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REGULAR ARTICLE

Electrochemical and Thermodynamic Investigation of Drug Solfarlem as a Potential Corrosion Inhibitor for Nickel-Phosphorus Coatings

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There are more materials being utilized as industry develops, and these materials are frequently exposed to severe corrosion while being employed. This depends on the material/corrosive pair and the environment in which these two elements come together; among the finest methods is the application of inhibitors to ensure their protection in contact with aggressive environments such as hydrochloric acid. This work describes the study of the corrosion behavior of a Nickel-Phosphorus coating in an acidic 1M HCl medium and the evaluation of the protective power of an organic compound. The Ni-P alloys were obtained by the electrodeposition technique from a solution containing Nickel Sulfate and Sodium Hypophosphite (NaH_2PO_2) on a steel substrate. The effect of drug Solfarlem as a corrosion inhibitor of Nickel-Phosphorus alloy in 1 M HCl was studied using electrochemical impedance spectroscopy, polarization curve and weight loss methods. Results show that the inhibition efficiency of this inhibitor by all methods was in good agreement and the optimal efficiency with a concentration of 6.10^{-5} mol/L was about 96%. The values for free energy of adsorption ΔG_{ads}, were calculated at each surface coverage, of the studied compound by applying the mathematical model. The adsorption of the inhibitor molecules corresponds to the Langmuir adsorption isotherm, followed by chemical adsorption on the surface of the Nickel-Phosphorus alloy.

Keywords: Nickel-Phosphorus alloy, Electrochemical techniques, Adsorption, Drug solfarlem.

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1. INTRODUCTION

There are more materials being utilized as industry develops, and these materials are frequently exposed to severe corrosion while being employed. This depends on the material/corrosive pair and the environment in which these two elements come together. Ni-P coatings are extensively employed in industrial applications, particularly in the automotive, aviation, and pipeline industries because of their mechanical magnetic properties and good resistance to corrosion and wear [1, 2].

Metals commonly exposed to industrial environments are more susceptible to attack in aggressive media. Acid solutions are employed in a wide range of industrial processes, and some of the most important fields of application are acid pickling, chemical cleaning and processing, ore production, and oil well acidification [3, 4]. The importance of protection against corrosion in acidic solutions is increased by the fact that materials cleaned with chemical acids corrode the metal after the corrosion products are removed. It has become necessary to look for ways to combat the corrosion of metals and alloys that come into contact with aggressive environments. The most commonly used methods are inhibitors, which effectively reduce unwanted destructive effects and prevent metal dissolution. The inhibitors most commonly used in acidic environments are organic compounds that contain nitrogen, sulfur, and oxygen atoms. In recent years, researchers have been interested in developing drugs that can inhibit metal corrosion in acidic media. [5-9].

The commercial drug Sulfarlem is an example of a new class of corrosion inhibitors. The aim and the objective of the present work is to investigate the efficiency of The commercial drug Sulfarlem as corrosion inhibitors of Ni-P alloy in acidic media, in order to study the influence of a sulfur atom and aromaticity, in the part of the molecule, we studied Sulfarlem as a corrosion inhibitor of Ni-P coatings in a 1 M HCl solution using weight loss and electrochemical techniques. The thermodynamic parameters obtained from these studies have also been reported.

2. MATERIALS AND METHODS

Nickel Phosphorus alloy were deposited on steel substrate. The bath composition and experimental conditions are mentioned in the article [10, 11].

This inhibitor was prepared from the commercial drug Solfarlem, which is a polymer-coated tablet. We melted 30 tablets in 50 mL of toluene after complete dissolution of the compound. After evaporation and recy-

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cling of toluene from the toxin, we obtained red and orange needles.

The concentration of hydrochloric acid (1 M HCl) was prepared by dilution of analytical grade HCl (37 %) in distilled water. The concentration of the inhibitor used ranged from (2 to 8) × 10^{-5} mM/L, and the electrolyte volume was 300 ml.

