The mathematical description of dopamine electrochemical oxidation, accompanied by its chemical and electrochemical polymerization

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Abstract: The electrooxidation of dopamine is accompanied by its chemical and electrochemical polymerization, and in which either the monomer or the polymer may be oxidized to the respective quinonic form, was investigated from the theoretical point of view. Dopamine is one of the important neurotransmitters in human and mammalian organisms. It is a precursor to epinephrine, which influences the cardiovascular, hormonal and renal functions. Its lack causes diseases like Parkinson, therefore, dopamine has been used as a drug for their treatment. On the other hand, its excess stimulates the sympathetic nervous system yielding the metabolic disorders and even schizophrenia. Thus, the development of the rapid and accurate method for its concentration measurement is very important. Dopamine is very popular as a monomer for synthesis of a conducting polymer – polydopamine, which is used as electrodes’ modifier in capacitors and in anticorrosive coatings. The electropolymerization of dopamine into polydopamine proceeds along with its traditional quinone-hydroquinonic oxidation. Both processes give their impact to the electrochemical behavior of dopamine during its electropolymerization. The mechanism’s complexity is also responsible for the electrochemical instabilities during electro-oxidation. In order to understand these instabilities it’s necessary to develop the mathematical model that is capable to describe the behavior of the system. It also helps us to esteem the influence of the electrochemical instabilities, by which it may be accompanied. The goal of this work is to describe an electrochemical oxidation and polymerization of dopamine that will provide an important connection between the electrochemical detection of biologically active compounds and their electropolymerization for electrode modification.

Keywords: dopamine; polydopamine; electrooxidation; electropolymerization; mathematical model.

Introduction

Dopamine is a hormone and one of the major neurotransmitters in human and mammalian organisms [1-4] that is synthesized in the body. It is a member of the catecholamine family of neurotransmitters in the brain and is a precursor to epinephrine (adrenaline) and norepinephrine (noradrenaline) hormones [5]. Its lack causes illnesses like Parkinson disease [6], therefore it can be used as a drug in medical protocols [7]. Its excess can cause different effects on sympathetic segment of neurosystem, metabolic disturbance and even schizophrenia [8]. Therefore, the development of a novel method that is capable to detect its concentration in a rapid, precise, accurate and sensitive way is very important task.

Chemically modified electrodes have various advantages, the main of which is the affinity between the modifier and the analyte, reason why they are one of the modern, cheap and tunable electroanalytic tools. For example, for detection of hydroquinonic compounds, various electrode modifiers of a different nature and composition were developed [9-15]. Some hydroquinonic and quinonic compounds may also serve as electrode...
modifies [16–17]. Dopamine is a polymerizable compound [18] and its polymer can be used as an electrode modifier similarly as of polyaniline [19]. Moreover, if the supporting electrolyte contains oxidizing ions, they can promote chemical polymerization of dopamine. In order to evaluate the effect of polymerization by the electrochemical (electroanalytical) process it’s necessary to investigate the system with polymerization-accompanied electrochemical oxidation of dopamine from mechanistic point of view.

Therefore, it’s important to develop and analyze the mathematical model that is capable to describe the behavior of the system. It will be useful us to evaluate the influence of the electrochemical instabilities that can be occurred [20–21].

The goal of this work is to evaluate by mechanistic way the influence of chemical and electrochemical polymerization of dopamine and introduce an important connection between the electrochemical detection of biologically active compounds and their electropolymerization.

System and its modeling

The dopamine polymerization is occurred in electrode potential lower than for benzolic compounds. It is due to the presence of donor groups (two hydroxyls and an ethylamine group) in the benzene ring, therefore, the number of chemical oxidants that is capable to promote polymerization, has to be higher compared to benzene. The bond between monomer units is formed either by participation of oxygen, or by creating C-C bond between positions 3 and 6 of the ring.

While the hydroquinonic structure is present (at least in part) in the polymer backbone, the polymer can be also oxidized further to its quinonic form. The oxidation of hydroquinonic monomer units makes the system similar to so called “polythiophene paradox” [22]. The electrochemical oxidation of the dopamine polymer is reversible in nature while overoxidation of the same polymer is irreversible. Therefore we can safely assume that these processes are different.

Taking in account the described assumptions, we are introducing three variables:

- \( c \) – the dopamine concentration in the pre-surface layer;
- \( \theta \) – dopamine coverage degree;
- \( \theta_p \) – polydopamine (chemically, or electrochemically obtained) coverage degree.

In order to simplify the mathematical model, we introduce several assumptions:

- the background electrolyte is present in an excess, therefore we can disregard the migration flow and the oxidizing dopant concentration change;
- the reactor is intensively stirred, so we can disregard the influence of convection flow;
- the concentration profile of dopamine in the pre-surface layer is linear;
- the thickness of pre-surface layer is constant and equal to \( \delta \).

