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# The Theoretical Evaluation of Clioquinol Electrochemical Determination over Squaraine Dye/CuS Composite

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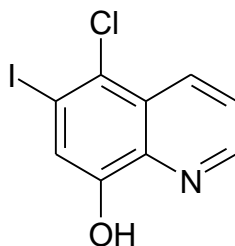
**Abstract:** In this work, the electrochemical determination of clioquinol on the electrode, modified by the squaraine dye complex with copper sulfide nanoparticles, is described. The electroanalytical process begins with the analyte hydrolysis, followed by its hybrid oxidation by the pyridinic nitrogen atom and the hydroxyquinole moiety. The indirect electropolymerization scenario, in its turn, isn't discarded either. The analysis of the correspondent model indicates the efficiency of the aquaraine dye/nano-CuS composite as an efficient electrode modifier for clioquinol electrochemical determination. The electroanalytical process is easy to conduct, and the analytical signal is easy to interpret. Contrarily to the acidic medium, the possibility of oscillatory and monotonic instability is less expressed.

**Keywords:** clioquinol, electrochemical sensor, squaraine dyes, copper (II) sulfide, stable steady-state

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## 1. Introduction

Clioquinol [1–4] (Fig. 1) is an antifungal, antibacterial and antiprotozoan drug. Its action is based on microorganism DNA inhibition. Moreover, it may be used for CoViD-19 treatment if combined with some zinc complexes.



**Figure 1.** Clioquinol structure.

The drug efficiency and action are dose-related. Moreover, as with other quinolone derivatives, it is highly toxic, provoking adverse effects like anorexia, weight loss, and muscle fragility [5–8]. Therefore, developing an efficient method for clioquinol detection and quantification is possible, and electroanalytical methods may serve as a good solution for this task [9–12].

Being a quinolone derivative, clioquinol may be either electrooxidized or electroreduced, which foresees the possibility for cathodic and anodic strategies. In the first case, the reaction may be realized by a nitrogen atom (yielding an N-oxide) or via a carbocyclic ring, substituted by donor substituents. In this process, the overvoltage generally occurs on bare electrodes, which is why the electrodes are modified by a specific material capable of reacting specifically with the analyte. Such electrodes are known as chemically modified (CME) [13–18].

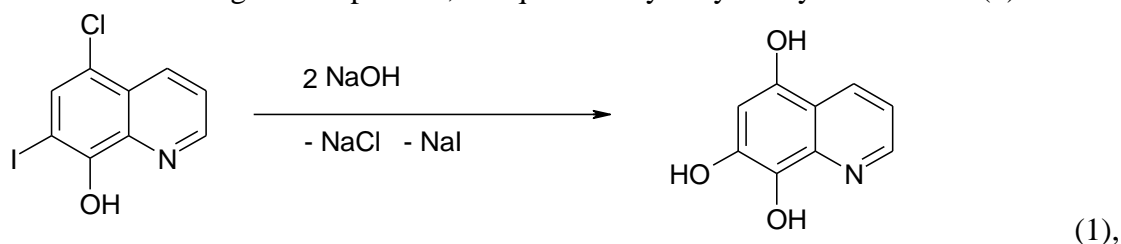
In the case of clioquinol, which contains a strong accepting pyridinic ring, the strong oxidants, like peroxides and cobalt (III) oxyhydroxide in pair with cobalt dioxide and bivalent copper compounds, including copper (II) sulfide, easily affordable in the form of the nanoparticles over an organic phase, which are oxidized in alkaline medium, yielding *in situ* trivalent copper compounds, are generally used for an anodic process. These oxidants are generally stabilized by conjugated organic oligo and macromolecular compounds like squaraine dye, conducting polymers, or carbon material.

Polymeric hybrid materials have been extensively used for the last two years in both anodic and cathodic processes [20, 21]. Moreover, those processes tend to be accompanied by electrochemical instabilities, which are capable of influencing the electrochemical equipment in general and the sensing properties of the concrete system. The analysis of the theoretical and experimental data [22–28] confirms that those instabilities are generally caused by ionic force, surface, and autocatalytical phenomena, such as electrode material conductivity, and are characteristic of different CME.

Therefore, in this work, we theoretically describe the possibility of the electrochemical determination of clioquinol over the composite containing squaraine dye and copper sulfide nanoparticles. This is realized by the mechanism suggestion, its mathematical description by developing and analyzing the mathematical model from the stability point of view, and the comparison between this system and similar ones [27, 28].

