


Some Theoretical Aspects of Lugduname Electrochemical Determination over an Undoped Poly(Naphthoquinone)

Volodymyr V. Tkach^{1,2,*} , Marta V. Kushnir¹ , Sílvia C. de Oliveira² , Vitalii V. Lystvan³ , Inna M. Dytynchenko³ , Adriano O. da Silva⁴ , Yana G. Ivanushko⁵ , Anzhelika F. Molodnanu⁵ , Olga V. Luganska⁶ , Petro I. Yagodynets¹ , Zholt O. Kormosh⁷ 

¹ Chernivtsi National University, 58000, Kotsyubyns'ky Str. 2, Chernivtsi, Ukraine; nightwatcher2401@gmail.com (V.V.T.), marta.v.kushnir@gmail.com (M.V.K.), ved1988mid@rambler.ru (P.I.Y.);

² Universidade Federal de Mato Grosso do Sul, Av. Sen. Felinto. Müller, 1555, C/P. 549, 79074-460, Campo Grande, MS, Brazil; scolive@gmail.com (S.C.O.);

³ Zhytomyr State University, 10000, Velyka Berdychivska Str., 40, Zhytomyr, Ukraine; lystvan@ukr.net (V.V.L.), dimchem1@gmail.com (I.M.D.);

⁴ Universidade Federal do Oeste do Pará, Campus Juriti, Rua V. de Souza Andrade, s/n, 68170-000, Juriti, PA, Brazil; ollympio@gmail.com (A.O.S.);

⁵ Bukovinian State Medical University, 58001, Teatralna Sq. 9, Chernivtsi, Ukraine; yana_iv@ukr.net, molodnanu.anzhelika@bsmu.edu.ua (A.F.M.);

⁶ Zaporizhzhia National University, 69600, Zhukovsky Str. 66, Zaporizhzhia, Ukraine; 130805olga@gmail.com (O.V.L.);

⁷ East Ukrainian National University, 43000, Voli Ave., 13, Lutsk, Ukraine; zholt-1971@ukr.net, kormosh@eenu.edu.ua (Z.O.K.);

* Correspondence: nightwatcher2401@gmail.com (V.V.T.);

Scopus Author ID 55758299100

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Abstract: This work evaluates the possibility of the sweetener lugduname electrochemical determination over a non-doped poly(naphthoquinone). Two reaction scenarios, including polymer doping and prorogation, are included in a mathematical model, developed and analyzed by linear stability theory and bifurcation analysis. Considering the double electric layer (DEL) influences of all electrochemical stages, the oscillatory behavior is more probable than similar cases. Nevertheless, the stability analysis confirms the poly(naphthoquinones) as an efficient electrode modifier for lugduname determination.

Keywords: lugduname; conducting polymers; doping; electropolymerization; electrochemical sensors; stable steady-state.

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1. Introduction

Lugduname (Fig. 1) is one of the most potent sweeteners known [1]. It is estimated to be up to 300 000 (three hundred thousand) times as sweet as common sugar. It was developed by the Université de Lyon (France) in 1996, and its name is derived from the Latin name of the city (Lugdunum). It is a part of the family of the guanidine derivatives of acetic acid, which are found to be potent sweeteners.

It is limitedly used as a sweetener by the food and pharmaceutical industries. Also, it is used to study taste responses in animals [2].

Nevertheless, its use is limited, as its toxicity essays have not been finished yet. Moreover, it contains toxic groups, like nitrile [3,4], the toxicity of which is dose-related.

Therefore, the development of a method for lugduname determination is actual [5], and the electrochemical methods could give it a good service.

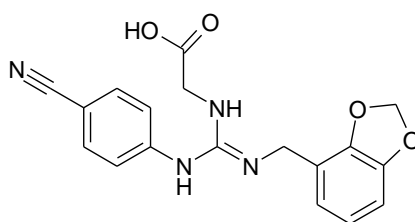


Figure 1. Lugduname.

As for now, no electrochemical methods for lugduname determination have been developed. Nevertheless, as lugduname contains electroactive functional groups, those methods may apply in cathodic and anodic ways.

