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Sucralose and its Genotoxic Ester Electrochemical Determination and Elimination by Conducting Polymer Cathode: A Mathematical Modeling

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Abstract: For the first time, the electrochemical determination and elimination of sucralose in the presence of its genotoxic ester by a cathodic process has been described theoretically. The electrochemical process occurs at a conducting-polymer cathode, providing efficient removal of both compounds. From the theoretical modeling of sucralose and 6-acetylsucralose determination and removal, it is possible to conclude that the conducting polymer, based on a monomer containing an amino group or pyridinic nitrogen, may be efficient in the determination and removal of both compounds.

Keywords: sucralose; 6-acetylsucralose; electrochemical sensor; cathodic removal; conducting polymers; electrochemical oscillations; stable steady-state.

1. Introduction

Sucralose (Figure 1) is one of the most used sweeteners in Portugal and throughout the European Union in the alimentary and pharmaceutical industries as a flavor corrector [1-5]. In the USA, it is also known by its registered trademark, Splenda ®. In Codex Alimentarius, it is registered as E955. It is a trichloro-substituted derivative of galactosucrose, which has twice the sweetness of saccharin, triple the sweetness of aspartame, and is up to a thousand times as sweet as the common sugar. When it comes to physical properties, free sucralose is a white, shiny, odorless substance that is soluble in water.

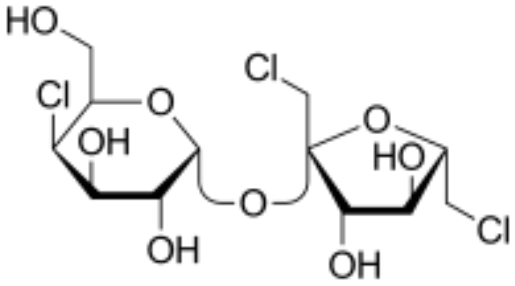

Figure 1. Sucralose.

Although sucralose is a derivative of natural compounds, it itself is not a natural compound. They synthesize it from sucrose in several steps (Figure 2).

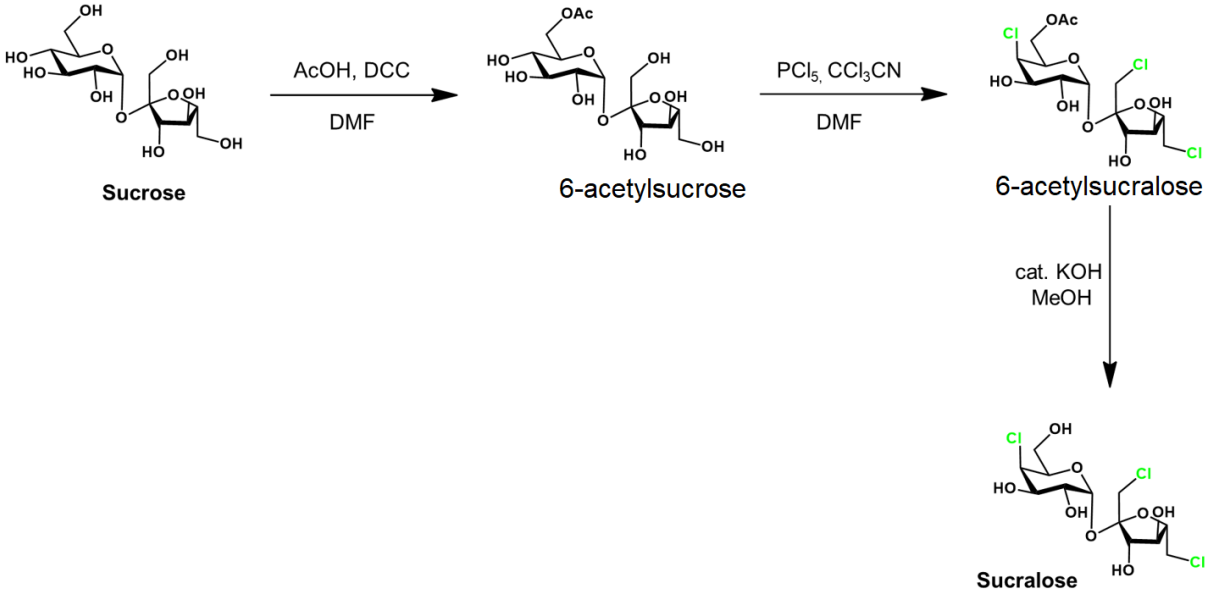

Figure 2. Sucralose synthesis from sucrose.

