

Theoretical Description for Necatorin Electrochemical Determination, Assisted by CoO(OH)/CoO₂ Redox Pair

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Abstract: The electrochemical determination of necatorin mycotoxin in a neutral medium has been investigated theoretically for the first time. As necatorin possesses three accepting moieties, including the pyridinic nitrogen heteroatoms and lactone ring, the anodic process would be realized by strong oxidants; thus, CoO(OH) as electrode modifier will be used as a reductant, producing *in situ* tetravalent cobalt oxide. Analysis of the corresponding mathematical model confirms the feasibility of using the CoO(OH)/CoO₂ redox pair to detect necatorin, with an easy and efficient analytical signal interpretation. As for the proper necatorin, it may be used as a green monomer for conducting polymers for electroanalytical and energy-conversion purposes.

Keywords: food safety; necatorin; cobalt (III) oxyhydroxide; electrochemical sensors; conducting polymers; electropolymerization; stable steady-state.

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1. Introduction

<https://nanobioletters.com/>

Lactarius turpis, also classified as *Lactarius necator* and popularly known as “ugly milkcap,” is a mushroom naturally found in Northern Europe and Siberia [1–4]. Depending on the culture of mushroom picking, it may be considered poor or inedible due to the presence of its specific mycotoxin, known as necatorin (Figure 1) [5–9]. As the major part of the derivatives of acridine, phenanthridine, and their aza-substituted compounds, they are mutagenic, and their mutagenesis is manifested in inchmeal, causing long-term toxicity. The short-term toxicity will be nearly analogous to that of Orellana [8,9]. For this reason, the development of an efficient method for necatorin electrochemical determination has become up to date [10–12], and the electrochemical sensors, yet used for similar compounds [13–18], could be efficiently used, due to their portability, precision, and rapidity.

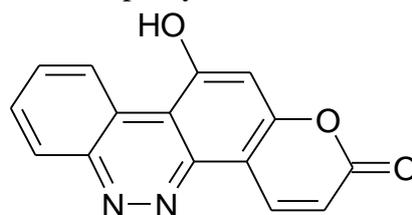
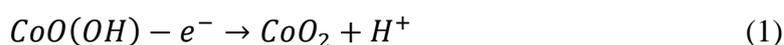


Figure 1. Necatorin.

Taking into account the presence of both donating (aromatic carbocyclic ring and phenolic hydroxyl) and accepting (lactone ring, both of the pyridinic nitrogens) functional groups, necatorin may be easily detected by both cathodic and anodic processes. Moreover, the presence of the conjugated system may indicate its potential for electropolymerization [19–25], which can be used for both energy conversion and electroanalysis.

Nevertheless, due to the presence of high-electron-accepting moieties, anodic oxidation will be more readily achieved when strong oxidants are used as electrode modifiers. Cobalt (III) oxyhydroxide, which nowadays has become more popular as an electrode modifier [26–30], may also be used, but the pair $\text{CoO}(\text{OH})/\text{CoO}_2$, involving an *in situ* formation of cobalt (IV) oxide (1):



The capability to oxidize necatorin under different conditions is strongly recommended.

The above-mentioned aspects are important for the mechanistic and dynamic behavior of the electroanalytical process. Moreover, the presence of electrochemical instabilities, which makes it difficult to interpret the analytical signal, may be associated with different oxidation scenarios involving $\text{CoO}(\text{OH})$ synthesis and electroanalytical properties [31,32]. Those instabilities are typical and may limit the electroanalytical and removal use of the electrochemical process. To foresee the possibility of electrochemical instabilities and the effects they may produce, it's necessary to investigate the process from a mechanical point of view and analyze its behavior theoretically.

The goal of this work is to evaluate the mechanical properties of necatorin during $\text{CoO}(\text{OH})/\text{CoO}_2$ -assisted electrooxidation, including the polymerization scenario. The theoretical investigation includes the development and analysis of the corresponding mathematical model, thereby permitting the analysis of its behavior as it is and in comparison with similar ones [33–35].