The corrosion tests were carried out using the gravimetric method by immersing the samples in an aggressive solution of HCl (1M) without and with different concentrations of inhibitor for a period of 15 days at room temperature. The samples were then removed and rinsed with distilled water, dried, and weighed. The inhibition efficiency IE (%) was calculated using Eq. (1)

$$\mathrm{IE} = [\frac{1-W}{W0}].100 \qquad (1)$$

Where: W_0 and W are Ni-P coatings corrosion rates (mg $\mathrm{cm}^{-2}\,\mathrm{h}^{-1}$) in the absence and presence of the inhibitor, respectively.

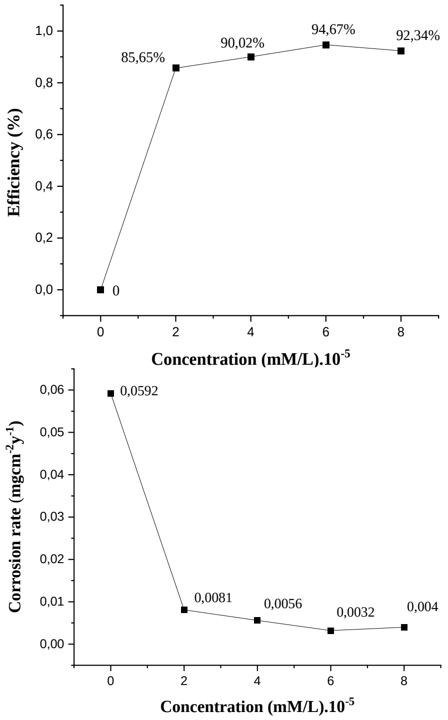

Fig. 1 – Inhibition efficiency and corrosion rate for Ni-P coating in 1 M HCl solution without and with different concentrations of inhibitor

Potentiodynamic polarization measurements were conducted at ambient temperature using an electrochemical measurement system controlled with a Potentiostat/Galvanostat pgz301 corrosion analysis. The tests were carried out in a three electrode cell filled with 500 ml of 1 M HCl solution, with the Ni-P as the working electrode (1 cm^2), a Pt auxiliary electrode and a statured calomel reference electrode. All potentials are reported Vs. SCE. For potentiodynamic polarization, the working electrode was in the form of a disc cut from steel and coated specimens with an exposed area of 1 cm^2. Calomel electrode was placed close to the working electrode to minimize Ohmic resistance. Potentiodynamic polarization tests were conducted with a scan rate of 20 mV/ min in a potential range from – 600 to – 200 mV. Corrosion rate (mg $\mathrm{cm}^{-2}\,\mathrm{y}^{-1}$), corrosion potential E_{corr} (mV) and corrosion current density i_{corr} (A dm^{-2}) have been calculated by using Tafel extrapolation technique provided by Volta master 4 software, the EIS measurements have been performed, using a same device worked and carried out in the frequency range of 10 KHz–10 mHz. The applied sinusoidal signal amplitude is 10 mV.

3. RESULTS AND DISCUSSION

3.1 Weight Loss Measurements

The corrosion parameters of the Ni-P alloy, obtained by the weight loss measurements with inhibitor concentrations ranging from 2.10^{-5} to 8.10^{-5} mM/L in a 1 M HCl solution, are listed in Table 1. As shown in Fig. 1, the corrosion rate was reduced with the addition of the inhibitor, and the inhibition efficiency increased as the inhibitor concentration increased.

Table 1 – Corrosion rate of Ni-P coating after 15 days of immersion in 1 M HCl at different inhibitor concentrations

	C_{inh} (mM/L)	Surface coverage θ	E (%)	Corrosion rate (mg $\mathrm{cm}^{-2}\,\mathrm{y}^{-1}$) × 10^{-2}
Ni-P	00	00	00	0.0592
Ni-P with inh	2.10^{-5}	0,8565	85,65	0.0081
	4.10^{-5}	0.9002	90,02	0.0056
	6.10^{-5}	0.9467	94,67	0.0032
	8.10^{-5}	0.9234	92,34	0,0040

