

The Theoretical Description of Sucralose and Lactic Acid Electrochemical Determination in Breast Milk Over a Norharmane-Insubosin Copolymer

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Abstract: The possibility of the electrochemical determination of sucralose and lactic acid on an anode modified by insubosin copolymer with norharmane. The sucralose and lactic acid peak separation is achieved not only by the differences in reaction mechanisms but also by the differences in the doping types due to the presence of the pyridinic nitrogen atoms of sucralose. The analysis of the corresponding model confirms the efficiency of the conducting copolymer of norharmane and insubosin as an efficient electrode modifier for sucralose and lactate electrochemical determination. However, the oscillatory behavior may be realized as highly probable.

Keywords: breast milk; sucralose; lactic acid; electrochemical determination; electrochemical sensor; electrochemical oscillations; stable steady-states.

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1. Introduction

Neonatal intestinal dysbiosis [1–4] is a pathological state characterized by failure in the initial colonization of the neonatal intestinal tract. The baby's gut microbiota is formed during pregnancy and up to a year after birth, based on the living microbes transferred from maternal microbiota and the host environment. Nevertheless, if used, some of the food components may cause problems with gut microbiota development and, further, metabolic pathologies in babies. One of them is sucralose [5–7].

Sucralose is a widely used sweetener generally considered safe [8–10]. Nevertheless, it is an environmentally unfriendly chloroorganic compound that tends to accumulate in the environment. Moreover, a recent investigation [5,6] has shown that the sucralose, if consumed by pregnant women, passes through a placental barrier, enters the breast milk, and thereby causes changes in the baby's gut microbiota, leading to metabolic disorders in babies and the possibility of type 2 diabetes development. It may also be used by pregnant women who smoke electronic cigarettes with sucralose added.

Taking this into account, the development of an efficient sucralose and lactic acid sensor for diabetic and obese pregnant women is actual [11–17], and, taking into account that both of the substances are electrochemically active, the electrochemical sensing may be an interesting approach for this task.

The use of chemically modified electrodes is an interesting, flexible, and modern tool for electroanalysis [18–24]. It permits an efficient peak separation of two or more analytes, the oxidation peaks for which overlap on bare electrodes. This is achieved by the reaction kinetic differences, which provide a more selective determination.

Nevertheless, the surface and double electric layer (DEL) conductivity changes may seriously affect the electrochemical efficiency of the sensor. This impact is manifested by oscillatory and monotonic instabilities. Their occurrence and influence on the behavior of the electroanalytical process [25–32] is an object of an *a priori* theoretical investigation, necessary for realization before the sensor comes into practical use (in a lab and in the field).

Therefore, the goal of our work is to investigate the behavior of the electrochemical sensor for sucralose and lactic acid determination in breast milk based on poly(norharmane-co-insubosin)-modified electrode. The corresponding mathematical model is to be developed and analyzed from the stability point of view, and the behavior of the system will be compared to similar ones [33–35].

2. Materials and Methods

Both monomers composing the conducting polymer (Figure 1) contain pyridinic nitrogen atoms, which possess basic properties.

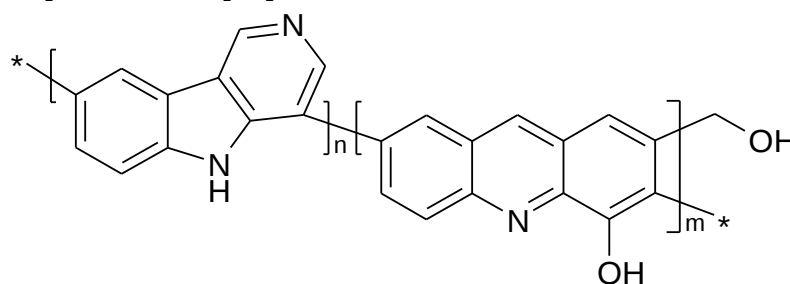


Figure 1. The approximate copolymer structure.

