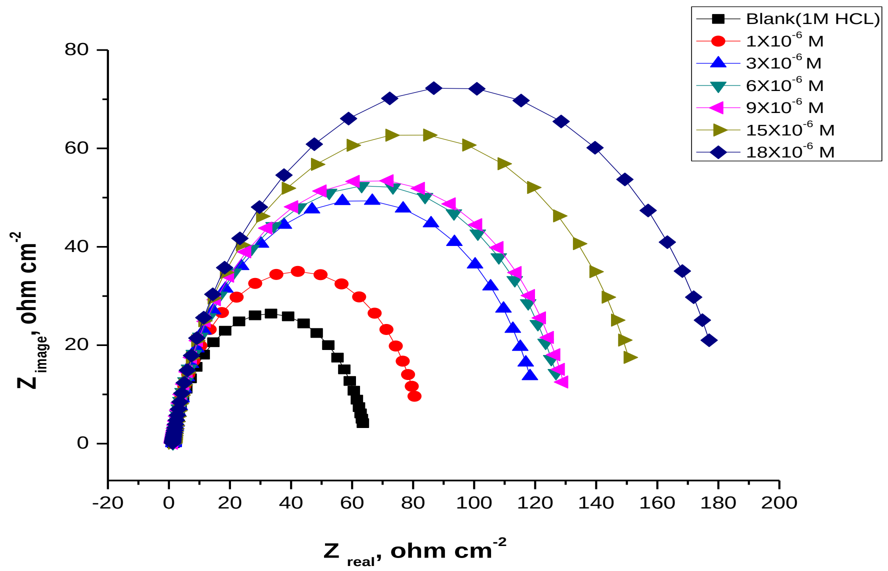


Figure 7a: The Nyquist plots for corrosion of CS in 1 M HCl in the absence and presence of different concentrations of inhibitor (2) at 25^{o}C

Slika 7a. Nyquist-ove krive za koroziju CS u 1 M HCl u odsustvu i prisustvu različitih koncentracija inhibitora (2) na 25°C

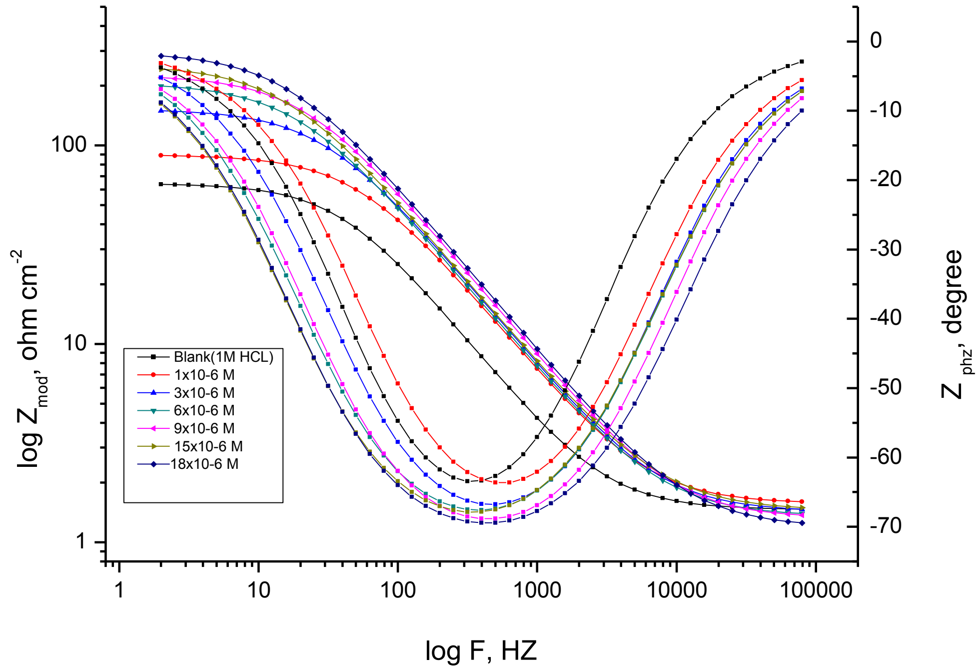


Figure 7b. The Bode plots for corrosion of CS in 1 M HCl in the absence and presence of different concentrations of inhibitor (2) at 25^{o}C

Slika 7b. Bode-ove krive za koroziju CS u 1 M HCl u odsustvu i prisustvu različitih koncentracija inhibitora (2) na 25°C

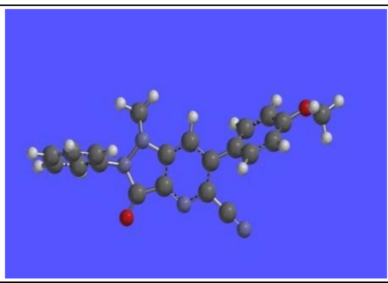
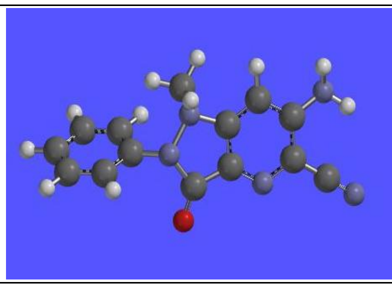
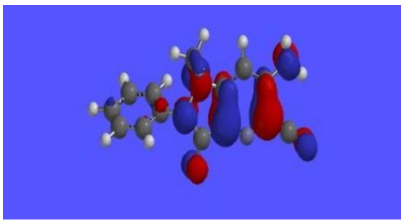
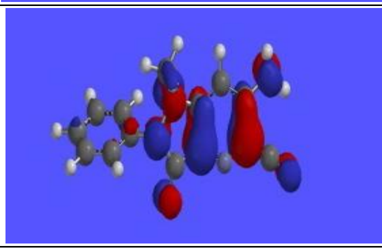
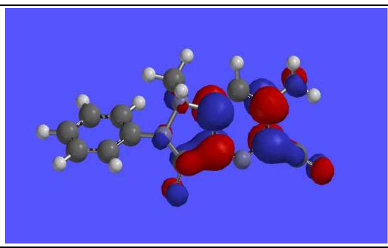
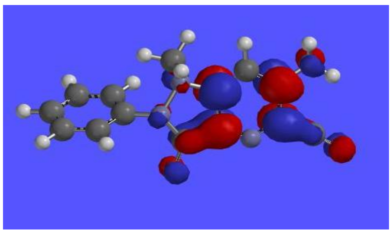
3.5. Quantum Chemical Study