3.2 Polarization Curves

The polarization test results of the Ni-P coating in 1 M HCl solution without and with the inhibitor at room temperature are presented and summarized in Fig. 2 and Table 2. According to the polarization curves results

Table 2 – Ni-P alloy corrosion polarization parameters in acid solutions with and without different inhibitor concentrations

	C_{inhi} (mM L^{-1})	E_{corr} (i = 0) mV	I_{corr} (µA cm^{-2})	β_a (mV)	β_c (mV)	R_p kΩ cm^2	T_{corr} (µm y^{-1})	IE (%)
Blank	00	– 267.4	2.0636	83.89	– 107.1	9.71	24.13	00
With inhibitor	2.10^{-5}	– 141.5	0.3522	88.40	– 111.8	13.56	14.50	82.93
	4.10^{-5}	– 119.3	0.1584	42.6	– 49.3	13.78	7.97	92.32
	6.10^{-5}	– 107.8	0.0755	43.4	– 55	14.35	7.22	96.34
	8.10^{-5}	– 104.6	0.1956	81.7	– 130.8	11.35	17.41	90.52

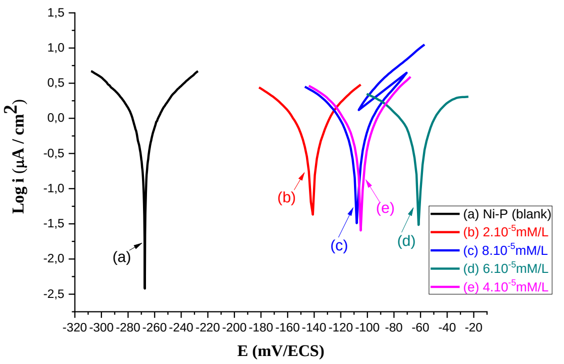

Fig. 2 – Ni-P coating polarization curves in 1 M HCl for different inhibitor concentrations

it can be concluded that the addition of the corrosion inhibitor, considerably reduces the corrosion current density until reaching the minimum value 0,0755 (μA cm^{-2}) corresponding to the shift in the corrosion potential towards positive values from – 267.4 mV/ECS without inhibitor to – 107.8 mV/ECS. This suggests that the inhibitor molecules form an adsorbed layer on the surface of the alloy, thus blocking the corrosion process. Its effect gradually increases with the addition of the inhibitor up to the concentration of 6 × 10^{-5} mM/L (minimum value).

3.3 Electrochemical Impedance Spectroscopy Measurements (EIS)

The experimental impedance results obtained for the Ni-P alloy in a 1M HCl solution without and with different concentrations of the inhibitor at room temperature are displayed in Nyquist (Fig. 3) and Bode (Fig. 4) plots.

The Nyquist diagrams obtained clearly show depressed half circles. The diameter of the capacitive loop increases with the increase in the concentration of the corrosion inhibitors, due to the progressive formation of a protective film on the surface of Ni-P alloy; this involves adsorption of inhibitors on the surface and formation of complexes [12, 13]. This corrosion process is controlled by the charge transfer phenomenon [14]. It is then followed by the material transfer phase which forms a barrier on the surface of the metal [15].

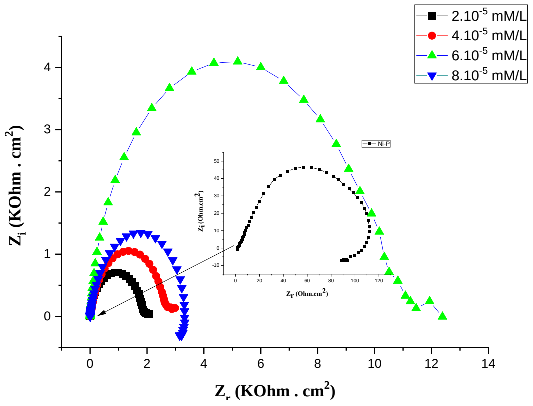

Fig. 3 – Nyquist diagrams of the Ni-P alloy in 1M HCl medium in the absence and presence of different concentrations of inhibitor

