

Theoretical Description for Pyrrole Assisted Cathodic Electropolymerization, Assisted by *In situ* Formed Selenite-Ion

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Abstract: The mathematical description for a novel economic and green cathodic indirect polypyrrole deposition has been suggested for the first time. The electrochemical cathodic process yields the Se/PPy composite, easy to obtain from the selenate waste solutions, providing a developed polymeric film formation with enhanced adhesion. The oscillatory behavior is possible if the selenate is inserted in the surface matrix. Nevertheless, it is far more probable than the direct anodic electropolymerization process.

Keywords: electropolymerization; polypyrrole; selenate; selenite; cathodic deposition; stable steady-state.

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1. Introduction

Polypyrrole is one of the most used conducting polymers over the last five decades [1–10]. It is one of the most used conducting polymers due to the low price of the polymer and the low polymerization potential. Its principal applications are corrosion protection, supercapacitors, sensors, and biosensors.

Polypyrrole may be synthesized in either chemical or electrochemical ways. The electropolymerization, which is preferable to obtain more conducting and flexible polymer coatings, maybe, in its turn, be realized either directly or indirectly.

The direct electropolymerization [11–17], in which the monomer interacts directly with the anodic current, is transformed into the radical-cation. As for the indirect electropolymerization, in which the pyrrole monomer interacts with the product of the electrochemical process, it is far less used [18–20]. Nevertheless, indirect electropolymerization seems to be more flexible and efficient for polypyrrole electrosynthesis

in acidic media. Moreover, the cathodic deposition, impossible for pyrrole direct electropolymerization, may also be realized.

For this purpose, a cathodic reaction [18,19] yields the initiator *in situ*. Furtherly, the pyrrole monomer is oxidized by the cathodic reaction product, yielding a polymer. In this case, a more developed polymer surface is obtained compared to those afforded by direct anodic electropolymerization in the same conditions. Also, the electrochemical instabilities seem to be more typical for direct electropolymerization [21–24] than for the indirect one.

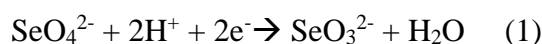
Nevertheless, the electrochemical behavior and mechanism may differ from one process to another. Also, the steady-state stability highly influences the resulting polymer (polymer composite morphology).

In this case, the possibility of selenite-assisted electrochemical cathodic polypyrrole deposition has been described. Polypyrrole deposition may be used for recycling selenate, which classifies this process and is economical and green. The correspondent mathematical model has been developed and analyzed using the linear stability theory and bifurcation analysis. The stability behavior of this system is thereby analyzed and compared with that of similar systems [25–28].

2. Materials and Methods

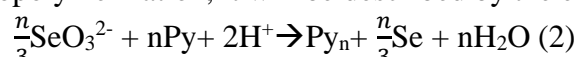
2.1. System and its modeling.

The conducting polymer deposition is realized in the acidic media. On the electrochemical stage, the selenate ion is reduced, yielding the selenite (1):

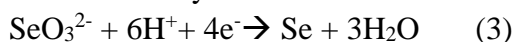


The oxidation potential of the $\text{SeO}_3^{2-}/\text{Se}$ redox pair is about 0,74 V (a bit lower than for $\text{Fe}^{3+}/\text{Fe}^{2+}$ redox pair (0,77 V) and higher than for pyrrole electrooxidation yielding radical-cation (0,5 V)), the reason why selenite is used as a specific oxidant in organic chemistry, converting acetaldehyde into glioxal.

As for the electropolymerization, it will be described by the equation(2):



Where n stands for the polymer chain length in pyrrolic units. Note that the pyrrole electropolymerization of n units, according to the Díaz mechanism, expels (2n-2) protons, so an additional 2 protons uptake is necessary to fulfill the semi reaction (3):



As the selenium is deposited over the electrode surface thereby being covered by the polypyrrole layer. It enhances the conducting polymer film adhesion over the cathode matrix.

So, to describe the behavior of this system in potentiostatic mode, we introduce three variables:

p – pyrrole monomer concentration in the pre-surface layer;

s –selenite matrix coverage degree;

h - protons' concentration in the pre-surface layer.

