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Cadmium and Nickel Removal from Wastewater by a Complex-forming Process on a Phenolic Conducting Polymer. A Theoretical Insight

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Abstract: The theoretical description of two bivalent metals (with the examples of nickel and cadmium), electrochemical removal, and determination by phenolic conducting polymer has been described. In this system, the conducting polymer acts in both aromatic and overoxidized forms, providing efficient metal cation removal. 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 phenolic conducting polymers for cadmium and nickel removal and immobilization into the polymer phase, where they are used in a composite. This is the keystone for the use of this process as a part of waste management and circular economy.

Keywords: waste management; circular economy; heavy metal removal; polyphenolic compounds; conducting polymer; overoxidation; stable steady state.

1. Introduction

Circular economy [1-4] is the economic model that is included in the doctrine of sustainable development. Contrarily to the traditional (linear) economy, the circular economy tends to reuse waste as a secondary raw material, providing the "green transfer" of the economy to an environmentally safer world.

In this regard, heavy metal ions [5-10] are of particular importance, as they are among the most toxic waste substances known, provoking mutagenesis, genotoxicity, and disruptions in homeostasis. They are also dangerous to the environment, which is why heavy metal removal and reuse are highly relevant [11-14], and the use of polyphenolic conducting polymers provides an efficient response. The formation of a phenolic complex in either the monomer or polymer phase has been described both experimentally and theoretically as an efficient means for determining and removing conducting polymers [15-19]. Nevertheless, no simultaneous use of neutral and overoxidized polyphenolic monomers has been reported, although the overoxidized polymer has been used to quantify heavy metals [20,21].

For this reason, the goal of our work is to theoretically investigate the possibility of the use of conducting (co)polymers for the removal of two bivalent heavy metal cations by both neutral and overoxidized polymer forms. For this purpose, the system's behavior will be described by a set of balance differential equations, which will be analyzed using linear stability theory and bifurcation analysis to determine the steady-state stability requirements, including oscillatory and monotonic instability conditions. Also, the behavior of the system will be compared with that of similar ones [18,19].

2. Materials and Methods

The types of complex-forming interaction of phenolic compounds and their polymers with metal cations have been described in previous works [15-19], and their interaction with metal cations has been depicted in [20,21]. Both interactions enhance the polymer's conductivity and enable the resulting composite for supercapacitors, energy conversion, and electroanalysis.

Taking certain assumptions and considering that the neutral polymer covers the entire electrode surface in the beginning, we describe the system´s behavior by a trivariate equation set (1):

$$\begin{cases} \frac{dc}{dt} = \frac{2}{\delta}\left(\frac{C}{\delta}(c_0 - c) - r_{c1} - r_{c2}\right) \\ \frac{dn}{dt} = \frac{2}{\delta}\left(\frac{N}{\delta}(n_0 - n) - r_{n1} - r_{n2}\right) \\ \frac{dp}{dt} = \frac{1}{P}(r_{O1} - r_{c2} - r_{n2} - r_{O2}) \end{cases} \quad (1)$$

Herein, c and n are cadmium and nickel cations pre-surface concentrations, C and N their diffusion coefficients, c_0 and n_0 stand for their pre-surface layer concentrations, p modified overoxidized polymer surface coverage degree, N is its maximal surface concentration, and the parameters r stand for the corresponding reaction rates, calculated as:

$$r_{c1} = k_{c1}c(1-p)\exp(-ac) \quad (2)$$

$$r_{c2} = k_{c2}cp\exp(-ac) \quad (3)$$

$$r_{n1} = k_{n1}n(1-p)\exp(-bn) \quad (4)$$

$$r_{n2} = k_{n1}np\exp(-bn) \quad (5)$$

$$r_{O1} = k_{O1}(1-p)\exp\left(\frac{xF\varphi_0}{RT}\right) \quad (6)$$

$$r_{O2} = k_{O2}p\exp\left(\frac{xF\varphi_0}{RT}\right) \quad (7)$$

Herein, the parameters k stand for the corresponding 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 is the number 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.

The system behavior is quite similar to that observed in similar systems but with slight differences involving both of the electrochemical stages. In these conditions, the heavy metal ion quantification is not possible, due to the reaction kinetics (the conditions for the realization of the electroanalytical process will be shown below). In the neutral medium, in which no autocatalytic behavior is observed during the conducting polymer overoxidation, the oscillatory behavior will be caused by DEL influences of metal cation removal and immobilization, as well as the electrochemical polymer transformation. Nevertheless, these influences do not impede the realization of steady-state electrochemical metal removal, as described in the next section.

