Materials International
Open Access Journal (ISSN: 2668-5728)

Article
Volume 6, Issue 3, 2024, 30
https://doi.org/10.33263/Materials63.030

Theoretical Description for Cadmium and Mercurium Economical and Green Electrochemical Sensing and Removal, Assisted by Conducting Polymer of Toxin from *L. Necator*

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Received: 9.01.2024; Accepted: 20.08.2024; Published: 10.09.2024

Abstract: For the first time, an economical and green methodology for cadmium and mercurium electrochemical analysis and removal has been proposed. The electrochemical sensing is based on the complexation of both metals in the polymer phase on the electrode with necatorin, a phenolic toxin from *L. Necator*, followed by the additional oxidation or overoxidation of the metalized polymer. The correspondent mathematical model has been developed and analyzed by means of linear stability theory and bifurcation analysis, and its investigation confirms the efficiency of the necatorin-based electrochemical process for both the removal and sense of the metals.

Keywords: cadmium; mercurium; necatorin; electrochemical analysis; complex formation; electrochemical sensing; electropolymerization; stable steady-state.

1. Introduction

The elements of the II B group of the Periodic table are among the most used by people [1-4]. Zinc powder is frequently used in the Kipp apparatus to obtain hydrogen in the lab and on a small industrial scale. Zinc compounds are used in pharmacy, dentistry, corrosion protection, energy conversion, etc. Cadmium and its salts were once used to produce yellow dyes for different purposes. As for mercurium, it is one of the seven metallic elements, anciently known as a component of vermillion pigment.

On the other hand, all of the metals in this group are considered "heavy metals" and possess high toxicity if their concentration exceeds the limit. Cadmium and mercurium compounds are especially toxic, as both of them damage the lungs and kidneys, which may cause death [5-8]. Moreover, both of the elements are considered toxic to the environment, which is the reason why the cadmium and mercurium compounds are utilized by special procedures [9-12]. For this reason, the development of an efficient method for quantifying and removing both cations is up to date, and the electrochemical techniques may be suitable for both purposes [13-18].

The modern methodology for heavy-metal detection, removal, and recuperation consists of its complexation with different compounds, including phenols and azo dyes [19-24]. One of these compounds may be necatorin (Figure 1), a toxin from *L. Necator* mushroom [25-28], and its polymer. It may form either 2D or 3D metal complexes, which may be used for heavy metal immobilization.

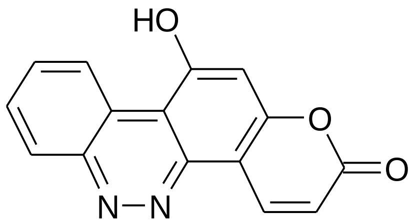

Figure 1. Necatorin.

Other mushroom polyphenols, including involution, orellanine, and A. Xanthodermus phenolic dyes, may also be used for this purpose. Nevertheless, necatorin complexes and their polymers are interesting as economic and green compounds for energy conversion and sensing.

The above-mentioned aspects are important for the mechanistic and dynamic behavior of the electroanalytical process. Moreover, the presence of electrochemical instabilities, which makes it difficult to interpret the analytical signal, may be associated with different oxidation scenarios, electropolymerization, and additional polymer oxidation or overoxidation [29-32]. Those instabilities are typical and may limit the electroanalytical and removal use of the electrochemical process. In order to foresee the possibility of the realization of the electrochemical instabilities, like the effect they may produce, it´s necessary to investigate the process from the mechanical point of view and analyze its behavior theoretically.

So, the goal of this work is the mechanical evaluation for necatorin-assisted cadmium and mercurium determination and removal based on complexation, and a complex polymer oxidation scenario is included. The correspondent mathematical model is developed and analyzed using linear stability theory and bifurcation analysis. The theoretical investigation includes comparing the behavior of this system with that of similar ones [33-35].

2. Materials and Methods

Both cadmium and mercurium react with necatorin molecules, yielding polymerizable complex compounds (both 2D and 3D). Their polymerization potential will be lower than that of the proper necatorin due to the donating character of the ion, which justifies the presence of the analytical signal. Nevertheless, considering the partial penalization of the necatorin polymer, it becomes more suitable to work with poly(necatorine), more capable of forming complexes with both of the metal cations due to more phenolic hydroxyls (Fig. 2).