Introducing some assumptions [28], we describe the system's behavior by the balance differential equation-set (3):

$$\begin{cases} \frac{dp}{dt} = \frac{2}{\delta} \left(\frac{\Delta}{\delta} (c_0 - c) - r_p \right) \\ \frac{ds}{dt} = \frac{1}{s} (r_r - r_p) \\ \frac{dh}{dt} = \frac{2}{\delta} \left(\frac{H}{\delta} (h_0 - c) - r_r - r_p \right) \end{cases} \quad (3)$$

Herein, Δ and H are diffusion coefficients, c_0 and h_0 are the pyrrole and protons bulk concentrations, S is the selenite maximal matrix concentration and the parameters r are the correspondent reaction rates, calculated as (4 – 5):

$$r_r = k_r (1 - s) h^2 \exp\left(-\frac{2F\phi_0}{RT}\right) \quad (4)$$

$$r_p = k_p s^{\frac{n}{3}} p^n h^2 \exp(-\alpha s) \exp(-\beta h) \quad (5)$$

Herein, the parameters k are the correspondent reaction rate constants, α and β are the parameters, describing the DEL capacitance changes during the chemical stages, F is the Faraday number, ϕ_0 is the potential slope in DEL, related to the zero-charge potential, R is the universal gas constant, and T is the absolute temperature of the solution.

The oscillatory behavior will be more probable in this system than in similar systems with cathodic polypyrroles electrodeposition. Nevertheless, it will be less probable than direct anodic electropolymerization in the same conditions, as shown below.

3. Results and Discussion

To describe the behavior of selenite-assisted cathodic polypyrrole electrodeposition in acidic media, we analyze the differential equation-set (3), considering the algebraic relations (4–5) using linear stability theory. The steady-state Jacobian matrix members for this system will 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:

$$a_{11} = \frac{2}{\delta} \left(-\frac{\Delta}{\delta} - nk_p s^{\frac{n}{3}} p^{n-1} h^2 \exp(-\alpha s) \exp(-\beta h) \right) \quad (7)$$

$$a_{12} = \frac{2}{\delta} \left(-\frac{n}{3} k_p s^{\frac{n}{3}-1} p^n h^2 \exp(-\alpha s) \exp(-\beta h) + ak_p s^{\frac{n}{3}} p^n h^2 \exp(-\alpha s) \exp(-\beta h) \right) \quad (8)$$

$$a_{13} = \frac{2}{\delta} \left(-2k_p s^{\frac{n}{3}} p^n h \exp(-\alpha s) \exp(-\beta h) + \beta k_p s^{\frac{n}{3}} p^n h^2 \exp(-\alpha s) \exp(-\beta h) \right) \quad (9)$$

$$a_{21} = \frac{1}{s} \left(-nk_p s^{\frac{n}{3}} p^{n-1} h^2 \exp(-\alpha s) \exp(-\beta h) \right) \quad (10)$$

$$a_{22} = \frac{1}{s} \left(-k_r h^2 \exp\left(-\frac{2F\phi_0}{RT}\right) + jk_r (1 - s) h^2 \exp\left(-\frac{2F\phi_0}{RT}\right) - \frac{n}{3} k_p s^{\frac{n}{3}-1} p^n h^2 \exp(-\alpha s) \exp(-\beta h) + ak_p s^{\frac{n}{3}} p^n h^2 \exp(-\alpha s) \exp(-\beta h) \right) \quad (11)$$

$$a_{23} = \frac{1}{s} \left(2k_r (1 - s) h \exp\left(-\frac{2F\phi_0}{RT}\right) - 2k_p s^{\frac{n}{3}} p^n h \exp(-\alpha s) \exp(-\beta h) + \beta k_p s^{\frac{n}{3}} p^n h^2 \exp(-\alpha s) \exp(-\beta h) \right) \quad (12)$$

$$a_{31} = \frac{2}{\delta} \left(-nk_p s^{\frac{n}{3}} p^{n-1} h^2 \exp(-\alpha s) \exp(-\beta h) \right) \quad (13)$$

$$a_{32} = \frac{2}{\delta} \left(k_r h^2 \exp\left(-\frac{2F\phi_0}{RT}\right) - jk_r (1 - s) h^2 \exp\left(-\frac{2F\phi_0}{RT}\right) - \frac{n}{3} k_p s^{\frac{n}{3}-1} p^n h^2 \exp(-\alpha s) \exp(-\beta h) + ak_p s^{\frac{n}{3}} p^n h^2 \exp(-\alpha s) \exp(-\beta h) \right) \quad (14)$$

$$a_{33} = \frac{2}{\delta} \left(-\frac{H}{\delta} - 2k_r(1-s)h \exp\left(-\frac{2F\varphi_0}{RT}\right) - 2k_p s^{\frac{n}{3}} p^n h \exp(-\alpha s) \exp(-\beta h) + \beta k_p s^{\frac{n}{3}} p^n h^2 \exp(-\alpha s) \exp(-\beta h) \right) \quad (15)$$

Taking into account the main diagonal elements (7), (11), and (15), we may conclude that the *oscillatory behavior* is possible in this system. Moreover, it is much more probable than for similar systems [25–28] due to the strong influence of ionic form transformations during electropolymerization.

