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Theoretical Description for Psilocin and Coprine Electrochemical Determination in Mushroom Pulp and Biological Liquids over Cobalt (II) Oxyhydroxide-Modified Electrode

Volodymyr V. Tkach [1,*], Tetiana V. Morozova [2], Marta V. Kushnir [1], Sílvio C. de Oliveira [3], Viktor V. Kryvetskyi [4], Inna I. Kryvetska [4], Igor V. Kryvetskyi [4], Igor G. Biryuk [4], Tetiana B. Sykyrytska [4], Yana G. Ivanushko [4], Alla V. Velyka [4], Petro I. Yagodynets [1,*], Adriano O. da Silva [5], Jarem R. Garcia [6], José Inácio Ferrão da Paiva Martins [7], Gennadii F. Tkach [8], Oleg P. Melnyk [8], Oleksii O. Melnyk [8], Maria V. Melnyk [8], Maria João Monteiro [9], Lilia O. Nikitchenko [10], Iryna G. Patseva [11], Vitalina Lukyanova [11], Liudmyla Mohelnytska [11]

1 Chernivtsi National University, 58001, Kotsyubynsky Str. 2, Chernivtsi, Ukraine
2 National Transport University, 02000, Omelianovych-Pavlenko Str. 1, Kyiv, Ukraine
3 Institute of Chemistry. Federal University of Mato Grosso do Sul, 79074 – 460, Av. Sen. Felinto Müller, 1555, Vila Ipiranga, Campo Grande, MS, Brazil
4 Bukovinian State Medical University, 58001, Teatralna Sq, 9, Chernivtsi, Ukraine
5 Federal University of the West of Pará, Juruti Campus, 68170 – 000, Rua Veríssimo de Souza Andrade, s/n, Juruti, PA, Brazil
6 State University of Ponta Grossa, Uvaranas Campus, Av. Gal. Carlos Cavalcanti, 4748, 84030-900, Ponta Grossa, PR, Brazil
7 Engineering Faculty of the University of Porto, 4200-465, Rua Dr. Roberto Frias, s/n, Porto, Portugal
8 National University of Life and Environmental Science of Ukraine, 03041, Heroiv Oborony Str, 15, Kyiv, Ukraine
9 University of Trás-os-Montes and Alto Douro, Quinta de Prados, 5001-801, Folhadela, Vila Real, Portugal
10 Vinnytsia State Pedagogical University, 21000, Ostroz'ky Str. 32, Vinnytsia, Ukraine
11 Zhytomyr Polytechnic State University, 10005, Chudnivs'ka Str. 103, Zhytomyr, Ukraine
* Correspondence: nightwatcher2401@gmail.com (V.V.T.), ved1988mid@rambler.ru (P. I.Y.)
Scopus Author ID 55758299100

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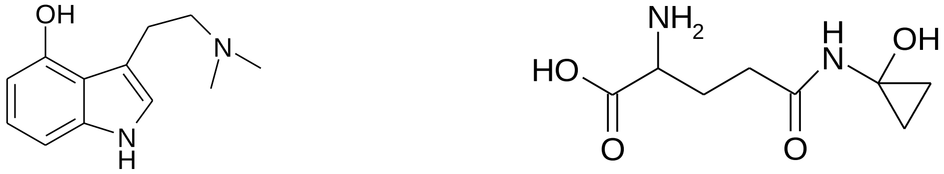
Abstract: The possibility of the electrochemical determination of poisonous mushroom toxins psilocin and coprine on CoO(OH)-modified electrode has been evaluated theoretically. The analysis of the correspondent mathematical model confirms the efficiency of the electrochemical sensor for detecting both mycotoxins in mushrooms and biological liquids for either investigation or diagnostic purposes. The linear dependence between the concentration of both of the analytes is established in a wide concentration range, providing an excellent analytical signal interpretation.

Keywords: poisonous mushroom intoxication; psilocin; coprine; cobalt (III) oxyhydroxide; electrochemical sensor; stable steady-state.

1. Introduction

Psilocin (Figure 1 to the left) is the main toxin of the poisonous psychedelic mushrooms *Ps. Mexicana* and *Ps. Cubensis is* responsible for its hallucinogenic effect [1 – 4]. Symptoms of psilocin toxicity include hypertension, tachycardia, visual problems, nausea, mydriasis, and disorientation.



(a) **(b)**

Figure 1. (a) Psilocin; **(b)** coprine structure.