The acylation in the first stage may also be made by diethylazodicarboxylate. The C4 epimerization (mutarotation) is realized in the second stage. This synthesis involves either the toxic reagents (DCC, DAAD, and PCl_5) or the intermediate (6-acetylsucralose), as mentioned below.

Despite being considered safe for use by diabetics and athletes, its harmful effects on human health and the environment are still unknown, and some of its negative effects have only begun to be studied now. A recent study involving pregnant and breastfeeding women [6-8] confirms that sucralose enters breast milk, causing irreparable damage to the development of the gut microbiota of the human fetus in the last months of pregnancy, as well as in neonates and babies, which is why its safety for use during pregnancy and breastfeeding is still questioned.

Moreover, as sucralose is nearly non-biodegradable, it accumulates in the environment [9,10]. Furthermore, when sucralose decomposes thermally or by certain bacteria, it forms toxic compounds such as dioxins and tetrachlorodibenzofurans. It should not be forgotten that sucralose is also part of the group of halogen organic compounds (Figure 3). Therefore, the development of a method to eliminate sucralose from the environment, particularly from sewage and groundwater, is a pressing need [11,12].

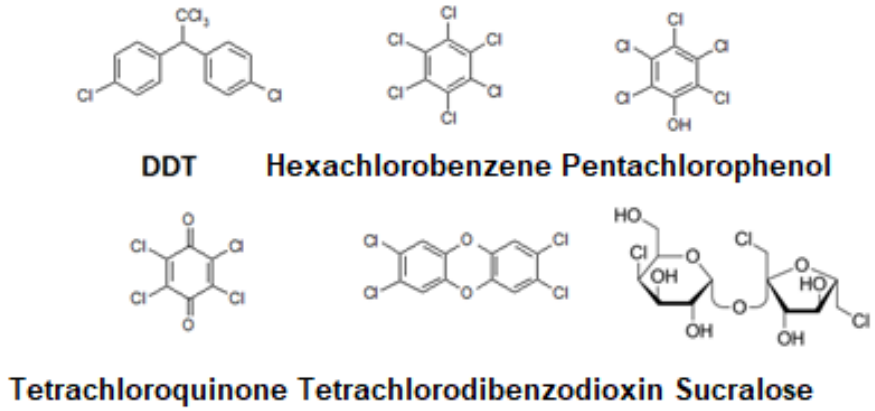

Figure 3. Sucralose is among the chloroorganic compounds.

It is also necessary to mention the presence of 6-acetylsucralose, the industrial precursor of the sweetener (Figure 2), in reasonable concentrations in samples of industrial sucralose. Moreover, its presence in the human intestine is likely, where sucralose is esterified at the C6-hydroxyl, which is the most active. Recent studies have proven the genotoxicity of 6-acetylsucralose [13,14].

Toxicological essays on sucralose esters showed that its genotoxic mechanism may be considered clastogenic (as it acts as an initiator of DNA structural ruptures). Even microscopic concentrations of 6-acetylsucralose, detected in industrial samples and beverages, exceed the safe threshold of 0.15 µg/person/day. 6-Acetylsucralose ester increased the expression of genes linked to inflammation, oxidative stress, and carcinogenesis, including MT1G and SHMT2 in intestinal epithelial cells. Another harmful action of the ester derivative is the inhibition of the activity of CYP1A2 and CYP2C19, proteins from the cytochrome P450 family, responsible for the transformation of several food substances into more readily available forms, leading to secondary toxic effects [13]. The increase in the genotoxicity of 6-acetylsucralose in relation to sucralose is due to the greater activity of the secondary organic chloride, linked to the C4 carbon atom (Figure 4), activated by the accepting action of the steric group. The activation of this same atom is also responsible for the mutagenic action of the substance.

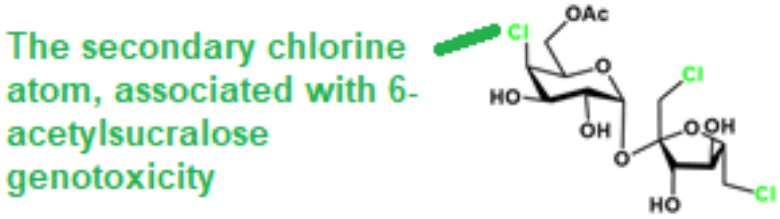

Figure 4. Secondary chlorine atom in 6-acetylsucralose.

For this reason, the electroanalytical detection of 6-acetylsucralose in the presence of sucralose is also an option to be considered, and, although the anodic process is also viable, the cathodic process would be more effective from an electroanalytical point of view, due to the

more pronounced difference in the behavior of 6-acetylsucralose and sucralose on the cathode than on the anode.