By this, in a neutral medium, the lactic acid may be efficiently immobilized by entering the polymer matrix by electrochemical doping, whereas the sucralose is immobilized by chemical doping, quaternizing the pyridinic nitrogen atom, as previously shown in [33–35]. Both reactions lead to the analyte immobilization on the polymer matrix, with their further oxidation. Nevertheless, their kinetics are different, which provides an efficient peak separation. Lactic acid, in the form of lactate-ion, is thereby oxidized to pyruvic acid, and the sucralose is oxidized by the unique remaining hydroxymethyl group.

Therefore, taking the assumptions accepted in [33–35], we describe the behavior of the system by the equation set (1):

$$\begin{cases} \frac{dl}{dt} = \frac{2}{\delta} \left(\frac{\Delta}{\delta} (l_0 - l) - r_{ld} \right) \\ \frac{ds}{dt} = \frac{2}{\delta} \left(\frac{S}{\delta} (s_0 - s) - r_{sd} \right) \\ \frac{dp}{dt} = \frac{1}{P} (r_{ld} + r_{sd} - r_1 - r_2) \end{cases} \quad (1)$$

In which l and s are lactic acid and sucralose concentrations in the pre-surface layer, l_0 and s_0 stand for their bulk concentrations, δ is the pre-surface layer thickness, Δ and S are the correspondent diffusion coefficients, p is the polymer modification degree, P is the modified polymer maximal surface concentration, and the parameters r stand for the correspondent reaction rates, calculated as:

$$r_{ld} = k_{ld} l (1 - p) \exp\left(\frac{vF\varphi_0}{RT}\right) \quad (2)$$

$$r_{sd} = k_{sd} s (1 - p) \exp(-as) \quad (3)$$

$$r_1 = k_1 p \exp\left(\frac{wF\varphi_0}{RT}\right) \quad (4)$$

$$r_2 = k_2 p \exp\left(\frac{xF\varphi_0}{RT}\right) \quad (5)$$

In which the parameters k are the corresponding reaction rate constants, v w and x are the numbers of electrons transferred during the electrochemical stages, a is the parameter relating DEL ionic force with the polymer quaternization with sucralose, F is the Faraday number, φ_0 is the zero-charge-related potential slope, R is the universal gas constant, and T is the absolute temperature.

The presence of accepting chlorine atoms in sucralose, like the different doping types, provides the peak separation. Therefore, despite the enhanced probability of the oscillatory behavior, the copolymer will be an efficient electrode modifier for sucralose and lactic acid determination in breast milk, as shown below.

3. Results and Discussion

In terms of stability evaluation of the system with sucralose and lactic acid electrochemical determination in breast milk, we investigate the equation set (1) by means of linear stability theory and expose the Jacobian steady-state matrix members as (6):

$$\begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix} \quad (6)$$

In which:

$$a_{11} = \frac{2}{\delta} \left(-\frac{\Delta}{\delta} - k_{ld} (1 - p) \exp\left(\frac{vF\varphi_0}{RT}\right) \right) \quad (7)$$

$$a_{12} = 0 \quad (8)$$

$$a_{13} = \frac{2}{\delta} \left(k_{ld} \exp\left(\frac{vF\varphi_0}{RT}\right) - j k_{ld} l (1 - p) \exp\left(\frac{vF\varphi_0}{RT}\right) \right) \quad (9)$$

$$a_{21} = 0 \quad (10)$$

$$a_{22} = \frac{2}{\delta} \left(-\frac{S}{\delta} - k_{sd} (1 - p) \exp(-as) + a k_{sd} s (1 - p) \exp(-as) \right) \quad (11)$$

$$a_{23} = \frac{2}{\delta} (k_{sd} s \exp(-as)) \quad (12)$$

$$a_{31} = \frac{1}{P} \left(k_{ld} (1 - p) \exp\left(\frac{vF\varphi_0}{RT}\right) \right) \quad (13)$$

$$a_{32} = \frac{1}{P} (k_{sd} (1 - p) \exp(-as) - a k_{sd} s (1 - p) \exp(-as)) \quad (14)$$

$$a_{33} = \frac{1}{p} \left(-k_{ld} \exp\left(\frac{vF\varphi_0}{RT}\right) + jk_{ld}l(1-p) \exp\left(\frac{vF\varphi_0}{RT}\right) - k_{sd}s \exp(-as) - k_1 \exp\left(\frac{wF\varphi_0}{RT}\right) + jk_1p \exp\left(\frac{wF\varphi_0}{RT}\right) - k_2 \exp\left(\frac{xF\varphi_0}{RT}\right) + jk_2p \exp\left(\frac{xF\varphi_0}{RT}\right) \right) \quad (15)$$