The similar symptoms, adding the malaise, agitation, palpitation, and even coma [5 – 8], are characteristic of the common inkcap (*Coprinopsis atramentaria*) associated with coprine mycotoxin (Figure 1 to the right). Coprine is an alcohol dehydrogenase inhibitor, which leads to the alcohol flush reaction. Taking into account that the intoxication symptoms of both of the mushrooms are similar, the development of the sensor, capable of quantifying and differentiating each of the two toxins, becomes necessary [9 – 12], and the electroanalytical techniques used for the similar substances [13 – 20], may be useful for this purpose.

Considering the structure of both psilocin and coprine, we may conclude that anodic oxidation is more viable to quantify both substances, as they contain donating functional groups. So, the chemically modified electrodes (CME), which are characterized by their affinity to the analytes [21 – 28] (by the principle of the lock and the key), may be easily used to quantify both. Cobalt (III) oxyhydroxide, a semiconducting material with well-developed electrochemical properties [29 – 38], may be easily used as an electrode modifier for quantifying both analytes.

In this work, we describe from the theoretical point of view the possibility of the electrochemical determination of psilocin and coprine on CoO(OH)-modified cathode in a neutral and lightly alkaline medium. This investigation includes the mechanistic stability analysis and comparison of the behavior of this system with that of similar ones [39 – 49].

2. Materials and Methods

Both psilocin and coprin may be oxidized by CoO(OH) by a hybrid mechanism. Moreover, the polymerization mechanism for psilocin is also possible, and it may include the participation of coprin as a dopant (especially in alkaline medium). For this reason, the analytes do interact in certain conditions, which changes the model in relation to similar systems [45 – 49].

Psilocin may be oxidized, yielding low- and high-molecular oxidation products. The low-molecular oxidation is caused by its phenolization, which results in the subsequent quinone formation. High-molecular oxidation is the assisted polymerization scenario, which is realized at relatively higher potentials.

Coprin is oxidized by an amino group or a cyclopropane ring via the Wagner reaction. On the other hand, coordinate ions in an alkaline medium may enter the growing poly(psilocybin chain).

Schematically, the process may be described in the Figure 2:

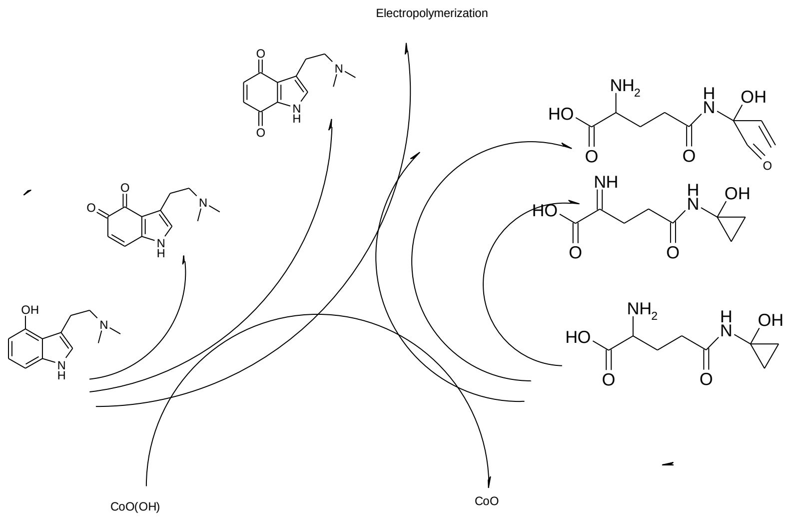


Figure 2. The scheme of the electroanalytical process.

Therefore, taking into account the above-cited statements and certain assumptions [45 – 49], we describe the behavior of this system by the trivariate equation-set (1):

$$\begin{cases} \frac{d\psi}{dt} = \frac{2}{\delta}\left(\frac{\Psi}{\delta}(\psi_0 - \psi) - r_{p1} - r_{p2} - r_p\right) \\ \frac{d\kappa}{dt} = \frac{2}{\delta}\left(\frac{K}{\delta}(\kappa_0 - \kappa) - r_{q1} - r_{q2} - r_p\right) \\ \frac{dc}{dt} = \frac{1}{C}\left(r_{p1} + r_{p2} + r_{q1} + r_{q2} + r_p - r_O\right) \end{cases} \quad (1)$$