3. Results and Discussion

In order to investigate the behavior of the system with cadmium and nickel cations elimination on both neutral and overoxidized polyphenol polymer, we analyze the balance differential equation set (1), considering the algebraic relations (2 – 7) by means of linear stability theory. The steady-state Jacobian matrix members may be described as:

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

Herein:

$$a_{11} = \frac{2}{\delta}\left(-\frac{C}{\delta} - k_{c1}(1-p)\exp(-ac) - k_{c2}c\exp(-ac) + a(k_{c1}c(1-p)\exp(-ac) + k_{c2}cp\exp(-ac))\right) \quad (9)$$

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

$$a_{13} = \frac{2}{\delta}(k_{c1}c\exp(-ac) - k_{c2}c\exp(-ac)) \quad (11)$$

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

$$a_{22} = \frac{2}{\delta}\left(-\frac{N}{\delta} - k_{n1}(1-p)\exp(-bn) - k_{n2}p\exp(-bn) + b(k_{n1}n(1-p)\exp(-bn) + k_{n2}np\exp(-bn))\right) \quad (13)$$

$$a_{23} = \frac{2}{\delta}(k_{n1}n\exp(-bn) - k_{n1}n\exp(-bn)) \quad (14)$$

$$a_{31} = \frac{1}{P}(k_{c1}(1-p)\exp(-ac) - k_{c2}c\exp(-ac) - a(k_{c1}c(1-p)\exp(-ac) - k_{c2}cp\exp(-ac))) \quad (15)$$

$$a_{32} = \frac{1}{P}\left(k_{n1}(1-p)\exp(-bn) - k_{n2}p\exp(-bn) - b(k_{n1}n(1-p)\exp(-bn) - k_{n2}np\exp(-bn))\right) \quad (16)$$

$$a_{33} = \frac{1}{P}\left(-k_{O1}(1-p)\exp\left(\frac{xF\varphi_0}{RT}\right) + jk_{O1}(1-p)\exp\left(\frac{xF\varphi_0}{RT}\right) - k_{n1}n\exp(-bn) - k_{n1}n\exp(-bn) - k_{c1}c\exp(-ac) - k_{c2}c\exp(-ac) - k_{O2}\exp\left(\frac{xF\varphi_0}{RT}\right) + jk_{O2}p\exp\left(\frac{xF\varphi_0}{RT}\right)\right) \quad (17)$$

Avoiding the cumbersome expression during the determinant analysis, we introduce new variables and rewrite the determinant as (18):

$$\text{Det J} = \frac{4}{\delta^2 V}\begin{vmatrix} -\kappa - \Xi - T & 0 & N - S \\ 0 & -\varphi - P - E & \Phi - D \\ \Xi - T & P - E & -N - \Phi - S - D - \Omega \end{vmatrix} \quad (18)$$

Considering that:

$$-Det\ J\begin{cases}>0, for\ steady-state\ stability\\ =0\ monotonic\ instability\end{cases} \quad (19)$$

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 (20):

$$\kappa(\varphi N+\varphi\Phi+\varphi S+\varphi D+\varphi\Omega+PN+PS+2PD+P\Omega+EN+2E\Phi+ES+E\Omega)+\Xi(\varphi\Phi+2\varphi S+\varphi D+\varphi\Omega+2PS+2PD+P\Omega+2E\Phi+2ES+E\Omega)+T(2\varphi N+\varphi\Phi+\varphi D+\varphi\Omega+2PN+2PD+P\Omega+2EN+2E\Phi+E\Omega)\begin{cases}>0, steady-state\ stability\\ =0, monotonic\ instability\end{cases} \quad (20)$$

Providing an efficient kinetically-controlled electrochemical system in which steady-state stability becomes easily obtained and maintained, ensuring its conversion into an 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, N, Ξ, T and Ω are positive. In the vast majority of cases, they both have positive signs, and considering that the other variables in the determinant are positive, it indicates a vast steady-state stability topological region.

The condition Det J=0 leads to *monotonic instability*. It may be seen as an N-shaped part of the steady-state voltammogram, which depicts the margin between the stable steady-states and unstable 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 realized beyond the detection limit in the case of the Hopf bifurcation realization. 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 a and j are positive, which corresponds to the DEL influences of the chemical and electrochemical stages. This factor is typical for similar systems [15 – 19] and may be described by the positivity of the elements $aa(k_{c1}c(1-p)\exp(-ac)+k_{c2}cp\exp(-ac))$ and $b(k_{n1}n(1-p)\exp(-bn)-k_{n2}np\exp(-bn))>0$, if a,b>0, like also $jk_{O1}(1-p)\exp\left(\frac{xF\varphi_0}{RT}\right)$ and $jk_{O2}p\exp\left(\frac{xF\varphi_0}{RT}\right)>0$, 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 [15-19].