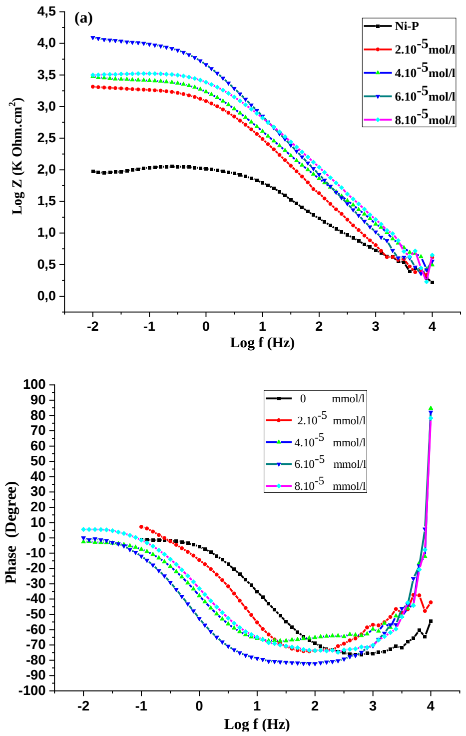

Fig. 4 – Bode plots of Ni-P coating in 1 M HCl with and without different inhibitor concentrations

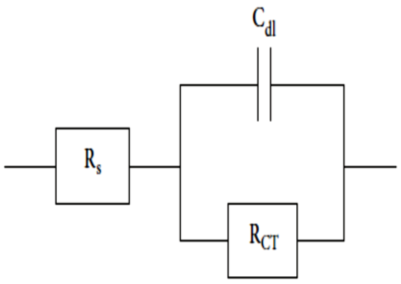

Fig. 5 – Equivalent circuit model

3.4 Adsorption Studies

In order to confirm the hypothesis on the interaction between the inhibitor and Ni-P coating surface, a mechanism of action by simple adsorption, thus blocking the active sites and consequently reducing the measured currents, the transfer resistances when recording the polarization curves and electrochemical impedances, respectively, are investigated by the adsorption isotherms.

The change in C/θ as a function of the inhibitor concentration specifies the adsorption isotherm that describes the system. The Langmuir isotherm for monolayer chemisorptions is given by the following equation (2) [16].

$$C_{\text{inb}}/\theta = 1/K_{\text{ads}} + C_{\text{inb}} \qquad (2)$$

Where K_{ads} is the equilibrium constant of the adsorption process. The plots of C_{inb}/θ. C_{inh} yield a straight line with a nearly unit slope, showing that the adsorption of inhibitors can be fitted to Langmuir adsorption as presented in Fig. 6. Adsorption equilibrium constant (K_{ads}) and free energy of adsorption ($\Delta G^{\circ}_{\text{ads}}$) were calculated using the relationships (3) and (4).

Table 3 – Ni-P coationg impedance parameters for in1M HCl solution with and without different inhibitor concentrations

	C_{inhi} (mM L^{-1}) × 10^{-5}	R_s (Ω cm^2)	R_{ct} (Ω cm^2)	C_{dl} (μF cm^2)	IE%
Blank	0	3.694	114.7	277.2	–
With Inhibitor	2	0.507	1.808 × 10^3	70.39	93.65
	4	8.259	2.685 × 10^3	59.29	95.72
	6	0.010	9.880 × 10^3	32.21	97.83
	8	3.093	3.435 × 10^3	37.05	96.66

Table 4 – Langmuir adsorption isotherm parameters for the adsorption of the inhibitor Ni-P coating surface

Method	Slope	R^2	K_{ads} (L mol^{-1}) × 10^5	K_L	ΔG_{ads} (Kj mol^{-1})
Polarization	1.02346	0.998	14.0845	0.0117	– 45.03

