

# Theoretical Description for Sucralose Cathodical Electrochemical Determination on the Conducting Polymer, Containing Pyridinic Nitrogen Atoms

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**Abstract:** A novel process for sucralose cathodically electrochemical determination has been suggested. In this process, the conducting polymer with pyridinic nitrogen atoms reacts specifically with sucralose, yielding a salt. This salt is thereby reduced gradually, yielding a less conducting macromolecule. The analysis of the correspondent mathematical model, realized through the linear stability theory and bifurcation analysis, shows that the oscillatory behavior is more probable to be realized due to charge changes and rearrangements during the electroanalytical process its influence on the surface and DEL conductivity. Nevertheless, this process may be efficient for the sucralose electrochemical determination.

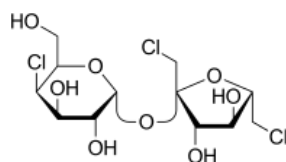
**Keywords:** sucralose; electrochemical sensor; conducting polymer; electrochemical oscillations; stable steady-state.

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## 1. Introduction

The chemically modified electrodes (CME) are one of the most modern and flexible tools in electroanalytical chemistry [1-4]. They provide a rapid, efficient, accurate and selective analyte determination due to their affinity to the analyte.

On the other hand, sucralose [5, 6] is one of the most-used sugar substitutes in the world. It is three times as sweeter as aspartame, twice as sweeter as saccharin, and 800 to 1000 times sweeter than sucrose [7, 8].



**Figure 1.** Sucralose.

It has been approved for use in the USA, in Canada, in Australia, and in the European Union [8]. Its chemical composition is related to that of carbohydrates. But, containing three chlorine atoms may present toxic effects like mutagenesis, carcinogenesis, provoke obesity, and growth of glucose levels [7-12]. Moreover, while stored in an inappropriate way, it may form dioxins, which are even more toxic compounds [13, 14]. The sucralose decomposition during baking may lead to the chloropropanoles formation [15]. So, the development of an efficient method for sucralose detection is actual [16-18], and the use of electrochemical methods for it would be very interesting.

The possibility of electrochemical sucralose determination has already been foreseen by us theoretically [19, 20] and confirmed experimentally [21]. A direct electrooxidation of sucralose was used either in the model or in the experiment. In work [19], sucralose immobilization on an acridine derivative capable of forming a quaternary salt has been suggested. The sucralose immobilization was foreseen to be followed by the electrochemical oxidation of hydroxyl groups of the sucralose units. Another opportunity could be an electrochemical reduction of sucralose salt with the correspondent pyridinic nitrogen compound, yet realized for paraquat and diquat [22, 23].

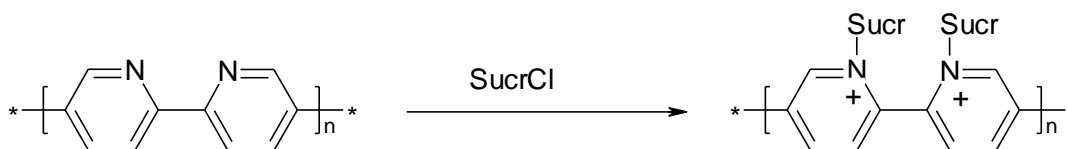
Nonetheless, the novel electrode modifiers' relation to the analytes may include the problems of mechanism interpretation, like the need to explain the electrochemical instabilities, yet described for the electrooxidation and reduction of organic compounds [24-28].

The mentioned problems may only be solved by analyzing a mathematical model capable of adequately describing the fluoxetine electrochemical determination. Moreover, it is also capable of comparing this system's behavior with similar processes by theoretical means.

So this work is aimed to describe the sucralose electrochemical determination theoretically over a conducting polymer containing pyridinic nitrogen atoms. In order to realize it, we suggest the reaction mechanism, developing and analyzing the mathematical model based on it. Also, the system's behavior has to be compared with similar electrochemical processes [19-21].

