

*Full Paper*

## **The Possibility of the Use of CoO(OH) as an Electrode Modifier for Hydrazine Detection and Its Mathematical Evaluation**

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**Abstract-** The possibility of the use of cobalt(III) oxyhydroxyde as an electrode modifier for hydrazine detection has been investigated. The mechanism for the electrochemical oxidation of hydrazine over CoO(OH) has been suggested, and the correspondent mathematical model was developed and analyzed. The optimal pH for electroanalytical efficiency of the system is between 7 and 12. Strongly alkaline media promote the direct hydrazine oxidation and may destroy the modifying material. The oscillatory and monotonic instabilities' possibility has been also investigated. These instabilities may be caused by surface and electrochemical factors.

**Keywords-** Hydrazine, Electroanalysis, Cobalt(III) oxyhydroxide, Electrooxydation, Stable steady-state

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## 1. INTRODUCTION

The chemically modified electrodes (CME) are one of modern and convenient tools, used for electroanalysis of different compounds [1-10]. Their advantages are the affinity to the analyte, overvoltage diminishing, efficiency and rapidity in analysis.

On the other hand, hydrazine is the simplest diamine, widely used in organic and pharmaceutical synthesis [11-14], and also as a fuel in fuel cells [15]. It is highly toxic compound, causing kidney and liver damages, capable to be absorbed by a human organism by different ways [16] (inclusively by skin), and to be formed in it by metabolism of different pharmaceutical compounds, like procarbazine [17]. Thus, the search of the hydrazine detection method capable to determinate it by rapid, efficient and precise way is, without a doubt, an actual problem [18] and the use of electrochemical analytical techniques is one of its possible solutions [19-20].

Many electrode modifiers have been developed for hydrazine detection, like metaloporphyrines [21-22], carbon paste and glassy carbon materials [23-25], titanium dioxide [26], or conducting polymers [27-30], curcumin[31], phthalocyanines [32], hematoxylin [33], phenoloaminoacids [34-35], metal oxide nanoparticles [36] and ferrocene derivatives [37], and cobalt (III) oxyhydroxide (CoO(OH)), p-type semiconductor, seen as an alternative to TiO<sub>2</sub>, and yet used as an electrode modifier for oxalate [37] and phenolic compounds [38] may be a good alternative.

Although, by far, there are no literature, concerning the use of CoO(OH) as a catalyst concretely for hydrazine electrooxidation, it is possible to predict this role for it, because the compounds, yet used for its oxidation [37-38], have the similar oxidation potential.

The principal difficulties of developing of new electroanalytical techniques are:

- Indecision in the most probable mechanism for electrochemical function of the sensor;
- Different stability of modifiers in different solutions;
- Possibility of the appearance of electrochemical instabilities, characteristic for the oxidation of different organic compounds, and for the synthesis of CoO(OH) [39-47].

These difficulties may be solved, when the suggestion of an electroanalytical technique has a rigid theoretical base, which may be given by the development and analysis of a mathematical model, capable to describe adequately its behavior. By this mean, it is also possible to compare the mechanism of similar electrochemical systems and evaluate the possibility of the use of certain material in electrochemical detection of a substance.

So, the aim of this work is to evaluate the possibility of the use of CoO(OH) in hydrazine electrooxidation. This aim includes the realization of next specific objectives:

- Development of the mathematical model, describing the system;

- The analysis of the model, by means of linear stability theory and bifurcation analysis;
- The interpretation of the modeling results, permitting to obtain the optimal analysis conditions.
- Comparison of the system's behavior with the behavior of similar systems [48-56]

## 2. SYSTEM AND ITS MODELING

Cobalt (II) oxide film is obtained chemically, or electrochemically, parting from a soluble cobalt (II) salt. This salt, like also hydrazine for probes, may be acquired from Sigma Aldrich or Merck in analytically pure concentrations.

