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The Theoretical Description for Ibotenic Acid and Muscimol Electrochemical Determination in Mushroom Pulp and Mushroom-based Alcoholic Beverages on Nano-CuS Composite with Conducting Polymer

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Abstract: In this article, the possibility for ibotenic acid and muscimol electrochemical determination on the electrode, modified by nano-CuS/conducting polymer composite, is theoretically described. The analytical signal is based on the reaction of the copper (III) sulfohydroxide with both of the analytes, and the reaction is realized by two parallel scenarios. The analysis of the correspondent model confirms the possibility of the oscillatory behavior caused by DEL influence of the electrochemical stage and chemical stage involving ibotenic acid. The chemical stages involving muscimol do not participate in the oscillatory behavior. The stability analysis confirms the easy realization of the linear dependence between the electrochemical parameter and concentration and, therefore, the facility in analytical signal interpretation.

Keywords: *Amanita muscaria*; ibotenic acid; muscimol; electrochemical sensor; conducting polymer; copper (II) sulfide; stable steady-state.

1. Introduction

Fly agaric, also known as *Amanita muscaria* [1–4], is one of the most widespread poisonous mushrooms in subtropical and moderate countries. It also becomes widespread in subequatorial and equatorial countries such as Panama, Colombia, and Venezuela [4]. Its poison is highly toxic, causing grave central nervous system disorders [5]. Nevertheless, it is still used in some alcoholic drinks in Russia, Finland, and Norway [6,7]. In Middle Ages, the vikings ate fly agaric into mushroom broth in order to enhance the spirit.

The fly agaric toxicity is generally caused by the presence of ibotenic acid [8–11] and their derivatives – muscimol and muscazone in the mushroom pulp (Figure 1).

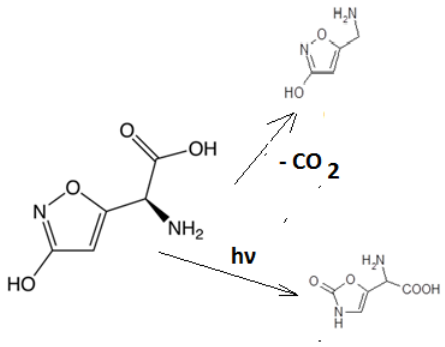

Figure 1. Ibotenic acid, muscimol and muscazone.

Being muscimol is the most dangerous among the three [12–14]. Moreover, being a biogenic amine, it stimulates the nervous system, causing the false happiness feeling and giving the reforce to the action of two other toxins. As for muscazone, a product of ibotenic acid photoisomerization, its action is less intensive but longer and somehow more intense (including partial blindness due to the methanol formation in its metabolism).

Considering the structure of all three compounds, we may conclude that cathodic reduction is the best strategy to detect all of the compounds electrochemically. Nevertheless, anodic oxidation may also be used. Muscimol will be oxidized at lower potentials than ibotenic acid and muscazone. For this purpose, chemically modified electrodes containing strong oxidants may be used [15–18].

Therefore, the composite material of CuS nanoparticles with a conducting polymer, in which the polymer is a mediator and stabilizer, and the CuS is the starting material for *in situ* anodic synthesis of trivalent copper as copper (III) sulfohydroxide by reaction (1):

$$CuS + OH^- - e^- \rightarrow CuS(OH) \qquad (1)$$

would be an interesting option. Such hybrid materials have already been used in electroanalytic[19–22] in either cathodic or anodic processes. Nevertheless, the electrochemical instabilities, typical for similar electroanalytical and electro synthetical processes [22–26], may impede the easy interpretation of the analytical signal or even lead to the electrochemical equipment breaking down. On the other hand, the electrochemical instabilities may reveal the presence of a substance or a specific reaction involving the electroactive compound.

The analysis of the theoretical and experimental data shows that the electrochemical instabilities in non-autocatalytic electrochemical systems are caused by the influences of

chemical and electrochemical processes on the double electric layer (DEL) ionic force, capacity, and conductivity, like also the electrode surface material impedance.

Therefore, the theoretical *a priori* investigation of the system´s behavior, capable of detecting the conditions of the most effective sensing interpretation, like also the conditions of the electrochemical instabilities, is an important stage for the development of new electrochemical sensors, especially for new compounds and involving new electrode modifiers. Also, this investigation, involving the development and analysis of a mathematical model, permits to compare the similar electroanalytical systems. Therefore, the aim of this work is to investigate, from the mechanistic theoretical point of view, the possibility of ibotenic acid and muscimol electrochemical determination on nano-CuS/conducting polymer composite. It involves the mechanism suggestion and mathematical model analysis and interpretation, including the comparison of this system's behavior with that of similar ones [27,28].

2. Materials and Methods

Copper (III) sulfohydroxide, a product of copper (II) sulfide *in situ* electrooxidation by reaction (1), is an aggressive oxidant, oxidizing both ibotenic acid and muscimol by two parallel scenarios:

- yielding an N-oxide;
- oxidizing amino group to imine.

