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Candesartan: a pharmaceutical product as effective corrosion inhibitor for Aluminum in Acidic Solution

ABSTRACT

The corrosion performance of aluminum in 1 M hydrochloric acid solution in the existence and absence of Candesartan drug was examined utilizing, potentiodynamic polarization (PDP) and AC impedance (EIS) tests. It was found that the inhibition efficacy (%IE) of Candesartan drug was influenced by its doses and temperature and reached to 92.9% at 40x10⁻⁶ M, 30°C. The adsorption isotherm agreed with Langmuir model. From Tafel data the investigated Candesartan drug acts as mixed kind inhibitor. The influence of temperature on the rate of dissolution and the thermodynamic parameters in the existence and absence of Candesartan drug were also, studied and explained. All tests gave similar results.

Keywords: Corrosion inhibition, aluminum, HCI, Candesartan drug, adsorption, impedance, polarization.

1. INTRODUCTION

Aluminum and its alloys have been the subject of numerous studies due to their high technological value and wide range of industrial applications, especially in the aerospace and household industries. On the other hand, AI and its alloys are corrosive and reactive materials. Aluminum corrosion resistance is based on the formation of a lightweight, adherent passive oxide film in a variety of environments. This amphoteric surface film dissolves significantly when the metal is exposed to high doses of acids or bases [1]. The key technique for preventing electrochemical corrosion of Al is to effectively separate the metal from corrosive medium. Corrosion inhibitors may be used to do this. Inhibitors are one of the most well-known methods of corrosion safety. The effectiveness of organic compounds used as inhibitors is affected not only by the atmosphere in which they function, but also by the nature of the metal surface, the electrochemical potential at the interface. The inhibitors work by the adsorption on the metal surfaces. The number of adsorption active centers

in a molecule, their charge density, molecular size, adsorption mode, formation of metallic complexes, are all factors to be consider [2,3]. Corrosion inhibitors with functional groups containing heteroatoms capable of donating lone pairs of electrons have been found to be especially useful [4-10]. Compounds with π -bonds, on the other hand, have good inhibitive properties because they allow electrons to interact with the metal surface. [11-15]. Both characteristics can, of course, be found in the same molecule, such as narcotics. The conventional approach to the selection of corrosion inhibitors has gradually changed in recent years, owing to the increasing concern and attention of the world towards environmental issues and environmental safety, as well as the dangerous effects of chemical use on ecological balance [16-19]. Recently research efforts have been done on the utilize of antibacterial drugs as corrosion inhibitors for carbon steel and Al in acidic and alkaline media [20-26]. The use of many types of drugs as corrosion inhibitors of various metals was collected by Gokhan Gece [27] in a review. The inhibitive effect some antibacterial drugs, namely Ampicillin, Cloxacillin, Flucloxacillin and Amoxicillin towards the corrosion of AI was investigated [28].

Among these cephalosporins, cefadroxil, cefazolin and cefalexin are utilized as corrosion inhibitors of Fe in acidic media [29-32]. Candesartan drug, which is the subject of this study, is non-toxic, inexpensive, and environmen-

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following chemical structure:

tally friendly medications used to treat hypertension. It has reactive centers like N, O atoms with lone pairs of electrons and aromatic rings with delocalized π - electron systems, which can aid adhesion to metal surfaces. Furthermore, it

has high molecular weight and is more likely to

efficiently cover more surface area of the metal,

avoiding corrosion. The drug Candesartan has the

2-ethoxy-3-[[4-[2-(2H-tetrazol-5-yl) phenyl] phenyl] methyl] benzimidazole-4-carboxylic acid. Molecular Weight = 440.45, Chemical Formula: $C_{24}H_{20}N_6O_3$, Hemijska formula - $C_{24}H_{20}N_6O_3$

2. EXPERIMENTAL DETAILS

In this analysis, AI metal with a purity of 99.99 percent was given from the "Al Company of Egypt, Nagh Ammady." "For anodic and cathodic Tafel polarization measurements, as well as AC impedance measurements, a cylindrical rod embedded in araldite with an exposed surface area of 1.0 cm² was used. Prior to each experiment, the surface of AI specimens was mechanically polished with different grades of emery papers, degreased with the alkaline solution (15 g Na_2CO_3 + 15 g Na₃PO₄ per liter), and rinsed with distilled water. GAMRY PCl4/300 electrochemical work station. Potentiostat/Galvanostat was utilized to calculate anodic and cathodic polarization. In three compartment cells, a saturated calomel reference electrode (SCE) and a platinum foil auxiliary electrode were used. A JASCO UV-vis 530 spectrophotometer was used to take both spectral measurements. Chemicals and reagents of analytical grade were used. The drug was supplied from a variety of pharmaceutical companies. In a stock solution, 1000 ppm" [33] were made by dissolving the required weight of drug (1 g) in a small amount of one molar HCI. Using a water thermostat set to ± 0.5 °C, measurements were taken at 30- 50 °C.