In the presence of oxy-hydroxides in alkaline medium, hydrazine is oxidized, like presented in [36] and [48]:



In strongly alkaline medium,  $\text{CoO}(\text{OH})$  is dissolved, to form a complex



So, to describe the electrooxidation of hydrazine in the presence of  $\text{CoO}(\text{OH})$ , we introduce three variables:

$c$  – the hydrazine concentration in the pre-surface layer;

$\theta$  – the  $\text{CoO}(\text{OH})$  surface concentration;

$a$  – the concentration of alkali in the pre-surface layer

To simplify the modeling, we suppose that the reactor is intensively stirred, so we can neglect the convection flow. Also we assume that the background electrolyte is in excess, so we can neglect the migration flow. The diffusion layer is supposed to be of a constant thickness, equal to  $\delta$ , and the concentration profile in it is supposed to be linear. It's also supposed that at the beginning of the reaction  $\text{CoO}$  covers the entire electrode surface.

The hydrazine enters the pre-surface layer by diffusion and reacts with  $\text{CoO}(\text{OH})$ . It may also enter to the non-promoted oxidation, described in [36], [48]. Thus, its balance equation will be rewritten as:

$$\frac{dc}{dt} = \frac{2}{\delta} \left( \frac{\Delta}{\delta} (c_0 - c) - r_2 - r_a \right) \quad (1)$$

In which  $r_2$  is the rate of the reaction (2),  $r_a$  is the autopromoted oxidation rate, and  $\Delta$  is the diffusion coefficient and  $c_0$ , the hydrazine concentration in bulk.

The cobalt oxyhydroxycompound is formed by the reaction (1) and enters in the reactions (2) and (3), participating to the electroanalytical act and dissolving. Thus, its balance equation will be rewritten as:

$$\frac{d\theta}{dt} = \frac{1}{\Gamma}(r_1 - r_2 - r_3) \quad (2)$$

In which  $\Gamma$  is  $\text{CoO(OH)}$  maximal concentration,  $r_1$  and  $r_3$  are the reaction rates of correspondent reactions (1) and (3).

The alkali enters the pre-surface layer by diffusion and enters in the reactions (1), (3), like also in the non-promoted hydrazine oxidation. So, it is possible to rewrite the balance equation for the alkali concentration as:

$$\frac{da}{dt} = \frac{2}{\delta} \left( \frac{D}{\delta} (a_0 - a) - r_1 - r_a - r_3 \right) \quad (3)$$

In which  $D$  is alkali diffusion coefficient and  $a_0$  is its concentration in the bulk of the solution.

The correspondent reaction rates may be calculated as:

$$r_1 = k_1 a (1 - \theta) \exp\left(\frac{F\varphi_0}{RT}\right) \quad (4)$$

$$r_2 = k_2 c \theta^2 \quad (5)$$

$$r_3 = k_3 a^3 \theta \exp(-g\theta) \quad (6)$$

$$r_a = k_a a^2 c \quad (7)$$

In which the parameters  $k$  are correspondent reaction rate constants,  $F$  is Faraday number,  $\varphi_0$  is the potential slope in double electric layer (DEL),  $R$  is universal gas constant  $T$  is an absolute temperature, and  $g$  is a variable, describing the interaction between the adsorbed particles.

On the general appearance, this model resembles, simultaneously, two types of the yet developed models for copper oxide nanoparticles' assisted hydrazine electrochemical detection [48] and for  $\text{CoO(OH)}$ -assisted electrochemical detection of oxalate [49] and phenolic compounds [50]. But, contrarily to the trivalent copper, involved in the electroanalytical process in [36,48], the trivalent cobalt is a more stable substance, from the oxidation-reduction point of view, and it influences the system's behavior. The common and different features of these systems will be described in the next section.