Also, it contains methylenedioxyphenylene group [6-8], similar to popular monomer ethylenedioxythiophene [9-12], it tends to polymerize or copolymerize. Therefore, the conducting polymer as an electrode modifier, yet used for similar substances [13,14], could be a suitable approach for lugduname electrochemical determination.

Nevertheless, the development of novel electroanalytical methods requires an *a priori* theoretical investigation. It helps to detect the conditions for the efficient electrochemical determination of the analyte [15-21] and the possibility for the electrochemical instabilities, typical for the organic electrooxidation, including electropolymerization [22-24].

Therefore, this work aims to evaluate the possibility of undoped poly(naphthoquinone)-assisted lugduname electrochemical determination. It includes the suggestion of the mechanism of the appearance of the analytical signal, the development and analysis (in terms of stability) of the correspondent mathematical model, the derivation of the conditions and requirements for the stability and the electrochemical instabilities, and the comparison of the behavior of this system with that of the similar processes [25-28].

2. Materials and Methods

2.1. System and its modeling.

Considering that the lugduname is an electrolyte (carbonic acid), the sweetener (in the form of a salt) may be intercalated into a polymer matrix. On the other hand, the sweetener may react with the polymer, prolonging its chain or even making a bridge between the polymer chains (Fig. 2).

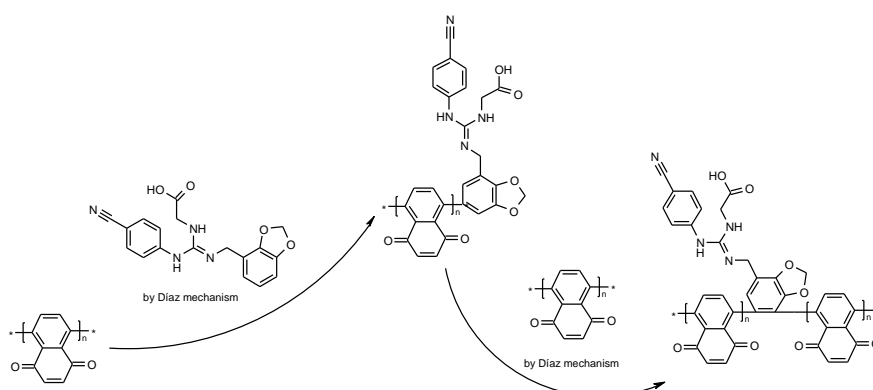


Figure 2. The scheme of “bridge” formation by lugduname.

This interaction may cause surface instability. As a matter of fact, EDOT has already been used to obtain “bridge polymers” not only for electroanalytic but also for OLEDs, OPEDs, capacitors, solar cells, etc. [11,12]. Therefore, this process could be interesting not only for electroanalytical but also for electrosynthetic purposes. The resulting polymer tends to be oxidized by guanidine fragment, yielding a benzo-dihydropyridine moiety by an intramolecular electrochemically assisted aromatic electrophilic substitution.

To describe this process in potentiostatic (amperometric) mode, we introduce two variables:

l —lugdunone concentration in the pre-surface layer;

p —modified polymer surface coverage degree.

Simplifying the modeling by some assumptions [25 – 28], we describe the system behavior by the bivariate equation-set:

$$\begin{cases} \frac{dl}{dt} = \frac{2}{\delta} \left(\frac{L}{\delta} (l_0 - l) - r_d - r_p \right) \\ \frac{dp}{dt} = \frac{1}{P} (r_d + r_p - r_o) \end{cases} \quad (1)$$

Where L is the diffusion coefficient, l_0 is the sweetener diffusion coefficient, P is the modified polymer maximal surface concentration, and the parameters r are the correspondent reaction rates, capable of being calculated as:

$$r_d = k_d l^x (1 - p)^y \exp(\alpha p) \exp\left(\frac{mF\varphi_0}{RT}\right) \quad (2)$$

$$r_p = k_p l^z (1 - p)^w \exp(\alpha p) \exp(\beta p) \exp\left(\frac{nF\varphi_0}{RT}\right) \quad (3)$$

$$r_o = k_o p \exp\left(\frac{2\lambda F\varphi_0}{RT}\right) \quad (4)$$

Herein, the parameters k are the correspondent reaction rate constants, x , y , z , and w are correspondent substances reaction orders, α and β are the parameters, describing the DEL ionic force and surface influences of the doping and polymerization reaction, λ is the number of the lugdunone moieties in the resulting polymer, F is the Faraday number, R is the universal gas constant, T is the absolute temperature and φ_0 is the potential slope, related to the zero-charge potential.