3. RESULTS AND DISCUSSION

3.1. Potentiodynamic polarization (PDP)) technique

It is possible to obtain important data about the kinetics of the cathodic and anodic reactions using the PDP technology [34]. Figure 1 demonstrations typical anodic and cathodic PDP diagrams for Al in 1M HCl in attendance and absence of varying Candesartan drug doses, the following parameters were obtained: electrochemical corrosion potential

(E_{corr}), corrosion current density (I_{corr}), polarization resistance (R_p) and cathodic and anode Tafel constants (β_c , β_a), respectively. From the values in Table 1, in the presence of different doses of Candesartan drug, there is a decrease in the current density and the decrease increases with increasing dose, indicating the formation of a layer from the drug molecules on the AI surface. Moreover, the values of the cathode and anode Tafel slopes have no sharp difference in the existence of changed doses from Candesartan drug, Fig. 1. This can be explained by the fact that the dissolution procedure is caused by covering the active sites located on the surface of the Al. This also indicates that the dissolution mechanism that is not affected by the presence of Candesartan drug [33]. If the variation in corrosion potential (E_{corr}) values is more than \pm 85 mV in the presence and absence of Candesartan drug, the inhibitor molecules are named anodic or cathodic type and this did not happen because, the potential of corrosion values displacement fewer than ±85 mV [35]. The difference in E_{corr} values is small (9 mV) and this assured that the Candesartan drug works as a mixed type of inhibitor. Finally, from Table (1) we can conclude that as the dose of Candesartan drug rises, the current density decreases, the %I increases, the corrosion rate reduced, and the polarization resistance raises due to the increase thickness of the adsorbed layer, Table 1.

The following equation was used to measure the %I from the corrosion current density (I_{corr}):

$$I\% = [1 - (I_{corr, add}/I_{corr, free})] \times 100$$
 (1)

where $I_{\text{corr,free}}$ and $I_{\text{corr,add}}$ are the corrosion current in the absence and presence of Candesartan drug, correspondingly.

[Inh.], ppm	-E _{corr} , mV	I _{corr} , mA cm ⁻²	-βc, mV dec ⁻¹	βa, mV dec ⁻¹	R_p , Ω cm ²	θ	1%
Blank	782	48.96	96	57	5.017		
10	790	10.61	83	45	11.920	0.657	74.1
20	791	9.85	85	45	12.970	0.682	79.9
30	788	7.18	80	43	13.300	0.703	85.3
40	789	5.14	80	43	14.880	0.737	89.5

Table 1. PDP data of AI in 1M HCI with and without altered doses of Candesartan drug at 30°CTabela 1. PDP podaci o AI u 1M HCI sa i bez izmenjenih doza leka Candesartan na 30°C

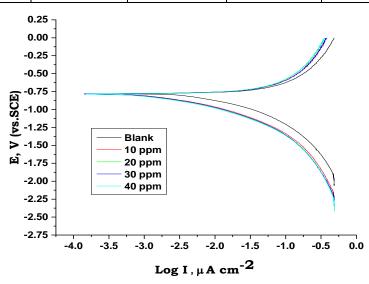


Figure 1. PDP curves for AI dissolution in 1M HCI with and without altered Candesartan drug doses at 30° C

Slika 1. PDP krive za rastvaranje AI u 1M HCl sa i bez izmenjenih doza leka Candesartan na 30°C

3.2. Adsorption isotherms

Candesartan adsorbed on the surface of the Al electrode is observed as a substitutional adsorption procedure between Candesartan in the aqueous phase (Org_{aq}) and H_2O molecules adsorbed on the Al surface [35].

$$Org_{(sol)} + x (H_2O)_{ads} \rightarrow Org_{(ads)} + x H_2O_{(sol)}$$
(2)

where x is the size ratio, that is, the amount of H_2O molecules exchanged by one organic molecule. Langmuir adsorption isotherm was by far the best match for the results. Plotting log C/ θ against log C yielded a straight line with a unit slope value, Fig. 2, suggesting that Candesartan drug adsorption on AI surface obeys Langmuir isotherm. These findings lead to the conclusion that there is no interaction among the adsorbed types.