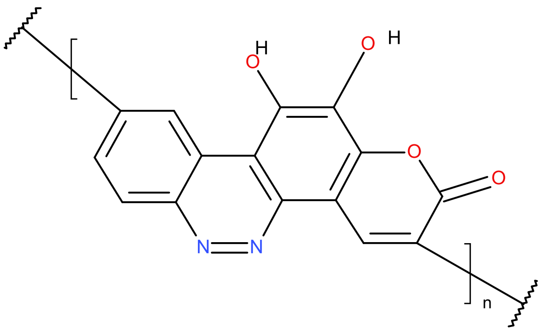

Figure 2. Phenolized poly(necatorin).

So, taking into account certain assumptions [33-35] and supposing that necatorin covers the entire electrode surface at the beginning, we describe the system's behavior by trivariant equation-set (1):

$$\begin{cases} \frac{dc}{dt} = \frac{2}{\delta}\left(\frac{C}{\delta}(c_0 - c) - r_c\right) \\ \frac{dm}{dt} = \frac{2}{\delta}\left(\frac{M}{\delta}(m_0 - m) - r_m\right) \\ \frac{dn}{dt} = \frac{1}{N}(r_c + r_m - r_o - r_O) \end{cases} \quad (1)$$

Herein, c and m are cadmium and mercurium cations pre-surface concentrations, C and M their diffusion coefficients, c_0 and m_0 stand for their pre-surface layer concentrations, n modified necatorin polymer surface coverage degree, N is its maximal surface concentration, and the parameters r stand for the correspondent reaction rates, calculated as:

$$r_c = k_c c(1-n)\exp(-ac) \quad (2)$$

$$r_m = k_m m(1-n)\exp(-bm) \quad (3)$$

$$r_o = k_o n \exp\left(\frac{xF\varphi_0}{RT}\right) \quad (4)$$

$$r_O = k_O n \exp\left(\frac{yF\varphi_0}{RT}\right) \quad (5)$$

Herein, the parameters k stand for the correspondent reaction rate constants, a and b are variables describing the dependence of double electric layer (DEL) electrophysical and electrochemical parameters on chemical stage kinetics, x and y are the numbers of electrons transferred during the electrochemical stage, F is the Faraday number, φ_0 is the zero-charge-related potential slope, R is the absolute gas constant, and T is the cell temperature.

Generally, the behavior of this system becomes similar to that of the ordinary electroanalytical system with two parallel electrochemical stages involving ion transformations without surface instability. The cyclic ionic form transformations will make the behavior more dynamic, augmenting the probability of the oscillatory and monotonic instabilities. Nevertheless, those instabilities do not strongly affect the system's efficiency, which remains high, as shown below.

3. Results and Discussion

In order to investigate the behavior of the system with cadmium and mercury cations determination and elimination on poisonous mushroom polyphenols polymer, we analyze the balance differential equation set (1), considering the algebraic relations (2 – 5) by means of linear stability theory. The steady-state Jacobian matrix members may be described as:

$$\begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix} \quad (6)$$

In which:

$$a_{11} = \frac{2}{\delta}\left(-\frac{C}{\delta} - k_c(1-n)\exp(-ac) + ak_c c(1-n)\exp(-ac)\right) \quad (7)$$

$$a_{12} = 0 \quad (8)$$

$$a_{13} = \frac{2}{\delta}(k_c c\exp(-ac)) \quad (9)$$

$$a_{21} = 0 \quad (10)$$

$$a_{22} = \frac{2}{\delta}\left(-\frac{M}{\delta} - k_m(1-n)\exp(-bm) + bk_m m(1-n)\exp(-bm)\right) \quad (11)$$

$$a_{23} = \frac{2}{\delta}(k_m m\exp(-bm)) \quad (12)$$

$$a_{31} = \frac{1}{N}(k_c(1-n)\exp(-ac) - ak_c c(1-n)\exp(-ac)) \quad (13)$$

$$a_{32} = \frac{1}{N}(k_m(1-n)\exp(-bm) - bk_m m(1-n)\exp(-bm)) \quad (14)$$

$$a_{33} = \frac{1}{N}\left(-k_c c\exp(-ac) - k_m m\exp(-bm) - k_o\exp\left(\frac{xF\varphi_0}{RT}\right) + jk_o n\exp\left(\frac{xF\varphi_0}{RT}\right) - k_o\exp\left(\frac{yF\varphi_0}{RT}\right) + jk_o n\exp\left(\frac{yF\varphi_0}{RT}\right)\right) \quad (15)$$

