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The Mathematical Model for Dantrolene Electrochemical Determination on VO(OH)' Modified Electrode

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Abstract: The possibility of dantrolene has been evaluated for the first time. Considering the presence of accepting groups, the electroanalytical process is carried out on a cathode, modified by vanadium (III) oxyhydroxide. The electroanalytical process is realized via a branched mechanism, which permits the detection of the drug concentration by multiple reduction peak depths. Both oscillatory and monotonic instability will be more likely to be realized due to the formation of ionic forms during some of the chemical stages. Nevertheless, the electroanalytical process is efficiently realized. The linear dependence between the electrochemical parameter and concentration is easily achieved, making the analytical signal easy to interpret.

Keywords: dantrolene; electrochemical sensor; vanadium (III) oxyhydroxide; electrochemical oscillations; stable steady-state

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1. Introduction

Dantrolene (Figure 1) [1-6] is a muscle relaxant that reduces the excitation-contraction coupling in muscle cells. It is a calcium channel blocker used to treat malignant hyperthermia, neuroleptic malignant syndrome, muscle spasticity, poisoning by 2-4-dinitrophenols, ecstasy drugs, and sting by some marine organisms, like the Portuguese man-of-war and blue sea dragon.

Dantrolene is synthesized from furfural by Meyerwein arylation and hydrazide formation.

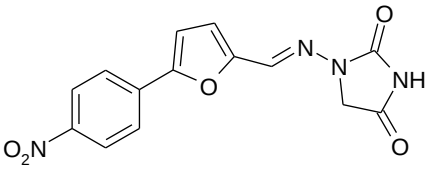

Figure 1. Dantrolene.

Although dantrolene is a hydantoin derivative, it does not manifest any antiepileptic activity. Its function is dose-related, and it might not be given to people with pre-existing liver, lung, or cardiovascular diseases [7-9]. Moreover, it is not recommended for pregnant and breastfeeding mothers or children up to 5 years old. Therefore, in order to control the dosage and action of the drug, it is necessary to develop an efficient and rapid analytical technique for dantrolene quantification [10-12] and the electroanalytical processes, yet used for similar compounds [13-20] may efficient in this case.

Possessing three accepting groups (nitro group, hydrazo group, hydantoin moiety), dantrolene is more suitable for electrochemical determination by cathodic processes, although anodic electrooxidation for electroanalytical uses is also possible. Electrochemical reduction involves proton and electron transfer. Using VO(OH) as a proton and electron transfer mediator allows us to avoid a strongly acidic medium in this reaction and stabilizes the system.

In this work, we describe theoretically the possibility of the electrochemical reduction of dantrolene on VO(OH)-modified cathode in neutral and mildly acidic medium. This investigation includes a mechanistic stability analysis and a comparison of this system's behavior with that of similar ones [21-32].

2. Materials and Methods

Dantrolene electrochemical reduction on vanadium oxyhydroxide will be realized firstly by hydantoin moiety, as this process will be most rapid. The reduction of two other groups is slower and will occur in the first reduction product. For this reason, the electroanalytical reduction of dantrolene will be realized by a hybrid scenario involving all electron-accepting moieties (Figure 2).

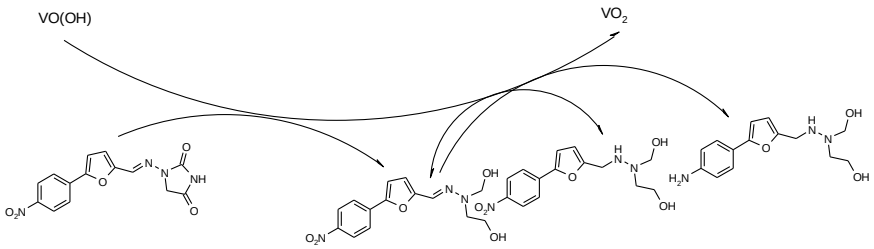

Figure 2. Schematical representation of dantrolene gradual electrochemical reduction on VO(OH)-modified electrode.