### 3. RESULTS AND DISCUSSION

The behavior of this system will be described by means of linear stability theory. The steady-state Jacobi functional matrix elements for it may be described 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:

$$a_{11} = \frac{2}{\delta} \left( -\frac{A}{\delta} - k_2\theta^2 - 2k_a a \right) \quad (9)$$

$$a_{12} = \frac{2}{\delta} (-2k_2\theta c) \quad (10)$$

$$a_{13} = \frac{2}{\delta} (-2k_a a c) \quad (11)$$

$$a_{21} = \frac{1}{\Gamma} (-k_2\theta^2) \quad (12)$$

$$a_{22} = \frac{1}{\Gamma} \left( -k_1 a \exp\left(\frac{F\varphi_0}{RT}\right) + Jk_1 a (1 - \theta) \exp\left(\frac{F\varphi_0}{RT}\right) - 2k_2 c \theta - k_3 a^3 \exp(-g\theta) + gk_3 a^3 \theta \exp(-g\theta) \right) \quad (13)$$

$$a_{23} = \frac{1}{\Gamma} \left( k_1 (1 - \theta) \exp\left(\frac{F\varphi_0}{RT}\right) - 3k_3 a^2 \theta \exp(-g\theta) \right) \quad (14)$$

$$a_{31} = \frac{2}{\delta} (-2k_a a) \quad (15)$$

$$a_{32} = \frac{2}{\delta} \left( k_1 a \exp\left(\frac{F\varphi_0}{RT}\right) - Jk_1 a (1 - \theta) \exp\left(\frac{F\varphi_0}{RT}\right) - k_3 a^3 \exp(-g\theta) + gk_3 a^3 \theta \exp(-g\theta) \right) \quad (16)$$

$$a_{33} = \frac{2}{\delta} \left( -\frac{D}{\delta} - 2k_a a - k_1 (1 - \theta) \exp\left(\frac{F\varphi_0}{RT}\right) - 3k_3 a^2 \theta \exp(-g\theta) \right) \quad (17)$$

In which J is the parameter, describing the influence of the electrochemical reaction on DEL.

Taking in account the  $a_{22}$  matrix element, it's possible to observe, that the addendums  $Jk_1 a (1 - \theta) \exp\left(\frac{F\varphi_0}{RT}\right)$  and  $gk_3 a^3 \theta \exp(-g\theta)$  are positive, so, the positive callback and, consecutively, the *oscillatory behavior* for this system is possible. As in [48-50], it is defined by two factors:

$Jk_1 a (1 - \theta) \exp\left(\frac{F\varphi_0}{RT}\right) > 0$ , when the CoO electrooxidation has strong influences on DEL rearrangement, so  $J > 0$ . It happens if CoO, in the conditions of the synthesis, is very strong reductant. In strongly alkaline media, CoO is oxidated rapidly, so these solutions contribute strongly to the oscillatory behavior.

The strongly alkaline media also contributes to such a behavior, when the rapid CoO(OH) dissolution is accompanied by attractive interaction of the film particles. So, the addendum  $gk_3 a^3 \theta \exp(-g\theta)$  is positive in this case. Both factors are characteristic for the behavior of CoO(OH) [49-50]. Yet in [48], that type of behavior is only defined by the first factor.

The oscillations are of relatively small amplitude and high frequency.

Applying the Routh-Hurwitz stability criterion, we obtain the *steady-state stability* condition for this system. To avoid the cumbersome expressions, we introduce new variables, so the matrix determinant may be represented as:

$$\frac{4}{\delta^2 \Gamma} \begin{vmatrix} -\kappa_1 - \mathcal{E} - \Lambda & -\Pi & -\xi \\ -\mathcal{E} & -\Lambda - \Pi - M & \Sigma - X \\ -\Lambda & \Lambda - M & -\kappa_2 - \xi - \Sigma - X \end{vmatrix} \quad (18)$$