It is possible to demonstrate that the differential equations’ set that are describing the electrochemical oxidation system can be defined as following:

\[
\begin{align*}
\frac{dc}{dt} &= \frac{2}{\delta} \left( \frac{\Delta}{\delta} (c_0 - c) + r_1 - r_3 \right) \\
\frac{d\theta}{dt} &= \frac{1}{G} \left( r_1 - r_1 - r_2 - r_{p1} - r_{p2} \right) \\
\frac{d\theta}{dt} &= \frac{1}{J} \left( r_{p1} + r_{p2} - r_3 \right)
\end{align*}
\]

where \( \Delta \) is the dopamine diffusion coefficient to from electrolyte to the surface, \( c_0 \) is its concentration in the solution, \( G \) and \( J \) are the maximal surface concentration of the dopamine and its polymer correspondingly, and the parameters \( r \) are the dopamine rates of adsorption \( (r_1) \), desorption \( (r_1) \), electrooxidation \( (r_2) \), chemical \( (r_{p1}) \) and electrochemical \( (r_{p2}) \) polymerization, and the rate of polydopamine oxidation \( (r_3) \). These rates can be express as following:

\[
\begin{align*}
  r_1 &= k_1 c (1 - \theta - \theta_p) \exp(\alpha \theta); \\
  r_1 &= k_1 \theta \exp(-\alpha \theta); \\
  r_2 &= k_2 \theta \exp\left(\frac{2F \gamma \theta}{RT}\right); \\
  r_{p1} &= k_p \theta^n \exp\left(\frac{2F \gamma \theta}{RT}\right) f(\theta_p); \\
  r_{p2} &= k_p \theta^n f(\theta_p); \\
  r_3 &= k_3 \theta_p \exp\left(\frac{jF \xi \theta_p}{RT}\right)
\end{align*}
\]

where the parameters \( k \) are the correspondent reaction rate constants, parameter \( \alpha \) is a variable, which describes the interaction between the dopamine adsorbed molecules, \( F \) is the Faraday number, \( z \) and \( j \) are the numbers of transferred electrons during the polymer formation and oxidation correspondingly, \( \gamma \) and \( \xi \) are parameters that describes the influences of the electrochemical processes on the double electric layer (DEL) capacitances, \( R \) is the universal gas constant, \( T \) is the absolute temperature of the solution, \( f \) is the function that describes the autocatalytic reaction of the dopamine polymerization and relates to the polymerization’s reaction order.
The electrochemical dopamine polymerization system is similar to “the polyaniline paradox” that combines nearly all variables. Its behavior, however, is slightly different, and will be discussed in the next section.

Results and discussion

In order to understand and investigate the behavior of the electrochemical system that includes dopamine’s electrooxidation and polymerization we apply a linear stability theory to equation set (1). Its steady-state Jacobian matrix members can be described as:

\[
\begin{pmatrix}
a_{11} & a_{12} & a_{13} \\
a_{21} & a_{22} & a_{23} \\
a_{31} & a_{32} & a_{33}
\end{pmatrix}
\]  

where variables can be described by Equations 9-17 (Figure 1).

\[
a_{11} = 2\left( \frac{\Delta}{\delta} - k_1(1 - \theta - \theta_p) \exp(\alpha \theta) \right)
\]

\[
a_{12} = 2\left( k_1 c \exp(\alpha \theta) - ak_1 c(1 - \theta - \theta_p) \exp(\alpha \theta) + k_{-1} \exp(-\alpha \theta) - \alpha k_{-1} \theta \exp(-\alpha \theta) \right)
\]

\[
a_{13} = 2\left( k_1 c \exp(\alpha \theta) \right)
\]

\[
a_{21} = \frac{1}{G} \left( k_1(1 - \theta - \theta_p) \exp(\alpha \theta) \right)
\]

\[
a_{22} = \frac{1}{G} \left( -k_1 c \exp(\alpha \theta) + ak_1 c(1 - \theta - \theta_p) \exp(\alpha \theta) - k_{-1} \exp(-\alpha \theta) + \alpha k_{-1} \theta \exp(-\alpha \theta) - \right.