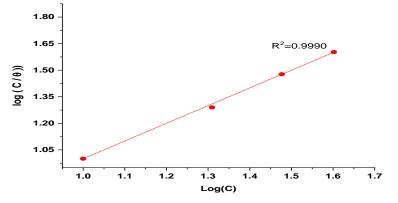


Figure 2. Langmuir adsorption isotherm achieved from PDP for AI dissolution in 1M HCI with and without altered doses of Candesartan drug at 30°C

Slika 2. Langmuirova adsorpciona izoterma postignuta iz PDP za rastvaranje AI u 1M HCl sa i bez izmenjenih doza leka Candesartan na 30°C

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3.3. Effect of temperature

PDP tests were utilized to assess the impact of temperature on the rate of dissolution of "Al in 1 M HCI containing 40 ppm of Candesartan drug over a temperature range of 30 to 50 °C. From PDP tests, the effect of improving temperature on the rate of corrosion (I_{corr}) and 1% was studied. The outcome data exposed that, on improving temperature there is a rise in the I_{corr} while 1% lowered i.e. the drug was adsorbed physically on Al surface. The activation energy (E_a) of the dissolution procedure was measured utilizing Arrhenius equation" [36]:

$$I_{\rm corr} = A \exp^{-Ea^*/RT}$$
(3)

where I_{corr} signifies the rate of dissolution, and T is kelvin temperature.

Figure 3 signifies Arrhenius drawn (log I_{corr} vs. 1/T) for unprotected and protected 1M HCl inclosing 40 ppm of the Candesartan drug. The E^*_a data can be observed from the slope of the straight lines. As in Table 2 the rise of the E^*_a in the existence of Candesartan drug is qualified to an appreciable lowered in the adsorption procedure of the Candesartan drug on the Al surface with improving the temperature and a conforming rise in the reaction rate due to the greater area of the metal that is exposed to the HCl [37].

 ΔS^* and ΔH^* for AI dissolution in 1M hydrochloric acid in the presence of 40 ppm of utilized Candesartan were achieved by applying the transition state equation:

$$log I_{corr} T = log (R/Nh + \Delta S^{*}/2.303R) + + (-\Delta H^{*}/2.303RT)$$
(4)

where "(h) is the Planck's constant and (N) gives the number of Avogadro. Fig. 4 shows straight lines resulting from a drawing log (I_{corr}/T) with 1000/T, where this figure shows the transitional state of the Candesartan drug.

The analysis of the results was obtained in Table 2, from Table 2 there is an increase in the E^*_a by improving the dose of Candesartan drug. This improve is due to the adsorption nature of Candesartan drug on the AI surface and corresponds to the physical adsorption of the molecules of Candesartan drug on AI surface. The results in Table 2 also showed that entropy values are negative in the existence of Candesartan drug". We conclude from this that the initial condition was less arranged than that found in the active molecules of Candesartan drug [38].

Table 2. Activation results for dissolution of AI in1M HCI at 40 ppm Candesartan drug

Tabela 2. Rezultati aktivacije za rastvaranje Al u 1M HCI na 40 ppm leka Candesartan

-ÄS [*] , J mol⁻K⁻¹	ÄH [*] , kJ mol⁻¹	Ea [*] , kJ mol⁻¹	Inhibitor
199.5	12.11	7.95	Free acid
194.7	25.04	17.73	drug

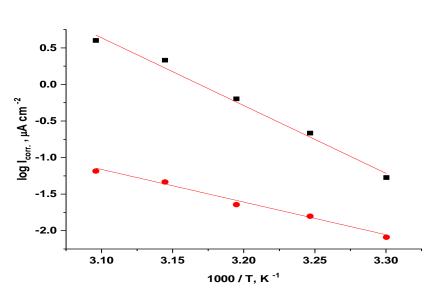


Figure 3. Log I_{corr} - 1/T plots for the dissolution of Al in 1M HCl at 40 ppm of the Candesartan drug Slika 3. Log Icorr - 1 / T dijagrami za rastvaranje Al u 1M HCl na 40 ppm leka Candesartan