Considering the absence of highly accepting carboxyl in muscimol, its oxidation will be given more easily and on lower anodic potential values (Figure 2).

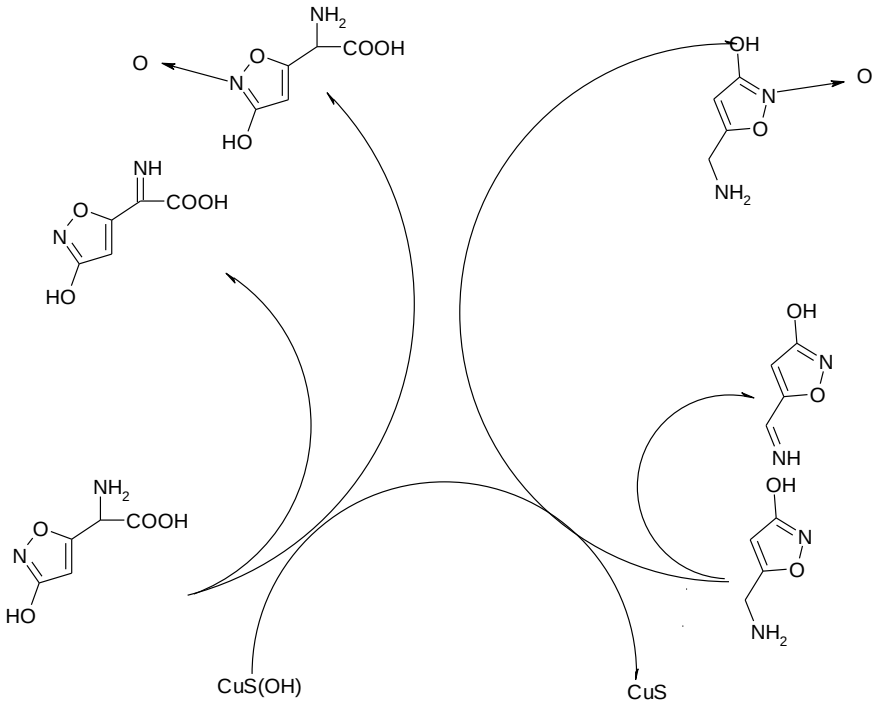

Figure 2. The scheme for the electroanalytical process.

Taking this into account and taking some assumptions [27,28], we describe the behavior of this system by a trivariant balance differential equation-set (2):

$$\begin{cases} \frac{dm_1}{dt} = \frac{2}{\delta}\left(\frac{M_1}{\delta}(m_{10} - m_1) - r_{11} - r_{12}\right) \\ \frac{dm_2}{dt} = \frac{2}{\delta}\left(\frac{M_2}{\delta}(m_{20} - m_2) - r_{21} - r_{22}\right) \\ \frac{dc}{dt} = \frac{1}{C}(r_{11} + r_{12} + r_{21} + r_{22} - r_1) \end{cases} \quad (2)$$

Herein, m_1 and m_2 are correspondently ibotenic acid and muscimol concentrations in the pre-surface layer, δ is the diffusion thickness, c is the copper sulfide matrix coverage degree, M_1 and M_2 are ibotenic acids and muscimol diffusion coefficients, m_{10} and m_{20} stand for the bulk concentration of each of the analytes, C is the copper sulfide maximal matrix concentration, and the parameters r stand for the correspondent reaction rates, calculated as:

$$r_{11} = k_{11}m_1(1-c)^2\exp(-ac) \quad (3)$$
$$r_{12} = k_{12}m_1(1-c)^2\exp(-ac) \quad (4)$$
$$r_{21} = k_{21}m_2(1-c)^2 \quad (5)$$
$$r_{22} = k_{22}m_2(1-c)^2 \quad (6)$$
$$r_1 = k_1 c\exp\left(\frac{F\varphi_0}{RT}\right) \quad (7)$$

Herein the parameters k correspond to the reaction rate constants, a is the parameter describing the influence of ionic forms transformations on DEL electrophysical properties, $F = N_A e$ is the Faraday constant, φ_0 stands for zero-charge-related potential slope in DEL, R is the universal gas constant, and T is the absolute temperature.

In this system, the oscillatory behavior on the chemical stage may be caused by the DEL impact of only one of two analytes analyte. Therefore, the electroanalytical process will be more stable and efficient than for ibotenic acid and muscazone, as shown below.

