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Diclofenac and Omeprazole Electrochemical Determination on Cobalt (III) Oxyhydroxide-Modified Electrode. A Theoretical Study

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Abstract: The theoretical description for diclofenac, and omeprazole electrochemical determination over CoO(OH)-modified electrode has been described. In a basic solution, phenolization and hydrolysis occur, and the solution is provided by cobalt (III) oxyhydroxide-based hydroxyl transfer. In the basic medium, the ionization degree of the analyte and its oxidation products is high, which enhances the probability of the oscillatory behavior. Also, the reaction mechanism may become branched, as both analytes tend to be oxidized by a hybrid mechanism, including parallel scenarios. The scenario becomes even more differentiated if the redox pair CoO(OH)/CoO₂ is used. Either way, the electrochemical determination of omeprazole and diclofenac on cobalt (III) oxyhydroxide may be efficient from the electroanalytical point of view.

Keywords: diclofenac; omeprazole; cobalt (III) oxyhydroxide; electrochemical sensors; stable steady-state.

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1. Introduction

Diclofenac (Fig. 1) is one of the most used NSAIDs [1–4]. It is frequently used as an anti-inflammatory drug and pain-killer for different pains. Its mechanism of action includes the formation of a phenolaminic derivative, which oxidizes to the correspondent quinonimine, scavenging the ROS and mitigating the inflammation process:

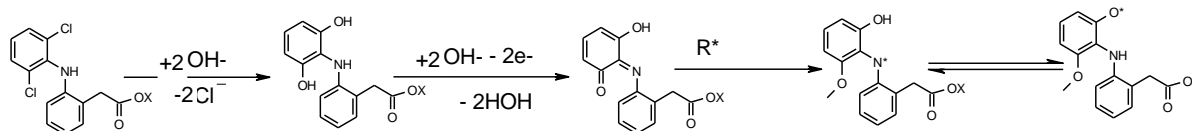


Figure 1. Diclophenac mechanism of action.

The counterion X depends on the pharmaceutical form used. The most used them are Na^+ , K^+ , and NR_4^+ , including ammonium and alkylammonium ions, being the ammonium salts usually added to creams and ointments.

On the other hand, if taken orally, diclofenac is hydrolyzed in the stomach, provoking gastritis and gastric ulcers. , due to the acidic hydrolysis, yielding a weak acid (Fig. 2).

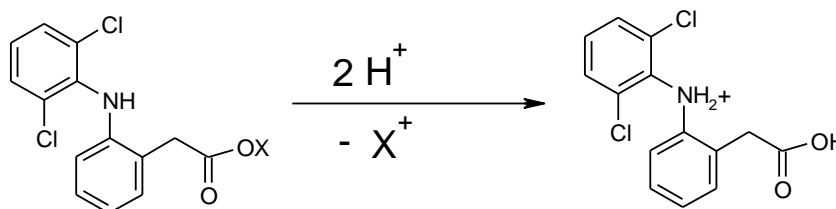


Figure 2. Acidic hydrolysis of diclophenac.

Also, its excessive concentrations, like long-time use, may lead to side effects like tiredness, slumber, nausea, and vomiting [5–8]. In this case, omeprazol (Fig. 3) [9–12] is used as a gastric proton pump to prevent and treat ulcer formation due to the presence of both pyrrolic and pyridinic nitrogen atoms. Omeprazole also provokes adverse effects like vertigo, nausea, diarrhea, and flatulence, which may cause discomfort [13–16]. Moreover, concentration control is also essential for successfully treating patients with liver and kidney insufficiency or older people. Thus, searching for a precise, exact, rapid, and sensitive method for quantifying omeprazole and diclofenac is a actual problem [17–20].

