Letters in Applied NanoBioScience
Open-Access Journal (ISSN: 2284-6808)

*Article*
**Volume 13, Issue 2, 2024, 58**
https://doi.org/10.33263/LIANBS132.058

# Theoretical Description for Melatonin Electrochemical Sensing on Cobalt(III) Oxyhydroxide-Modified Zeolite Matrix During the Modification Interchange

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Scopus Author ID 55758299100
**Received: 28.02.2023; Accepted: 28.05.2023; Published: 30.06.2024**

**Abstract:** For the first time, the electroanalytical system for melatonin electrochemical determination over cobalt (III) oxyhydroxide-modified zeolite matrix during the modification interchange has been theoretically described. The analysis of the correspondent mathematical model confirms the efficiency of the conducting polymer for melatonin determination. On the other hand, the oscillatory and monotonic instabilities are possible but of reduced probability, as the ionization degree of melatonin in neutral and lightly alkaline media is reduced.

**Keywords:** melatonin; electrochemical sensor; chemically modified electrodes; cobalt(III)oxyhydroxide; electrochemical oscillations; stable steady-state

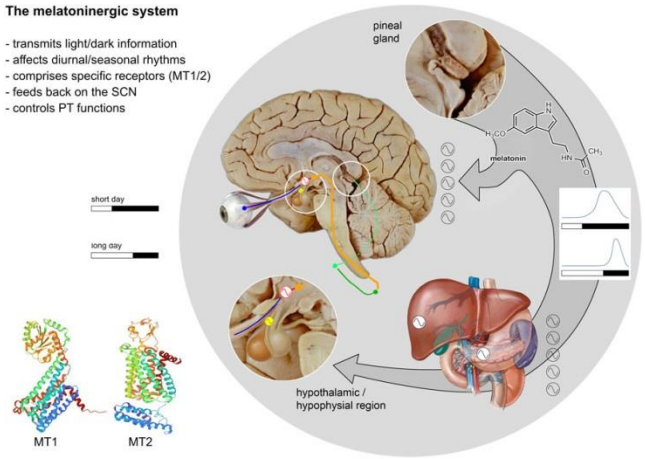


## 1. Introduction

Neurohormone melatonin [1 – 5] is produced by the pineal gland, also known as *epiphysis cerebri*. This endocrine gland located in the posterior aspect of the cranial fossa in the brain (Fig. 1A) plays a key role in the circadian rhythm (day-night cycle) of sleep and wakefulness and the seasonal rhythm. Melatonin secretion pattern is dictated by light exposure. Serum concentrations of melatonin are low during daylight hours but increase and reach a maximum in the darkness. The melatonin concentration in baby organisms tends to be regularized at 3 months, maximal between midnight and 8 a.m. The duration of melatonin secretion depends linearly on the darkness interval. When light is perceived in the retina by the nerve cells (rods and cones), specialized ganglia take the impulses generated by the light to the suprachiasmatic nucleus (SCN) of the hypothalamus [1]. In mammals, melatonin elicits its

function principally through two specific receptors called MT1 and MT2 (Fig. 1A). MT1 is highly expressed in the SCN and the hypophysial pars tuberalis (PT), an important interface for the control of seasonal functions. The expression of MT2 is more widespread.

**A**




**B**

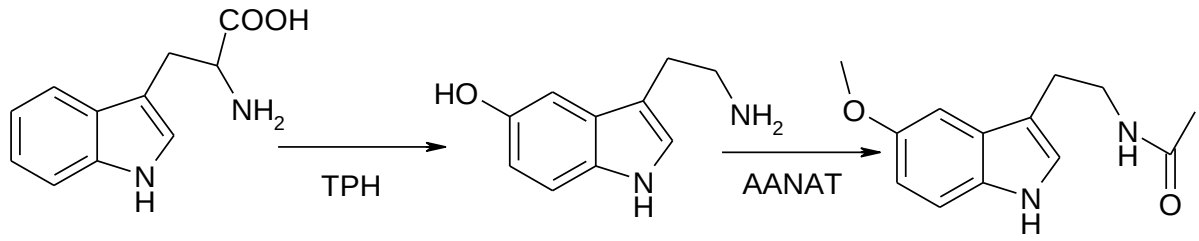



**Figure 1. (A)** The metaloninergic system in the circadian and seasonal rhythms. Reproduced from **(B)** Synthesis of melatonin.