3. Results and Discussion

We investigate the behavior of the system with ibotenic acid and muscimol determination over CuS/conducting polymer composite by means of the linear stability theory, describing the steady-state Jacobian members as (8):

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

In which:

$$a_{11} = \frac{2}{\delta}\left(-\frac{M_1}{\delta} - k_{11}(1-c)^2\exp(-ac) + ak_{11}m_1(1-c)^2\exp(-ac) - k_{12}(1-c)^2\exp(-ac) + ak_{12}m_1(1-c)^2\exp(-ac)\right) \quad (9)$$

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

$$a_{13} = \frac{2}{\delta}(2k_{12}m_1(1-c)\exp(-ac) + 2k_{12}m_1(1-c)\exp(-ac)) \quad (11)$$

$$a_{21} = 0 \quad (12)$$

$$a_{22} = \frac{2}{\delta}\left(-\frac{M_2}{\delta} - k_{21}(1-c)^2 - k_{22}(1-c)^2\right) \quad (13)$$

$$a_{23} = \frac{2}{\delta}(2k_{21}m_2(1-c) + 2k_{22}m_2(1-c)) \quad (14)$$

$$a_{31} = \frac{1}{C}(k_{11}(1-c)^2\exp(-ac) - ak_{11}m_1(1-c)^2\exp(-ac) + k_{12}(1-c)^2\exp(-ac) - k_{12}m_1(1-c)^2\exp(-ac)) \quad (15)$$

$$a_{32} = \frac{1}{C}(k_{21}(1-c)^2 + k_{22}(1-c)^2) \quad (16)$$

$$a_{33} = \frac{1}{C}\left(-2k_{12}m_1(1-c)\exp(-ac) - 2k_{12}m_1(1-c)\exp(-ac) - 2k_{21}m_2(1-c) - 2k_{22}m_2(1-c) - k_1\exp\left(\frac{nF\varphi_0}{RT}\right) + jk_1c\exp\left(\frac{nF\varphi_0}{RT}\right)\right) \quad (17)$$

Taking into account the Jacobian main diagonal elements (6), (10), and (14), it is possible to conclude that, as this diagonal contains the elements capable of being positive, the positive callback may be characteristic of this electrochemical process. Therefore, the Hopf bifurcation, correspondent to the oscillatory behavior, becomes possible.

In this system, the oscillatory behavior is caused by the (co)action of two factors, such as in [27 - 28]. Besides the factor of the influence of the electrochemical stage on the surface and pre-surface layer electrophysical properties (ionic force, conductivity, and capacitance), described by the positivity of the element $jk_1c\exp\left(\frac{nF\varphi_0}{RT}\right) > 0$, if j>0, the oscillatory behavior will also be caused by ionic form transformation on the chemical stage, described by the positivity of the elements $ak_{11}m_1(1-c)^2\exp(-ac)$ and $ak_{12}m_1(1-c)^2\exp(-ac)$, involving the reactions of the ibotenate-ion.

It´s important to mention that in neutral and mildly alkaline mediums, the second analyte, muscimol, isn´t ionized. Therefore, in this system, the oscillatory behavior will be defined by the chemical transformation of only one analyte – ibotenic acid in the form of ibotenate-ion.

As for the steady-state stability, its topological region is more narrow than in the simplest case but wider than in the case of two ionizing analytes. Applying the Routh-Hurwitz criterion, we rewrite the Jacobian matrix as (18):

$$\frac{4}{\delta^2 C}\begin{vmatrix} -\kappa-\Xi & 0 & \Sigma \\ 0 & -\lambda-\Lambda & T \\ \Xi & \Lambda & -\Sigma-T-\Omega \end{vmatrix} \quad (18)$$

Introducing new variables in order to avoid cumbersome expressions.

Opening the straight brackets and resolving the inequation Det J<0, salient from the criterion, and changing the signs to the opposite, we obtain the necessary requisite for steady-state stability (19):

$$\kappa(\lambda\Sigma + \lambda T + \lambda\Omega + \Lambda\Sigma + \Lambda\Omega) + \Xi(\lambda T + \lambda\Omega + \Lambda\Omega) > 0 \quad (19)$$

This inequity describes an efficient electroanalytical process, which is both diffusion and kinetically controlled. Moreover, taking into account the absence of side reactions, compromising the analyte and modifier stability, it will depend on linear dependence between the analytes concentration and electrochemical parameter (current and(or) potential).

As for the detection limit, it will correspond to the monotonic instability, depicting the margin between the stable steady-state and unstable state. Its condition may be exposed as Det J=0 or (19):

$$\kappa(\lambda\Sigma + \lambda T + \lambda\Omega + \Lambda\Sigma + \Lambda\Omega) + \Xi(\lambda T + \lambda\Omega + \Lambda\Omega) = 0 \quad (20)$$

A similar scenario will be given in the case of muscimol and muscazone electrochemical determination. Nevertheless, it will be simplified for muscazone due to only one oxidation possibility against two for muscimol and ibotenic acid. This behavior is

characteristic till a certain pH value, at which the amido ester group in muscazone is hydrolyzed. The behavior of this system will be described in one of our next works.

4. Conclusions

From the analysis of the system with ibotenic acid and muscimol determination on CuS/Conducting Polymer composite, it is possible to conclude that the mentioned composite is an efficient electrode modifier for ibotenic acid and muscimol determination. The electroanalytical process is both diffusion and kinetically controlled, being less stable than for the simplest case but more stable than for two ionizing analytes. As for the oscillatory behavior, its probability is high but lower than for the case of ibotenic acid and muscazone.

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Conflicts of Interest

The authors declare no conflict of interest.

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