

Comparison of the corrosion inhibition by drugs for the corrosion of Nickel in hydrochloric acid

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Abstract: The effect of inhibitor concentration on the corrosion process has been investigated of nickel and nickel copper alloy in 0.05 M HCl at 25±1°C using electrochemical (Tafel polarization method, electrochemical impedance spectroscopy measurements) in presence of different drugs. Inhibition efficiency of the investigated compounds was found to depend on the concentration and the nature of the inhibitors. It was found that the results of these drugs on the metal surface obeys Langmuir's adsorption isotherm. The results indicated that the inhibitors could adsorb on the metal surface firmly through the heteroatoms, with excellent corrosion inhibition performance. The data obtained from electrochemical techniques are in a good agreement.

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1. Introduction

Nickel is an active metal with respect to copper in electromotive force (emf) series. In presence of oxygen, it gets attacked by dilute acids and undergoes pitting corrosion in chloride medium. In deaerated condition, it protects the base metal by forming a stable passive film of Ni(OH)₂. When alloyed with copper, which is known to have very good pitting resistance in sea water, improves its corrosion resistance under reducing environment and pitting attack in seawater.

Particularly alloy containing 60–70 wt.% Cu in Ni shows very high resistance to impingement attack and addition of 2 wt.% Fe is known to further improve its impingement resistivity⁽¹⁾. However, under cathodic protection condition alloy like commercial Monel (70 Ni–30 Cu) does not show pitting in flowing sea water. It finds application for sea water condenser tubes, pump shafts and valve trim. Alloy like, Monel-400 (67 Ni, 30 Cu, 2 Fe, 0.03 C) is used mainly in hot water tanks and for various equipment in the chemical industry and fluorination plant⁽²⁾.

The electrochemical behavior of Cu–Ni alloys in acidic chloride medium was investigated. Commercial Cu–Ni alloys were investigated using potentiodynamic techniques, complemented by electrochemical impedance spectroscopy. The influence of alloy composition, chloride ion concentration and immersion time on the electrochemical response of the alloys was analyzed. Results of present investigations with pure metals (Cu and Ni) are also considered in this paper for the sake of comparison. Potentiodynamic measurements reveal that the increase in nickel content decreases the

corrosion rate of the alloy and when the nickel content exceeds 30%, an increase in the corrosion rate was recorded. Also, the corrosion current density increases with increasing the concentration of chloride ions up to 0.6 M.⁽³⁾

The purpose of the present paper is to explore the inhibition effect of some drugs containing heteroatoms towards the corrosion of Nickel and Nickel copper alloy in hydrochloric acid, to throw some light on the mechanism of inhibition.

2. Experimental methods

2.1. Materials

The nickel metal with purity of 99.999 provided by (Johnson and Matthey England) used for all measurements.

2.2. Inhibitors

The drugs used in this study are available, easily available as lab indicators and their structures are listed in Table (1).

2.3. Solutions

The aggressive solutions, 0.05 M HCl were prepared by dilution of BDH grade (98 %) HCl with demineralized water. The concentration range of the inhibitors used was from 1x 10⁻² M to 1 x10⁻⁵ M.

2.4. Procedures used for corrosion measurements

2.4.1. Polarization measurements

Polarization experiments were carried out in a conventional three-electrode cell with a platinum counter electrode and a saturated calomel electrode (SCE) coupled to a fine Luggin capillary as the reference electrode. The working electrode was in the form of a square cut from copper embedded in epoxy resin of polytetrafluoroethylene (PTFE) so that the flat

surface was the only surface in the electrode. The working surface area was $1.0 \times 1.0 \text{ cm}^2$. Tafel polarization curves were obtained by changing the electrode potential automatically from -1000 to +1500 mV at open circuit potential with a scan rate of 5 mVs^{-1} . Stern-Geary method⁽⁴⁾ used for the determination of corrosion current is performed by extrapolation of anodic and cathodic Tafel lines to a point which gives $\log i_{\text{corr}}$ 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⁽⁵⁾:

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

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

Table (1): Structure formula of used inhibitors.

