

# The Mathematical Modeling for Mercury Electroanalytical Determination and Removal by Green Conducting Polymer, Based on Toxic Mushroom Compounds

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**Abstract:** This work presents a theoretical evaluation of mercury electroanalytical quantification and removal from wastewater by oxidation in the presence of a conducting polymer derived from poisonous mushrooms. As their main toxins are phenolic, electro(co)polymerization and the complexation of the phenolic toxins with both mercury cations in the polymer phase are possible. The mathematical modeling of the system's behavior confirms its efficiency for the electrochemical determination and removal of heavy metals from natural waters and wastewater. The procedure is foreseen to be realized in an economical and green manner.

**Keywords:** heavy metals; poisonous mushrooms; *Cortinarius orellanus*; *Paxillus involutus*; *Agaricus xanthodermus*; conducting polymers; electrochemical removal; electrochemical sensors; electrochemical oscillations; stable steady-state.

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## 1. Introduction

Mercury is one of seven metals known in antiquity [1–4]. It was frequently found in the free state as liquid drops in ores, but generally, it was traditionally obtained from vermillion. Vermillion dye has been used in the territory of modern Turkey since the VIII millennium B.C. The vermillion deposits in China have been explored since the IV millennium B.C. Ancient Egyptian vessels containing either vermillion or pure mercury were found in pyramids and dated to at least the 16th century B.C.

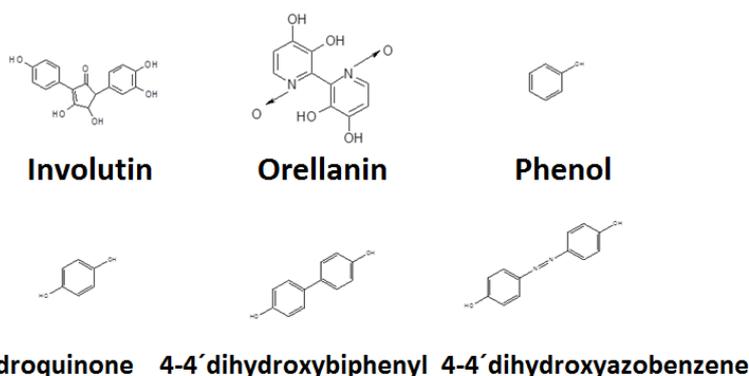
The alchemists designed the symbol of this metal to resemble the planet Mercury, which is why it is called Mercury in English, Spanish, and Portuguese. The Latin name “hydrargyrum” was first used by Plinius Major and has a Greek origin (from the Greek expression meaning “water silver” or “liquid silver”, which is also related to the English “quicksilver”).

Nevertheless, both organic and inorganic mercury compounds are highly toxic [5–10]. Its toxicity is related to the toxic mercurium organic compounds, which affect the nervous, gastrointestinal, and immune systems. The WHO lists mercury among the 10 most (eco)toxic elements, which is why mercury determination and decontamination are among the most important detoxification processes in the world. Electrochemical methods may be interesting for this purpose [11–16].

As mercury is a liquid metal, cathodic decarburization may not be applicable, and the most efficient strategy is to remove mercury electrochemically, transforming it into a stable complex compound that may then be electropolymerized, taking the mercurium ions into the polymer phase.

In order to implement this function, the anode will be modified by a conjugated dye [17], its polymer, or another conducting polymer possessing complex-forming functional groups. Those monomers and polymers are generally synthetic [18–25], but natural analogues may replace them.

In this regard, toxic mushrooms may be an interesting source of monomers. Besides being highly renewable, they do not participate in the trophic chain of human and most animal organisms. It's important to mention the mushrooms *Cortinarius Orellanus*, *Paxillus Involutus*, and the yellow stainer *Agaricus Xanthodermus*, which contain phenolic compounds capable of being used as monomers for economical and green conducting polymers (Figure 1).



**Figure 1.** The phenolic mushroom toxins are potentially used as monomers.

Therefore, in this work, we theoretically investigate the use of conducting (co)polymers derived from mushroom phenols for both mercury ion quantification and removal. 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 [25–28].

## 2. Materials and Methods

We evaluate the electroanalytical system in which both mercury cations form complexes with mushroom polyphenols. As the mercury ions are donors, the complex compounds will have lower polymerization potential than the monomer, thereby taking the mercury complex into the stable polymer phase, which won't influence the solution anymore.

Because the polymerization peak depends on the mercury concentration, this reaction will be used to quantify both mercury ions.