2. Materials and Methods

Necatorin may be evaluated simultaneously as either Cinnoline, aza[10]phenanthridine, and coumarin derivatives. For this reason, both anodic and cathodic scenarios may be applied to it. Moreover, electropolymerization is also possible due to the formation of an ion-radical growth center by both electrophilic and nucleophilic mechanisms. If anodic electropolymerization (direct or assisted) is performed, poly(necatorin) will be configured as shown in Figure 2.

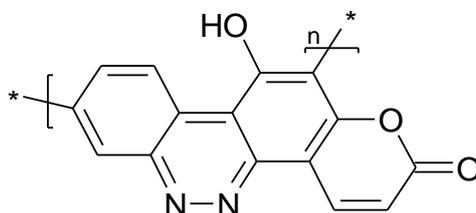


Figure 2. Possible poly(necatorin) structure.

The resulting polymer maintains the N-N bond, which may mediate proton transfer. Moreover, it may be oxidized by N-oxidation and phenolization. The realization of “the polythiophene paradox,” in which the newly synthesized polymer becomes partly overoxidized, isn’t discarded.

For this reason, the electroanalytical process is shown in Figure 3.

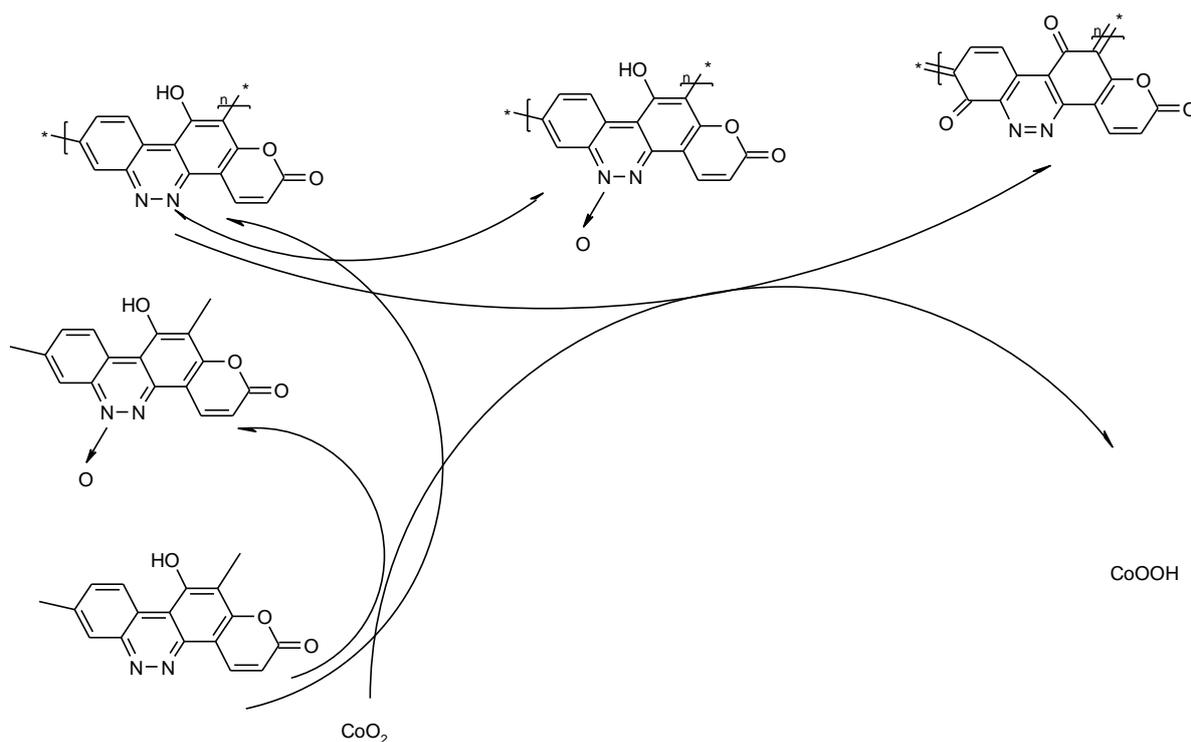


Figure 3. Necatorin CoO(OH)/CoO₂ assisted electrochemical oxidation.

