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The Theoretical Description for Paracetamol and Naproxen Electrochemical Determination, Assisted by Conducting Polymer Composite with Cobalt (III) Oxyhydroxide

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Abstract: In this work, the possibility for the simultaneous electrochemical determination of naproxen and paracetamol over the electrode, modified by conducting polymer composite with cobalt (III) oxyhydroxide, is analyzed for the first time. The correspondent mathematical model has been developed and analyzed by means of linear stability theory and bifurcation analysis. It has been shown that the electrochemical analysis of both substances may be easily conducted and interpreted at moderate electrode potentials. As for the oscillatory behavior, it is more probable than in the simplest cases due to the ionic forms' transformations during the naproxen determination and their impact on DEL capacitance.

Keywords: naproxen; paracetamol; cobalt (III) oxyhydroxide; conducting polymers; electrochemical sensors; electrochemical oscillations; stable steady-state

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1. Introduction

Paracetamol or acetaminophen (Figure 1 to the left) [1-4] is an acylated phenolamine, introduced to the pharmaceutical practice in 1893 by Von Mering as an analgesic drug for

adults and children. Its action is based on prostaglandin synthesis inhibition in the central nervous system. Nevertheless, its side effects include liver intoxication leading even to death [5]. Therefore, developing new methods for its determination is really actual [6 - 8].

On the other hand, naproxen [9-12] is a non-steroid analgesic drug based on 2- β -naphthyl propionic acid. Its action is based on cyclo oxidase inhibition. It is used to treat muscular pains, convulsions, and edemas. Nevertheless, it is contraindicated to babies and children till 2 years old, like people with asthma, gastrointestinal diseases, and gastric ulcers. The side effects include sleepiness, fatigue, and depression [13-15]. So, the development of precise and exact methods for its determination in different conditions is actual [16-20].

Figure 1. Paracetamol (left side) and naproxen (right side).

Taking into account the structures of paracetamol and naproxen, it is possible to conclude that both of them are electrochemically active, so the electroanalytical processes are applicable to them [21-35]. Moreover, in certain conditions, their electropolymerization, yielding a conducting polymer, is also possible due to the aromatic nature of both of the analytes.

One of the electrode modifiers suitable for their electrochemical determination could be cobalt(III) oxyhydroxide. It is a p-type semiconductor, suggested as an alternative to titanium dioxide, but with more flexible electrochemical behavior [36-42].

Nevertheless, the use of novel electrode modifiers with novel analytes may be impeded by: the indecision concerning the exact mechanism of electrochemical reaction; the necessity of determination of the parameter region, correspondent to the most efficient active substance and mediating action; the presence of electrochemical instabilities, yet described for the CoO(OH) synthesis [43 - 45].

The mentioned problems may only be solved by analyzing a mathematical model capable of adequately describing ephedrine electrochemical determination. Moreover, it is also capable of comparing this system's behavior with that of similar ones without any experimental essay.

So, the goal of this work is the mechanistic theoretic analysis of the naproxen and paracetamol electrochemical determination, assisted by CoO(OH)-modified conducting polymer electrode. In order to achieve it, we realize the specific goals:

- the suggestion of the mechanism of the reaction consequence, leading to the appearance of an analytical signal;
- development of the balance equation mathematical model, correspondent to the electroanalytical system;
- analysis and interpretation of the model in terms of the electroanalytical use of the system;

- the seek for the possibility of electrochemical instabilities and for the factor causing them:
- the comparison of the mentioned system's behavior with similar ones [46-49].

2. Materials and Methods

In the electrochemical sensors, based on hybrid organic-inorganic materials, in which the organic or carbon matrix hosts the inorganic nanoparticles, the inorganic material enters in direct contact with the analyte, being an active substance. The conjugated conducting polymer, for its turn, acts as an electron transfer mediator.

In the case of CoO(OH)-assisted paracetamol and naproxen electrochemical determination, paracetamol is oxidized by the quinone-hydroquinonic mechanism, ceding two protons and two electrons. Yet, naproxen electrooxidation may be realized in two manners:

by tertiary carbon hydroxylation;

by ortho-position carbon phenolization.

The macromolecular oxidation of both of the compounds is possible at higher potentials. Schematically, the mechanism for the oxidation of both compounds may be represented in Figure 2.

$$\begin{array}{c} O\\ O\\ H\\ N\\ O\\ CoO(OH) \end{array}$$

Figure 2. The schematical representation of the electroanalytical process.