In this case, the electroanalytical system resembles the dye-assisted electropolymerization of an electrochemically synthesized monomer [25,26], but somehow more dynamic due to the surface instability. Nevertheless, the systems remain electroanalytical efficient, as shown below.

3. Results and Discussion

To investigate the system's behavior with the electrochemical determination of lugdunone on an undoped naphthoquinone, we analyze the equation-set (1) using the linear stability theory. The steady-state Jacobian matrix members may be expressed as:

$$\begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{pmatrix} \quad (5)$$

where:

$$a_{11} = \frac{2}{\delta} \left(-\frac{L}{\delta} - x k_d l^{x-1} (1 - p)^y \exp(\alpha p) \exp\left(\frac{mF\varphi_0}{RT}\right) - z k_p l^{z-1} (1 - p)^w \exp(\alpha p) \exp(\beta p) \exp\left(\frac{nF\varphi_0}{RT}\right) \right) \quad (6)$$

$$a_{12} = \frac{2}{\delta} \left(y(1-p)^{y-1} \exp(\alpha p) \exp \frac{mF\varphi_0}{RT} - \alpha(1-p)^y \exp(\alpha p) \exp \frac{mF\varphi_0}{RT} - jk_d l^x (1-p)^y \exp(\alpha p) \exp \frac{mF\varphi_0}{RT} + wk_p l^z (1-p)^{w-1} \exp(\alpha p) \exp(\beta p) \exp \frac{nF\varphi_0}{RT} - \alpha k_p l^z (1-p)^w \exp(\alpha p) \exp(\beta p) \exp \frac{nF\varphi_0}{RT} - jk_p l^z (1-p)^w \exp(\alpha p) \exp(\beta p) \exp \frac{nF\varphi_0}{RT} \right) \quad (7)$$

$$a_{21} = \frac{1}{p} \left(xk_d l^{x-1} (1-p)^y \exp(\alpha p) \exp \frac{mF\varphi_0}{RT} + zk_p l^{z-1} (1-p)^w \exp(\alpha p) \exp(\beta p) \exp \frac{nF\varphi_0}{RT} \right) \quad (8)$$

$$a_{22} = \frac{1}{p} \left(-y(1-p)^{y-1} \exp(\alpha p) \exp \frac{mF\varphi_0}{RT} + \alpha(1-p)^y \exp(\alpha p) \exp \frac{mF\varphi_0}{RT} + jk_d l^x (1-p)^y \exp(\alpha p) \exp \frac{mF\varphi_0}{RT} - wk_p l^z (1-p)^{w-1} \exp(\alpha p) \exp(\beta p) \exp \frac{nF\varphi_0}{RT} + \alpha k_p l^z (1-p)^w \exp(\alpha p) \exp(\beta p) \exp \frac{nF\varphi_0}{RT} + \beta k_p l^z (1-p)^w \exp(\alpha p) \exp(\beta p) \exp \frac{nF\varphi_0}{RT} + jk_p l^z (1-p)^w \exp(\alpha p) \exp(\beta p) \exp \frac{nF\varphi_0}{RT} - k_o \exp \frac{2\lambda F\varphi_0}{RT} + jk_o p \exp \frac{2\lambda F\varphi_0}{RT} \right) \quad (9)$$

The main singular point conditions may be joined in the table below (Table 1).

Table 1. The principal singular point condition.

| | |
|--------------------------------|-----------------|
| Steady-state stability | Tr J<0, Det J>0 |
| Oscillatory instability | Tr J=0, Det J>0 |
| Monotonic instability | Tr J<0, Det J=0 |

To simplify the determinant analysis by avoiding the cumbersome expressions, we introduce the determinant, rewriting the determinant as:

$$\frac{4}{\delta^2 p} \begin{vmatrix} -\kappa - \varepsilon & \Omega \\ \varepsilon & -\Omega - \Lambda \end{vmatrix} \quad (10)$$

Taking into account the second line of the table, the oscillatory behavior condition is:

$$\begin{cases} -\frac{2}{\delta} (\kappa + \varepsilon) - \frac{1}{p} (\Omega + \Lambda) = 0 \\ \kappa\Omega + \kappa\Lambda + \varepsilon\Lambda > 0 \end{cases} \quad (11)$$

The main condition for oscillatory behavior is Tr J=0. It may be satisfied if the main diagonal of the Jacobian matrix contains the positive elements responsible for the positive callback (as the negative callback is characteristic of all the systems).