no	Structure	IUPAC Name & Chemical Formula	Molecular Weight (g/mol)
1		([2-[(5-nitro-1,3-thiazol-2-yl)carbamoyl]phenyl]ethanoate) C ₁₂ H ₉ N ₃ O ₅ S	307.283
2		3-([2-(diaminomethyleneamino)thiazol-4-yl]methylthio) N'-sulfamoylpropanimidamide C ₈ H ₁₅ N ₇ O ₂ S ₃	337.449
3		2-(2,4-Difluorophenyl)-1,3-bis(1H-1,2,4-triazol-1-yl)propan-2-ol C ₁₃ H ₁₂ F ₂ N ₆ O	306.271
4		1-[2-[(4-chloro-phenyl) methoxy]-2-(2,4-dichlorophenyl)ethyl]-1H-imidazole mononitrate C ₁₈ H ₁₅ Cl ₃ N ₂ O	381.684

2.4.2. Electrochemical impedance spectroscopy measurements

Impedance measurements were carried out in frequency range from 100 kHz to 10 mHz 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:

$$C_{\text{dl}} = 1 / (2 \pi f_{\text{max}} R_{\text{ct}}) \quad (2)$$

The inhibition efficiencies and the surface coverage (θ) obtained from the impedance measurements are defined by the following relation:

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

where R_{ct}° and R_{ct} are the charge transfer resistance in the absence and presence of inhibitor, respectively.

3. Results and Discussion

3.1 Potentiodynamic polarization technique

Potentiodynamic polarization curves of nickel in 0.05M hydrochloric acid in the absence and presence of different concentrations of some drugs compounds at 25°C are illustrated in Figs (1,2,3,4). The numerical values of the variation of corrosion current density (i_{corr}), corrosion potential (E_{corr}), Tafel slopes (β_a and β_c), degree of surface coverage (θ) and inhibition efficiency (%IE) with the concentrations of different drugs compounds are given in Tables (2,3,4,5).

The data of these figs (1,2,3,4) indicate that:

1- The cathodic and anodic curves obtained exhibit Tafel-type behavior. Addition of these organic compounds increased both cathodic and anodic overvoltages and caused mainly parallel displacement to the more negative and positive values, respectively.

2- The corrosion current density (i_{corr}) decreases with increasing the concentration of these compounds, which indicates that the presence of these compounds retards the dissolution of nickel in 0.05 M HCl solution and the degree of inhibition

depends on the concentration and type of the inhibitor present.

3- The order of the increase in the inhibition efficiency for the additives is:

compound(3) > compound(2) > compound(4) > compound(1)

Which was consistent with the sequence obtained from open circuit determinations?

4- The investigated compounds appeared to act as mixed type inhibitors, as it is shown from Figs. (1,2,3,4) where both cathodic and anodic polarization curves are influenced by the presence of the inhibitive compounds in the corrosive media. In addition, the cathodic and anodic Tafel lines were more or less parallel. In other words, the inhibitor did not change the slope of the cathodic or anodic polarization curves, as is evident from Tables (2,3,4,5). This indicates that the hydrogen evolution and metal dissolution reactions were activation controlled and the addition of inhibitors did not modify the mechanism of these processes⁽⁶⁻⁷⁾. The inhibitive action of these investigated compounds occurs by blocking the active sites on the electrode surface and consequently decreasing the surface area available for the electrochemical reactions.

5- The corrosion potential (E_{corr}) does not remarkably shift in presence of these investigated compounds, therefore these compounds can be described as mixed type inhibitors for nickel in 0.05M HCl solution⁽⁸⁾.