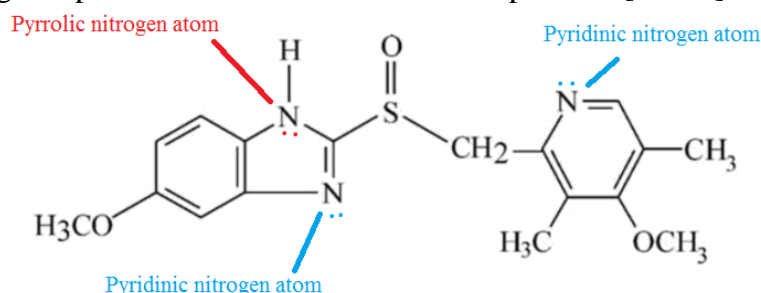


Figure 3. Omeprazole.

In our previous works, we investigated the electrochemical determination of diclofenac, and omeprazole separately [21–24]. Both cathodic and anodic processes may be compatible with those analytes, and in the case of the anodic oxidation, cobalt (III) oxyhydroxide, yet used for similar analytes [25–32], maybe a suitable anode modifier. Nevertheless, there are some objections concerning the anodic oxidation mechanisms and critical phenomena accompanying the electroanalytical process [33–35].

For this reason, this work aims to investigate *a priori* the possibility for omeprazole and diclofenac electrochemical determination on cobalt (III) oxyhydroxide-modified electrode, which includes the mechanism suggestion and the correspondent mathematical model investigation with the comparison of the behavior of this system with that of the similar ones [21–24, 33–35].

2. Materials and Methods

In a lightly alkaline medium, omeprazole and diclophenac diclofenac become hydrolized, hydrolyzed, and phenolized, with phenolization considered the main scenario. As for the carboxyl, it will be present in the salt form. Also, some phenolic hydroxyls may be electrooxidized to the corresponding quinones.

For this reason and considering that cobalt (III) oxyhydroxide may act as the hydroxyl donor, the mechanism for the electrooxidation of omeprazole and diclofenac will be described in Fig. 4:

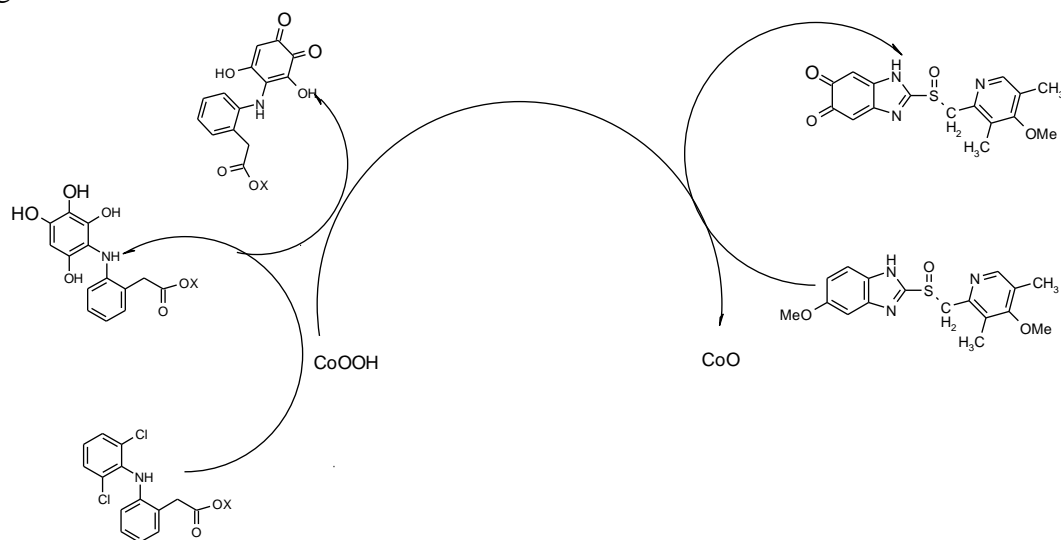


Figure 4. The schematic presentation of the electroanalytical process.

More oxidation scenarios, including the N-oxidation and S-oxidation, are included if the $\text{CoO}(\text{OH})/\text{CoO}_2$ redox pair is used. By this, tetravalent cobalt will thereby act as a strong oxidant.