In previous works, the electroanalytical behavior of sucralose in anode and cathode processes was theoretically evaluated [15-18]. However, the possibility of including 6-acetylsucralose was not considered.

On the other hand, the development of new electrosynthesis, electroanalysis, and electrochemical conversion processes requires *a priori* theoretical investigation of the system's behavior. This investigation allows you to address problems such as the uncertainty surrounding some details of the electroanalytical process (e.g., how electroreduction is carried out under specific conditions and which modifiers could be used) and the possibility of instability characteristic of the electrooxidation of organic compounds, including electropolymerization [19-21].

Therefore, the general objective of this work is to evaluate the performance of the electrochemical detection of 6-acetylsucralose in the presence of sucralose through a cathodic process, assisted by conducting polymer containing the basic groups (organic amine and/or pyridinic nitrogen atoms). Furthermore, the behavior of this system will be compared with that of similar systems [15-18].

2. Materials and Methods

Both sucralose and 6-acetylsucralose may be reduced in acidic conditions with dechlorination. In this process, the protons replace organic chloride (1 – 2):

$$SucrCl_3 + 3H^+ + 6e^- \rightarrow SucrH_3 + 3Cl^- \quad (1)$$

Which Sucr stands for sucralose fragment.

However, the carboxylic group will be cathodically reduced in a faster step than dechlorination. The summary reduction reaction is given as (2):

$$SucrCl_3COCH_3 + 8H^+ + 11e^- \rightarrow C_2H_5OH + H_2O + SucrH_3 + 3Cl^- \quad (2)$$

If both of the substances are immobilized on the polymeric phase, which is achieved by the use of basic groups (amines and/or pyridinic nitrogen) in the polymer composition, both of the electrochemical stages (1 – 2) are realized in the polymer phase, providing an efficient peak separation.

Taking this into account and taking some assumptions [15-21], we describe the behavior of this system by a trivariate balance differential equation set (3):

$$\begin{cases} \frac{da}{dt} = \frac{2}{\delta}\left(\frac{\Delta}{\delta}(a_0 - a) - r_1\right) \\ \frac{ds}{dt} = \frac{2}{\delta}\left(\frac{S}{\delta}(s_0 - s) - r_2\right) \\ \frac{dp}{dt} = \frac{1}{P}(r_1 + r_2 - r_{r1} - r_{r2}) \end{cases} \quad (3)$$

Herein, a and s are acetylsucralose and sucralose pre-surface concentrations, a_0, and s_0 are their corresponding bulk concentrations, Δ and S stand for diffusion coefficients, δ is the pre-surface layer thickness, p is the modified polymer surface coverage degree, V is its maximal concentration, and the parameters r are the corresponding reaction rates, calculated as (4 – 7):

$$r_1 = k_1 a(1 - p)\exp(-ba) \quad (4)$$

$$r_2 = k_2 s(1-p)\exp(-bs) \quad (5)$$

$$r_{r1} = k_{r1} p \exp\left(-\frac{xF\varphi_0}{RT}\right) \quad (6)$$

$$r_{r2} = k_{r2} p \exp\left(-\frac{yF\varphi_0}{RT}\right) \quad (7)$$

Herein, the parameters k stand for the corresponding reaction rate constants, b is the parameter relating DEL electrochemical and electrophysical properties to ionic forms interchange during the chemical immobilization, x, and y are the numbers of electrons transferred during the electrochemical stage, F=N_A*e is the Faraday number, φ_0 is the zero-charge-related potential slope, R is the universal gas constant, and T is the absolute reaction temperature.

In relation to the use of vanadium oxyhydroxide, the probability of oscillatory and monotonic instability is enhanced. The conducting polymer will be efficient for the electrochemical detection and removal of sucralose and 6-acetylsucralose, as shown below.

3. Results and Discussion

We investigate the behavior of the system during electroanalytical detection of sucralose and its 6-acetyl derivative using linear stability theory. The stationary elements of the Jacobi functional matrix can be calculated as (8):

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

Sendo:

$$a_{11} = \frac{2}{\delta}\left(-\frac{\Delta}{\delta} - k_1(1-p)\exp(-ba) + bk_1 a(1-p)\exp(-ba)\right) \quad (9)$$