Opening the determinant brackets, the steady-state stability requirement will be rewritten as:

$$-\kappa_1(\Lambda\kappa_2 + \Pi\kappa_2 + M\kappa_2 + \Lambda\xi + \Pi\xi + M\xi + \Pi\Sigma + 2M\Sigma + 2\Lambda X + \Pi X) - \mathcal{E}(\Lambda\kappa_2 + M\kappa_2 + 2M\xi + 2M\Sigma + 2\Lambda X + \Pi X) - \Lambda(\Lambda\kappa_2 + \Pi\kappa_2 + M\kappa_2 + 2M\Sigma + 2\Lambda X + 2\Pi X) < 0 \quad (19)$$

As the parameter values outside the brackets are always negative, and nearly all the expressions inside them (except those containing the CoO(OH) formation parameter  $\Lambda$  and CoO(OH) dissolution parameter  $M$ , that may have negative values) are positive, the inequation (19) is satisfied, so, in the case of the positivity of  $\Lambda$  and  $M$ , defining the absence, or fragility of DEL influences of the electrosynthesis of CoO(OH), and the CoO(OH) particles repulsion during its dissolution, the steady-state will be stable.

The reaction, thus, will be controlled by alkali and hydrazine diffusion, as the slowest process. The diffusion addendums are majority in the inequation (19).

As in similar systems [49-50], in very high pH conditions, although the steady-state is stable, it won't be electroanalytically efficient (see the table from [50]). Moreover, the velocity of direct hydrazine oxidation by alkali, absent in the cases of [49-50] will be the main process of hydrazine outlet, due to CoO(OH) dissolution. The steady-state destabilization probability is higher. So, the optimal pH for the analysis is lightly alkaline, close to neutral ( $7 < \text{pH} < 12$ ), and the pH-dependence of steady-state efficiency is corresponding, in general features, the Table 1[50]. In this pH range, CoO(OH) is an efficient catalyst for hydrazine electrooxidation.

When the stabilizing and destabilizing factors are equivalent, the *monotonic instability* occurs. It is corresponding to the steady-state stability margin.

Its condition, for this system, is:

$$-\kappa_1(\Lambda\kappa_2 + \Pi\kappa_2 + M\kappa_2 + \Lambda\xi + \Pi\xi + M\xi + \Pi\Sigma + 2M\Sigma + 2\Lambda X + \Pi X) - \mathcal{E}(\Lambda\kappa_2 + M\kappa_2 + 2M\xi + 2M\Sigma + 2\Lambda X + \Pi X) - \Lambda(\Lambda\kappa_2 + \Pi\kappa_2 + M\kappa_2 + 2M\Sigma + 2\Lambda X + 2\Pi X) = 0 \quad (20)$$

In this point various steady-states coexist, and each one is unstable. It is destroyed, when the conditions are changed.

If the direct oxidation is very much slower, than a CoO(OH)-assisted, its rate may be neglected and the system will be described by the model, similar to that of [49,50].

When the CoO(OH) configuration is changed, during the process, an additional reaction, similar to an isomerization, is realized. Both configurations are soluble in alkaline media,

forming the same complex. This reaction influences the DEL, and its case will be evaluated in one of our next works.

#### 4. CONCLUSIONS

From the theoretical evaluation of the possibility of use of CoO(OH) as an electrode modifier for hydrazine oxidation it's possible to conclude that:

- The oscillatory behavior in the case of CoO(OH)-assisted hydrazine oxidation is possible, and it is more probable, than in case of the use of trivalent copper. The alkalinity of the solution affects directly the oscillatory behavior.
- The steady-state stability is warranted by repealing interaction of CoO(OH) particles and the fragility of DEL influences of the electrochemical reaction. The reaction is diffusion controlled. In highly alkaline solution, the steady-state will be stable, but won't be electroanalytically efficient. The optimal pH for the analysis is  $7 < \text{pH} < 12$ .
- The monotonic instability for this system is possible, and it is realized in the case of the equality of stabilizing and destabilizing effects in DEL.
- In the case of CoO(OH) configuration change, the behavior of the system is more complicated, because this change influences DEL and oscillatory behavior.

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