Taking this into account and accepting certain assumptions [21-28], we describe the behavior of this system by the trivariate equation-set, exposed as:

$$\begin{cases} \frac{dc}{dt} = \frac{2}{\delta}\left(\frac{\Delta}{\delta}(c_0 - c) - r_1\right) \\ \frac{dc*}{dt} = \frac{2}{\delta}(r_1 - r_{21} - r_{22}) \\ \frac{dv}{dt} = \frac{1}{V}(r_1 + r_{21} + r_{22} - r_r) \end{cases} \quad (1)$$

Herein, c is dantrolene pre-surface concentration, Δ is its diffusion coefficient, δ is the diffusion-layer thickness, c_0 is dantrolene bulk concentration, c* is dantrolene first reduction product pre-surface concentration, v is vanadium dioxide surface coverage degree, V is its maximal surface concentration, and the parameters r stand for the correspondent reaction rates, calculated as (2-5):

$$r_1 = k_1 c(1 - v)^8 \exp(-\alpha c) \quad (2)$$

$$r_{21} = k_{21} c*(1 - v)^6 \exp(-\beta c*) \quad (3)$$

$$r_{22} = k_{22} c*(1 - v)^2 \exp(-\beta c*) \quad (4)$$

$$r_r = k_r v \exp\left(-\frac{F\varphi_0}{RT}\right) \quad (5)$$

Herein, the parameters k stand for the correspondent reaction rate constants, α and β stand for the parameters, relating the double electric layer (DEL) electrochemical and electrophysical properties to ionic forms transformation during the chemical stages involving dantrolene and its first reduction product correspondently, 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.

Considering that the ionic compound transformation occurs in this system, the oscillatory behavior will be more probable than in the simple case and of equal probability to similar systems [21-28], in which the steady-state stability topological region remains wide.

This corresponds to the efficient dantrolene determination in mildly acidic and neutral mediums, as shown below.

3. Results and Discussion

We investigate the steady-state stability for the system with dantrolene VO(OH)-assisted determination by linear stability theory and describe the steady-state Jacobian elements as (6):

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

in which:

$$a_{11} = \frac{2}{\delta}\left(-\frac{\Delta}{\delta} - k_1(1 - v)^8 \exp(-\alpha c) + \alpha k_1 c(1 - v)^8 \exp(-\alpha c)\right) \quad (7)$$

$$a_{12} = 0 \quad (8)$$

$$a_{13} = \frac{2}{\delta}(8k_1 c(1 - v)^7 \exp(-\alpha c)) \quad (9)$$

$$a_{21} = \frac{2}{\delta}(k_1(1 - v)^8 \exp(-\alpha c) - \alpha k_1 c(1 - v)^8 \exp(-\alpha c)) \quad (10)$$

$$a_{22} = \frac{2}{\delta}\left(-k_{21}(1 - v)^6 \exp(-\beta c*) - k_{22}(1 - v)^2 \exp(-\beta c*) + \beta(k_{21} c*(1 - v)^6 \exp(-\beta c*) + k_{22} c*(1 - v)^2 \exp(-\beta c*))\right) \quad (11)$$

$$a_{23} = \frac{2}{\delta}(-8k_1 c(1 - v)^7 \exp(-\alpha c) + 6k_{21} c*(1 - v)^5 \exp(-\beta c*) + 2k_{22} c* \exp(-\beta c*)) \quad (12)$$

$$a_{31} = \frac{1}{V}(k_1(1-v)^8 \exp(-\alpha c) - \alpha k_1 c(1-v)^8 \exp(-\alpha c)) \quad (13)$$

$$a_{32} = \frac{1}{V}\left(k_{21}(1-v)^6 \exp(-\beta c*) + k_{22}(1-v)^2 \exp(-\beta c*) - \beta(k_{21} c*(1-v)^6 \exp(-\beta c*) + k_{22} c*(1-v)^2 \exp(-\beta c*))\right) \quad (14)$$

$$a_{33} = \frac{1}{V}\left(-8k_1 c(1-v)^7 \exp(-\alpha c) - 6k_{21} c*(1-v)^5 \exp(-\beta c*) - 2k_{22} c* \exp(-\beta c*) - k_r \exp\left(-\frac{F\varphi_0}{RT}\right) + jk_r v \exp\left(-\frac{F\varphi_0}{RT}\right)\right) \quad (15)$$