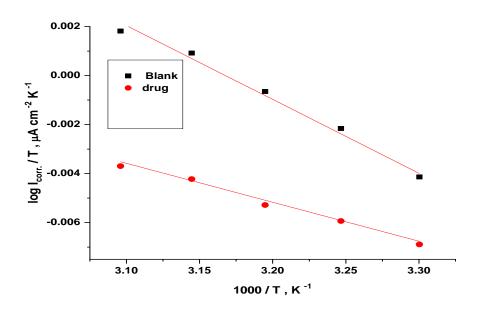


Figure 4. Log (I_{corr}/T)- (1/T) plots for the dissolution of Al in 1M HCl at 40 ppm of the Candesartan drug Slika 4. Log (Icorr / T) - (1 / T) grafikoni za rastvaranje Al u 1M HCl na 40 ppm leka Candesartan

3.4. AC Impedance test

The corrosion of AI in 1M HCI in the presence of examined Candesartan drug was investigated by EIS test at 30 °C after dipping 20 min. "Figs 5 and 6 show Nyquist and Bode curves in the presence and the absence of Candesartan drug, respectively. In both uninhibited and inhibited solutions, all Nyquist plots seem to have a single capacitive loop. The impedance data of AI dissolution in 1M HCI was examined using the fitting circuit model shown in Fig.7 [39].

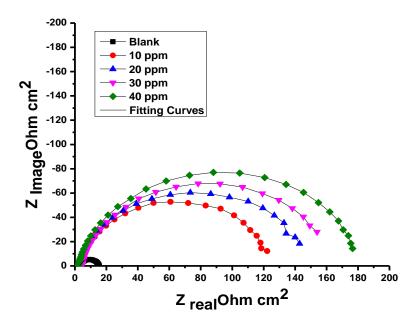


Figure 5. The Nyquist plots for AI in 1M HCl attendance and absence of altered doses of Candesartan drug at 30°C

Slika 5. Nikuist-ove krive za Al u 1M HCl u prisustvu i odsustvu izmenjenih doza leka Candesartan na 30°C

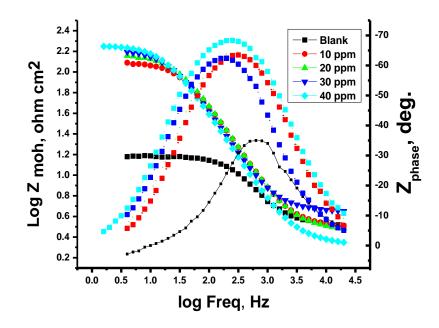


Figure 6. The Bode plots for AI in 1M HCI with and without altered doses of Candesartan drug at 30°C Slika 6. Bode krive za AI u 1M HCI sa i bez izmenjenih doza leka Candesartan na 30°C

Figure 7 shows the region of low frequency, and in the presence of "Candesartan drug, the impedance values increase compared to the absence of Candesartan drug, confirming the observed corrosion inhibition of the drug used. Candesartan drug dose also increases, the frequency range increases, and the phase angle reaches its maximum level, suggesting the successful adsorption of this drug molecules on the Al surface [40].

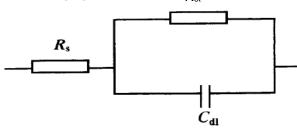


Figure 7. Equivalent circuit model used to fit impedance data

Slika 7. Ekvivalentni model kola koji se koristi za uklapanje podataka o impedansi

This may be because of the heterogeneity of the Al surface in the presence of the drug (Candesartan compound). The radius of the circle increases as the dose of the Candesartan drug increases, and thus the charge transfer resistance in corrosion reactions increases, according to the Nyquist curves. From all the above, there is high resistance established as the result of adsorption of the Candesartan molecule at the Al - solution/interface"". From the following equation the capacitance double layer (C_{dl}) was calculated:

$$C_{dl} = 1/\left(2\pi f_{max}R_{ct}\right) \tag{5}$$

Table 3 shows that as the dose of Candesartan drug increases, the values of C_{dl} decrease, "which can be explained by a decrease in the local dielectric constant and/or a rise in the thickness of the electrical double layer [41]. This is due to the adsorption of the Candesartan molecule on the Al/solution interface. Charge transfer, which is primarily responsible for Al corrosion, is depicted by impedance schemes that are almost semicircular in nature" [42,43].

