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The Theoretical Description for the CoO(OH)-Assisted Electrochemical Determination of Tobacco Nitrosocompounds

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Abstract: The possibility for the tobacco nitrosocompounds electrochemical determination on cobalt (III) oxyhydroxide is evaluated theoretically. If the redox pair $CoO(OH)/CoO_2$ is used, two possibilities of the electrochemical reaction involve the nitroso group oxidation or N-oxidation. As for N-nitrosoanabatine, another oxidation reaction involving the isolated double bond is possible. All of the reactions are efficient from an electroanalytical point of view, providing efficient analytical signal interpretation. The sensor may be used to measure either global nitrosyl compounds concentration or the concentration of the specific organic nitrosyls from tobacco, like N-nitrosonornicotine, N-nitrosoanabasine, and N-nitrosoanabatine. As for the oscillatory behavior, its probability becomes low in neutral media and augments in an alkaline medium and even more in an acidic medium, due to the interaction of ionic forms with a double electric layer.

Keywords: tobacco-specific nitrosamines; N-nitrosocompounds; conducting polymers; cobalt (III) oxyhydroxide; electrochemical sensors; electrochemical oscillations; stable steady-state.

1. Introduction

Organic nitrosocompounds [1–4] are among the most toxic mutagens and teratogens in the world. They may have a natural, semisynthetic, or synthetic origin. Either C- or especially N-nitrosyls are considered aggressive pollutants for the environment.

Generally, the nitroso- and nitrosamino compounds are obtained in organisms by nitrate and nitrite metabolism in acidic media, yielding a strong electrophile, the nitrosyl cation:

$$NO_3^- + 4H^+ + 2e^- \rightarrow NO^+ + 2H_2O \quad (1)$$

$$NO_2^- + 2H^+ \rightarrow NO^+ + H_2O \quad (2)$$

Which thereby participates in the electrophilic reactions in the organism, yielding toxic derivatives. The same process also occurs in plants, including tobacco, where the alkaloid secondary amine reacts with the nitrosyl cation, yielding the specific tobacco nitrosamines, the most characteristic of which are described in Figure 1.

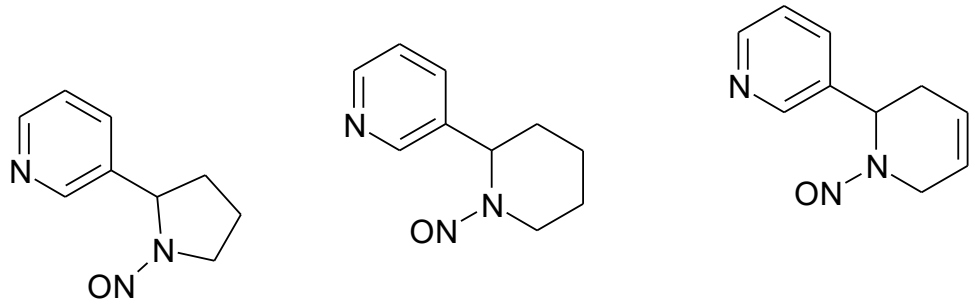

Figure 1. N-nitrosonornicotine, N-nitrosoanabasine, and N-nitrosoanabatine.

All of them appear either in natural conditions or during the tobacco curing and processing [5–9]. They are highly toxic not only for those who actively smoke but also for those who smoke passively, which is why their determination is highly relevant [10–14], and the electrochemical sensing may be an interesting response to this question.

Being an intermediary compound in the nitrogroup electrochemical reduction to an aminogroup, the nitrosyl moiety is electrochemically active [15–18], thereby being capable of participating in either anodic or cathodic processes. Moreover, the pyridinic ring is also electrochemically active, as it possesses a nitrogen atom and aromatic fragments [19–21].

Therefore, as a semiconducting electroactive material, the cobalt(III) oxyhydroxide may be used as an electrode modifier for tobacco nitrosamines electrochemical determination [22–26]. Moreover, both CoO/CoO(OH) and CoO(OH)/CoO_2 redox pairs may be used. Nevertheless, the electrochemical sensitivity depends highly on tobacco nitrosamines or their oxidation products ionization in a double electric layer (DEL), which may highly impact the analytical signal interpretation and the electroanalytical process stability [27–32]. This impact may only be detailed by an *a priori* theoretical investigation involving the mechanism suggestion and the development and analysis of the corresponding mathematical model [33–35].