Besides the element $jk_r(1-s)h^2 \exp\left(-\frac{2F\varphi_0}{RT}\right) > 0$, typical for nearly all of the electropolymerization and polymer modification systems, the elements $\beta k_p s^{\frac{n}{3}} p^n h^2 \exp(-\alpha s) \exp(-\beta h) > 0$, if $\beta > 0$ and $\alpha k_p s^{\frac{n}{3}} p^n h^2 \exp(-\alpha s) \exp(-\beta h) > 0$, if $\alpha > 0$, describing the DEL influences of the selenite and protons transformation during the electropolymerization. The oscillatory behavior in this system is more probable than in the similar cathodic electrodeposition systems, but even though less probable than for the direct anodic electropolymerization, in which the monomer interaction with the electrode plays its specific role. The oscillation amplitude and frequency will strongly depend on the solution composition, including the pH.

As for the steady-state stability, its realization will correspond to a wider topological area than in the case of the anodic electropolymerization, indicating a more developed polymer surface formation. Nevertheless, the polymer properties will strongly depend on pH.

Applying the Routh-Hurwitz criterion to the equation-set (3) and rewriting the Jacobian determinant as (16):

$$\frac{4}{\delta^2 S} \begin{vmatrix} -\kappa - \mathcal{E} & -\Sigma & -\Lambda \\ -\mathcal{E} & P - \Sigma & T - \Lambda \\ -\mathcal{E} & -P - \Sigma & -\eta - T - \Lambda \end{vmatrix} \quad (16)$$

Which, taking into account the determinant properties, will be rewritten as:

$$\frac{4}{\delta^2 S} \begin{vmatrix} -\kappa - \mathcal{E} & -\Sigma & -\Lambda \\ 0 & 2P & 2T \\ -\mathcal{E} & -P - \Sigma & -\eta - T - \Lambda \end{vmatrix} \quad (17)$$

Opening the straight brackets and applying the $\text{Det } J < 0$ requisites, salient from the criterion, we obtain the steady-state stability requirement (18):

$$-\kappa(2P\Sigma - 2P\eta - 2P\Lambda) - \mathcal{E}(2P\Sigma - 2P\eta - 2\Sigma T) < 0 \quad (18)$$

or (19):

$$\kappa(2P\Sigma - 2P\eta - 2P\Lambda) + \mathcal{E}(2P\Sigma - 2P\eta - 2\Sigma T) > 0 \quad (19),$$

Which is easily satisfied, especially when both protons and matrix selenate are in excess relatively to the pyrrole. The same factor provided efficient nitrosyl-assisted polypyrrole formation (nitrate excess to pyrrole and protons). Therefore, it will also be important in this system.

The polymerization process will be both diffusion and kinetically controlled, providing an efficient developed polymer surface, easily used for different purposes.

As for the monotonic instability, it separates the unstable states from the steady-state stability region, being, therefore, its margin. Its condition will be expressed as $\text{Det } J = 0$, or (20):

$$\kappa(2P\Sigma - 2P\eta - 2P\Lambda) + \mathcal{E}(2P\Sigma - 2P\eta - 2\Sigma T) > 0 \quad (20)$$

This model describes the possibility of pyrrole electropolymerization in the conditions of the absence of the second electropolymerizable compound. If present, that compound

influences the system by its homopolymerization or copolymerization with pyrrole. This case will be described in our next papers.

4. Conclusions

The theoretical description for the selenite-assisted conducting polymer formation shows us that this process leads to the appearance of a well-developed polymeric material, which is more suitable than direct electropolymerization. The oscillatory behavior is more probable than for similar systems due to the strong DEL influence of the transformed ions. Nevertheless, steady-state stability is easy to obtain and maintain. Excess selenate and protons versus monomer stabilize the system behavior, providing a more efficient polymer formation.

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Conflicts of Interest

The authors declare no conflict of interest.

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