In the simplest scenario, in which the electro(co)polymerization of the analytes does not occur, they are incapable of interacting with each other.

Taking into account the mentioned above, in order to describe the electrochemical process with the naproxen and paracetamol electrochemical determination, we introduce three variables:

- p paracetamol concentration in the pre-surface layer;
- n naproxen concentration in the pre-surface layer;
- c cobalt (II) oxide polymer matrix coverage degree.

To simplify the modeling, we suppose the reactor is intensively stirred, so we can neglect the convection flow. Also, we assume that the background electrolyte is in excess, so

we can neglect the migration flow. Finally, the diffusion layer is supposed to be of a constant thickness equal to δ , and its concentration profile is supposed to be linear.

It is possible to show that the behavior of this system will be described by a trivariate equation set, expressed as:

$$\begin{cases} \frac{dp}{dt} = \frac{2}{\delta} \left(\frac{P}{\delta} (p_0 - p) - r_p \right) \\ \frac{dn}{dt} = \frac{2}{\delta} \left(\frac{N}{\delta} (n_0 - n) - r_{n1} - r_{n2} \right) \\ \frac{dc}{dt} = \frac{1}{c} \left(r_p + r_{n1} + r_{n2} - r_1 \right) \end{cases}$$
(1)

Herein, P and N are paracetamol and naproxen diffusion coefficients, p0 and n0 are their bulk concentrations, C is the maximal CoO matrix concentration, and the parameters r are the correspondent reaction rates, calculated as:

$$r_p = k_p p (1 - c)^2 (2)$$

$$r_{n1} = k_{n1}p(1-c)^2 \exp(-ap) \tag{3}$$

$$r_{n2} = k_{n2}p(1-c)^2 \exp(-ap)$$
 (4)

$$r_1 = k_1 c \exp\left(\frac{F\varphi_0}{RT}\right) \tag{5}$$

In which the parameter k stand for the correspondent reaction rate constants, the parameter a relates the ionic forms transformation with the DEL electrophysical and electrochemical properties, F is the Faraday number, φ_0 is the potential slope, corresponding to the zero-charge potential, R is the universal gas constant, and T is the absolute temperature.

In this model, considering that the ionic compound's ionization is less intense than the carboxylic compound, its DEL effect is neglected. This factor will also be included in the case of more acidic phenolic compounds (e.g., salicylic acid), polyphenolic compounds, and(or) more basic solutions. Either way, the naproxen ionic form transformation impacts the DEL ionic force and capacitance, increasing the possibility of oscillatory behavior. Nevertheless, the stable steady-state is easily obtained and maintained, providing the efficient peak separation of naproxen and paracetamol, alongside the successful analytical signal interpretation, as shown below.

3. Results and Discussion

To investigate the system's behavior with naproxen electrochemical determination, we analyze equation-set (1) using linear stability theory. The steady-state Jacobian matrix elements may be described as:

$$\begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix} \tag{6}$$

In which

$$a_{11} = \frac{2}{\delta} \left(-\frac{P}{\delta} - k_p (1 - c)^2 \right) \tag{7}$$

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

$$a_{13} = \frac{2}{\delta} \left(2k_p (1 - c) \right) \tag{9}$$

$$a_{21} = 0 (10)$$

$$a_{22} = \frac{2}{\delta} \left(-\frac{N}{\delta} - k_{n1} (1 - c)^2 \exp(-ap) + ak_{n1} p(1 - c)^2 \exp(-ap) - k_{n2} (1 - c)^2 \exp(-ap) + ak_{n2} p(1 - c)^2 \exp(-ap) \right)$$
(11)
$$a_{23} = \frac{2}{\delta} (2k_{n1} p(1 - c) \exp(-ap) + 2k_{n2} p(1 - c) \exp(-ap))$$
(12)
$$a_{31} = \frac{1}{c} (k_p (1 - c)^2)$$
(13)
$$a_{32} = \frac{1}{c} (k_{n1} (1 - c)^2 \exp(-ap) - ak_{n1} p(1 - c)^2 \exp(-ap) + k_{n2} (1 - c)^2 \exp(-ap) - ak_{n2} p(1 - c)^2 \exp(-ap))$$
(14)
$$a_{33} = \frac{1}{c} \left(-2k_p (1 - c) - 2k_{n1} p(1 - c) \exp(-ap) - 2k_{n2} p(1 - c) \exp(-ap) - k_1 \exp\left(\frac{F\varphi_0}{PT}\right) + jk_1 c \exp\left(\frac{F\varphi_0}{PT}\right) \right)$$
(15)

For the oscillatory behavior to be realized, the presence of the positive addendums, correspondent to the positive callback, in the main diagonal is necessary.