The penalized and oxidized poly(necatorin) structure will gain some properties of the overoxidized conducting polymer. Although its conductivity will diminish after phenolizing overoxidation, it may still be useful for electroanalytical determinations in proton-leaving processes and those based on complex formation.

Therefore, taking certain assumptions [33–35], we describe the behavior of this process by a trivariate equation set (1).

$$\begin{cases} \frac{dn}{dt} = \frac{2}{\delta} \left(\frac{N}{\delta} (n_0 - n) - r_p - r_{O1} \right) \\ \frac{dp}{dt} = \frac{2}{\delta} (r_p - r_{O2} - r_{ph}) \\ \frac{dc}{dt} = \frac{1}{c} (r_d + r_{O1} + r_{O2} + r_{ph} - r_r) \end{cases} \quad (2)$$

Herein, n and p are necatorin and its polymer pre-surface layer concentrations, n_0 stands for necatorin bulk concentration, Δ is its diffusion coefficient, c and C are cobalt oxyhydroxide coverage degree and maximal surface concentration, to which it is referred, and the parameters r stand for the chemical and electrochemical reaction rates:

$$r_{O1} = k_{O1}n(1 - c) \quad (3)$$

$$r_p = k_p c^x (1 - c)^y \quad (4)$$

$$r_{O2} = k_{O2}p(1 - c)^n \quad (5)$$

$$r_{ph} = k_{ph}p^w (1 - c)^z \quad (6)$$

$$r_r = k_r s \exp\left(\frac{F\phi_0}{RT}\right) \quad (7)$$

Herein, k stand for the correspondent unit reaction rates (rate constants), n is the average number of the monomer units in the polymer, w , x , y , and z are polymerization and phenolization reaction order, F is the Faraday number, ϕ_0 is the zero-charge related potential slope, R is the universal gas constant and T is the absolute temperature.

In this system, a neutral medium is used to minimize the destabilizing influence of ionization of both necatorin and its polymer. For this reason, the probability of the oscillatory behavior will be reduced, and the proper process will be more efficient and easier to reproduce, as shown below.

3. Results and Discussion

Herein, the behavior of the necatorin electrochemical determination over CoO(OH)-modified electrode in pair with cobalt dioxide in a neutral medium is described by the equation set (1), including the algebraic relations (2–7) therein. According to the linear stability theory, the steady-state Jacobian matrix members are described as (8):

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

Herein:

$$a_{11} = \frac{2}{\delta} \left(-\frac{N}{\delta} - k_{O1}(1 - c) - xk_p c^{x-1} (1 - c)^y \right) \quad (9)$$

$$a_{12} = 0 \quad (10)$$

$$a_{13} = \frac{2}{\delta} (k_{O1}n(1 - c) + yk_p n^x (1 - c)^{y-1}) \quad (11)$$

$$a_{21} = \frac{2}{\delta} (xk_p n^{x-1} (1 - c)^y) \quad (12)$$

$$a_{22} = \frac{2}{\delta} (-k_{O2}(1 - c)^n - wk_{ph}p^{w-1} (1 - c)^z) \quad (13)$$

$$a_{23} = \frac{2}{\delta} (nk_{O2}p(1 - c)^{n-1} + zk_{ph}p^w (1 - c)^{z-1}) \quad (14)$$

$$a_{31} = \frac{1}{c} (k_{O1}(1 - c) + xk_p c^{x-1} (1 - c)^y) \quad (15)$$

$$a_{32} = \frac{1}{c} (k_{O2}(1 - c)^n + wk_{ph}p^{w-1} (1 - c)^z) \quad (16)$$

$$a_{33} = \frac{1}{c} \left(-k_{O1}n(1 - c) - yk_p n^x (1 - c)^{y-1} - nk_{O2}p(1 - c)^{n-1} - zk_{ph}p^w (1 - c)^{z-1} - k_r \exp\left(\frac{F\phi_0}{RT}\right) + jk_r s \exp\left(\frac{F\phi_0}{RT}\right) \right) \quad (17)$$