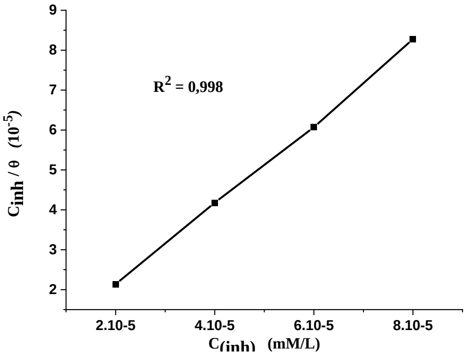

Fig. 6 – Langmuir isotherm for inhibitor adsorption on Ni-P coatings surface

$$K_{\text{ads}} = 1/C_{\text{inb}} \times [\theta/(1-\theta)] \quad (3)$$

$$\Delta G^{\circ}_{\text{ads}} = 2.3RT \log (55.5\, K_{\text{ads}}) \quad (4)$$

Where R is the universal gas constant, T is the absolute temperature, and the molar heat of adsorption of water is 55.5.

The K_{ads} and R^2 values are listed in Table 4, where R^2 is the correlation coefficient, which is the degree of fit between the experimental data and the isotherm equation.

The curve in Fig. 6 is linear with a best correlation coefficient equal to 0.998, which shows that the adsorption of the inhibitor on the alloy surface in a hydrochloric medium obeys the Langmuir adsorption isotherm.

The negative values of $\Delta G^{\circ}_{\text{ads}}$ indicate spontaneity of the adsorption process and stability of the adsorbed layer on the metal surface, are due to charge sharing or charge transfer of inhibitor molecules on the surface of the metal (chimisorption) [17, 18]. Literature reveals that if ΔG_{ads} is between 20 and 40 kJ mol^{-1}, the adsorption could proceed via (mixed adsorption) and if it is more than 40 kJ mol^{-1}, the adsorption type would be chemisorptions [19]. K_L values confined between 0 and 1 indicate that the inhibitors have achieved adsorption [20].

4. CONCLUSION

On the basis of experimental results in this work, the following conclusions can be drawn:

1) The inhibition efficiency increases with the increase of Sulfarem drug concentration.

2) Electrochemical results indicated that the Sulfarem drug is an affective green inhibitor of Ni-P alloy and the adsorption was a typical chemisorption. Moreover, the IE (%) values obtained from the polarization test reached up to 96.0 % for 6.10^{-5} mM/L, which agrees well with those gained by EIS.

3) The adsorption of Sulfarem drug on Ni- P alloy surface is found to obey the Langmuir adsorption isotherm.

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Електрохімічне та термодинамічне дослідження препарату Сольфарлем як інгібітора корозії для нікель-фосфорних покриттів

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З розвитком промисловості використовується все більше матеріалів, які часто піддаються сильній корозії. Це залежить від пари матеріал/корозійний і середовища, в якому два елементи поєднуються. Одним з найкращих методів є застосування інгібіторів для забезпечення їх захисту при контакті з агресивними середовищами, такими як соляна кислота. У даній роботі описано дослідження корозійної поведінки нікель-фосфорного покриття в кислому середовищі 1М HCl та оцінка захисної здатності органічної сполуки. Сплави Ni-P отримано методом електроосадження з розчину, що містить сульфат нікелю та гіпофосфіт натрію (NaH_2PO_2)на сталевій підкладці. Вивчено дію препарату Сольфарлем як інгібітора корозії нікель-фосфорного сплаву в 1 M HCl за допомогою спектроскопії електрохімічного імпедансу, поляризаційної кривої та методів втрати маси. Результати показують, що ефективність інгібування цього інгібітора всіма методами добре узгоджується, а оптимальна ефективність із концентрацією 6,10–5,00 моль/л становила близько 96%. Значення вільної енергії адсорбції ΔG_{ads}, розраховували для кожного покриття поверхні досліджуваної сполуки із застосуванням математичної моделі. Адсорбція молекул інгібіторів відповідає ізотермі адсорбції Ленгмюра з подальшою хімічною адсорбцією на поверхні нікель-фосфорного сплаву.

Ключові слова: Нікель-фосфорний сплав, Електрохімічні методи, Адсорбція, Препарат Сольфарлем.