So, the goal of our work is to evaluate, from the theoretical point of view, the behavior of the system with specific tobacco nitrosamines determination, assisted by CoO(OH)-modified electrode by use of CoO(OH)/CoO_2 redox pair. The corresponding reaction mechanism is suggested, and the corresponding mathematical model is developed

and analyzed from a stability point of view in order to infer the condition of the most efficient sensing and the realization of the instabilities. Also, the behavior of this system will be compared to that of similar ones [33–35].

2. Materials and Methods

In the case of the use of the $CoO(OH)/CoO_2$ redox pair, cobalt (III) oxyhydroxide is oxidized, yielding cobalt dioxide as (3):

$$CoO(OH) + OH^- - e^- \rightarrow CoO_2 + H_2O \quad (3)$$

Being a strong oxidant, cobalt (IV) oxide will thereby oxidize TSNA by either the nitroso group or the pyridinic nitrogen atom. Yet for the anabatine molecule, the third oxidation scenario involves the isolated double bond of the tetrahydropyridine moiety. All of the oxidation scenarios are joined in Figure 2.

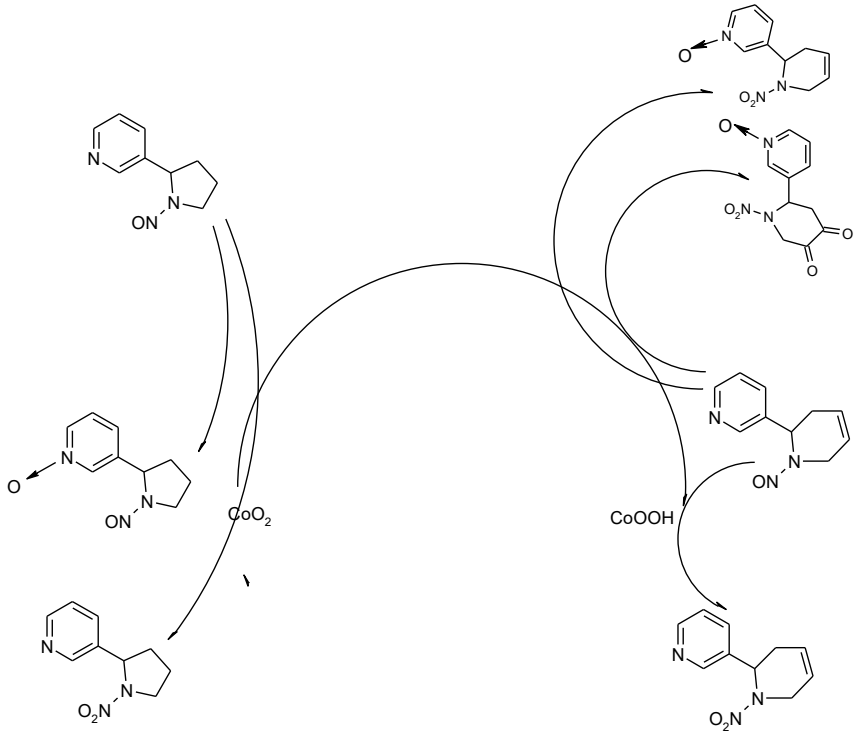

Figure 2. The scheme of the electroanalytic process of N-nitrosonornicotine and N-nitrosoanabatine.