## 2. Materials and Methods

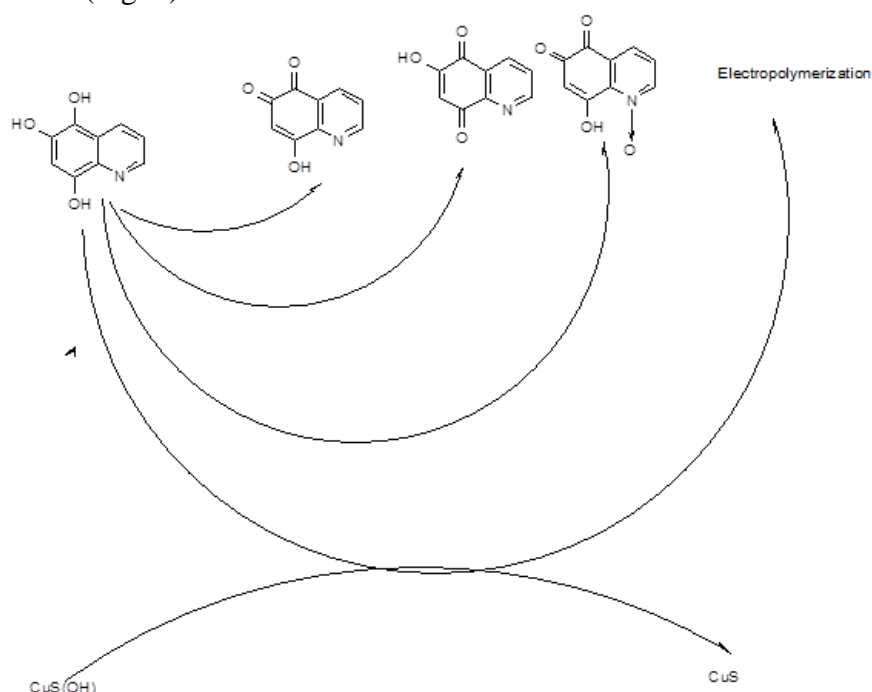
In the first stage of the process, clioquinol is hydrolyzed by the reaction (1):



and yields a hydroxyquinole moiety, condensed with the pyridinic ring, whereas the cobalt sulfide is oxidized in an alkaline medium, yielding a strong, highly energetic oxidant – copper (III) sulfohydroxide (2):



Then, copper sulfohydroxide oxidized the clioquinol hydrolysis products by four parallel scenarios, including  $\alpha$ -hydroquinonic,  $\gamma$ -hydroquinonic, N-oxidation, and indirect electropolymerization (Fig. 2):



**Figure 2.** The scheme of the electroanalytical process.

Therefore, taking some assumptions [27, 28], we describe the system's behavior by a trivariant equation-set (3):

$$\begin{cases} \frac{dq}{dt} = \frac{2}{\delta} \left( \frac{\Delta}{\delta} (q_0 - q) - r_h \right) \\ \frac{dh}{dt} = \frac{2}{\delta} (r_h - r_1 - r_2 - r_N - r_P) \\ \frac{dc}{dt} = \frac{1}{C} (r_1 + r_2 + r_N + r_P - r_3) \end{cases} \tag{3}$$

Herein,  $q$  and  $h$  are clioquinol and its hydrolysis product concentrations,  $q_0$  stands for clioquinol bulk concentration,  $\Delta$  stands for clioquinol diffusion coefficient,  $C$  is the copper sulfide maximal matrix concentration,  $c$  is its surface coverage degree, and the parameters  $r$  stand for the correspondent reaction rates, calculated as:

$$r_h = k_h q \exp(-aq) \tag{4}$$

$$r_1 = k_1 h(1 - c)^2 \quad (5)$$

$$r_2 = k_2 h(1 - c)^2 \quad (6)$$

$$r_N = k_N h(1 - c)^3 \quad (7)$$

$$r_p = k_p h^n (1 - c)^{2n-2} \quad (8)$$

$$r_3 = k_3 c \exp\left(\frac{F\varphi_0}{RT}\right) \quad (9)$$

Herein, the parameters  $k$  stand for the correspondent reaction rate constants,  $n$  is the polymer chain length,  $F$  stands for the Faraday number,  $\varphi_0$  is the zero-charge-related DEL potential slope,  $R$  is the universal gas constant, and  $T$  is the solution absolute temperature.

As the pyridinic nitrogen atom is not ionized in alkaline media, the ionic form transformation is given only during the analyte hydrolysis. Therefore, DEL is less affected than in the acidic medium. Moreover, the stability topological area is widened, which favors the easy analytical signal interpretation, as shown below.