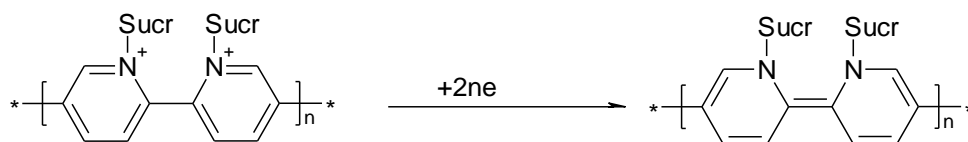
## 2. System and its Modeling

In the first stage, the sucralose reacts with the polymer, yielding a quaternary salt (Fig. 2):



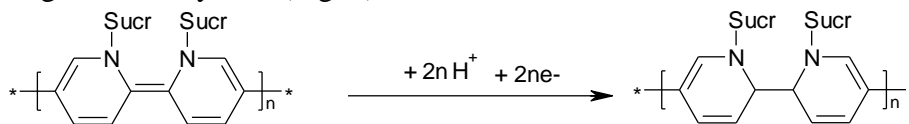
**Figure 2.** Quaternary salt formation

As the cathodic current is applied to the polymer, the conducting polymer is thereby reduced. The charge distribution is changed, and the double bond conjugated system is modified (Fig. 3):



**Figure 3.** First electrochemical reduction stage

By this, the deformed conjugated material is non-aromatic and, therefore, can reduce, interrupting the bond system (Fig. 4).



**Figure 4.** Second electrochemical reduction stage

As for the material conductivity increases in the first stage due to the salt formation and decreases in the chemical and electrochemical stages.

Taking into account the mentioned statements, we describe the behavior of this system by the trivariate balance differential equation set, formed by the variables:

- s – sucralose concentration in the pre-surface layer;
- p – polymer salt surface coverage degree;
- q – quinonized polymer surface coverage degree.

Accepting certain assumptions [28], we describe the behavior of this system will be described by three balance equations, written as (1):

$$\begin{cases} \frac{ds}{dt} = \frac{2}{\delta} \left( \frac{\Delta}{\delta} (s_0 - s) - r_s \right) \\ \frac{dp}{dt} = \frac{1}{P} (r_s - r_{r1}) \\ \frac{dq}{dt} = \frac{1}{Q} (r_{r1} - r_{r2}) \end{cases} \quad (1)$$

Herein,  $s_0$  is the sucralose bulk concentration, P and Q are corresponding polymeric forms maximal surface concentrations, and the parameters r are the corresponding reaction rates, calculated as:

$$r_s = k_s s^{2n} (1 - p - q) \exp(-as) \quad (2)$$

$$r_{r1} = k_{r1} p \exp\left(-\frac{2nF\varphi_0}{RT}\right) \quad (3)$$

$$r_{r2} = k_{r2} q \exp\left(-\frac{2nF\varphi_0}{RT}\right) \quad (4)$$

Herein, the parameters k are the correspondent rate constants, a is the variable describing the chemical stage influence to the DEL and surface conductivity, n is the polymer chain length, F is the Faraday number,  $\varphi_0$  is the potential slope, related to the zero-charge potential, R is the universal gas constant, and T is the absolute temperature.

As all of the three stages, which compose the process, influence the double electric layer, the oscillatory and monotonic instabilities are capable of realizing with more probability. Nevertheless, the electroanalytical process will remain efficient, as shown below.

### 3. Results and Discussion

In order to describe the behavior of the electrochemical system with sucralose cathodically determination over a conducting polymer with pyridinic nitrogen, we analyze the equation-set (1) by means of linear stability theory. The steady-state Jacobian matrix members for this system will be described as (5):

$$\begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix} \quad (5)$$

where:

$$a_{11} = \frac{2}{\delta} \left( -\frac{\Delta}{\delta} - 2nk_s s^{2n-1} (1-p-q) \exp(-as) + ak_s s^{2n} (1-p-q) \exp(-as) \right) \quad (6)$$

$$a_{12} = \frac{2}{\delta} (k_s s^{2n} \exp(-as)) \quad (7)$$

$$a_{13} = \frac{2}{\delta} (k_s s^{2n} \exp(-as)) \quad (8)$$

$$a_{21} = \frac{1}{p} (2nk_s s^{2n-1} (1-p-q) \exp(-as) - ak_s s^{2n} (1-p-q) \exp(-as)) \quad (9)$$

$$a_{22} = \frac{1}{p} \left( -k_s s^{2n} \exp(-as) - k_{r1} \exp\left(-\frac{2nF\phi_0}{RT}\right) + jk_{r1}p \exp\left(-\frac{2nF\phi_0}{RT}\right) \right) \quad (10)$$

$$a_{23} = \frac{1}{p} \left( -k_s s^{2n} \exp(-as) - lk_{r1}p \exp\left(-\frac{2nF\phi_0}{RT}\right) \right) \quad (11)$$

$$a_{31} = 0 \quad (12)$$

$$a_{32} = \frac{1}{q} \left( k_{r1} \exp\left(-\frac{2nF\phi_0}{RT}\right) + jk_{r1}p \exp\left(-\frac{2nF\phi_0}{RT}\right) \right) \quad (13)$$

$$a_{33} = \frac{1}{q} \left( -lk_{r1}p \exp\left(-\frac{2nF\phi_0}{RT}\right) - k_{r2} \exp\left(-\frac{2nF\phi_0}{RT}\right) + lk_{r2}q \exp\left(-\frac{2nF\phi_0}{RT}\right) \right) \quad (14)$$