Avoiding cumbersome expressions, we describe the Jacobian matrix as (18) :

$$\frac{4}{\delta^2 S} \begin{vmatrix} -\nu - \varepsilon - \Sigma & 0 & T + Y \\ \varepsilon & -P & T - Y \\ \varepsilon + \Sigma & P & -T - Y - \Omega \end{vmatrix} \quad (18)$$

Opening the brackets, we obtain the determinant value, calculated as (19):

$$P(2\nu Y + \nu\Omega + \varepsilon\Omega + \Sigma\Omega - 3\varepsilon T + 2\varepsilon Y - \Sigma T) \begin{cases} > 0, \text{linear dependence range} \\ = 0, \text{detection limit} \end{cases} \quad (19)$$

When $-\text{Det } J > 0$, the Routh-Hurwitz criterion confirms steady-state stability, enabling efficient electrochemical determination of bisphenols. The wide stability region also makes this system suitable for electroanalytical sensing. This condition is generally satisfied when the kinetic parameter Ω is positive, as it usually is, and, when combined with other positive determinant variables, it defines a broad, stable operating region. The process is largely kinetically controlled.

In the absence of side reactions or factors that could compromise the analyte or modifier—beyond those specified by the mechanism—the electrochemical response exhibits linearity with analyte concentration. This ensures reliable signal interpretation, which is critical for necatorin monitoring. Electrosynthetically, this corresponds to the formation of a well-developed polymer surface that serves as an electrode modifier for both electroanalytical applications and energy conversion systems.

The condition $\text{Det } J = 0$ corresponds to the detection limit, manifested as monotonic instability. It appears as an N-shaped region in the steady-state voltammogram, marking the boundary between stable and unstable states and reflecting steady-state multiplicity. In other words, multiple unstable steady states coexist at this point.

The primary condition for a Hopf bifurcation, which leads to oscillatory behavior, is the presence of positive elements on the main diagonal of the Jacobian, which drive the positive feedback. Unlike the cases reported in [33–35], here only one main-diagonal term can be positive: $jk_r s \exp\left(\frac{F\varphi_0}{RT}\right) > 0$, provided that $k_r > 0$. This term represents cyclic changes in conductivity in the electroanalytical cell during the electrochemical stage, corresponding to frequent, small-amplitude oscillations.

A poly (necatorin)-modified electrode may be used in either anodic or cathodic electroanalytical systems, mediating proton and electron transfer and immobilizing an analyte via doping. It may also be used to determine heavy metal concentrations in neutral and acidic solutions. Acidic solutions are also suitable for cathodic electroanalytical systems.

In basic solutions, an ester fragment is destroyed, which is frequently used to desintoxicate the mushroom. In a basic medium, it is recommended to use a conducting polymer as an electrode modifier. This system is similar to that used for sotolon determination and will be analyzed in the continuation.

4. Conclusions

Analysis of necatorin electrochemical determination using a $\text{CoO}(\text{OH})$ -modified electrode in pair with CoO_2 indicates that the process is efficient, with a clear linear relationship between analyte concentration and the electrochemical response in a kinetically controlled system. A neutral pH is recommended for the anodic process. At the same time, oscillatory behavior is less likely than in alkaline media and occurs only due to changes in the double-layer structure during the electrochemical stage. Poly(necatorin)-modified electrodes are

suitable for both electroanalytical and energy conversion applications at neutral or mildly acidic pH values, particularly for cathodic processes.

