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Cathodic Determination of Naproxene and Paracetamol in Biological Liquids and Wastewater: A Theoretical Insight

Volodymyr V. Tkach [1,*], Marta V. Kushnir [1], Sílvio C. de Oliveira [2], Yana G. Ivanushko [3], Viktor V. Kryvetskyi [3], Inna I. Kryvetska [3], Igor V. Kryvetskyi [3], Petro I. Yagodynets [1,*], Adriano O. da Silva [4], Nataliia P. Derevianko [5], Mykhailo P`. Zavhorodnii [5], Nataliia S. Serhata [5], Mykola O. Serhatyi [5], Kateryna V. Bandurina [5], Tetiana V. Baryshok [5], Vira M. Odyntsova [6], Mykola P. Krasko [6], Ivan M. Bilai [6], Andrii I. Bilai [6], Valerii P. Moroz [7], Laziz N. Niyazov [8], Tetiana V. Morozova [9], Jarem R. Garcia [10], José Inácio Ferrão da Paiva Martins [11], Zholt O. Kormosh [12], Alla V. Grekova [13], Ianina F. Burdina [13], Oleg P. Melnyk [14], Oleksii O. Melnyk [14], Maria V. Melnyk [14], Maria João Monteiro [15]

1 Chernivtsi National University, 58001, Kotsyubynsky Str. 2, Chernivtsi, Ukraine
2 Institute of Chemistry. Federal University of Mato Grosso do Sul, 79074 – 460, Av. Sen. Felinto Müller, 1555, Vila Ipiranga, Campo Grande, MS, Brazil
3 Bukovinian State Medical University, 58001, TeatralnaSq, 9, Chernivtsi, Ukraine
4 Federal University of the West of Pará, Juruti Campus, 68170 – 000, Rua Veríssimo de Souza Andrade, s/n, Juruti, PA, Brazil
5 Khortytska Natsionalna Navchalno-Reabilitatsiyna Akademiya, 69000, Naukove Mistechko, 59, Khortytsia Island, Zaporizhzhia, Ukraine
6 Zaporizhzhia State Medical University, 69600, Mayakovsky Ave. 24, Zaporizhzhia, Ukraine
7 National Pharmaceutical University, 61000, Pushkinska Str. 57, Kharkiv, Ukraine
8 Abu Ali Ibn Sino Bukhara State Medical Institute, 705018, Navoi Str., 1, Bukhara, Uzbekistan
9 National Transport University, 02000, Omelianovych-Pavlenko Str. 1, Kyiv, Ukraine
10 State University of Ponta Grossa, Uvaranas Campus, Av. Gal. Carlos Cavalcanti, 4748, 84030-900, Ponta Grossa, PR, Brazil
11 Engineering Faculty of the University of Porto, 4200-465, Rua Dr. Roberto Frias, s/n, Porto, Portugal
12 Volyn National University, 43000, Voli Ave., 13, Lutsk, Ukraine
13 Odesa National Medical University, 65000, Valikhovsky Ln. 2, Odesa, Ukraine
14 National University of Life and Environmental Science of Ukraine, 03041, Heroiv Oborony Str, 15, Kyiv, Ukraine
15 Universidade de Trás-os-Montes e Alto Douro, Quinta de Prados, 5000-801 Vila Real, Portugal * Correspondence: nightwatcher2401@gmail.com (V.V.T.), ved1988mid@rambler.ru (P. I.Y.) ;

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Abstract: The possibility of the cathodical determination of naproxene and paracetamol has been theoretically analyzed. The process may be realized in neutral and mildly acidic media, in which the conducting polymer is recommended as an electrode modifier. From the steady-state stability point of view, the electroanalytical process is stable and efficient. Nevertheless, the topological zone of the linear dependence between the analytes´ concentration and electrochemical parameter becomes dependent on the conducting polymer nature being different for ionic and non-ionic monomers.

Keywords: naproxene; paracetamol; electrochemical sensors; conducting polymers; stable steady-state.

1. Introduction

Paracetamol or acetaminophen (Figure 1 to the left) [1-4] is an acylated phenolamine, introduced to the pharmaceutical practice in 1893 by Von Mering as an analgesic drug for adults and children. Its action is based on the inhibition of prostaglandin synthesis in the central nervous system. Nevertheless, its side effects include the liver intoxication leading even to the death [5]. Therefore, the development of new methods for its determination is really actual [6-8].