In terms of the *oscillatory behavior*, this system will be analogous to the other systems, involving sucralose and pyridinic nitrogen heteroatoms [33–35] and the electrooxidation of a doped analyte within the polymer matrix. The oscillations' amplitude and frequency will be highly dependent on the DEL ionic force, which, in turn, is directly linked to the background electrolyte composition, including pH.

The Hopf bifurcation conditions, corresponding to the oscillatory behavior, are foreseen for the case of the presence of the positive elements in the Jacobian main diagonal. In this system, there are four main diagonal elements, capable of being positive, being only one, $ak_{sd}s(1-p) \exp(-as) > 0$, if $a > 0$, referent to the chemical stage, more concretely, to the chemical doping of polymer by sucralose by a pyridinic nitrogen atom, yet observed in [33–35].

The other positive elements refer to each one of the electrochemical stages and their influences on the DEL ionic force and related electrophysical properties. These elements are $jk_{ld}l(1-p) \exp\left(\frac{vF\varphi_0}{RT}\right) > 0$, $jk_1p \exp\left(\frac{wF\varphi_0}{RT}\right) > 0$, $jk_2p \exp\left(\frac{xF\varphi_0}{RT}\right) > 0$, if $j > 0$, analogous to the systems [33–35].

As for the *steady-state stability*, it will be assured in the wide topological region, corresponding to the linear dependence between the electrochemical parameter and concentration. As neither the analyte nor the electrode modifier undergoes a side reaction, compromising their stability, the steady-state stability corresponds to easy analytical signal interpretation.

The mathematical condition for this stability is defined by the Routh-Hurwitz criterion. In order to apply it to the equation set (1), we introduce new variables, thereby rewriting the Jacobian determinant as (16):

$$\frac{4}{\delta^2 p} \begin{vmatrix} -\kappa - \Xi & 0 & \Sigma \\ 0 & -\xi - P & T \\ \Xi & P & -\Sigma - T - \Omega \end{vmatrix} \quad (16)$$

Being the steady-state stability requirement, thereby expressed as (17) by the Det $J < 0$ condition, salient from the criterion, after changing the signs to the opposite ones:

$$\kappa(\xi\Sigma + \xi T + \xi\Omega + P\Sigma + P\Omega) + \Xi(\xi T + \xi\Omega + P\Omega) > 0 \quad (17)$$

Corresponding to mostly diffusion-controlled electroanalytical systems.

The detection limit will be defined by *monotonic instability*, for which multiple unstable steady states coexist. This margin between stable, steady-states and unstable states corresponds to the nullity of the determinant, or (18):

$$\kappa(\xi\Sigma + \xi T + \xi\Omega + P\Sigma + P\Omega) + \Xi(\xi T + \xi\Omega + P\Omega) > 0 \quad (18)$$

The neutral or mildly acidic medium, corresponding to milk, is the most favorable for this process. In a strongly acidic medium, the pyridinic nitrogen atoms will be highly protonized, which will affect the behavior as described in [33]. In the absence of lactic acid, sucralose concentration will be measured by the process, analogous to that described in [34,35].

This process is also suitable for sucralose determination in soft drinks, where it is frequently added with real sugars. The selectivity is achieved by pyridinic nitrogen quaternization, which isn't realized with natural carbohydrates. The behavior of this system may also be described by the model, analogous to that expressed by the equation set (1).

4. Conclusions

In the theoretical evaluation of the system with sucralose and lactic acid determination in breast milk, it was possible to conclude that the conducting polymer with pyridinic nitrogen atoms will be a selective and efficient electrode modifier for the determination of both of the analytes. In terms of the oscillatory behavior, this system will be analogous to that with the sucralose determination over a heterocyclic polymer with pyridinic nitrogen atoms. Besides breast milk, this sensor may also be used for non-alcoholic soft drinks.

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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