Taking this into account and accepting some assumptions [21–24], we describe the system's behavior by the trivariant equation-set as:

$$\begin{cases} \frac{da}{dt} = \frac{2}{\delta} \left(\frac{A}{\delta} (a_0 - a) - r_{a1} - r_{a2} \right) \\ \frac{do}{dt} = \frac{2}{\delta} \left(\frac{O}{\delta} (o_0 - o) - r_o \right) \\ \frac{dc}{dt} = \frac{1}{c} (r_{a1} + r_{a2} + r_o - r_r) \end{cases} \quad (1)$$

Herein A and O are the diclofenac and omeprazole diffusion coefficients, a_0 and o_0 are their bulk concentrations, δ is the diffusion layer thickness, C is the maximal cobalt (II) oxide surface coverage degree, and the parameters r are the correspondent reaction rates, calculated as:

$$r_{a1} = k_{a1} a (1 - c)^6 \exp(-la) \quad (2)$$

$$r_{a2} = k_{a2} a (1 - c)^4 \exp(-la) \quad (3)$$

$$r_o = k_o o (1 - c)^3 \exp(-lo) \quad (4)$$

$$r_r = k_r c \exp\left(\frac{F\phi_0}{RT}\right) \tag{5}$$

Herein, the parameters k are the correspondent reaction rate constants, l is the double electric layer (DEL) ionic force-related parameter, describing the impact of the ionic forms transformations, F is the Faraday number, ϕ_0 is the potential slope related to the zero-charge potential, R is the universal gas constant, and T is the absolute temperature.

In a basic medium, ionization and phenolization impact the DEL ionic force and related parameters (ion activity, pre-surface layer conductivity), which causes electrochemical instabilities in the system. Nonetheless, the electrochemical determination of omeprazole and diclofenac in a lightly alkaline medium, assisted by cobalt (III) oxyhydroxide, is an efficient process, as shown below.

3. Results and Discussion

To describe the system with the electroanalytical determination of diclofenac, and omeprazole in basic media over CoO(OH)-modified electrode, we analyze the equation-set (1) using linear stability theory. The steady-state Jacobian matrix members will be described as (6):

$$\begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix} \tag{7}$$

In which:

$$a_{11} = \frac{2}{\delta} \left(-\frac{A}{\delta} - k_{a1}(1-c)^6 \exp(-la) + lk_{a1}a(1-c)^6 \exp(-la) - k_{a2}(1-c)^4 \exp(-la) + lk_{a2}a(1-c)^4 \exp(-la) \right) \tag{7}$$

$$a_{12} = 0 \tag{8}$$

$$a_{13} = \frac{2}{\delta} (6k_{a1}a(1-c)^5 \exp(-la) + 4k_{a2}a(1-c)^3 \exp(-la)) \tag{9}$$

$$a_{21} = 0 \tag{10}$$

$$a_{22} = \frac{2}{\delta} \left(-\frac{o}{\delta} - k_o(1-c)^3 \exp(-lo) + lk_o o(1-c)^3 \exp(-lo) \right) \tag{11}$$

$$a_{23} = \frac{2}{\delta} (3k_o o(1-c)^2 \exp(-lo)) \tag{12}$$

$$a_{31} = \frac{1}{c} (k_{a1}(1-c)^6 \exp(-la) - lk_{a1}a(1-c)^6 \exp(-la) + k_{a2}(1-c)^4 \exp(-la) - lk_{a2}a(1-c)^4 \exp(-la)) \tag{13}$$

$$a_{32} = \frac{1}{c} (k_o(1-c)^3 \exp(-lo) - lk_o o(1-c)^3 \exp(-lo)) \tag{14}$$

$$a_{33} = \frac{1}{c} \left(-6k_{a1}a(1-c)^5 \exp(-la) - 4k_{a2}a(1-c)^3 \exp(-la) - 3k_o o(1-c)^2 \exp(-lo) - k_r \exp\left(\frac{F\phi_0}{RT}\right) + jk_r c \exp\left(\frac{F\phi_0}{RT}\right) \right) \tag{15}$$

Observing the main diagonal elements (7), (11), and (15), we can conclude that, as they contain positive addendums, the positive callback they describe is possible. This callback is necessary for the Hopf bifurcation, which corresponds to the oscillatory behavior. The

oscillatory pattern will be similar to that described in [21–24, 33–35] and dependent on background electrolyte composition, including the pH.

