The Theoretical Description for the Electrochemical Determination of Anti-COVID-19 Drug Umifenovir, Assisted by a Poly(squaraine dye-co-naphthoquinones) Composite with CoO(OH), Paired with CoO2

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Abstract: For the first time, the system with anti-COVID19 drug umivenofir electrochemical determination, assisted by a Squaraine Dye – CoO(OH)/CoO2 composite, has been described theoretically. The mechanism contains hydrolysis, chemical, and electrochemical oxidation stages. The correspondent mathematical model has been developed and analyzed using linear stability theory and bifurcation analysis. Despite the branched character of the mechanism, the composite of Squaraine Dye with CoO(OH)/CoO2 may be an efficient electrode modifier for the umivenofir determination. However, the oscillatory behavior may be possible, but only because of double electric layer influences on the electrochemical stage.

Keywords: COVID-19; umifenovir; electrochemical sensors; squaraine dye; cobalt(III)oxyhydroxide; stable steady-state.

1. Introduction

Umifenovir [1 – 4] (Fig. 1), sold under the brand name Arbidol is an antiviral drug for treating different types of influenza. It is used mostly in Russia and China. Recently, its efficacy against COVID-19 (sole and alongside darunavir) has been claimed by the study conducted in the mentioned countries [5–11]. Its mechanism of action consists of preventing the contact between the virus and host cells, inhibiting the viral envelope from contacting the cell membrane of the target cell. Therefore, the infection of the target cell is impeded. Besides influenza, it has already been used to treat Zaire ebolavirus [12], Kaposi´s sarcoma-associated herpesvirus [13].
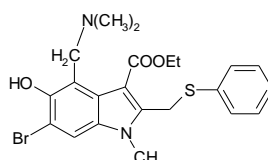
Figure 1. Umifenovir.

Nevertheless, the drug is not widely used in Western democratic countries. Most of the articles describing its investigation were published in Russian, making it difficult to analyze their quality by the WHO. US, the Food and Drug Administration. Moreover, even within Russia, its efficacy of Umifenovir has been questioned [14 – 15]. Also, the side effects are not known well. Therefore, the development of a method capable of detecting rapidly and efficiently the concentration of Umifenovir is an actual task [16 – 18].

Considering the composition of Umifenovir (as an indolic derivative, substituted by electroactive groups), it is possible to conclude that it has to be electrochemically active. Similar compounds [19-28] (both drugs and pesticides) have already been used as analytes in electrochemical sensors, involving the chemically modified electrodes with hybrid materials. In these materials, the redox-active metal oxides are stabilized by conjugated oligomers (squaraine dyes) and polymers, acting as stabilizers and mediators [29-35].

Nevertheless, the organic electrooxidation processes (including the electropolymerization) tend to be accompanied by electrochemical instabilities, similar to those observed in BZ and BR reactions. These instabilities include the oscillatory changes in electrochemical potential and monotonic instabilities [36-38] and influence the analytical signal interpretation and reproducibility.

Therefore, in this system, we investigate the system with the electrochemical determination of Umifenovir, assisted by the composite of poly (squaraine dye-co-naphthoquinones) composite with cobalt (III) oxyhydroxide, paired with the cobalt dioxide. In order to realize this task, we realize the specific goals like the suggestion of the most probable mechanism for the reaction sequence, leading to the electroanalytical signal realization, development and analysis of the mathematical model, correspondent to the mechanism, derivation of the steady-state stability conditions and oscillatory and monotonic instability requirements and compare the behavior of this system with that of the similar ones [39–42].