### 3. Results and Discussion

We analyze the system with clioquinol electrochemical determination on squaraine dye/CuS composite by analyzing the equation-set (3) using the linear stability theory and, for this purpose, expose the Jacobian matrix members as (10):

$$\begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix} \quad (10)$$

Herein:

$$a_{11} = \frac{2}{\delta} \left( -\frac{\Delta}{\delta} - k_h \exp(-aq) + ak_h q \exp(-aq) \right) \quad (11)$$

$$a_{12} = 0 \quad (12)$$

$$a_{13} = 0 \quad (13)$$

$$a_{21} = \frac{2}{\delta} (k_h \exp(-aq) - ak_h q \exp(-aq)) \quad (14)$$

$$a_{22} = \frac{2}{\delta} (-k_1(1 - c)^2 - k_2(1 - c)^2 - k_N(1 - c)^3 - nk_p h^{n-1} (1 - c)^{2n-2}) \quad (15)$$

$$a_{23} = \frac{2}{\delta} (2k_1 h(1 - c) + 2k_1 h(1 - c) + 3k_N h(1 - c)^2 + (2n - 2)k_p h^n (1 - c)^{2n-3}) \quad (16)$$

$$a_{31} = 0 \quad (17)$$

$$a_{32} = \frac{1}{c} (k_1(1 - c)^2 + k_2(1 - c)^2 + k_N(1 - c)^3 + nk_p h^{n-1} (1 - c)^{2n-2}) \quad (18)$$

$$a_{33} = \frac{1}{c} \left( -2k_1 h(1 - c) - 2k_1 h(1 - c) - 3k_N h(1 - c)^2 - (2n - 2)k_p h^n (1 - c)^{2n-3} - k_3 \exp\left(\frac{F\varphi_0}{RT}\right) - jk_3 c \exp\left(\frac{F\varphi_0}{RT}\right) \right) \quad (19)$$

From the Jacobian main diagonal elements (11), (15), and (19), it is possible to conclude that the positive callback, necessary for the Hopf bifurcation, describing the oscillatory behavior, is possible, as the mentioned elements possess positive addendums.

In this system, the oscillatory behavior is more probable than in the simplest case, but either way, it is less probable than in the acidic medium. It is caused by two factors against one in the simplest case, but the DEL ionic force change factor is only manifested in the hydrolysis reactions, contrarily to other chemical stages. Also, the oscillatory behavior will be caused by the change of DEL and surface ionic force and conductivity in the electrochemical stage. Mathematically, it is manifested by the positivity of the elements  $ak_hq \exp(-aq)$  and  $-jk_3c \exp\left(\frac{F\varphi_0}{RT}\right)$ . The oscillation frequency and amplitude will depend on the background electrolyte composition.

Avoiding the cumbersome expressions during the steady-state stability investigation using the Routh-Hurwitz criterion, we rewrite the Jacobian determinant as (20):

$$\frac{4}{\delta^2 C} \begin{vmatrix} -\kappa - \mathcal{E} & 0 & 0 \\ \mathcal{E} & -\Sigma & P \\ 0 & \Sigma & -P - \Omega \end{vmatrix} \quad (20)$$

And express the stability requisite by opening the straight brackets and applying the  $\text{Det}J < 0$  condition, salient from the criterion, as (21):

$$-\Sigma\Omega(\kappa + \mathcal{E}) < 0 \quad (21)$$

The inequation (21) is warranted to be satisfied if the hydrolytic parameter  $\mathcal{E}$  and the electrooxidation parameter  $\Omega$  are positive, which is characteristic of most real systems. Therefore, this inequation describes an efficient diffusion and kinetically controlled electroanalytical system.

Considering that no side reaction compromising the analyte and(or) modifier stability is realized in this system, the steady-state stability will correspond to the linear dependence between the electrochemical parameter and concentration, providing efficient analytical signal interpretation.

The detection limit is defined by the monotonic instability, which depicts the margin between the stable steady states and unstable states. It is described mathematically by the nullity of the Jacobian determinant, or (22):

$$-\Sigma\Omega(\kappa + \mathcal{E}) = 0 \quad (22)$$

If stronger oxidants are used, the clioquinol will also be N-oxidized and electropolymerized. The assisted electropolymerization will be thereby realized as electrocopolymerization, and the Jacobian elements (12), (13), and (17) won't be nil. As for the proper equation-set (3), it will be rewritten as (23):

$$\begin{cases} \frac{dq}{dt} = \frac{2}{\delta} \left( \frac{\Delta}{\delta} (q_0 - q) - r_h - r_N - r_P \right) \\ \frac{dh}{dt} = \frac{2}{\delta} (r_h - r_1 - r_2 - r_N - r_P) \\ \frac{dc}{dt} = \frac{1}{c} (r_1 + r_2 + r_N + r_P - r_3) \end{cases} \quad (23)$$

This case will be analyzed in one of our next works.

## 4. Conclusions

From the analysis of the system with the electrochemical determination of clioquinol over the Squaraine Dye/CuS composite, it is possible to conclude that the electroanalytical process is efficient. Moreover, it is more efficient in the alkaline medium than in the acidic medium. Although oscillatory behavior is possible, it is less probable than acidic solutions. As for the electroanalytical process, it is either diffusion or kinetically controlled, and the electroanalytical signal is easy to interpret.

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## Conflicts of Interest

The authors declare no conflict of interest.

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