Author Contributions

Conceptualization, V.V.T., T.V.M., J.I.F.P.M., Y.G.I., P.I.Y., M.V.K. and M.P.K., methodology, V.V.T., T.V.M., J.I.F.P.M., Y.G.I., P.I.Y., M.V.K.; validation, V.V.T., T.V.M., J.I.F.P.M., Y.G.I., P.I.Y., M.V.K., A.O.H., S.C.O., V.V.K., I.I.K., V.I. K., I.G.B., T.B.S., Y.G.I., M.G.B., A.O.S., J.R.G., G.F.T., O.P.M., M.V.M., O.O.M., M.J.M., V.M.O. and M.P.K. formal analysis, V.V.T., T.V.M., J.I.F.P.M., Y.G.I., P.I.Y., M.V.K., A.O.H., S.C.O., V.V.K., I.I.K., V.I. K., I.G.B., T.B.S., Y.G.I., M.G.B., A.O.S., J.R.G., G.F.T., O.P.M., M.V.M., O.O.M., M.J.M., L.O.N., I.G.P., V. L., L.M., V.M.O. and M.P.K.; investigation, V.V.T., T.V.M., J.I.F.P.M., Y.G.I., P.I.Y., M.V.K., A.O.H., S.C.O., V.V.K., I.I.K., V.I. K., I.G.B., T.B.S., Y.G.I., M.G.B., A.O.S., J.R.G., G.F.T., O.P.M., M.V.M., O.O.M., M.J.M., L.O.N., I.G.P., V. L., L.M., V.M.O. and M.P.K.; resources, V.V.T., T.V.M., J.I.F.P.M., Y.G.I., P.I.Y., M.V.K., A.O.H., S.C.O., V.V.K., I.I.K., V.I. K., I.G.B., T.B.S., Y.G.I., M.G.B., A.O.S., J.R.G., G.F.T., O.P.M., M.V.M., O.O.M., M.J.M., L.O.N., I.G.P., V. L., L.M., V.M.O. and M.P.K.; data curation, V.V.T., T.V.M., J.I.F.P.M., Y.G.I., P.I.Y., M.V.K., A.O.H., S.C.O., V.V.K., I.I.K., V.I. K., I.G.B., T.B.S., Y.G.I., M.G.B., A.O.S., J.R.G., G.F.T., O.P.M., M.V.M., O.O.M., M.J.M., L.O.N., I.G.P., V. L., L.M., V.M.O. and M.P.K.; writing—original draft preparation, V.V.T., T.V.M., J.I.F.P.M., Y.G.I., P.I.Y., M.V.K., A.O.H., S.C.O., V.V.K., I.I.K., V.I. K., I.G.B., T.B.S., and M.P.K.; writing—review and editing, V.V.T., T.V.M., J.I.F.P.M., Y.G.I., P.I.Y., M.V.K., A.O.H., S.C.O., V.V.K., I.I.K., V.I. K., I.G.B., T.B.S., Y.G.I., M.G.B., A.O.S., J.R.G., G.F.T., O.P.M., M.V.M., O.O.M., M.J.M., L.O.N., I.G.P., V. L., L.M., V.M.O. and M.P.K.; visualization, V.V.T., T.V.M., J.I.F.P.M., Y.G.I., P.I.Y., M.V.K., A.O.H., S.C.O., V.V.K., I.I.K., V.I. K., I.G.B., T.B.S., Y.G.I., M.G.B., A.O.S., J.R.G., G.F.T., O.P.M., M.V.M., O.O.M., M.J.M., L.O.N., I.G.P., V. L., L.M., V.M.O. and M.P.K.; supervision, P.I.Y., Y.G. I., J.R. G., J. I. F. P. M.; project administration, V.V.T.. All authors have read and agreed to the published version of the manuscript.

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Data supporting the findings of this study are available upon reasonable request from the corresponding author.

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Conflicts of Interest

The authors declare no conflict of interest.

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