Taking into account the main diagonal elements (6), (10), and (14), we may conclude that the oscillatory behavior, *in this case*, is possible. Moreover, it will be more probable than for similar systems due to the somehow more expressive influence of all three reaction stagers to double the electric layer and surface conductivity and resistance.

These elements are  $lk_{r2}q \exp\left(-\frac{2nF\phi_0}{RT}\right) > 0$  if  $l>0$ ,  $jk_{r1}p \exp\left(-\frac{2nF\phi_0}{RT}\right) > 0$  if  $j>0$ , describing the DEL influences of two electrochemical stages, and  $ak_s s^{2n} (1-p-q) \exp(-as) > 0$  if  $a>0$ , describing the analogous influences of the chemical stage.

The oscillations' frequency and amplitude are dependent on the background electrolyte composition, directly related to DEL structure and conductivity. Nevertheless, the proper oscillations are expected to be frequent and of small amplitude.

In order to apply the Routh-Hurwitz criterion without cumbersome expressions, we rewrite the matrix determinant as (15):

$$\frac{2}{\delta N_1 N_2} \begin{vmatrix} -\kappa - \mathcal{E} & \Sigma & \Sigma \\ \mathcal{E} & -\Gamma - \Sigma & -T - \Sigma \\ 0 & -\Phi - \Gamma & -\Omega - T - \Sigma \end{vmatrix} \quad (15)$$

Opening the brackets and applying the requisite  $\text{Det } J < 0$ , salient from the criterion, we obtain the steady-state stability condition, exposed as:

$$-\kappa(\Gamma\Omega + \Sigma\Omega + \Sigma T + \Sigma^2 - \Phi T - \Phi\Sigma) - \mathcal{E}(\Gamma\Omega + \Sigma\Omega + \Sigma T - \Phi T + \Phi\Sigma) < 0 \quad (16)$$

This describes an electroanalytical efficient process, which is either diffusion or kinetically controlled. The transition to diffusion- or kinetically controlled mode is realized by changing the electrolyte composition, analyte concentration, and electrode shape.

The steady-state stability topological zone has to be narrower than in the more simple cases described in [19-21]. Nevertheless, it remains relatively wide, providing a wide concentration interval of sensitive sucralose determination. As no side reactions capable of compromising the analyte or modifier stability are typical for this case, the steady-state stability will correspond to the linear dependence between the analyte concentration and electrochemical parameter.

As for the detection limit, it will be described by the monotonic instability. It delimits the stable steady-states from unstable states, and its condition will be  $\text{Det } J=0$ , or (17):

$$-\kappa(\Gamma\Omega + \Sigma\Omega + \Sigma T + \Sigma^2 - \Phi T - \Phi\Sigma) - \varepsilon(\Gamma\Omega + \Sigma\Omega + \Sigma T - \Phi T + \Phi\Sigma) = 0 \quad (17)$$

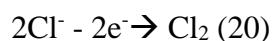
Also, if the reaction is realized in acidic media, sucralose fragment tends to dehalogenize, yielding a chloride ion. By this, the model will be transformed into (18):

$$\begin{cases} \frac{ds}{dt} = \frac{2}{\delta} \left( \frac{\Delta}{\delta} (s_0 - s) - r_s \right) \\ \frac{dp}{dt} = \frac{1}{P} (r_s - r_{r1}) \\ \frac{dq}{dt} = \frac{1}{Q} (r_{r1} - r_{r2} - r_d) \end{cases} \quad (18),$$