Herein, ψ and κ are the psilocin and coprine pre-surface concentrations, ψ_0 and κ_0 stand for their bulk concentrations, Ψ and K are their diffusion coefficients, δ is the pre-surface layer thickness, c is the cobalt (II) oxide surface coverage degree, C is its maximal surface concentration and the parameters r stand for the correspondent reaction rates, calculated as (2 – 7):

$$r_{p1} = k_{p1}\psi(1-c)^4 \quad (2)$$

$$r_{p2} = k_{p2}\psi(1-c)^4 \quad (3)$$

$$r_p = k_p\psi^x(1-c)^y\kappa^z \exp(-\alpha\psi)\exp(-\beta\kappa) \quad (4)$$

$$r_{q1} = k_{q1}\kappa(1-c)^4 \exp(-\beta\kappa) \quad (5)$$

$$r_{q2} = k_{q2}\kappa(1-c)^4 \exp(-\beta\kappa) \quad (6)$$

$$r_O = k_O c \exp\frac{F\varphi_0}{RT} \quad (7)$$

Herein, the parameters k stand for the correspondent reaction rate constants, x, y, and z are the polymerization reaction rates, α and β are variables describing the DEL influence of the chemical stages, F is the Faraday number, φ_0 is the zero-charge-related DEL potential slope, R is the universal gas constant, and T is the absolute temperature.

It is important to mention that coprine, as an amino acid, affects DEL during the chemical stages, whereas psilocin only affects its forming radical cation during assisted electropolymerization. This effect may be responsible for the oscillatory behavior observed beyond the detection limit. Nevertheless, this influence does not impede the effective use of this system, as shown below.

3. Results and Discussion

We describe the behavior of the system with psilocin and coprine electrochemical determination over CoO(OH) by means of linear stability theory and expose the equation-set steady-state Jacobian elements as:

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

In which:

$$a_{11} = \frac{2}{\delta}\left(-\frac{\Psi}{\delta} - k_{p1}(1-c)^4 - k_{p2}(1-c)^4 - xk_p\psi^{x-1}(1-c)^y\kappa^z \exp(-\alpha\psi)\exp(-\beta\kappa) + \alpha k_p\psi^x(1-c)^y\kappa^z \exp(-\alpha\psi)\exp(-\beta\kappa)\right) \quad (9)$$

$$a_{12} = \frac{2}{\delta}\left(-zk_p\psi^x(1-c)^y\kappa^{z-1}\exp(-\alpha\psi)\exp(-\beta\kappa)\right) + \beta k_p\psi^x(1-c)^y\kappa^z \exp(-\alpha\psi)\exp(-\beta\kappa) \quad (10)$$

$$a_{13} = \frac{2}{\delta}\left(4k_{p1}\psi(1-c)^3 + 4k_{p2}\psi(1-c)^3 + yk_p\psi^x(1-c)^{y-1}\kappa^z \exp(-\alpha\psi)\exp(-\beta\kappa)\right) \quad (11)$$

$$a_{21} = \frac{2}{\delta}\left(-k_p(1-c)^4 - xk_p\psi^{x-1}(1-c)^y\kappa^z \exp(-\alpha\psi)\exp(-\beta\kappa)\right) \quad (12)$$

$$a_{22} = \frac{2}{\delta}\left(-\frac{K}{\delta} - k_{q1}(1-c)^4\exp(-\beta\kappa) + \beta k_{q1}\kappa(1-c)^4\exp(-\beta\kappa) - k_{q2}(1-c)^4\exp(-\beta\kappa) + \beta k_{q2}\kappa(1-c)^4\exp(-\beta\kappa) - \left(zk_p\psi^x(1-c)^y\kappa^{z-1}\exp(-\alpha\psi)\exp(-\beta\kappa)\right) + \beta k_p\psi^x(1-c)^y\kappa^z\exp(-\alpha\psi)\exp(-\beta\kappa)\right) \quad (13)$$

$$a_{23} = \frac{2}{\delta}\left(4k_{q1}\kappa(1-c)^3\exp(-\beta\kappa) + 4k_{q2}\kappa(1-c)^3\exp(-\beta\kappa) + yk_p\psi^x(1-c)^{y-1}\kappa^z\exp(-\alpha\psi)\exp(-\beta\kappa)\right) \quad (14)$$

$$a_{31} = \frac{1}{C}\left(k_{p1}(1-c)^4 + k_{p2}(1-c)^4 - xk_p\psi^{x-1}(1-c)^y\kappa^z\exp(-\alpha\psi)\exp(-\beta\kappa) - \alpha k_p\psi^x(1-c)^y\kappa^z\exp(-\alpha\psi)\exp(-\beta\kappa)\right) \quad (15)$$

$$a_{32} = \frac{1}{C}\left(k_{p1}(1-c)^4 + k_{p2}(1-c)^4 - xk_p\psi^{x-1}(1-c)^y\kappa^z\exp(-\alpha\psi)\exp(-\beta\kappa) - \alpha k_p\psi^x(1-c)^y\kappa^z\exp(-\alpha\psi)\exp(-\beta\kappa)\right) \quad (16)$$

$$a_{33} = \frac{1}{C}\left(-k_{p1}\psi(1-c)^3 - 4k_{p2}\psi(1-c)^3 - yk_p\psi^x(1-c)^{y-1}\kappa^z\exp(-\alpha\psi)\exp(-\beta\kappa) - 4k_{q1}\kappa(1-c)^3\exp(-\beta\kappa) - 4k_{q2}\kappa(1-c)^3\exp(-\beta\kappa) - k_O\exp\frac{F\varphi_0}{RT} + jk_O c\exp\frac{F\varphi_0}{RT}\right) \quad (17)$$