Avoiding the cumbersome expression during the determinant analysis, we introduce new variables and rewrite the determinant as (15):

$$\text{Det J} = \frac{4}{\delta^2 V}\begin{vmatrix} -\kappa - \Xi & 0 & N \\ 0 & -\varphi - P & \Phi \\ \Xi & P & -N - \Phi - \Omega \end{vmatrix} \quad (16)$$

Considering that:

$$-Det\ J\begin{cases} > 0, for\ steady - state\ stability \\ = 0\ monotonic\ instability \end{cases} \quad (17)$$

Opening the brackets, applying the Det J<0 requisite, salient from the criterion, and changing the signs to the opposite, we rewrite the condition set as (18):

$$-\kappa(\varphi N + \varphi\Phi + \varphi\Omega + PN + P\Omega) - \Xi(2\varphi N + \varphi\Phi + \varphi\Omega + 2PN + P\Omega)\begin{cases} > 0, curve\ linearity \\ = 0, detection\ limit \end{cases} \quad (18)$$

If –Det J>0, the Routh-Hurwitz stability criterion is valid, and the steady-state is thereby stable, providing an efficient steady-state metal cations determination and removal, ensuring its conversion into economic and green material. Moreover, the wide stability region confirms the efficiency of this electrochemical system for both electroanalytical and environmental remediation purposes.

This criterion is readily satisfied if the kinetic parameters P and Ω are positive. In the vast majority of the cases, they both have positive signs and considering that the other variables in the determinant are positive, it indicates the vast steady-state stability topological region. The electroanalytical process is mostly kinetically controlled.

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 important for mercury 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 part of the steady-state voltammogram, depicts the margin between stable and unstable states, and corresponds to steady-state multiplicity. In other words, multiple steady-states, each one unstable, coexist at this point.

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

Observing the main diagonal elements (7), (11), and (15), we may observe that the oscillatory behavior becomes possible if the kinetic parameters a and j are positive, which corresponds to the DEL influences of the chemical and electrochemical stages. This factor is typical for similar systems [27,28] and may be described by the positivity of the elements $ak_c c(1-n)\exp(-ac)$ and $bk_m m(1-n)\exp(-bm) > 0$, if a,b>0, like also $jk_o n\exp\left(\frac{xF\varphi_0}{RT}\right)$ and $jk_O n\exp\left(\frac{yF\varphi_0}{RT}\right) > 0$, if j>0. These elements describe the positive callback, and this callback will depend on the system´s characteristics. For example, the oscillation frequency and amplitude will depend on the background electrolyte composition, which has been proven experimentally and theoretically [33-35].

This model is valid for the anodic section for the heavy-metal cation removal, given by membrane electrolysis [33]. This process becomes more efficient as it combines the classical metal cathodic deposition with the complex formation of necatorin polymer.

4. Conclusions

From the behavior investigation of the system with the cadmium and mercury cations electrochemical detection and removal, assisted by a conducting polymer based on *L. Necator* toxin, it was possible to conclude that it may be an excellent electrode modifier for both processes. Despite the increase in the oscillatory and monotonic instability probability, the analytical signal is easy to interpret, and the steady-state stability is easy to obtain and maintain, providing an excellent metal cation recycling into an economical and green material suitable for sensing and energy conversion.

Funding

This research received no external funding.

Acknowledgments

Volodymyr V. Tkach acknowledges the Engineering Faculty of the University of Porto and the University of Trás-os-Montes and Alto Douro for their support during these difficult times for Ukraine and its research.

Conflicts of Interest

The authors declare no conflict of interest.

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