2. Materials and Methods

2.1. System and its modeling.

The process of the electrochemical determination of the Umifenovir, assisted by the poly(squaraine dye-co-naphthoquinones)-$CoO(OH)/CoO_2$ composite is similar to that

observed for the pyriproxyfen pesticide [39], but somehow more branched. It is represented in Fig. 2:
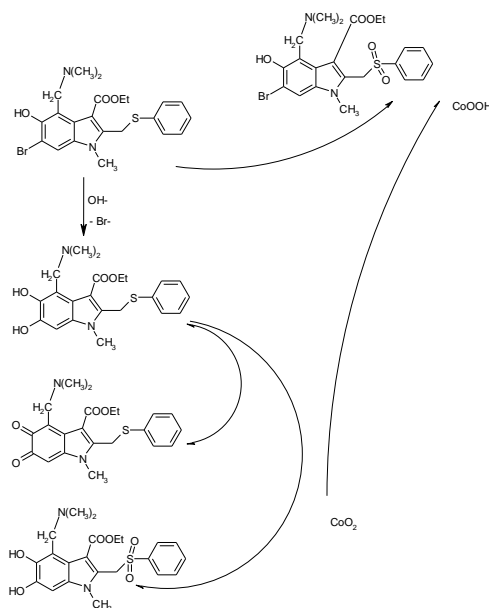
Figure 2. Umifenovir electrochemical determination scheme.

The Cobalt dioxide is regenerated on the electrochemical stage:

$$CoO(OH) + OH^- - e^- \rightarrow CoO_2 + H_2O \qquad (1)$$

As Umifenovir is a ternary amine, it has its proper basicity, promoting hydrolysis or CoO_2 regeneration reactions.

So, in order to describe the behavior of the electroanalytical system with Umifenovir electrochemical determination, we introduce three variables:

u – Umifenovir concentration in the pre-surface layer;

u* - Umifenovir hydrolysis product concentration in the pre-surface layer;

c – cobalt (III) oxyhydroxide matrix coverage degree.

To simplify the modeling, we suppose that the reactor is intensively stirred to neglect the convection flow. Also, we assume that the background electrolyte is in excess of neglecting the migration flow. The diffusion layer is supposed to be of a constant thickness, equal to δ. The concentration profile in it is supposed to be linear. Also, we assume that the oxidation products do not interfere in the reaction.

Therefore, the system´s behavior will be described by balance equation-set (2):

$$\begin{cases} \frac{du}{dt} = \frac{2}{\delta}\left(\frac{\Delta}{\delta}(u_0 - u) - r_h - r_{21}\right) \\ \quad \frac{du*}{dt} = \frac{2}{\delta}(r_h - r_{22} - r_{23}) \\ \frac{dc}{dt} = \frac{1}{C}(r_{21} + r_{22} + r_{23} - r_1) \end{cases} \qquad (2)$$

Herein, Δ is the diffusion coefficient, u_0 is the bulk drug concentration, C is the cobalt oxyhydroxide maximal surface concentration, and the parameters r stand for the correspondent reaction rates, calculated as:

$$r_{21} = k_{21}u\,(1 - c)^4 \qquad (3)$$
$$r_h = k_h u \qquad (4)$$
$$r_{22} = k_{22}u*(1 - c)^2 \qquad (5)$$
$$r_{23} = k_{23}u*(1 - c)^4 \qquad (6)$$

$$r_1 = k_1 c \exp\left(\frac{F\varphi_0}{RT}\right) \tag{7}$$

In which k are respective reaction rate constants, F is the Faraday number, R is the universal gas constant, and T is the absolute temperature.

Contrarily to the system with pyriproxyfen electrochemical determination [39], the system will be somehow more dynamic. Nevertheless, the model's analysis shows that the electroanalytical system has to be efficient, as shown below.

3. Results and Discussion

In order to describe the behavior of the system with Umifenovir electrochemical determination, assisted by Poly(Squaraine Dye-co-Naphthoquinone)–CoO(OH) composite, we analyze the equation-set (2) using linear stability theory. The steady-state Jacobian matrix members may be described as:

$$\begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix} \tag{8}$$

where:

$$a_{11} = \frac{2}{\delta}\left(-\frac{\Delta}{\delta} - k_h - k_{21}(1-c)^4\right) \tag{9}$$