The rate-limiting concentration of enzyme serotonin N-acetyltransferase (AANAT), which enables the synthesis of melatonin by N-acylation of the neurotransmitter serotonin, is low during daylight time (Fig. 1B). Neurotransmitter serotonin is, in turn, derived from the essential aminoacid called tryptophan. Tryptophan hydroxylase (TPH) is the enzyme responsible for the hydroxylation (i.e., phenolization) of tryptophan in position 5 (Fig. 1B), which is the initial and rate-limiting step in the synthesis of serotonin. In the pineal gland, serotonin undergoes acylation (i.e., amidation) and then methylation (etherification) to yield the end product, melatonin. Melatonin then reacts with G-protein receptors to implement the biological effects. These receptors are found in the SCN, the retina, and the *pars tuberalis* in the adenohypophysis. Receptors are also scattered around various areas of the brain.

Besides regulating sleepiness, melatonin is an antioxidant, recovering the epithelial cells under ultraviolet irradiation [6 – 8]. It is also responsible for recovering the neurons affected by Alzheimer's disease and ischemia. This is the typical function of indolic derivatives, substituted by donor substituent. In the works [9 – 10], the function of melatonin as an antioxidant in wine and grape culture, in which melatonin assumes partly the function of polyphenolic derivatives, augmenting their content in grape juice and wine, has been shown. Nevertheless, the melatonin concentration in wines is strictly regulated due to the coaction of melatonin with alcohol [11]. As for the main function of melatonin, its pharmaceutical forms help people to prevent or reduce the jet lag effects, especially for those traveling in a west-east

direction for five or more time zones or in an east-west direction, crossing the International Date Line.

Considering the importance of melatonin for the human organism, developing a method capable of detecting its concentration efficiently is timely and of extreme relevance [11,12]. This molecule's Two major attributes make it a particularly attractive candidate for an electroanalytical technique working as a solution for this task. As aforementioned, melatonin is an indolic derivative substituted by a donor group. Therefore, it is active toward anodic oxidation [13 –15]. Moreover, melatonin has already been polymerized, yielding a conducting polymer [16,17], and the use of polymelatonin in electroanalytic has been referred to in the literature.

In this work, the possibility of cobalt oxyhydroxide (CoO(OH))-assisted melatonin determination during the modification interchange is described theoretically. The modification interchange may be considered the first stage of CoO(OH) oxidation [18 – 20]. The correspondent mathematical model is analyzed through linear stability theory and bifurcation analysis to derive the best sensing function requisites, as well as the oscillatory and monotonic instability conditions. The behavior of this system is compared with those of similar ones [21 – 28].

## 2. System and its modeling

In this work, we investigated the case in which β-CoO(OH), embedded in a zeolite matrix, paired with cobalt monoxide (CoO), undergoes modification interchange during the electroanalytical process described by the following equations:

$$CoO + OH^- \rightarrow \beta\text{-}CoO(OH) + e^- \quad (1)$$

$$\beta\text{-}CoO(OH) \rightarrow \gamma\text{-}CoO(OH)^* \text{ (more active)} + e^- \quad (2)$$

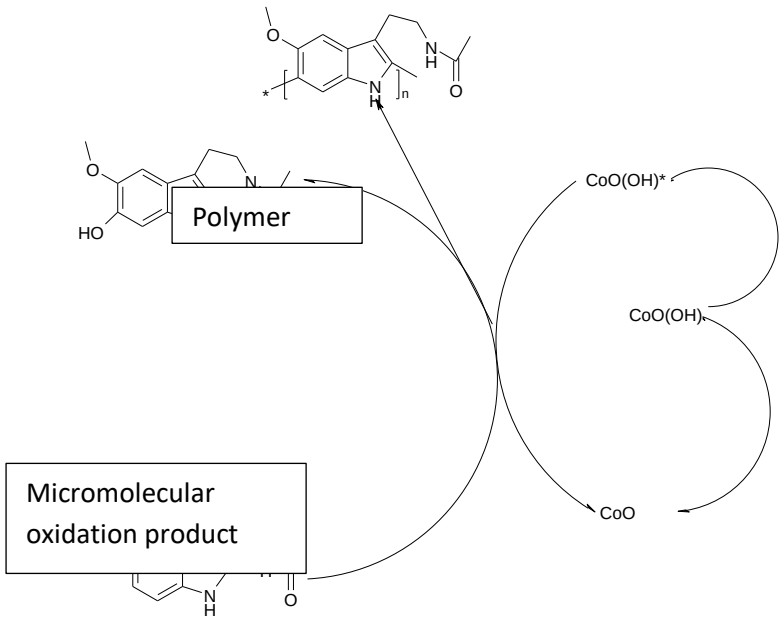



**Figure 2.** The schematic representation of an electroanalytical process.

The more electroactive γ-CoO(OH)* species reacts with melatonin, oxidizing it by both micro- and macromolecular scenarios.