Table 3. EIS parameters for dissolution of AI in 1M HCI at altered doses of Candesartan drug at 30°C

Tabela 3. EIS parametri za rastvaranje Al u 1M HCl u izmenjenim dozama leka Candesartan na 30°C

Conc., ppm	$C_{dl,} \mu F \ cm^{-2}$	$R_{t_{i}}\Omega~cm^{2}$	θ	۱%
Blank	44.57	11.36		
10	28.82	109.4	0.896	89.6
20	28.97	125.8	0.910	91.0
30	26.19	138.8	0.918	91.8
40	27.04	159.8	0.929	92.9

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3.5. Surface morphology

The 3D micrographs of AI surface acquired from AFM technique with and without drug are depicted in Fig. (8). Fig. 8 a shows AI surface after immersed in 1M HCI solution without drug, average roughness is 202.39 nm. Fig. 8b shows AI surface after submerged in 1M HCl with 40 ppm drug, average roughness is 116.45 nm. The average roughness of the drug is less than without drug suggested better adsorption of the drug on Al surface.

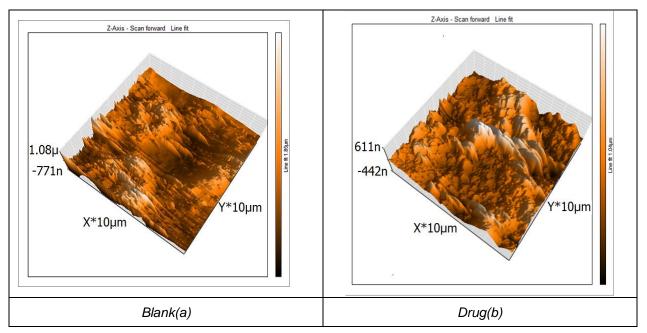


Figure 8. AFM 3D image of AI surface immersed in 1M HCI and in 40 ppm of the drug at 30°C Slika 8. AFM 3D slika površine AI uronjene u 1M HCI i u 40 ppm leka na 30°C

3.6. Corrosion inhibition mechanism

The adsorption of examined Candesartan drug at AI surface can happened among its active sites of N and O atoms in addition to a π electron interaction of the benzene nucleus with unshared p of Al atoms [44]. According to Vijh and Desai et al [45-47], the potential of zero charge (pzc) of Al in acidic media is negative and is equal to -0.4 V, so, the pure AI is negatively charged at E_{ocp} . Candesartan drug can be protonated in 1M HCI medium and become cationic molecule. So, the cationic form of the protonated Candesartan drug molecules can adsorb on the negative charge metal surface by electrostatic attraction forces forming physical adsorption. Candesartan drug is efficient inhibitor because it has 6 N and 3 O atoms active sites, 3 benzene rings, 2 hetero five-member ring and has high molecular size that covers more surface area when adsorbed on it.

4. CONCLUSIONS

The investigated Candesartan drug in 1M HCl solution can used as effective corrosion inhibitor for Al in acid medium. Candesartan drug has an inhibition capacity of 92.9 percent at 40 ppm. The

percentage of inhibition increases by raising Candesartan drug dose on the other hand, it decreases by raising the temperature. This suggested that Candesartan drug is physically adsorbed on AI surface. The adsorption of Candesartan drug on AI surface was found to obey Langmuir adsorption model. Tafel curves confirm that the drug is a mixed type of inhibitor. The results showed that this drug has proven to be an important, environmentally friendly one and lowcost inhibitor.

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IZVOD

Candesartan: farmaceutski proizvod kao efikasni inhibitor korozije za aluminijum u kiselom rastvoru

Ispitivane su performanse korozije aluminijuma u 1M rastvoru hlorovodonične kiseline u prisustvu i odsustvu leka Candesartan primenom postupaka potenciodinamičke polarizacije (PDP) i AC impedanse (EIS). Utvrđeno je da su na efikasnost inhibicije (% IE) leka Candesartan uticale njegove doze i temperatura i dostigla je 92,9% pri 40x10⁻⁶ M na 30°C. Izoterma adsorpcije se slaže sa Langmuirovim modelom. Prema podacima Tafela, ispitivani lek Candesartan deluje kao inhibitor mešovite vrste. Takođe, proučavan je i objašnjen uticaj temperature na brzinu rastvaranja i termodinamičke parametre u prisustvu i odsustvu leka Candesartan. Svi testovi su dali slične rezultate.

Ključne reči: inhibicija korozije, aluminijum, HCl, lek Candesartan, adsorpcija, impedansa, polarizacija.

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