\left. - k_2 \exp\left( \frac{2F\gamma\theta}{RT} \right) - \gamma k_2 \theta \exp\left( \frac{2F\gamma\theta}{RT} \right) - nk_p \theta^{n-1} \exp\left( \frac{zF\gamma\theta}{RT} \right)f(\theta_p) - \gamma k_p \theta^n \exp\left( \frac{zF\gamma\theta}{RT} \right)f(\theta_p) \right)
\]

\[
a_{23} = \frac{1}{G} \left( -k_1 c \exp(\alpha \theta) - f'(\theta_p) (k_p \theta^n) \exp\left( \frac{zF\gamma\theta}{RT} \right) + k_p \theta^n \right)
\]

\[
a_{31} = 0
\]

\[
a_{32} = \frac{1}{J} \left( nk_p \theta^{n-1} \exp\left( \frac{zF\gamma\theta}{RT} \right)f(\theta_p) + \gamma k_p \theta^n \exp\left( \frac{zF\gamma\theta}{RT} \right)f(\theta_p) \right)
\]

\[
a_{33} = \frac{1}{J} \left( f'(\theta_p) (k_p \theta^n) \exp\left( \frac{zF\gamma\theta}{RT} \right) + k_p \theta^n) - k_3 \exp\left( \frac{jF\xi\theta_p}{RT} \right) - \xi k_p \theta_p \exp\left( \frac{jF\xi\theta_p}{RT} \right) \right)
\]

\[
(-\xi - \Xi)(\Lambda \Sigma + \Omega \Sigma + P \Sigma + PY + PK - K \Lambda - K \Omega - KP) + \Xi(PY - \Lambda K + \Lambda \Sigma) < 0
\]

\[
(-\xi - \Xi)(\Lambda \Sigma + \Omega \Sigma + P \Sigma + PY + PK - K \Lambda - K \Omega - KP) + \Xi(PY - \Lambda K + \Lambda \Sigma) = 0
\]

Figure 1. The series of Equations 9-17 and 30-31.
Observing the expressions (8), (12) and (15), it’s possible to demonstrate the oscillatory behavior of the system. Moreover, it is even more likely occurrence compare to similar systems [20-21]. It happens because the main matrix diagonal contains more positive elements compare to mathematical models for similar systems [20-21, 23-24].

The oscillatory behavior will be observed under following conditions:

- attraction between the adsorbed molecules that are represented by the positivity of the element $+ak_c(1-\theta - \theta_p)\exp(\alpha \theta)$ and of $+ak_c\theta \exp(-\alpha \theta)$.
- strong influence of electrochemical processes on DEL capacitances. This cause is common to all similar systems and represent the electrochemical instability the oscillation amplitudes and electrolyte’s composition [20]. In the case of dopamine three electrochemical processes are taking place that can cause changes to electrolyte’s conductivity (increases or decreases). Therefore, the DEL capacitance will have its value altered, which will cause the electrochemical oscillations. Mathematically, they are described by the positivity of the elements

$$-\partial \Theta \exp\left(\frac{jF\xi \Theta \Theta_p}{RT}\right), \quad -\gamma k_2 \theta \exp\left(\frac{2F\gamma \Theta}{RT}\right)$$

(18) \hspace{1cm} (19)

and

$$-\gamma k_p \Theta^n \exp\left(\frac{zF\gamma \Theta}{RT}\right)f(\theta_p)$$

(20)

which make part of the main diagonal elements $a_{22}$ and $a_{33}$.

- the autocatalysis during polymer formation can occur because of rapid transference of an olygomer and a polymer compare to a monomer. This factor as a cause of the oscillatory behavior occurs during the “polythiophene paradox”. Mathematically it can be described by the positivity of the element

$$f'(\theta_p)(k_p \Theta^n \exp\left(\frac{zF\gamma \Theta}{RT}\right)+k_p \Theta^n)$$

(21)

The oscillatory behavior has three possible causing factors compare to the two factors that are existing in the general [23]. Ther factors are being similar to the polythiophene paradox [24].

In order to evaluate the steady-state stability we apply the Routh-Hurwitz stability criterion to the differential equations’ set (1). To avoid the appearance of the cumbersome expressions we introduce new variables:

$$\begin{align*}
\Delta/\delta &= \kappa \quad (22) \\
k_1(1-\theta - \theta_p)\exp(\alpha \theta) &= \Xi \quad (23) \\
k_c\exp(\alpha \theta) - ak_c(1-\theta - \theta_p)\exp(\alpha \theta) + k_c\exp(-\alpha \theta) - ak_c\theta \exp(-\alpha \theta) &= \Lambda \quad (24) \\
k_c\exp(\alpha \theta) &= Y \quad (25) \\
k_2\exp\left(\frac{2F\gamma \Theta}{RT}\right) + \gamma k_2 \theta \exp\left(\frac{2F\gamma \Theta}{RT}\right) &= \Omega \quad (26) \\
nk_p \Theta^n - \exp\left(\frac{zF\gamma \Theta}{RT}\right)f(\theta_p) + \gamma k_p \Theta^n \exp\left(\frac{zF\gamma \Theta}{RT}\right)f(\theta_p) &= P \quad (27) \\
k_3\exp\left(\frac{JF\xi \Theta \Theta_p}{RT}\right) - \partial \Theta \exp\left(\frac{JF\xi \Theta \Theta_p}{RT}\right) &= \Sigma \quad (28)
\end{align*}$$

so the Jacobian determinant can be described as following:

$$\frac{2}{DGJ}\begin{vmatrix}
-\kappa - \Xi & \Lambda & Y \\
\Xi & -\Lambda - \Omega - P & -Y - K \\
0 & P & K - \Sigma
\end{vmatrix}$$