The macromolecular scenario provides the synthesis of a conducting polymer, similar to the electropolymerization techniques used in electroanalytics, as described in the literature [16,17]. In this case, a hybrid composite or metal-organic framework (MOF) with interesting properties may be formed, depending on the initial morphological state of CoO(OH) (powder, film, or nanoparticles).

The micromolecular scenario may occur by phenolization of the melatonin ortho-position in relation to the methoxyl group in neutral and alkaline media, yielding a co-product proton or water, respectively. Further oxidation of the micromolecular oxidation product and its participation in the electropolymerization scenario will be evaluated in future work.

Taking the aforementioned into account, we describe the behavior of the melatonin oxidation over CoO(OH)-modified anode during modification interchange by an equation set of three variables:

m – melatonin concentration in the pre-surface layer;
c – main cobalt(III) oxyhydroxide modification surface coverage degree;
c* - activated cobalt(III) oxyhydroxide surface coverage degree.

Taking some assumptions [25 – 28], we describe the system´s behavior by the balance equation-set (3):

$$\begin{cases} \frac{dm}{dt} = \frac{2}{\delta}\left(\frac{\Delta}{\delta}(m_0 - m) - r_m - r_p\right) \\ \frac{dc}{dt} = \frac{1}{C}(r_o - r_e) \\ \frac{dc*}{dt} = \frac{1}{C*}(r_e - r_m - r_p) \end{cases} \quad (3)$$

Herein, $\Delta$ is the diffusion coefficient, $m_0$ is the bulk melatonin concentration, C e C* are maximal surface coverage degrees of each form of cobalt (III) oxyhydroxide, and the parameters r are the correspondent reaction rates, calculated as:

$$r_m = k_m m c *^2 \quad (4)$$

$$r_p = k_m m^x c *^y \exp(-am) \quad (5)$$

$$r_o = k_o(1 - c - c *) \exp\frac{F\varphi_0}{RT} \quad (6)$$

$$r_e = k_e c \exp\frac{F\varphi_0}{RT} \quad (7)$$

Herein, the parameters k are correspondent reaction rate constants, x and y are polymerization reaction orders, a is a variable describing the impact of the assisted electropolymerization on the double electric layer (DEL) during the growth center creation, $F=N_A*e$ is the Faraday number, $\varphi_0$ is the zero-charge-related potential slope, R is the universal gas constant, and T is the absolute temperature.

In this case, only the melatonin CoO(OH)-assisted electropolymerization reaction affects the DEL ionic force, directly related to its ionic composition and activity, capacitance, impedance, and conductivity. Other reactions affecting it are purely electrochemical. For this reason, this system becomes more stable than similar ones [25 – 28]. Therefore, we are led to conclude that CoO(OH) may be an efficient electrode modifier during the modification interchange, as demonstrated below.

## 3. Results and Discussion

We investigate the electroanalytical determination of melatonin over CoO(OH) during modification interchange by analyzing the equation-set (3) alongside the algebraic relations (4 – 7) by means of linear stability theory. The steady-state Jacobian matrix elements may be described as (8):

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

Where:

$$a_{11} = \frac{2}{\delta}\left(-\frac{\Delta}{\delta} - k_m c*^2 - x k_m m^{x-1} c*^y \exp(-am) + a k_m m^x c*^y \exp(-am)\right) \quad (9)$$

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

$$a_{13} = \frac{2}{\delta}(-2mc* \exp(-am) - y k_m m^x c*^{y-1} \exp(-am)) \quad (11)$$

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

$$a_{22} = \frac{1}{C}\left(-k_o \exp\frac{F\varphi_0}{RT} + jk_o(1 - c - c*)\exp\frac{F\varphi_0}{RT} - k_e \exp\frac{F\varphi_0}{RT} + jk_e c \exp\frac{F\varphi_0}{RT}\right) \quad (13)$$

$$a_{23} = \frac{1}{C}\left(-k_o \exp\frac{F\varphi_0}{RT} + lk_o(1 - c - c*)\exp\frac{F\varphi_0}{RT} - k_e \exp\frac{F\varphi_0}{RT} + lk_e c \exp\frac{F\varphi_0}{RT}\right) \quad (14)$$

$$a_{31} = \frac{1}{C*}(-k_m c*^2 - x k_m m^{x-1} c*^y \exp(-ac*)) \quad (15)$$

$$a_{32} = \frac{1}{C*}\left(k_e \exp\frac{F\varphi_0}{RT} - jk_e p \exp\frac{F\varphi_0}{RT}\right) \quad (16)$$

$$a_{33} = \frac{1}{C*}\left(k_e \exp\frac{F\varphi_0}{RT} - lk_e c \exp\frac{F\varphi_0}{RT} - 2mc* - y k_m m^x c*^{y-1} \exp(-ac*) + a k_m m^x c*^y \exp(-ac*)\right) \quad (17)$$