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

$$a_{13} = \frac{2}{\delta}(k_1 a \exp(-ba)) \quad (11)$$

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

$$a_{22} = \frac{2}{\delta}\left(-\frac{S}{\delta} - k_2(1-p)\exp(-bs) + bk_2 s(1-p)\exp(-bs)\right) \quad (13)$$

$$a_{23} = \frac{2}{\delta}(k_2 s \exp(-bs)) \quad (14)$$

$$a_{31} = \frac{1}{P}(k_1(1-p)\exp(-ba) - bk_1 a(1-p)\exp(-ba)) \quad (15)$$

$$a_{32} = \frac{1}{P}(k_2(1-p)\exp(-bs) - bk_2 s(1-p)\exp(-bs)) \quad (16)$$

$$a_{33} = \frac{1}{P}\left(-k_1 a\exp(-ba) - k_2 s\exp(-bs) - k_{r1}\exp\left(-\frac{xF\varphi_0}{RT}\right) - k_{r2} p\exp\left(-\frac{yF\varphi_0}{RT}\right) + j\left(k_{r1} p\exp\left(-\frac{xF\varphi_0}{RT}\right) + k_{r2} p\exp\left(-\frac{yF\varphi_0}{RT}\right)\right)\right) \quad (17)$$

Avoiding the cumbersome expressions during the determinant analysis, we rewrite the Jacobian determinant as (18):

$$\frac{4}{\delta^2 P}\begin{vmatrix} -\kappa - \Xi & 0 & T \\ 0 & -\xi - \Sigma & \Phi \\ \Xi & \Sigma & -T - \Phi - \Omega \end{vmatrix} \quad (18)$$

Opening the straight brackets, applying both the Routh-Hurwitz stability criterion and monotonic instability condition, and changing the signs to the opposite, we obtain the requirement set, expressed as (19):

$$\kappa(\xi T + \xi \Phi + \xi \Omega + \Sigma T + \Sigma \Omega) + \Xi(\xi \Phi + \xi \Omega + \Sigma \Omega) \begin{cases} > 0, & linear\ dependence \\ = 0, & detection\ limit \end{cases} \quad (19)$$

If –Det J>0, the Routh-Hurwitz stability criterion is valid, and the steady-state is thereby stable, providing an efficient steady-state determination of both of the chloroorganic compounds. Moreover, the wide stability region lets us use this system in pharmaceutical formulations and biological liquids, as well as in the environment. The polymerization scenario also provides efficient pesticide removal and immobilization in the polymer phase.

This criterion is readily satisfied if the kinetic parameters $\Sigma, \Xi,$ and Ω are positive. In the vast majority of cases, both have positive signs, and given that the other variables in the determinant are positive, this indicates a large, steady-state stability topological region. The electroanalytical process is mostly kinetically controlled, but with the possibility to transition to diffusion-controlled mode if pH → 0.

In the absence of the side reactions or other factors capable of compromising the analyte and (or) modifier stability, excluding the reactions foreseen by the mechanism, the linearity between the electrochemical parameter and concentration is observed, providing an efficient analytical signal interpretation, which is really important for drug concentration monitoring.

The condition Det J=0 corresponds to the detection limit, manifested by the *monotonic instability*. It may be seen as an N-shaped feature in the steady-state voltammogram, which depicts the boundary between stable and unstable steady states and corresponds to the steady-state multiplicity. In other words, multiple steady-states, each one unstable, coexist at this point.

As for the oscillatory behavior, it is observed beyond the detection limit in the case of the Hopf bifurcation. Its realization requires the presence of the positive-callback-related positive addenda in the main diagonal elements.

Observing the main diagonal elements (9), (13), and (17), we may observe that the oscillatory behavior becomes possible if the kinetic parameters b and j are positive, which corresponds to the DEL influences of all the chemical and electrochemical stages. This factor is typical for similar systems [15-21] and may be described by the positivity of the elements $bk_1a(1-p)\exp(-ba) > 0$ and $bk_2s(1-p)\exp(-bs) > 0$ if b>0, describing ionic forms cyclic transformations during the sucralose derivatives immobilization, and $j\left(k_{r1}p\exp\left(-\frac{xF\varphi_0}{RT}\right) + k_{r2}p\exp\left(-\frac{yF\varphi_0}{RT}\right)\right) > 0$, if j>0, describing the similar cyclic phenomena on both DEL and the surface during the electrochemical stages. These elements describe the positive callback, and this callback will depend on the system´s characteristics. For example, the oscillation frequency and amplitude depend on the background electrolyte composition, as demonstrated experimentally and theoretically [15-21].

As for the anodic process, different modifiers can also assist it. Nevertheless, as both analytes tend to hydrolyze in strongly alkaline media, yielding the same compound, a neutral medium is strongly recommended.