Taking into account the behavior of both analytes and accepting some assumptions [33–35], we describe the behavior of this system by a trivariate equation set (4):

$$\begin{cases} \frac{dn}{dt} = \frac{2}{\delta}\left(\frac{\Delta}{\delta}(n_0 - n) - r_{11} - r_{12}\right) \\ \frac{da}{dt} = \frac{2}{\delta}\left(\frac{A}{\delta}(a_0 - a) - r_{21} - r_{22} - r_w\right) \\ \frac{dc}{dt} = \frac{1}{C}(r_{11} + r_{12} + r_{21} + r_{22} + r_w - r_3) \end{cases} \quad (4)$$

In which n and a are nitrosonornicotine and nitrosoanabatine concentrations in the pre-surface layer, n_0, and a_0 are the correspondent analytes bulk concentrations, Δ and A are the correspondent diffusion coefficient, c is the cobalt (III) oxyhydroxide surface coverage degree, C is its maximal surface concentration, and the parameters r are the correspondent reaction rates, calculated as:

$$r_{11} = k_{11} n (1 - c)^2 \quad (5)$$

$$r_{12} = k_{11} n (1 - c)^2 \exp(-\lambda n) \quad (6)$$

$$r_{21} = k_{21} a (1 - c)^2 \quad (7)$$

$$r_{22} = k_{21}a(1-c)^2 \exp(-\mu a) \quad (8)$$

$$r_w = k_w a(1-c)^2 \quad (9)$$

$$r_3 = k_3 c \exp\left(\frac{F\varphi_0}{RT}\right) \quad (10)$$

In which the parameters k are the corresponding reaction rate constants, λ and μ stand for the parameters relating the DEL ionic force and related electrophysical properties with the N-acynitroform formation during the oxidation, F is the Faraday number, φ_0 stands for zero-charge-related potential slope, R is the universal gas constant, and T is the absolute temperature.

In basic media, for which this model is valid, the oscillatory behavior is more probable than for the neutral media, due to the formation of pseudoacid ion as oxidation product (see the similar systems for perilartine electrochemical determination [33–35]), but less probable than for acidic media, due to the total ionization of the pyridinic ring in both of the compounds. Either way, the CoO(OH)-assisted electrooxidation may be proven efficient, as shown below.

3. Results and Discussion

We investigate the behavior of the system of TSNA electrooxidation in the CoO(OH)/CoO_2-modified anode by analyzing the equation set (4) by means of linear stability theory and expose the Jacobian steady-state members as (11):

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

In which:

$$a_{11} = \frac{2}{\delta}\left(-\frac{\Delta}{\delta} - k_{11}(1-c)^2 - k_{11}(1-c)^2\exp(-\lambda n) + \lambda k_{11}(1-c)^2\exp(-\lambda n)\right) \quad (12)$$

$$a_{12} = 0 \quad (13)$$

$$a_{13} = \frac{2}{\delta}(2k_{11}n(1-c) + 2k_{11}n(1-c)\exp(-\lambda n)) \quad (14)$$

$$a_{21} = 0 \quad (15)$$

$$a_{22} = \frac{2}{\delta}\left(-\frac{\Delta}{\delta} - k_{21}(1-c)^2 - k_{21}\exp(-\mu a) + \mu k_{21}\exp(-\mu a) - k_w(1-c)^2\right) \quad (16)$$

$$a_{23} = \frac{2}{\delta}(2k_{21}a(1-c) + 2k_{21}a(1-c)\exp(-\mu a) + 2k_w a(1-c)) \quad (17)$$

$$a_{31} = \frac{1}{C}(k_{11}(1-c)^2 + k_{11}(1-c)^2\exp(-\lambda n) - \lambda k_{11}(1-c)^2\exp(-\lambda n)) \quad (18)$$

$$a_{32} = \frac{1}{C}(k_{21}(1-c)^2 + k_{21}\exp(-\mu a) - \mu k_{21}\exp(-\mu a) + k_w(1-c)^2) \quad (19)$$

$$a_{33} = \frac{1}{C}\left(-2k_{11}n(1-c) - 2k_{11}n(1-c)\exp(-\lambda n) - 2k_{21}a(1-c) - 2k_{21}a(1-c)\exp(-\mu a) - 2k_w a(1-c) - k_3\exp\left(\frac{F\varphi_0}{RT}\right) + jk_3 c\exp\left(\frac{F\varphi_0}{RT}\right)\right) \quad (20)$$