Close analysis of the main diagonal elements (9), (13), and (17), allows us to infer that the oscillatory behavior in this system is possible. Nevertheless, it is less probable than for the electropolymerization of somehow more ionic monomers, such as aminoindoles, indolic sulfoacids, and polyphenolic derivatives, due to the absence of the impact of the micromolecular oxidation on DEL. Only assisted electropolymerization impacts the double layer due to the transformation of ionic forms.

In order for the Hopf bifurcation to take place, it is necessary to have positive elements on the main diagonal. These elements are responsible for the positive callback, and in this system they are $a k_m m^x p*^y \exp(-ap*) > 0$, if a>0, $jk_o(1 - p - p*)\exp\frac{wF\varphi_0}{RT} > 0$ and $jk_e p \exp\frac{vF\varphi_0}{RT} > 0$, if j>0, as well as $-lk_e p \exp\frac{vF\varphi_0}{RT} > 0$, if l<0. These elements are characteristic of electropolymerization and sensing systems involving CoO(OH) and/or

intrinsically conducting polymers and describe oscillatory behavior dependent on the background electrolyte composition, as observed experimentally [15 – 18] and theoretically [19 – 21].

We investigate the steady-state stability by applying the Routh-Hurwitz criterion to the equation set (3). We simplify the Jacobian analysis by rewriting its determinant as (18):

$$\frac{2}{\delta PP*}\begin{vmatrix} -\kappa - \Lambda & 0 & -T \\ 0 & -\Sigma - P & -\Phi + M \\ -\Lambda & P & -M - T \end{vmatrix} \quad (18)$$

and avoiding cumbersome expressions.

Opening the brackets and applying the Det J<0 condition derived from the criterion, we obtain the steady-state stability requisite as (19):

$$-\kappa(\Sigma M + \Sigma T + PT + P\Phi) - \Lambda(\Sigma M + P\Phi) < 0 \quad (19),$$

or, after changing signs (20):

$$\kappa(\Sigma M + \Sigma T + PT + P\Phi) + \Lambda(\Sigma M + P\Phi) > 0 \quad (20),$$

The inequity (20) is ensured if the variables P, Φ, M, and T remain positive. Really, if we suppose that the above-mentioned parameters are positive, the left side of the inequity (20) will be shifted to more positive values, diminishing the deviations and stabilizing the steady state. The electroanalytical process will be both diffusion and kinetically controlled but receive a higher impact from the diffusion factor than the electroanalytical process involving the excited conducting polymer as an anode modifier.

From the electroanalytical point of view, the steady-state stability corresponds to the linear current-concentration dependence, corresponding to the best analytical signal interpretation. From the electrosynthetical point of view, it corresponds to a well-developed polymeric composite surface. The stable, steady state is easy to obtain and maintain on a vast topological zone till the detection limit is reached.

This limit is described by the monotonic instability, depicting a margin between the stable steady-states and unstable states. It is related to the condition of Det J=0, or (21):

$$\kappa(\Sigma M + \Sigma T + PT + P\Phi) + \Lambda(\Sigma M + P\Phi) = 0 \quad (21)$$

The low-molecular melatonin oxidation product may participate in electropolymerization at higher anodic potentials. Moreover, it tends to undergo hydrolytic oxidation to the correspondent quinonic compound. For this case, model (3) will be rewritten, and this process will be described in our next works.

## 4. Conclusions

From the theoretical analysis of melatonin CoO(OH)-assisted electrochemical determination and electropolymerization during the modification interchange, it was possible to conclude that this process is an efficient kinetically controlled electroanalytical and electrosynthetical system. The oscillatory behavior in this system is possible but less probable

than for more ionic analytes and monomers. Either way, the oscillation frequency and amplitude depend strongly on the background electrolyte nature due to DEL impact on electrochemical stages and melatonin electropolymerization.

## Funding

This research received no external funding.

## Acknowledgments

Volodymyr V. Tkach acknowledges the Faculty of Engineering of the University of Porto and the University of Trás-os-Montes and Alto Douro for their support in these difficult times for Ukraine and its research.

## Conflicts of Interest

The authors declare no conflict of interest.

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