4. Conclusions

From the analysis of the process with the cathodic detection of sucralose and its 6-acetylsucralose ester, assisted by conducting polymer with basic groups (amino groups or pyridinic nitrogen), it is possible to conclude that in the present process, the polymer facilitates the obtaining and maintenance of the stable, steady state in this system, due to the efficient immobilization of the analyte. The electrochemical process is mostly kinetically controlled, passing to diffusion-controlled mode at low pH. The oscillatory behavior is possible in this system. Periodic effects on the structure of the double electric layer cause it. These effects are observed at both chemical and electrochemical stages.

Author Contributions

Conceptualization, T.V.M.; V.V.T.; L.V.R.; O.V.H.; L.N.N.; P.I.Y.; Z.Z.Y.; O.O.P.; N.Y.C.; S.P.V.; Y.G. I.; I.O.M.G. and G.M.P.; methodology, V.V.T.; M.J.M.; I.O.M.G.; A.O.S. V.M.O.; S.C.O.; O.O.V.; M.P.K.; B.S.S.; D.M.M.; J.R.G.; J.I.F.P.M.; Z.O.K.; A.V.G.; I.F.B.; N.A.S.; N.M.S.; N.M.K.; V.O.K..; validation, V.V.T.; I.O.M.G.; M.J.M.; A.O.S. V.M.O.; S.C.O.; O.O.V.; J.I.F.P.M.; Z.O.K.; A.V.G.; I.F.B.; I.M.K. and L.O.K.; formal analysis, V.V.T.; M.J.M.; A.O.S. V.M.O.; S.C.O.; O.O.V.; M.P.K.; B.S.S.; D.M.M.; J.R.G.; N.M.G.; N.P.D.; J.I.F.P.M.; Z.O.K.; A.V.G.; I.F.B.; N.A.S.; N.M.S.; N.M.K.; V.O.K.; I.O.M.G. and T.V.M.; investigation, V.V.T.; M.J.M.; A.O.S. V.M.O.; S.C.O.; O.O.V.; M.P.K.; B.S.S.; D.M.M.; J.R.G.; J.I.F.P.M.; Z.O.K.; A.V.G.; I.F.B.; N.A.S.; N.M.S.; N.M.K.; V.O.K.. and T.V.M.; resources, V.V.T.; I.O.M.G.; M.J.M.; A.O.S. V.M.O.; S.C.O.; O.O.V.; M.P.K.; B.S.S.; D.M.M.; J.R.G.; J.I.F.P.M.; Z.O.K.; A.V.G.; I.F.B.; N.A.S.; N.M.S.; N.M.K.; N.M.G.; N.P.D.; V.O.K.. and T.V.M.; data curation, V.V.T.; M.J.M.; A.O.S. V.M.O.; S.C.O.; O.O.V.; M.P.K.; B.S.S.; D.M.M.; J.R.G.; I.O.M.G.; J.I.F.P.M.; Z.O.K.; A.V.G.; I.F.B.; M.V.K.; N.A.S.; N.M.S.; N.M.K.; V.O.K.; N.M.G.; N.P.D. and T.V.M.; writing—original draft preparation, V.V.T.; M.J.M.; A.O.S. V.M.O.; S.C.O.; O.O.V.; M.P.K.; B.S.S.; D.M.M.; N.M.G.; N.P.D.; J.R.G.; J.I.F.P.M.; Z.O.K.; A.V.G.; I.F.B.; M.V.K.; N.A.S.; N.M.S.; N.M.K.; V.O.K.. and T.V.M.; writing—review and editing, V.V.T.; M.J.M.; A.O.S. V.M.O.; S.C.O.; O.O.V.; M.P.K.; B.S.S.; I.O.M.G.; D.M.M.; J.R.G.; J.I.F.P.M.; Z.O.K.; A.V.G.; N.M.G.; N.P.D.; I.F.B.; M.V.K.; N.A.S.; N.M.S.; N.M.K.; V.O.K.; I.M.K.; L.O.K. and T.V.M.; visualization, V.V.T.; M.J.M.; A.O.S.; I.O.M.G.; V.M.O.; S.C.O.; O.O.V.; M.P.K.; B.S.S.; D.M.M.; J.R.G.; J.I.F.P.M.; Z.O.K.; A.V.G.; I.F.B.; M.V.K.; N.A.S.; N.M.S.; N.M.K.; V.O.K.; I.M.K.; L.O.K. and T.V.M.;; supervision, V.V.T.; M.J.M.; I.O.M.G.;.; J.R.G.; J.I.F.P.M.; Z.O.K.; P.I.Y. and T.V.M.;; project administration, V.V.T.; M.J.M.; I.O.M.G. and J.I.F.P.M.. All authors have read and agreed to the published version of the manuscript.

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Informed Consent Statement

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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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