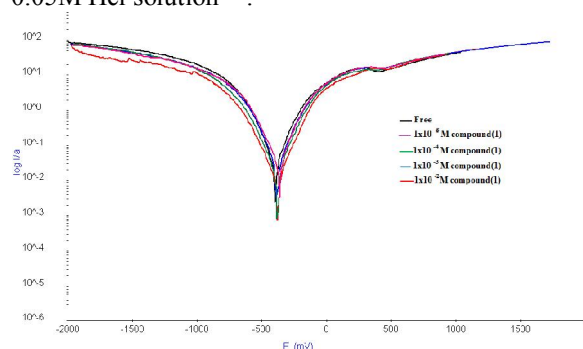


Fig (1): Potentiodynamic polarization curves for Nickel electrode in 5×10^{-2} M HCl in absence and presence of different concentrations of compound (1) at 25 °C

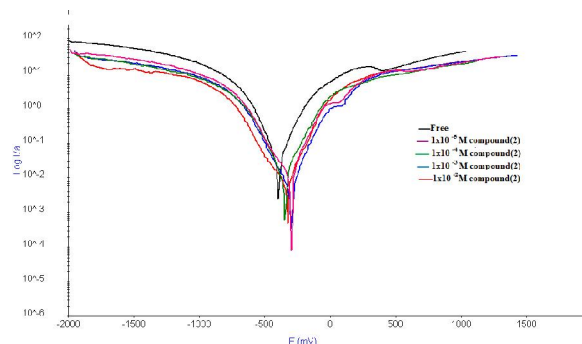


Fig (2): Potentiodynamic polarization curves for Nickel electrode in 5×10^{-2} M HCl in absence and presence of different concentrations of compound (2) at 25 °C

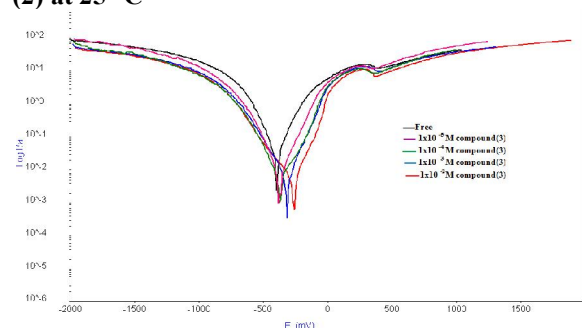


Fig (3): Potentiodynamic polarization curves for Nickel electrode in 5×10^{-2} M HCl in absence and presence of different concentrations of compound (3) at 25 °C

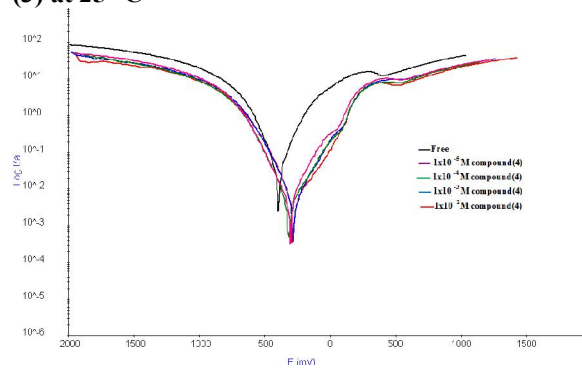


Fig (4): Potentiodynamic polarization curves for Nickel electrode in 5×10^{-2} M HCl in absence and presence of different concentrations of compound (4) at 25 °C

Table (2): Corrosion parameters obtained from potentiodynamic polarization curves of Nickel in 0.05 M HCl containing various concentrations of compound(1), at 25 °C:

Compound	Conc, M	$-E_{corr}$ mV, vs SCE	i_{corr} A/Cm ⁻²	β_c V/dec	β_a V/dec	θ	% IE
Blank	0.0	570	0.0334	0.0345	0.0323	-----	-----
Compound d (1)	1×10^{-2}	316	0.0143	0.0332	0.0255	0.57	57
	1×10^{-3}	767	0.0127	0.0426	0.0228	0.32	32
	1×10^{-4}	567	0.0189	0.0639	0.0204	0.43	43
	1×10^{-5}	371	0.0320	0.0342	0.0298	0.04	40

Table (3): Corrosion parameters obtained from potentiodynamic polarization curves of Nickel in 0.05 M Hcl containing various concentrations of compound(2), at 25 °C:

Compound	Conc, M	$-E_{\text{corr}}$ mV, vs SCE	i_{corr} mA/Cm ⁻²	β_c mV/dec	β_a mV/dec	θ	% IE
Blank	0.0	570	0.0334	0.0345	0.0323	-----	-----
Compound d(2)	1×10^{-2}	291	0.0060	0.0030	0.0075	0.82	82
	1×10^{-3}	364	0.0080	0.0046	0.0054	0.76	76
	1×10^{-4}	395	0.0104	0.0079	0.0019	0.68	68
	1×10^{-5}	331	0.0130	0.0063	0.0143	0.60	60

Table (4): Corrosion parameters obtained from potentiodynamic polarization curves of Nickel in 0.05 M Hcl containing various concentrations of compound(3), at 25 °C

Compound	Conc, M	$-E_{\text{corr}}$ mV, vs SCE	i_{corr} mA/Cm ⁻²	β_c mV/dec	β_a mV/dec	θ	% IE
Blank	0.0	570	0.0334	0.0345	0.0323	-----	-----
Compound d(3)	1×10^{-2}	074	0.0036	0.0034	0.0038	0.88	85
	1×10^{-3}	312	0.0054	0.0064	0.0044	0.80	80
	1×10^{-4}	483	0.0089	0.0087	0.0031	0.72	72
	1×10^{-5}	331	0.0160	0.0104	0.0035	0.52	52

Table (5): Corrosion parameters obtained from potentiodynamic polarization curves of Nickel in 0.05 M Hcl containing various concentrations of compound(4), at 25 °C

Compound	Conc, M	$-E_{\text{corr}}$ mV, vs SCE	i_{corr} mA/Cm ⁻²	β_c mV/dec	β_a mV/dec	θ	% IE
Blank	0.0	570	0.0334	0.0345	0.0323	-----	-----
Compound d(4)	1×10^{-2}	286	0.0045	0.0054	0.0036	0.85	85
	1×10^{-3}	288	0.0087	0.0090	0.0085	0.73	73
	1×10^{-4}	237	0.0133	0.0040	0.0027	0.60	60
	1×10^{-5}	315	0.0153	0.0035	0.0050	0.54	54

3.2. Electrochemical Impedance Spectroscopy (EIS)

Electrochemical Impedance Spectroscopy (EIS) is a nondestructive electrochemical method that is used to evaluate the electrochemical properties of electrode and electrode/electrolyte interface. EIS provides a new method to characterize the film coverage on the electrode, which is related to charge transfer resistance (R_{ct}). The interface capacitance can also be used to determine the film quality⁽⁹⁻¹²⁾. It is known that the coverage of an organic substance on the metal surface depends not only on the structure of the organic substance and the nature of the metal, but also on the experimental conditions such as immersion time and concentration of adsorbent.

Impedance is the general term similar to resistance and is used where complex behavior of one or more electrical circuit elements exists. The complex function is expressed in one or both of the two graphical representations called Bode and Nyquist plots. From the Nyquist plot it is possible to extract

few electrochemical parameters such as solution resistance, polarization resistance and total resistance. Also it is possible to determine the number of time constants involved in the electrochemical reaction by looking at the shape of the Nyquist plot. Each semicircle in the Nyquist plot is characteristic of a time constant involved in the electrochemical process. A Nyquist plot does not show any frequency value although some definite frequency was used to get the impedance at each data point. To overcome this shortcoming, a Bode plot was developed to indicate exactly what frequency was used to create a data point.

From the Bode plot it is possible to read impedance with respect to frequency and also the involvement of capacitance and resistance in the electrochemical reaction from the phase angle and frequency, allowing us to make a good judgment on resistor, capacitor and inductor and design the equivalent electric circuit accordingly. The rate of transfer of charged particles within the electrolyte,

electrode and at electrode/electrolyte interface depends on the resistance of the electrolyte, electrode and on the rate of reaction at the interface. Surface structural defects, crystallographic orientation, inclusion of foreign species can also influence the local electric field. EIS always applies a small potential disturbance in order to maintain the pseudo linearity in the cell's response. For small electrical signals the response is always pseudo linear with a small change in phase between the applied and response signal.