Avoiding the cumbersome expression during the determinant analysis, we introduce new variables and rewrite the determinant as (16):

$$\text{Det J} = \frac{4}{\delta^2 C}\begin{vmatrix} -\kappa - \Xi & 0 & \Lambda \\ \Xi & -P & -\Lambda + \Phi \\ \Xi & P & -\Lambda - \Phi - \Omega \end{vmatrix} \quad (16)$$

which, considering the determinant properties, will be rewritten as (17)

$$\text{Det J} = \frac{4}{\delta^2 C}\begin{vmatrix} -\kappa - \Xi & 0 & \Lambda \\ \Xi & -P & -\Lambda + \Phi \\ 2\Xi & 0 & -2\Lambda - \Omega \end{vmatrix} \quad (17)$$

Considering that:

$$-Det\ J \begin{cases} > 0, for\ steady - state\ stability \\ = 0\ monotonic\ instability \end{cases} \quad (18)$$

Opening the brackets, applying the Det J<0 requisite, salient from the criterion, and changing the signs to the opposite, we rewrite the condition set as (19):

$$-P(2\Lambda\kappa + 2\Lambda\Omega + \Xi\Omega) \begin{cases} > 0, curve\ linearity \\ = 0, monotonic\ instability \end{cases} \quad (19)$$

If –Det J>0, the Routh-Hurwitz stability criterion is valid, and the steady-state is thereby stable, providing an efficient steady-state drug determination. Moreover, the wide stability region allows us to use this system in pharmaceutical formulations, biological liquids, and the environment.

This criterion is readily satisfied if the kinetic parameters P, Λ, and Ω are positive. In the vast majority of the cases, they both have positive signs, and considering that the other variables in the determinant are positive, it indicates the vast steady-state stability topological region. The electroanalytical process is mostly kinetically controlled.

In the absence of the side reactions or other factors capable of compromising the analyte and (or) modifier stability, excluding the reactions foreseen by the mechanism, the linearity between the electrochemical parameter and concentration is observed, providing an efficient analytical signal interpretation, which is important for drug concentration monitoring.

The condition Det J=0 corresponds to the detection limit, manifested by the *monotonic instability*. It may be seen as an N-shaped part of the steady-state voltammogram, which depicts the margin between the stable steady-states and unstable states and corresponds to the steady-state multiplicity. In other words, multiple steady-states, each one unstable, coexist at this point.

As for the oscillatory behavior, it is realized beyond the detection limit in the case of the Hopf bifurcation realization. Its realization requires the presence of the positive-callback related positive addendums in main diagonal elements.

Observing the main diagonal elements (11), (15), and (19), we may observe that the oscillatory behavior becomes possible if the kinetic parameters α, λ, and j are positive, which corresponds to the DEL influences of the chemical and electrochemical stages. This factor is typical for similar systems [21-28] and may be described by the positivity of the elements

$\alpha k_1 c(1-v)^8 \exp(-\alpha c)$, if α>0, $\beta(k_{21} c*(1-v)^6 \exp(-\beta c*) + k_{22} c*(1-v)^2 \exp(-\beta c*)) > 0$, if $\beta > 0$ and $jk_r v \exp\left(-\frac{F\varphi_0}{RT}\right) > 0$, if j>0. These elements describe the positive callback, and this callback will depend on the system´s characteristics. For example, the oscillation frequency and amplitude depend on the background electrolyte composition, proven experimentally and theoretically [21-28].

Dantrolene determination as a sodium salt (in a more soluble form) is given on conducting polymer matrix. In this case, the process is anodes anodic, preceded by dantrolene CP doping. This case is similar to that observed in [21-28] and will be evaluated in our next papers.

4. Conclusions

From the theoretical description of dantrolene electrochemical cathodic determination by VO(OH)-modified cathode, it has been possible to conclude it may be an excellent modifier for drug quantification in different neutral and mildly acidic media. The electroanalytical process is mostly kinetically controlled. The oscillatory behavior in this system may be caused by the influence of DEL on electrochemical and chemical stages. The system may be used as an electroanalytical, providing efficient analytical signal interpretation.

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Conflicts of Interest

The authors declare no conflict of interest.

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