As in [21–24, 33–35], the factors causing the oscillatory behavior are DEL-related and manifest the cyclic influences of both chemical and electrochemical stages on DEL ionic force. They are described by the positivity of the elements $lk_{a1}a(1 - c)^6 \exp(-la) > 0$, $lk_{a2}a(1 - c)^4 \exp(-la) > 0$, $k_o o(1 - c)^3 \exp(-lo) > 0$, if $l > 0$ and $jk_r c \exp\left(\frac{F\phi_0}{RT}\right) > 0$, if $j > 0$, and are generally frequent and of small amplitude.

As for the *steady-state stability*, its condition includes the nullity or negativity of the addendums mentioned above. Simplifying the analysis of the determinant, to facilitate the Routh-Hurwitz criterion application, we introduce the new variables and rewrite it as (16):

$$\frac{4}{\delta^2 c} \begin{vmatrix} -\kappa - \Lambda & 0 & T \\ 0 & -\omega - \Sigma & P \\ \Lambda & \Sigma & -T - P - \Omega \end{vmatrix} \quad (16)$$

Opening the brackets and applying the Det $J < 0$ requirement, salient from the criterion, we expose the stability requirement as (17):

$$\kappa(\omega T + \omega P + \omega \Omega + \Sigma T + \Sigma \Omega) + \Lambda(\omega P + \omega \Omega + \Sigma \Omega) > 0 \quad (17)$$

This will correspond to both diffusion and kinetically controlled electroanalytical systems [21–24, 33–35], in which, as no side reactions are possible for either analyte or electrode modifier, the steady-state stability will be correspondent to the linear dependence between the electrochemical parameter and concentrations of both of the drugs.

The requirement (17) will be realized if the kinetic parameters Ω , T , and P are positive (which is warranted if the addendums mentioned above are negative) and, despite corresponding to a bit narrower topological region, will define the efficient analytical signal interpretation.

As for the *detection limit*, it will correspond to the monotonic instability, corresponding to static or saddle-node bifurcation. Its main requisite is Det $J = 0$, or (18):

$$\kappa(\omega T + \omega P + \omega \Omega + \Sigma T + \Sigma \Omega) + \Lambda(\omega P + \omega \Omega + \Sigma \Omega) = 0 \quad (18)$$

With the higher alkaline pH, diclofenac will be detected indirectly as its hydrolysis product. Moreover, it will be even more ionized, which makes the sensor less efficient in those conditions.

The same model will apply to the simultaneous determination of the pair of analogous drugs (e.g., aceclofenac and lansoprazole). Also, if the CoO(OH)/CoO₂ redox pair is used, the oxidation mechanism becomes more hybridic due to a stronger oxidant as an active form. This will make the system's behavior similar to that of the Ag₂O₂ [24].

4. Conclusions

From the system with the electrochemical determination of omeprazole and diclofenac on a CoO(OH)-modified electrode, it was possible to conclude that this electroanalytical system will be both diffusion and kinetically controlled. The oscillatory behavior in this case will be more probable than in the most straightforward situation due to the transformation of the ionic forms in DEL. Also, although the linear dependence between the electrochemical parameter and concentration of both drugs is realized in a more narrow parameter region, CoO(OH) is an efficient electrode modifier for omeprazole and diclofenac simultaneous electrochemical determination. The same modifier may also be applied to analogous drugs.

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Conflicts of Interest

The authors declare no conflict of interest.

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