Taking into account the main diagonal elements (12), (16), and (20), important for the positive callback, described by the positive addendums in these elements, we may see that it contains three elements, capable of being positive. Those elements are $\lambda k_{11}(1-c)^2\exp(-\lambda n) > 0$, if $\lambda > 0$, $\mu k_{21}\exp(-\mu a) > 0$, if $\mu > 0$, describing the positive callback during the DEL influences of the chemical stages and $jk_3 c\exp\left(\frac{F\varphi_0}{RT}\right) > 0$ if j>0, describing the positive callback during the DEL influences of the electrochemical stage. This callback is manifested in the *oscillatory behavior.*

The oscillation amplitude and frequency will depend on the background electrolyte composition, as in [29–32]. Moreover, the oscillatory behavior probability will be directly

dependent on pH. In neutral pH, in which the nitrosyl oxidation product becomes more ionized, the parameters λ and μ are set equal to zero; thus, the expressions $\mu k_{21}\exp(-\mu a) = \lambda k_{11}(1-c)^2\exp(-\lambda n) = 0$, and the exponential expressions $\exp(-\mu a) = \exp(-\lambda n) = 1$, excluding two of three potentially positive elements, leaving only one, related to the electrochemical stage, such as in [33–35].

We simplify the *steady-state stability analysis,* reexposing the Jacobian determinant as (21):

$$\begin{vmatrix} -\kappa-\Xi & 0 & P \\ 0 & -\alpha-\Lambda & T \\ \Xi & \Lambda & -P-T-\Omega \end{vmatrix} \quad (21)$$

Opening the brackets and applying the Det J<0 conditions, inferred from the criterion, we can prove the presence of an efficient diffusion-controlled electroanalytical system, in which the steady-state stability is easy to obtain and maintain, and the steady-state stability requisite (22):

$$\kappa(\alpha P+\alpha T+\alpha\Omega+\Lambda P+\Lambda\Omega)+\Xi(\alpha T+\alpha\Omega+\Lambda\Omega)>0 \quad (22)$$

The requisite (22) is satisfied in a vast topological parameter region, being thereby correspondent to the linear dependence between the electrochemical parameter and the TSNA concentrations, as neither analytes nor the modifier undergoes the side reactions, capable of affecting their stability unless foreseen by the electroanalytical process. The requisite (22) is more likely to be satisfied in neutral than in the alkaline and even more than in an acidic medium.

As for the *detection limit,* described by the monotonic instability, it delimits the margin between the stable steady-states and unstable states. Its realization condition will be thereby given as (23):

$$\kappa(\alpha P+\alpha T+\alpha\Omega+\Lambda P+\Lambda\Omega)+\Xi(\alpha T+\alpha\Omega+\Lambda\Omega)=0 \quad (23)$$

In the *acidic media,* the equation set (4) will remain intact, but the rate expressions will be rewritten as:

$$r_{11} = k_{11}n(1-c)^2\exp(-\lambda n) \quad (24)$$
$$r_{12} = k_{11}n(1-c)^2\exp(-\lambda n) \quad (25)$$
$$r_{21} = k_{21}a(1-c)^2\exp(-\mu a) \quad (26)$$
$$r_{22} = k_{21}a(1-c)^2\exp(-\mu a) \quad (27)$$
$$r_{w} = k_{w}a(1-c)^2\exp(-\mu a) \quad (28)$$

The system will thereby become similar to that described in [35] and even less stable than in the alkaline or neutral medium. Therefore, the neutral or neutralized (in the case of the acidic or basic electrode modifier component, if composite material is used) medium is the most suitable for CoO(OH)-assisted TSNA electrochemical determination; the neutral medium is preferred.

4. Conclusions

From the analysis of the system with CoO(OH)-assisted TSNA electrochemical determination, it is possible to conclude that this process is a highly efficient diffusion-controlled process in which the linear dependence between the electrochemical parameter and concentration is easily obtained and maintained. The easy interpretation of the analytical signal is given mostly in a neutral medium, in which neither the analytes nor their oxidation products are not ionized and, thereby, their ionic forms do not influence the DEL ionic force and related electrophysical properties, making it less probable the oscillatory behavior.

Author Contributions

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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