The corrosion behavior of nickel in 0.05 M HCl solution in the absence and presence of different concentrations of these organic compounds was investigated by the EIS technique at 25±1°C. Figs (5,6,7,8) show the Nyquist plots for copper in 0.05 M HCl solution in the absence and presence of different concentrations of investigated these organic compounds at 25±1°C. The obtained Nyquist impedance diagrams in most cases does not show perfect semicircle, generally attributed to the frequency dispersion⁽¹³⁾ as a result of roughness and in homogenates of the electrode surface. The data reveal that, each impedance diagram consists of a large capacitive loop with low frequencies dispersion (inductive arc). This inductive arc is generally attributed to anodic adsorbed intermediates controlling the anodic process^(14,15). By following this, inductive arc was disregarded.

The impedance spectra of the different Nyquist plots were analyzed by fitting the experimental data to a simple equivalent circuit model as given in Figs.(9,10), which includes the solution resistance R_s and the double layer capacitance C_{dl} which is placed in parallel to the charge transfer resistance R_{ct} ⁽¹⁶⁾.

In 0.05 M HCl with the presence of various concentrations of inhibitors, the impedance diagrams show the same trend (one capacitive loop), however, the diameter of this capacitive loop increases with increasing concentration.

The main parameters deduced from the analysis of Nyquist diagram are:

The resistance of charge transfer R_{ct} (diameter of high frequency loop)

The capacity of double layer C_{dl} which is defined as:

$$C_{dl} = \frac{1}{2 \pi f_{max} R_{ct}} \quad (4)$$

where f is the frequency, the surface coverage (θ) and the inhibition efficiency obtained from the impedance measurements are defined by the following relations:

$$\theta = \left(1 - \frac{R_{ct}^0}{R_{ct}} \right) \quad (5)$$

$$\% IE = \left(1 - \frac{R_{ct}^0}{R_{ct}} \right) \times 100 \quad (6)$$

Where R_{ct}^0 and R_{ct} are the charge transfer resistance in the absence and presence of different concentrations of inhibitor, respectively.

The impedance data such as C_{dl} , R_{ct} , θ and %IE were estimated by assuming Randles circuit Fig(9) is given in Table (6,7,8,9).

From the impedance data given in Tables (6,7,8,9), we conclude that:

The value of R_{ct} increases with the increasing the concentration of the inhibitors and hence, the increase in the corrosion inhibition efficiency in acidic solution. This can be attributed to the formation of protective film on the metal solution interface⁽¹⁷⁾.

The existence of single semicircle showed that single charge transfer process occurred during dissolution of nickel which unaffected by the presence of investigated compounds.

The %IE obtained from EIS measurements are nearly close to those obtained from polarization.

The order of inhibition efficiency obtained from EIS measurements is:

Compound(3) > compound(2) > compound(4) > compound(1).

According to Figure (10), the charge transfer resistance determines the corrosion rate and represents the resistance between the metal/OHP (outer Helmholtz plane). Figure (10) describes the potential distributions on the metal/solution interface and proposed electrical equivalent circuit diagram for the corrosion system blank solution.

The analysis of Nyquist from experimental data is done by using the circuit in Figure (9), in which R_s represents the electrolyte resistance, R_{ct} represents the charge-transfer resistance and the constant phase element (CPE).

Nyquist impedance plots obtained for the nickel electrode at respective corrosion potentials after 15 min immersion in 0.05 M HCl in presence and absence of various concentrations of compound(1), compound(2), compound(3) and compound(4). These diagrams exhibits a single semi-circle shifted along the real impedance (Z_r) axis. These diagrams show single capacitive loop, which is attributed to charge transfer of the corrosion process, and the diameters of the loops increase with the increase of concentrations.