Avoiding the cumbersome expression during the determinant analysis, we introduce new variables and rewrite the determinant as (18):

$$\text{Det J} = \frac{4}{\delta^2 C}\begin{vmatrix} -\lambda - \Xi - \Sigma & -T & \Lambda + \Pi \\ -\Sigma & -\xi - P - T & \Phi + \Pi \\ \Xi + \Sigma & P + T & -\Lambda - \Phi - \Pi - \Omega \end{vmatrix} \quad (18)$$

Considering that:

$$-Det\,J\begin{cases} > 0, for\ steady-state\ stability \\ = 0\ monotonic\ instability \end{cases} \quad (19)$$

Using the determinant properties, we simplify it to (20):

$$\frac{4}{\delta^2 C}\begin{vmatrix} -\lambda - \Xi - \Sigma & -T & \Lambda + \Pi \\ -\Sigma & -\xi - P - T & \Phi + \Pi \\ \Xi & -\xi & -\Lambda - \Omega \end{vmatrix} \quad (20)$$

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 (21):

$$\Xi(T\Phi + T\Pi - \xi\Lambda - P\Lambda - T\Lambda - \xi\Pi - P\Pi - T\Pi) + \xi(-\Lambda\Sigma + \Phi\lambda + \Phi\Xi + \Phi\Sigma + \Pi\lambda + \Phi\Xi) + (\Lambda + \Omega)(\lambda\xi + \lambda P + \lambda T + \Xi\xi + \Xi P + \Xi T + \Sigma\xi + \Sigma P + \Sigma T) > 0 \quad (21)$$

If –Det J>0, the Routh-Hurwitz stability criterion is valid, and the steady-state is thereby stable, providing an efficient electrochemical detection of both toxins. Moreover, although the stability region is narrower than in similar systems [45 – 49], let's use this system as an electroanalytical for sensing purposes due to the efficient analytical signal interpretation. The electroanalytical process is kinetically controlled.

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, depicts the margin between stable and unstable states, and corresponds to 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.

These elements are parent to those observed in similar systems [45 – 49] and correspond to DEL influences of both chemical and electrochemical stages. It is important to mention that psilocin only influences the DEL electrophysical parameters, leading to the oscillatory behavior, during the assisted electropolymerization, whereas coprine does influence it, the reason why there are various coprine-related Jacobian main diagonal positive addendums: $\beta k_{q1}\kappa(1-c)^4\exp(-\beta\kappa) > 0$, $\beta k_{q2}\kappa(1-c)^4\exp(-\beta\kappa) > 0$ $\beta k_p\psi^x(1-c)^y\kappa^z\exp(-\alpha\psi)\exp(-\beta\kappa) > 0$, if β>0 with only one for psilocin: $\alpha k_p\psi^x(1-c)^y\kappa^z\exp(-\alpha\psi)\exp(-\beta\kappa) > 0$, if α>0. All of these elements, like also the DEL electrochemical impact-related addendum $jk_Oc\exp\frac{F\varphi_0}{RT} > 0$, positive if j>0, will define the dependence of the oscillation frequency and amplitude of the background electrolyte composition, which has been proved experimentally and theoretically for similar systems [42 – 49].

A similar model may be used to detect coprin with other phenolic mycotoxins, like involutin and orellanin. Nevertheless, as both involution and orellanine contain the hydroquinonic moiety, their polymerization potential will be equal to or lower than that of psilocin, providing fewer energy losses.

The same process may be used to synthesize an ecologically safe conducting polymer composite, which is convenient from the point of view of green chemistry and circular economy.

4. Conclusions

From the theoretical description for the electrochemical determination of two mycotoxins, psilocin and coprine, on a CoO(OH)-modified anode, it has been possible to conclude it may be an excellent modifier for the removal and quantification of both mycotoxins. The electrochemical removal is kinetically controlled. The oscillatory behavior in this system may be caused by DEL influence in both electrochemical and chemical stages. Nevertheless, coprine oxidation has more impact on DEL electrophysical properties than psilocin micromolecular oxidation. The same system has electroanalytical and electrosynthetical values in the polymerization conditions.

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Conflicts of Interest

The authors declare no conflict of interest.

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