$$a_{12} = 0 \tag{10}$$

$$a_{13} = \frac{2}{\delta}(4k_{21}(1-c)^3) \tag{11}$$

$$a_{21} = 0 \tag{12}$$

$$a_{22} = \frac{2}{\delta}(-k_{22}(1-c)^2 - k_{23}(1-c)^4) \tag{13}$$

$$a_{23} = \frac{2}{\delta}(2k_{22}u*(1-c) + 4k_{23}u*(1-c)^3) \tag{14}$$

$$a_{31} = \frac{1}{C}(4k_{21}(1-c)^3) \tag{15}$$

$$a_{32} = \frac{1}{C}(k_{22}(1-c)^2 + k_{23}(1-c)^4) \tag{16}$$

$$a_{33} = \frac{1}{C}\left(-4k_{21}(1-c)^3 - 2k_{22}u*(1-c) - 4k_{23}u*(1-c)^3 - k_1\exp\left(\frac{F\varphi_0}{RT}\right) + jk_1c\exp\left(\frac{F\varphi_0}{RT}\right)\right) \tag{17}$$

As in similar systems [39-42], the oscillatory behavior is possible in this system. Nevertheless, as in a bit more simple system with pyriproxyfen electrochemical determination [39], the unique factor responsible for oscillatory behavior is the electrochemical stage's influence on a double electric layer capacitance and conductivity. It is described by the positivity of the element $jk_2 s\exp\left(\frac{F\varphi_0}{RT}\right) > 0$ if j>0. The oscillations are expected to be frequent and of small amplitude and are expected to be observed beyond the detection limit.

In order to investigate the steady-state stability, we apply the Routh-Hurwitz stability criterion to the equation-set (2). Avoiding the cumbersome expressions, we introduce new variables, rewriting the determinant as:

$$\frac{4}{\delta^2 C}\begin{vmatrix} -\kappa - \Xi & 0 & P \\ 0 & -\Omega - \Sigma & T + \Phi \\ \Xi & \Omega + \Sigma & -P - T - \Phi - \Lambda \end{vmatrix} \tag{18}$$

Opening the brackets and applying the condition of Det J<0, salient from the criterion, we obtain the steady-state stability requisite, exposed as:

$$-\kappa(\Omega P + \Omega\Lambda + \Sigma P + \Sigma\Lambda) - \Xi\Lambda(\Omega + \Sigma) < 0 \tag{19}$$

Describing an efficient, although a bit more dynamic, diffusion-controlled electroanalytical system, analogous to [39]. The requisite (19) is warranted to be satisfied within the unique condition of the positivity of the electrochemical parameter Λ (as the rest of the variables are always positive).

Considering that no side reaction, capable of compromising the analyte and(or) modifier stability is present in this case, it is possible to conclude that the steady-state stability will be electroanalytical efficient. It corresponds to the linear dependence between the current and Umifenovir concentrations, and it is satisfied for the relatively vast parameter topological region. Therefore, the composite of the copolymer of squaraine dye with naphthoquinones with CoO(OH) may be an efficient electrode modifier for the electrochemical determination of Umivenofir.

The detection limit is correspondent to the margin between the stable steady-states and unstable states. Its condition is correspondent to the nullity of the determinant and may be exposed as:

$$-\kappa(\Omega P + \Omega\Lambda + \Sigma P + \Sigma\Lambda) - \Xi\Lambda(\Omega + \Sigma) = 0 \qquad (20)$$

The model described here is valid for neutral or mildly basic solutions. The ester group also tends to hydrolyze in more fundamental solutions, yielding a more ionic compound, the salt. This will affect the DEL ionic force and may cause the oscillatory behavior, similar to those observed in [36] and foreseen in [40 – 42].

4. Conclusions

From the analysis of the system with the Umivenofir electrochemical determination as an anodic process, assisted by CoO(OH)–Poly (Squaraine Dye- Co-Naphthoquinone) composite, it is possible to conclude that; The system's behavior is more dynamic, while compared to the simplest analogous system, due to the direct analyte interaction with the modifier, parallel to the indirect; The electroanalytical process tends to be diffusion controlled with the easy realization of linear dependence between the concentration and the current; The oscillatory behavior tends to have less probability of being realized than similar systems, due to the impossibility of DEL influence of the electrochemical stage.

Conflicts of Interest

The authors declare no conflict of interest.

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