(29)

Opening the brackets and applying to the determinant the requirement $Det J < 0$ that is derived from the criterion we can obtain the steady-state stability condition that is described in Equation 30 (Figure 1).

The topological area of satisfaction of the inequation (30) is less than in the similar cases, including even the case of the polythiophene paradox [20-24]. Nevertheless, the steady-state stability is easy to maintain as it will be warranted to be stable if:

- the repulsion between the adsorbed particles that is described by the positivity of the parameter $\Lambda$ when the parameter $\alpha$ is negative. Together with the satisfaction of the conditions exposed below, the element $\Lambda \Sigma$ is maintained positive, and the left-side expression of the inequation is more negative;
- the influence of the electrochemical processes to the DEL capacitances that is described by the positivities of the parameters $\Omega$, $\Sigma$ and $\Lambda$. Each parameter corresponds to the certain electrochemical process – electrooxidation ($\Omega$), electropolymerization ($\Sigma$) and the polymer electrooxidation ($\Lambda$). In the case of the positivity of the these parameters the expression $\Lambda \Sigma + \Omega \Sigma + P \Sigma + P \Sigma$ will have the a positive value, and it will “push” the left-side expression of the
inequality (30) to more negative values, resulting in its satisfaction;

• the absence or fragility of autocatalytic effect during electropolymerization. If the autocatalysis isn’t realized, \( f(0) = \text{const} \) and \( f(0) = 0 \), which annihilates the elements containing the parameter \( K \), the nullity or negativity of which “pushes” the left-side expression of the inequality (30) to more negative values, satisfying the steady-state stability condition.

Depending on dopamine concentration and on electrode’s shape the electrochemical process can be diffusion-controlled or adsorption-controlled.

It will correspond to a linear dependence between an electrochemical parameter and a dopamine concentration from the electroanalytical point of view. It will correspond to a polymer formation from electro synthetic point of view.

The monotonic instability is also probable if the destabilizing and stabilizing influences are equal and it relates to a detection limit from the electroanalytical point of view. It will be caused by an autocatalysis and its conditions can be described by Equation 31 (Figure 2).

Not only dopamine, but also other compounds that are having a hydroquinonic structural characteristics and active sites for electropolymerization can undergo the process that is described in this work. For example, acetaminophen (paracetamole) can be also polymerized [25]. Its electrochemical detection on poly- (aniline blue) electrodes was reported [26] and described theoretically [27]. The section 2 reported electropolymerization that was accompanied by electrochemical detection of paracetamol over polymeric surface. Our model can be used to described polycetaminophen electrooxidation.

Conclusions

The analysis of dopamine electrooxidation that is accompanied by chemical and electrochemical polymerization allow us to conclude that:

• the stable steady-state, despite to the narrower parameter topological zone, can be easily maintain. The factors, which are warranting the steady-state stability, are repulsion between particles, fragility of DEL influences of electrochemical processes and the absence or fragility of autocatalysis

• depending on dopamine concentration, the electrode area and the presence of active sites, the process will be controlled by diffusion or by adsorption;

• the oscillatory behavior in this case is more probable than in the cases of electropolymerization. It caused by a surface, an electrochemical and an autocatalytical factors;

• the monotonic instability of this system can appear and can be caused by autocatalysis in electropolymerization.

Notes

The authors declare no conflict of interest.

Author contributions. The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

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Математичний опис електроокиснення допаміну, що супроводжується його хімічною та електрохімічною полімеризацією

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Резюме: Цікавий випадок електроокиснення допаміну, що супроводжується його хімічною та електрохімічною полімеризацією, у вимовах якої як меномер, так і полімер можуть електрохімічно окиснюватися, досліджено з теоретичної точки зору. Допамін – один із найважливіших нейротрансмітерів в людських організмах, а також в організмах інших щетинистих тварин. Він є ключовим регулятором функції серцево-судинної системи, гормонального апарату, нирок тощо. Його нестача стає причиною зниження дії адаптивних систем, які регулюють функцію відповідних органів

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