On the other hand, naproxen [9–12] is a non-steroid analgesic drug based on 2-β-naphthylpropionic acid. Its action is based on cyclooxidases inhibition. It is used in the treatment of muscular pains, convulsions, and edemas. Nevertheless, it is contraindicated to babies and children till 2 years old, like also to people with asthma, gastrointestinal diseases, and gastric ulcers. The side effects include sleepiness, fatigue, and depression [13–15]. So, the development of the precise and exact methods for its determination in different conditions is actual (Figure 1) [16–20].

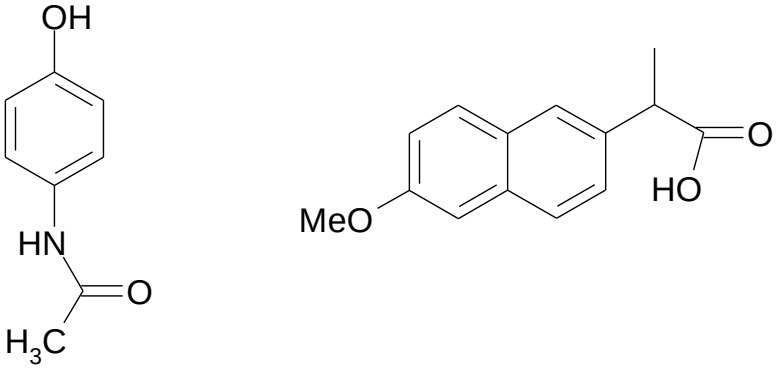

Figure 1. Paracetamol (left side) and naproxen (right side).

Taking into account the structures of paracetamol and naproxen, it is possible to conclude that both of them are electrochemically active, so the electroanalytical processes are applicable to them [21–35]. Both cathodic and anodic processes may be applicable, the cathodical being the most compatible with both of them due to the presence of strong, accepting functional groups.

The electrochemical reduction may be given in a neutral or acidic medium on a conducting-polymer-modified electrode [28–35], suitable for the proton transfer mediation. Conducting polymers are suitable electrode modifiers, as they are capable of combining plastic properties with metal conductivities, which is the reason why they are called synthetic metals. Moreover, they possess tunable properties, adjusted by monomer (carbo- or heterocyclic compound or a dye), polymerization technique (direct or indirect, anodic or cathodic), dopant, and doping degree chosen. For this reason, they are popular in electroanalytics and widely used in sensors and biosensors.

Nevertheless, the double electric layer (DEL) effect on the electrochemical process and its stability manifestation will also depend on the monomer nature, and it will strongly influence the sensor response. For this reason, the goal of our work is to evaluate theoretically the behavior of the electrochemical determination of naproxene and paracetamol in the acid medium over conducting polymer. This theoretical evaluation will include the suggestion of the most probable reaction mechanism, development, and analysis of the correspondent mathematical model, valid for the polymers of both ionic and non-ionic monomers, and also to compare the behavior of this system with that of the similar ones [36–42].

2. Materials and Methods

In an acidic medium, the conducting polymer becomes reduced, and the reduced form provides the proton transfer to both of the analytes. Both naproxen and paracetamol become reduced by the carbonyl group. Nevertheless, paracetamol may also be reduced with CO – NH bond cleavage, permitting two scenarios of its reduction. Schematically, the electroanalytical process is described in the Figure 2.

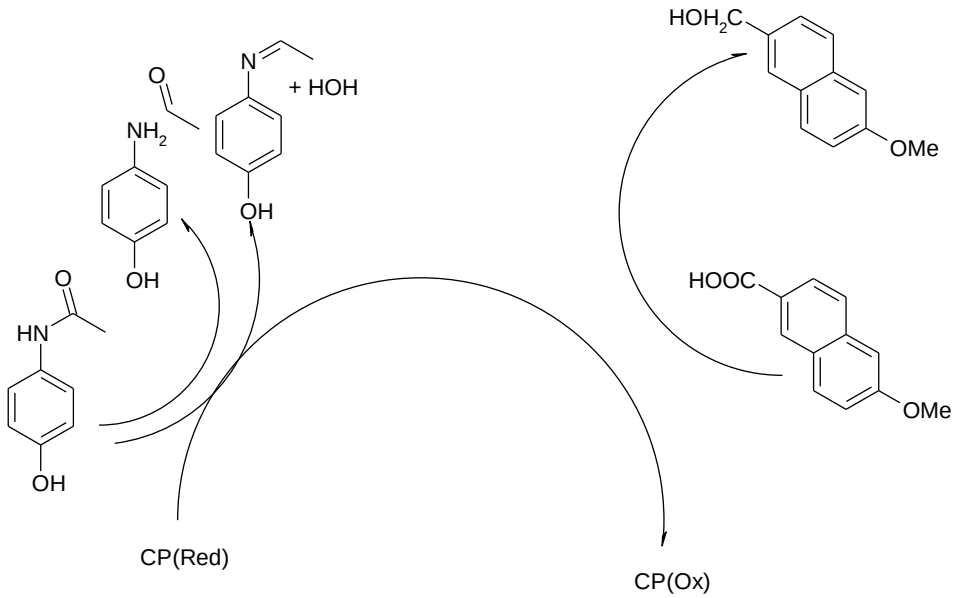

Figure 2. The scheme of the electroanalytical process.