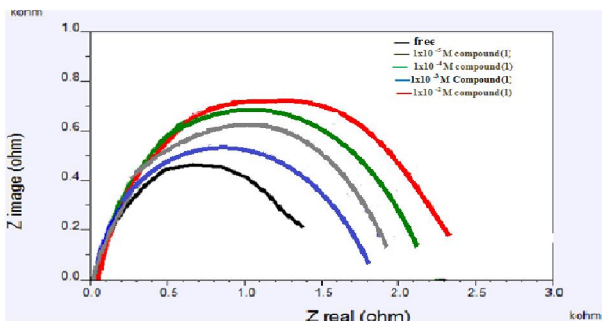


Fig (5): Potentiostatic EIS Nyquist for nickel electrode in 5×10^{-2} M HCl in absence and presence of different concentrations of compound (1) at 25°C

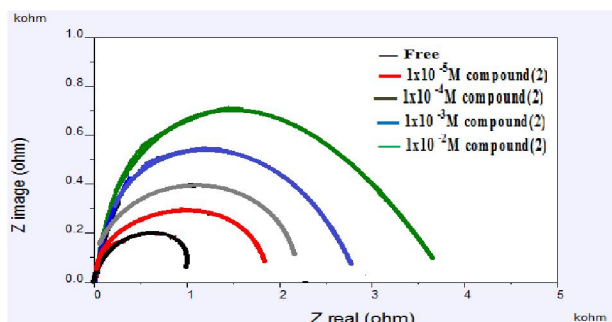


Fig (6): Potentiostatic EIS Nyquist for nickel electrode in 5×10^{-2} M HCl in absence and presence of different concentrations of compound (2) at 25°C

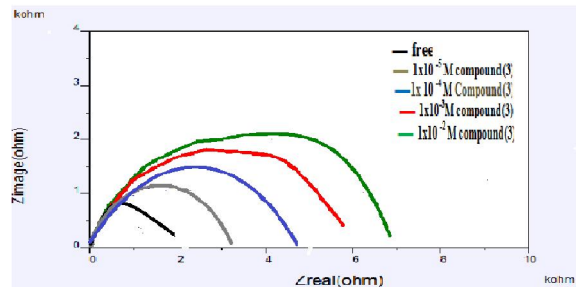


Fig (7): Potentiostatic EIS Nyquist for nickel electrode in 5×10^{-2} M HCl in absence and presence of different concentrations of compound (3) at 25°C

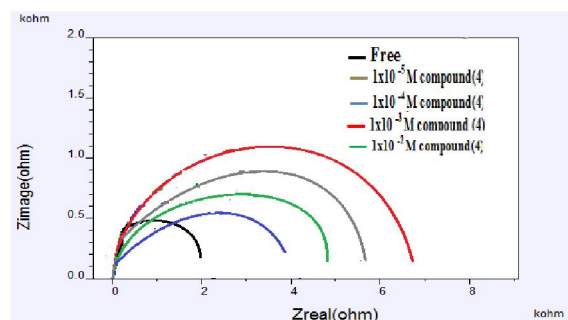


Fig (8): Potentiostatic EIS Nyquist for nickel electrode in 5×10^{-2} M HCl in absence and presence of different concentrations of compound (4) at 25°C

Table (6): EIS data of nickel in 0.05 M HCl in the absence and presence of different concentrations of compound(1), at 25°C :

Conc. M	$R_s, \Omega\text{cm}^2$	N	$R_{ct}, \Omega\text{cm}^2$	C_{dl}, Fcm^{-2}	θ	%IE
free	72.1	0.9999	1460	2.57×10^{-4}	-	-
1×10^{-2}	103.5	0.8989	2060	4.65×10^{-5}	0.29	29
1×10^{-3}	46.2	0.9900	5870	4.06×10^{-4}	0.75	75
1×10^{-4}	137	0.9699	2178	1.51×10^{-4}	0.33	33
1×10^{-5}	197	0.9399	2413	1.25×10^{-3}	0.39	39

Table (7): EIS data of nickel in 0.05 M HCl in the absence and presence of different concentrations of compound (2), at 25°C :