As mentioned above, this system does not have any electroanalytical application due to the kinetics of its interaction with the polymer. Nevertheless, altering the conditions may be used for the electrochemical determination of the heavy metal cations one by one. In this case, the system will be described by the trivariate equation (21):

$$\begin{cases}\frac{dc}{dt}=\frac{2}{\delta}\left(\frac{C}{\delta}(c_0-c)-r_{c1}-r_{c2}\right)\\ \frac{dp}{dt}=\frac{1}{P}(r_{O1}-r_{c2}-r_{O2})\\ \frac{dp*}{dt}=\frac{1}{P*}(r_{O2}+r_{c2}-r_{O3})\end{cases} \quad (21)$$

Its analysis confirms the similarity between these systems and will be described in our next works.

4. Conclusions

From the behavior investigation of the system with the cadmium and nickel cations electrochemical removal towards the neutral and overoxidized polyphenolic conducting polymer phase, it was possible to conclude that the process is highly efficient due to the facile steady-state formation. Despite the increase in the probability of oscillatory and monotonic instability, the metal cations are readily removed, and steady-state stability is readily achievable and easy to maintain, providing an excellent means of recycling metal cations into an economical, green material suitable for sensing and energy conversion. This material may also be used in supercapacitors.

Author Contributions

Conceptualization, T.V.M.; V.V.T.; M.V.K.; Z.H.A.; Z.Z.Y.; F.A.U.; validation, V.V.T.; T.V.M.; M.V.K.; Z.H.A.; H.B.J.; G.B.K; F.A.U.; Y.G.I.; J.I.F.P.M.; J.R.G.; G.M.P.; N.K.; O.O.P.; O.M.H.; G.T.P. N.Y.C.; S.P.V.; P.I.Y.. and Z.Z.Y..; formal analysis, V.V.T.; T.V.M.; M.V.K.; Z.H.A.; H.B.J.; G.B.K; F.A.U.; Y.G.I.; J.I.F.P.M.; J.R.G.; G.M.P.; N.K.; O.O.P.; O.M.H.; G.T.P. N.Y.C.; S.P.V.; P.I.Y.. and Z.Z.Y..; investigation, V.V.T.; T.V.M.; M.V.K.; Z.H.A.; H.B.J.; G.B.K; F.A.U.; Y.G.I.; J.I.F.P.M.; J.R.G.; G.M.P.; N.K.; O.O.P.; O.M.H.; G.T.P. N.Y.C.; S.P.V.; P.I.Y.. and Z.Z.Y..; data curation, V.V.T.; T.V.M.; M.V.K.; Z.H.A.; H.B.J.; G.B.K; F.A.U.; Y.G.I.; J.I.F.P.M.; J.R.G.; G.M.P.; N.K.; O.O.P.; O.M.H.; G.T.P. N.Y.C.; S.P.V.; P.I.Y.. and Z.Z.Y..; writing—original draft preparation, V.V.T.; T.V.M.; M.V.K.; Z.H.A.; H.B.J.; G.B.K; F.A.U.; Y.G.I.; J.I.F.P.M.; J.R.G.; G.M.P.; N.K.; O.O.P.; O.M.H.; G.T.P. N.Y.C.; S.P.V.; P.I.Y.. and Z.Z.Y..; writing—review and editing, V V.V.T.; T.V.M.; M.V.K.; Z.H.A.; H.B.J.; G.B.K; F.A.U.; Y.G.I.; J.I.F.P.M.; J.R.G.; G.M.P.; N.K.; O.O.P.; O.M.H.; G.T.P. N.Y.C.; S.P.V.; P.I.Y.. and Z.Z.Y..; visualization, V.V.T.; T.V.M.; M.V.K.; Z.H.A.; H.B.J.; G.B.K; F.A.U.; Y.G.I.; J.I.F.P.M.; J.R.G.; G.M.P.; N.K.; O.O.P.; O.M.H.; G.T.P. N.Y.C.; S.P.V.; P.I.Y.. and Z.Z.Y..; supervision, V.V.T.; T.V.M.;; P.I.Y.. and Z.Z.Y..; project administration, V.V.T.; J.I.F.P.M.. 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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