And mathematically, considering certain suppositions [36–42], by a trivariant balance equation-set (1):

$$\begin{cases} \frac{dn}{dt} = \frac{2}{\delta}\left(\frac{N}{\delta}(n_0 - n) - r_n\right) \\ \frac{dp}{dt} = \frac{2}{\delta}\left(\frac{P}{\delta}(p_0 - p) - r_{p1} - r_{p2}\right) \\ \frac{dm}{dt} = \frac{1}{M}\left(r_n + r_{p1} + r_{p2} - r_r\right) \end{cases} \quad (1)$$

This, compared to the anodic route [36], is characterized by the role changes of naproxen and paracetamol in terms of the hybridity of the chemical transformation mechanism.

Herein, P and N are paracetamol and naproxen diffusion coefficients, p_0 and n_0 are their bulk concentrations, m is the modified macromolecule surface coverage degree, M is the maximal modified macromolecule surface concentration, and the parameters r are the correspondent reaction rates, calculated as:

$$r_n = k_n n(1-m)^4 \exp(\alpha m) \quad (2)$$

$$r_{p1} = k_{p1} p(1-m)^2 \exp(\alpha m) \quad (3)$$

$$r_{p2} = k_{p2} p(1-m)^2 \exp(\alpha m) \quad (4)$$

$$r_r = k_r m^x \exp\left(-\frac{yF\varphi_0}{RT}\right) \quad (5)$$

In which the parameters k stand for the correspondent reaction rate constants, the parameter α relates the ionic forms transformation with the DEL and surface electrophysical and electrochemical properties:

$$\alpha \begin{cases} = 0, if\ the\ monomer\ is\ not\ ionized\ in\ acidic\ media \\ \neq 0, if\ the\ monomer\ becomes\ ionized\ in\ acids \end{cases} \quad (6)$$

x stands for the polymer, F is the Faraday number, y is the number of transferred electrons, φ_0 is the potential slope corresponding to the zero-charge potential, R is the universal gas constant, and T is the absolute temperature.

The behavior of the system will be slightly similar to that described in [36] but with the analyte role changes in the reaction mechanism. This will foresee the efficiency of the electroanalytical process. On the other hand, the topological zone for linear dependence between the electrochemical parameter and concentration will be dependent on the monomer nature, as shown below.

3. Results and Discussion

We describe the electrochemical behavior of the electroanalytical process of cathodic CP-assisted naproxene and paracetamol determination by linear stability theory and expose the steady-state Jacobian matrix members as (7):

$$\begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix} \quad (7)$$

Herein:

$$a_{11} = \frac{2}{\delta}\left(-\frac{N}{\delta} - k_n(1-m)^4 \exp(\alpha m)\right) \quad (8)$$

$$a_{12} = 0 \quad (9)$$

$$a_{13} = \frac{2}{\delta}(4k_n n(1-m)^3 \exp(\alpha m) - \alpha k_n n(1-m)^4 \exp(\alpha m)) \quad (10)$$

$$a_{21} = 0 \quad (11)$$

$$a_{22} = \frac{2}{\delta}\left(-\frac{P}{\delta} - k_{p1}(1-m)^2 \exp(\alpha m) - k_{p2}(1-m)^2 \exp(\alpha m)\right) \quad (12)$$

$$a_{23} = \frac{2}{\delta}\left(2k_{p1}p(1-m)^{\square} \exp(\alpha m) - \alpha k_{p1}p(1-m)^2 \exp(\alpha m) + 2k_{p2}p(1-m)^{\square} \exp(\alpha m) - \alpha k_{p2}p(1-m)^2 \exp(\alpha m)\right) \quad (13)$$

$$a_{31} = \frac{1}{M}(k_n(1-m)^4 \exp(\alpha m)) \quad (14)$$

$$a_{32} = \frac{1}{M}\left(k_{p1}(1-m)^2 \exp(\alpha m) + k_{p2}(1-m)^2 \exp(\alpha m)\right) \quad (15)$$

$$a_{33} = \frac{1}{M}\left(-4k_n n(1-m)^3 \exp(\alpha m) + \alpha k_n n(1-m)^4 \exp(\alpha m) - 2k_{p1}p(1-m)^{\square} \exp(\alpha m) + \alpha k_{p1}p(1-m)^2 \exp(\alpha m) - 2k_{p2}p(1-m)^{\square} \exp(\alpha m) + \alpha k_{p2}p(1-m)^2 \exp(\alpha m) - xk_r m^{x-1} \exp\left(-\frac{yF\varphi_0}{RT}\right) + jk_r m^x \exp\left(-\frac{yF\varphi_0}{RT}\right)\right) \quad (16)$$

For the oscillatory behavior to be realized, the presence of the positive addendums, which correspond to the positive callback, in the main diagonal is necessary.