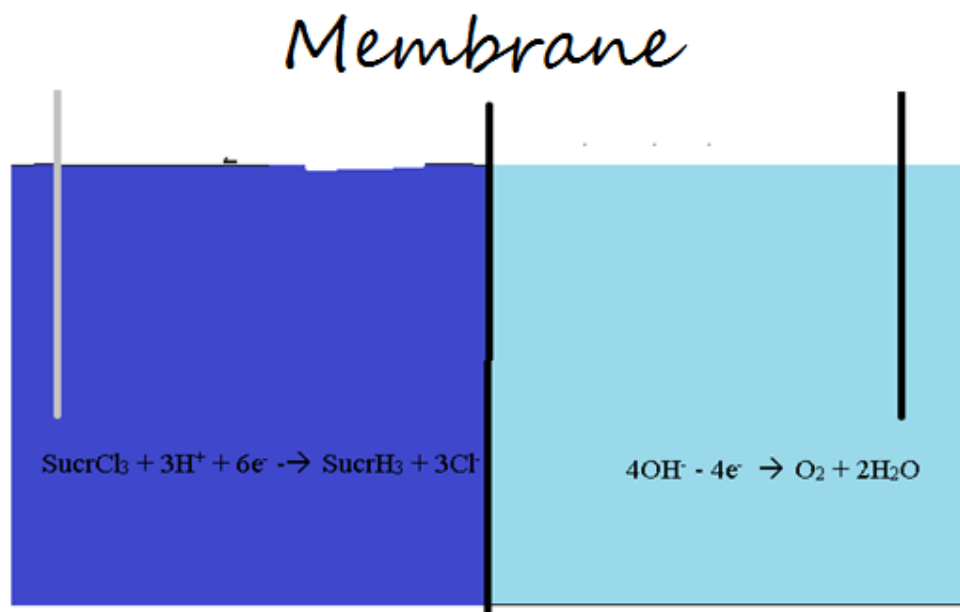
where  $r_d$  is the dehalogenation rate, described as:

$$r_d = k_d q \exp\left(-\frac{4nF\phi_0}{RT}\right) \quad (19)$$

As a chloride ion is formed during the dehalogenation, the membrane, impeding the chlorine evolution (20)

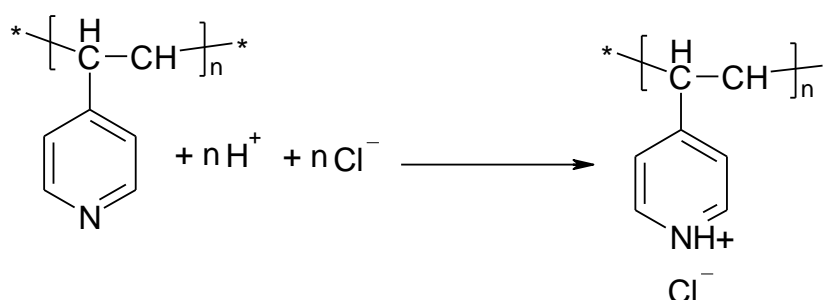


will be installed in order to separate the cathodic and anodic solutions. By this, the dehalogenation cell will be designed as in Figure 5:



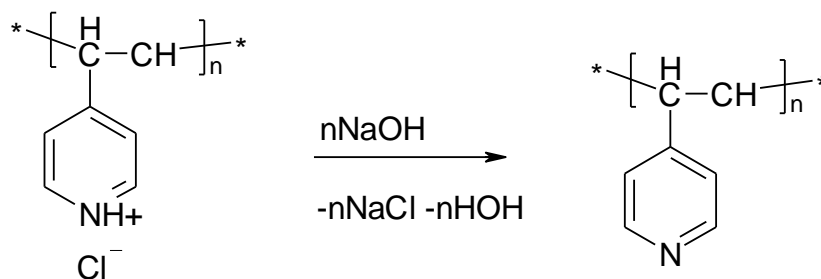
**Figure 5.** Sucralose dehalogenation cell.

The process will be thereby similar to [28]. The membrane is made of polyvinyl pyridines (PVP), which retain the chloride-ion by salt formation (Fig. 6):



**Figure 6.** Chlorine retention by PVP membrane

As for the membrane regeneration, it is realized as on the Figure 7:



**Figure 7.** Membrane regeneration

By maintenance of the sucralose elimination process, membrane confection, and regeneration rhythms, an economical and green sucralose elimination process is conducted.

#### 4. Conclusions

From the theoretical description of sucralose electrochemical determination over a conducting polymer containing the pyridinic nitrogen, it has been possible to conclude that the mechanism consists of the chemical and two gradual electrochemical stages, leading to the appearance of a determined analytical signal. The polymer may serve as an excellent modifier for sucralose quantification. The stable steady-state is maintained easily. The system is electroanalytical efficient. Depending on the electrolyte composition, electrode size, and the analyte concentration, the process may be diffusion- or kinetically controlled. The oscillatory behavior in this system is possible, being caused only by DEL influences of both electrochemical processes, like also of the chemical stage.

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#### Conflicts of Interest

The authors declare no conflict of interest.

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