It is known that the energy of highest occupied molecular orbital (E_{HOMO}) often associated with the electron donating ability of the molecules. High values of E_{HOMO} indicate a tendency of the molecule to donate electrons to act with acceptor molecules with low-energy, empty molecular orbital. Similarly, the energy of lowest unoccupied molecular orbital (E_{LUMO}) represents the ability of the molecule to accept electrons. The lower value of E_{LUMO} suggests that the molecule accepts electrons more probably [34]. The calculated quantum chemical indices, E_{HOMO}, E_{LUMO}, energy gap (ΔE) and dipole moment (μ), of investigated compounds are calculated and are shown in Table 5. Inhibition efficiency increases with increasing values of E_{HOMO}, dipole moment and with decreasing values of E_{LUMO}. The results seem to indicate, that charge transfer from the inhibitor takes place during the adsorption on the metal surface. Increasing values of E_{HOMO} and may facilitate adsorption and hence, inhibition by influencing the transport process through the adsorbed layer [35]. Similar relations were found between the inhibition efficiency and the energy gap ΔE [36]. Lower values of the energy gap will render good inhibition, because the energy to remove an electron from the last occupied orbital will be low. The dipole moment is another way to obtain data on electronic distribution in a molecule and is one of the properties more used traditionally to discuss and rationalize the structure and reactivity of many chemical systems [37]. The values of E_{HOMO} show the relation 1>2 for this property. In addition, the values of the energy gap ΔE show the relation 2 >1 for this property. The results of Table 5 show that the values of μ (dipole moment) decreases in the following order: 2 >1. Some authors showed that an increase of the dipole moment leads to decrease of inhibition and vice versa, suggesting that lower values of dipole moment will favor accumulation of inhibitor in the surface layer [38]. In contrast, the increase in the dipole moment can lead to increase of inhibition and vice versa [39, 40], which could be related to the dipole-dipole interaction of molecules and metal surface. The higher the value of μ obtained is coherent with the second explanation indicating stronger dipole-dipole interactions of inhibitor molecules and metallic surface.

Table 5. The calculated quantum chemical parameters for investigated inhibitors

Tabela 5. Izračunati hemijski parametri za ispitivane inhibitore

	Compound 1	Compound 2
$-E_{HOMO}$, (ev)	9.16	9.41
$-E_{LUMO}$, (ev)	1.18	1.31
ΔE, (eV mol^{-1})	7.96	8.10
μ, (Debye)	7.45	6.86
Molecular weight	356.38	265.28

	Compound 1	Compound 2
Structure		
E_{HOMO},		
E_{LUMO},		

3.5. Mechanism of corrosion inhibition

A clarification of mechanism of inhibition requires full knowledge of the interaction between the protective compound and the metal surface. Many of the organic corrosion inhibitors have at least one polar unit with atoms of nitrogen, sulphur, oxygen and phosphorous. It has been reported that the inhibition efficiency decreases in the order O > N > S > P. In addition iron is well known for its co-ordination affinity to heteroatom bearing ligands [34]. In HCl acid medium, molecule exist as protonated species and it is assumed that Cl^- ions are first adsorbed on the metal surface and the net positive charge on the metal surface enhances the specific adsorption of chloride ions [41]. Generally, in acid solution the inhibition of metallic corrosion occurs through (i) electrostatic interaction of protonated molecules with already adsorbed chloride ions (ii) donor-acceptor interactions between the π-electrons of aromatic ring and vacant d-orbital of surface iron atoms (iii) interaction between unshared electron pairs of heteroatoms and vacant d-orbital of iron surface atoms [42]. In the present study, the values of ΔG^o_{ads} are less than 40 kJ mol^{-1}. Hence, it shows the adsorption of the inhibitor molecules on the surface of CS predominantly takes place by the physical and chemical adsorption. Compound 1 is more effective corrosion inhibitor than compound 2. This is due to it has higher molecular size, which may cover larger area from CS surface and also has one additional phenyl ring in its structure.

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IZVOD

SMANJENJE KOROZIJE UGLJENIČNOG ČELIKA U KISELOJ SREDINI DODATKOM DERIVATA ANTIPIRINA

Apsorbciona i inhibicijska efikasnost nekih derivata antipirina na koroziju ugljeničnog čelika u 1M HCl procjenjena je pomoću tri elektrohemijske tehnike (merenjem impedanse (EIS), elektro-hemijske frekvencije (EFM) i potentiodinamičke polarizacije).
Rezultati polarizacije su pokazali da ovi derivati deluju kao inhibitori mešovitog tipa. Ovi derivati se apsorbuje na površinu ugljeničnih čelika prema Langmuirovom adsorpcionom modelu. EIS rezultati su pokazali da je došlo do pada u kapacitivnosti dvojnog sloja (CDL) i povećanje otpornosti prenosa punjenje (RCT). Tri različite tehnike dale su saglasne rezultate.
Ključne reči: *inhibicija korozije, ugljenični čelik, HCl, derivati antipirina.*

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