Taking into account the Jacobian main-diagonal elements (8), (12), and (16), it is possible to conclude that the oscillatory behavior in this system is possible. Nevertheless, its probability will be highly dependent on the nature of the monomer on which the polymer is based.

If the monomer is basic and, thereby, ionized in an acidic medium, $\alpha \neq 0$, and the possible positivity of the elements $\alpha k_n n(1-m)^4 \exp(\alpha m) > 0$, $\alpha k_{p1}p(1-m)^2 \exp(\alpha m) > 0$ and $\alpha k_{p2}p(1-m)^2 \exp(\alpha m) > 0$, augments the probability of the oscillatory behavior due to DEL and surface influences of the conducting polymer ionicity and ionic form transformations during the chemical stages of the electroanalytical process. This corresponds to similar systems [36–42], in which the oscillatory behavior probability, such as the oscillation frequency and

amplitudes, depend on the background electrolyte composition directly related to the DEL ionic force.

Yet if the monomer is not ionized in acidic solutions, $\alpha = 0$, and all of the mentioned elements become set to zero. The oscillatory behavior is still possible, but it may only be caused by the DEL influences of the electrochemical stage, described by the positivity of $jk_r m^x \exp\left(-\frac{yF\varphi_0}{RT}\right)$ if j>0.

As for the steady-state stability, its condition results from the Routh-Hurwitz criterion application to the equation set (1). Avoiding the cumbersome expressions, we introduce new variables, rewriting the Jacobian determinant as (17):

$$\frac{4}{\delta^2 C}\begin{vmatrix} -\kappa - \Xi & 0 & T \\ 0 & -\xi - \Sigma & P \\ \Xi & \Sigma & -T - P - \Omega \end{vmatrix} \quad (17)$$

Opening the brackets and applying the Det J<0, salient from the criterion, we obtain the steady-state stability requisite, expressed after changing the signs as (18):

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

defining an efficient electroanalytical process controlled either by diffusion or reaction kinetics. The transition to pure diffusion or purely kinetic controlled mode will depend not only on electrode shape and analyte concentration but also on conducting polymer composition.

The requisite (18) is warranted to be satisfied in the case of the positivity of the parameters Ω and Σ (the rest of the variables are always positive), defining the polymer chemical (if any) and electrochemical transformation influences on DEL. The requisite (18) is satisfied in a vast parameter topological region, but it becomes narrower if the monomer becomes ionized in an acidic medium.

As no reactions capable of compromising the analyte or modifier stability are characteristic for this case, the steady-state stability will correspond to the linear dependence between the current and concentration. Also, as the condition (18) is readily satisfied for a vast parameter region, the electrochemical process will be efficient from an analytical point of view.

As for the detection limit, it is defined by the monotonic instability, conditioned by the requisite Det J=0, or (19):

$$\kappa(\xi T + \xi P + \xi\Omega + \Sigma T + \Sigma\Omega) + \Xi(\xi P + \xi\Omega + \Sigma\Omega) = 0 \quad (19)$$

In this system, different unstable states coexist. The system chooses one of them. If the conditions are altered, it is thereby destroyed and may not be regenerated if the condition (18) is restored. This is defined by the saddle node or static bifurcation.

The same system may be used to determine the vet drugs in milk and meat. Their pH is moderately or mildly acidic, up to neutral, and the vet anti-inflammatory drugs have a similar composition, which is the reason why the conducting polymer sensor may be applicable as a cathode for vet drug determination in animal-based food.

4. Conclusions

From the theoretical evaluation of CP-assisted cathodic naproxene and paracetamol determination in acidic solutions, it was possible to conclude that this process is efficient. Nevertheless, the analytical signal interpretation becomes easier, and the linear range wider if a conducting polymer of a non-ionized monomer is chosen. The use of ionized monomers also augments the probability of oscillatory and monotonic instabilities in the system.

Author Contributions

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

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Conflicts of Interest

The authors declare no conflict of interest.

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