Taking into account the Jacobian main-diagonal elements (7), (11), and (15), it is possible to conclude that the oscillatory behavior in this system is possible. Moreover, it is more probable than in the simplest systems [47 – 49], in which the unique factor causing the oscillatory is the DEL impact on the electrochemical stage, described by the positivity of the addendum $jk_1c\exp\left(\frac{F\varphi_0}{RT}\right) > 0$ if j>0.

The additional factor causing the oscillatory behavior is the DEL ionic force and conductivity cyclic changes caused by the ionic form transformations during the naproxen determination. They are described by the positivity of the elements $ak_{n1}p(1-c)^2 \exp(-ap)$ and $ak_{n2}p(1-c)^2 \exp(-ap)$ if a>0. All of the mentioned elements are dependent on background electrolyte composition, and so are the amplitude and frequency of the oscillations.

Moreover, the parameter a tends to be nil if the working solution pH decreases. Therefore, in the less basic media, close to neutral, the oscillatory behavior will be less probable than in more basic.

As for the steady-state stability, its condition results from the Routh-Hurwitz criterion applied to the equation set (1). Avoiding the cumbersome expressions, we introduce new variables, rewriting the Jacobian determinant as (16):

$$\frac{4}{\delta^2 c} \begin{vmatrix}
-\kappa - \Xi & 0 & T \\
0 & -\xi - \Sigma & P \\
\Xi & \Sigma & -T - P - O
\end{vmatrix}$$
(16)

Opening the brackets and applying the Det J<0, salient from the criterion, we obtain the steady-state stability requisite, expressed after changing the signs as (17):

$$\kappa(\xi T + \xi P + \xi \Omega + \Sigma T + \Sigma \Omega) + \Xi(\xi P + \xi \Omega + \Sigma \Omega) > 0 \tag{17},$$

defining an efficient electroanalytical process controlled either by diffusion or reaction kinetics. The transition to pure diffusion or purely kinetic-controlled mode will depend on electrode shape and analyte concentration.

The requisite (17) is warranted to be satisfied in the case of the positivity of the parameters Ω and Σ (the rest of the variables are always positive), defining the naproxen ionic form transformations and CoO electrooxidation influences on DEL. The requisite (17) is satisfied in a vast parameter topological region.

As no reactions capable of compromising the analyte or modifier stability are characteristic for this case, the steady-state stability will correspond to the linear dependence between the current and concentration. Also, as condition (17) is readily satisfied for a vast parameter region, the electrochemical process will be the efficient analytical point of view.

As for the detection limit, it is defined by the monotonic instability, conditioned by the requisite Det J=0, or (18):

$$\kappa(\xi T + \xi P + \xi \Omega + \Sigma T + \Sigma \Omega) + \Xi(\xi P + \xi \Omega + \Sigma \Omega) = 0$$
(18)

In this system, different unstable states coexist. The system chooses one of them. If the conditions are altered, it is thereby destroyed and may not be regenerated if the condition (18) is restored. This is defined by the saddle-node or static bifurcation.

The peak separation by CoO(OH)/CP composite is achieved by the difference in rate constants for each of the possible interactions between the analytes and the electrode modifier, like also by the presence of different donating and accepting groups in the conjugated system of each of the analytes. This factor becomes important also in their electro(co)polymerization case, evaluated in one of our next works.

4. Conclusions

From the theoretical description for naproxen and paracetamol determination over cobalt (III) oxyhydroxide – conducting polymer composite, it has been possible to conclude that this is an efficient electroanalytical process controlled by both diffusion and kinetics. The peak separation is easy to achieve, and the analytical signal interpretation is easy to obtain and maintain. As for the oscillatory behavior, it is caused by DEL ionic force effects of the electrochemical stage and one of the chemical stages.

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Conflicts of Interest

The authors declare no conflict of interest.

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