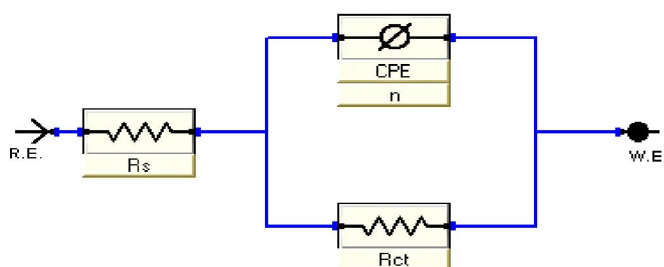
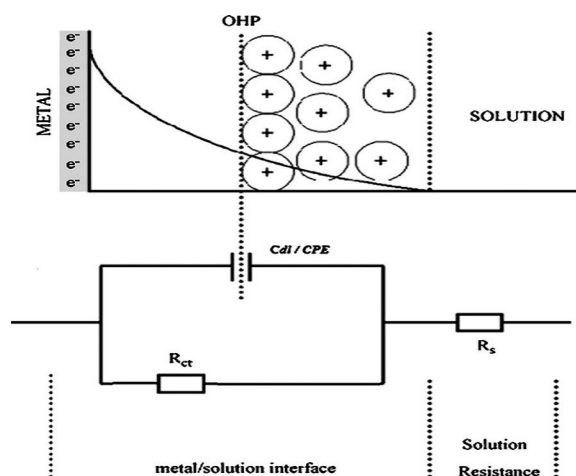
Conc. M	$R_s, \Omega\text{cm}^2$	N	$R_{ct}, \Omega\text{cm}^2$	C_{dl}, Fcm^{-2}	θ	%IE
free	72.1	0.9999	1460	2.57×10^{-4}	-	-
1×10^{-2}	101.4	0.9599	5026	5.06×10^{-5}	0.70	71
1×10^{-3}	162.3	0.9999	4552	8.564×10^{-5}	0.67	67
1×10^{-4}	181.5	0.9999	3563	1.11×10^{-4}	0.59	59
1×10^{-5}	214.4	0.9999	2862	1.43×10^{-4}	0.48	50

Table (8): EIS data of nickel in 0.05 M HCl in the absence and presence of different concentrations of compound (3), at 25 °C:

Conc. M	$R_s, \Omega\text{cm}^2$	N	$R_{ct}, \Omega\text{cm}^2$	C_{dl}, Fcm^{-2}	θ	%IE
free	72.1	0.9999	1460	2.57×10^{-4}	-	-
1×10^{-2}	306.8	0.9999	6780	5.235×10^{-5}	0.78	80
1×10^{-3}	293.6	0.9999	6034	4.327×10^{-5}	0.75	76
1×10^{-4}	264.1	0.9999	4438	8.756×10^{-5}	0.67	67
1×10^{-5}	242.5	0.9999	4388	5.168×10^{-5}	0.66	66

Table (9): EIS data of nickel in 0.05 M HVI in the absence and presence of different concentrations of compound (4), at 25 °C:

Conc. M	$R_s, \Omega\text{cm}^2$	N	$R_{ct}, \Omega\text{cm}^2$	C_{dl}, Fcm^{-2}	θ	%IE
free	72.1	0.9999	1460	2.57×10^{-4}	-	-
1×10^{-2}	134	0.9999	2800	3.797×10^{-5}	0.47	47
1×10^{-3}	228.7	0.9999	3830	7.061×10^{-5}	0.61	61
1×10^{-4}	153	0.9999	2620	4.70×10^{-5}	0.45	45
1×10^{-5}	258.4	0.9999	4128	9.734×10^{-5}	0.64	64

**Fig(9): Equivalent circuit used to model impedance data in 0.05 M HCl solutions, OHP, outer Helmholtz plane; C_{dl} , double layer capacitance; R_s , solution resistance; R_{ct} , charge-transfer resistance.****Fig(10): Equivalent circuit model used to fit the impedance spectra**

4. Conclusions

- 1- The tested drugs compounds establish a very good inhibition for Nickel in HCl solution.
- 2- drug compounds inhibit Nickel corrosion by adsorption on its surface and act better than the passive oxide film.
- 3- The inhibition efficiencies of the tested drug compounds increase with increasing of their concentrations.
- 4- Double layer capacitances decrease with respect to blank solution when the inhibitor added.
- 5- Polarization data shows that the used inhibitors act as mixed-type inhibitor in 0.05 M HCl.
- 6- The values of inhibition efficiencies obtained from the different independent quantitative techniques used are in good agreement and showed the validity of the results.

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