In this system, the main diagonal element a_{22} contains six positive addendums, correspondent to the three factors causing oscillatory behavior in the system:

$\alpha(1-p)^y \exp(\alpha p) \exp \frac{mF\varphi_0}{RT} > 0$ and $\alpha k_p l^z (1-p)^w \exp(\alpha p) \exp(\beta p) \exp \frac{nF\varphi_0}{RT} > 0$ if $\alpha > 0$ depict the DEL influences of the polymer doping and chain propagation and bridging with the conducting polymer ionic forms' transformation;

$\beta k_p l^z (1-p)^w \exp(\alpha p) \exp(\beta p) \exp \frac{nF\varphi_0}{RT} > 0$ if $\beta > 0$, depicting the oscillatory influence of the surface instabilities caused by the attraction between macromolecules during the "bridging". That surface instability has already been observed either experimentally [22] or theoretically [26,27];

$jk_p l^z (1-p)^w \exp(\alpha p) \exp(\beta p) \exp \frac{nF\varphi_0}{RT} > 0$; $jk_d l^x (1-p)^y \exp(\alpha p) \exp \frac{mF\varphi_0}{RT} > 0$ and $jk_d l^x (1-p)^y \exp(\alpha p) \exp \frac{mF\varphi_0}{RT} > 0$, if $j > 0$, describe the instabilities caused by cyclic polymer capacitance and conductivity impact during the electrochemical reactions.

The oscillatory behavior will be more probable than similar systems [25-28]. The oscillation frequency and amplitude will be highly dependent on the composition of the background electrolyte. Nevertheless, the oscillatory instability is manifested far beyond the detection limit.

Taking into account the first line of the table, the steady-state stability requirement is exposed as:

$$\begin{cases} -\frac{2}{\delta}(\kappa + \mathcal{E}) - \frac{1}{P}(\Omega + \Lambda) < 0 \\ \kappa\Omega + \kappa\Lambda + \mathcal{E}\Lambda > 0 \end{cases} \quad (12)$$

It is readily satisfied if the parameters α , β , and j have nil or negative values, and the DEL influences aren't strong enough to destabilize the system. Suppose the mentioned parameters are nil or negative. In that case, the left side of the first inequation will have more negative values, and the left side of the second inequation will have more positive values, stabilizing the system.

As no side reactions capable of destabilizing the modifier or analyte are present in the system, the steady-state stability will correspond to the linear dependence between the lugduname concentration and the current. The electroanalytical process is efficient and both diffusion and kinetic-controlled.

As for the detection limit, its condition is described by the monotonic instability. It denominates the margin between the stable steady-states and unstable states, correspondent to (13)

$$\begin{cases} -\frac{2}{\delta}(\kappa + \mathcal{E}) - \frac{1}{P}(\Omega + \Lambda) < 0 \\ \kappa\Omega + \kappa\Lambda + \mathcal{E}\Lambda = 0 \end{cases} \quad (13)$$

The resulting polymer is also capable of being used as an electrode modifier for electroanalysis. One of the analytes, which may be used in such a system, is sucralose – another sweetener, 300 times less sweet than the lugduname. This electroanalytical system will be described in one of our next works.

4. Conclusions

A theoretical analysis of the possibility of lugduname electrochemical determination has been given. Assisted by an undoped poly(naphthoquinone) confirms that the system is electroanalytical efficient. The electroanalytical process is either diffusion or kinetically controlled. The system's behavior is more dynamic than that of similar systems due to the presence of both surface and electrochemical instabilities. Three factors are capable of causing oscillatory behavior. Nevertheless, the electrochemical oscillations may be realized far beyond the detection limit and do not influence the electroanalytical process.

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Conflicts of Interest

The authors declare no conflict of interest.

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