For this reason, taking some assumptions [25–28], we describe the potentiostatic behavior of this system by a trivariate balance differential equation set (1):

$$\begin{cases} \frac{dh}{dt} = \frac{2}{\delta} \left( \frac{H}{\delta} (h_0 - h) - r_{h1} \right) \\ \frac{dh_2}{dt} = \frac{2}{\delta} \left( \frac{H_2}{\delta} (h_{20} - h_2) - r_{h2} \right) \\ \frac{dp}{dt} = \frac{1}{P} (r_{h1} + r_{h2} - r_o) \end{cases} \quad (1)$$

herein,  $h$  and  $h_2$  are the pre-surface concentrations of both the mercury cations,  $h_0$  and  $h_{20}$  are their bulk concentrations,  $H$  and  $H_2$  are their diffusion coefficients,  $\delta$  is the pre-surface layer thickness,  $p$  is the phenolic complex surface coverage degree,  $P$  stands for its maximal surface concentration, and the parameters  $r$  describe the reaction rates:

$$r_{h1} = k_{h1} h (1 - p) \exp(-ah) \quad (2)$$

$$r_{h2} = k_{h2} h_2 (1 - p) \exp(-bh_2) \quad (3)$$

$$r_o = k_o p \exp\left(\frac{zF\phi_0}{RT}\right) \quad (4)$$

in which the parameters  $k$  are the corresponding reaction rate constants,  $a$  and  $b$  are variables describing, correspondingly, the DEL impact of the ionic forms transformation during the chemical stages,  $z$  is the number of transferred electrons,  $F$  is the Faraday number,  $\phi_0$  is the potential slope related to the zero-charge potential,  $R$  is the universal gas constant, and  $T$  is the absolute temperature.

Generally, the behavior of this system becomes similar to that of the ordinary electroanalytical system involving ion transformations without surface instability. Cyclic ionic form transformations will make the behavior more dynamic, increasing the likelihood of 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

To investigate the behavior of the system with mercury cation determination and elimination on poisonous mushroom polyphenol monomers during their polymerization, we analyze the balance differential equation set (1), considering the algebraic relations (2–4) using linear stability theory. The steady-state Jacobian matrix members may be described as:

$$\begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix} \quad (5)$$

in which:

$$a_{11} = \frac{2}{\delta} \left( -\frac{H}{\delta} - k_{h1}(1-p) \exp(-ah) + ak_{h1}h(1-p) \exp(-ah) \right) \quad (6)$$

$$a_{12} = 0 \quad (7)$$

$$a_{13} = \frac{2}{\delta} (k_{h1}h \exp(-ah)) \quad (8)$$

$$a_{21} = 0 \quad (9)$$

$$a_{22} = \frac{2}{\delta} \left( -\frac{H_2}{\delta} - k_{h2}(1-p) \exp(-bh_2) + bk_{h2}h_2(1-p) \exp(-bh_2) \right) \quad (10)$$

$$a_{23} = \frac{2}{\delta} (k_{h2}h_2 \exp(-bh_2)) \quad (11)$$

$$a_{31} = \frac{1}{P} (k_{h1}(1-p) \exp(-ah) - ak_{h1}h(1-p) \exp(-ah) r_O) \quad (12)$$

$$a_{32} = \frac{1}{P} (k_{h2}(1-p) \exp(-bh_2) - bk_{h2}h_2(1-p) \exp(-bh_2)) \quad (13)$$

$$a_{33} = \frac{1}{P} \left( -k_{h1}h \exp(-ah) - k_{h2}h_2 \exp(-bh_2) - k_O \exp\left(\frac{zF\phi_0}{RT}\right) + jk_O p \exp\left(\frac{zF\phi_0}{RT}\right) \right) \quad (14)$$

Introducing new variables to streamline the analysis, the determinant can be rewritten as equation (15)

$$\text{Det } J = \frac{4}{\delta^2 V} \begin{vmatrix} -\kappa - \varepsilon & 0 & N \\ 0 & -\phi - P & \Phi \\ \varepsilon & P & -N - \Phi - \Omega \end{vmatrix} \quad (15)$$

Considering that:

$$-Det J \begin{cases} > 0, \text{ for steady - state stability} \\ = 0 \text{ monotonic instability} \end{cases} \quad (16)$$

After expanding the brackets, applying the  $\text{Det } J < 0$  criterion, and inverting the signs, the condition set is rewritten as (17).

$$-\kappa(\phi N + \phi \Phi + \phi \Omega + PN + P\Omega) - \varepsilon(2\phi N + \phi \Phi + \phi \Omega + 2PN + P\Omega) \begin{cases} > 0, \text{ curve linearity} \\ = 0, \text{ detection limit} \end{cases} \quad (17)$$

This criterion is readily satisfied when the kinetic parameters P and  $\Omega$  are positive. In most cases, both parameters are indeed positive, and since the other variables in the determinant are also positive, this indicates a large region of steady-state stability. The electroanalytical process is therefore predominantly kinetically controlled.

In the absence of side reactions or other factors that could compromise the stability of the analyte or modifier—excluding the reactions predicted by the mechanism—the linear relationship between the electrochemical parameter and concentration is observed. This ensures efficient interpretation of the analytical signal, which is particularly important for monitoring mercury concentrations.

The condition  $\text{Det } J=0$  corresponds to the detection limit, manifested as monotonic instability. This can be visualized as an N-shaped segment of the steady-state voltammogram, representing the boundary between stable and unstable steady states and corresponding to steady-state multiplicity. In other words, multiple unstable steady states coexist at this point.

Oscillatory behavior, on the other hand, occurs beyond the detection limit and is associated with a Hopf bifurcation. Its occurrence requires the presence of positive-feedback-related terms in the main diagonal. Observing the main diagonal elements (6), (10), and (14), oscillatory behavior becomes possible when the kinetic parameters aaa and jjj are positive,

reflecting DEL effects from both the chemical and electrochemical stages. This feature is typical for similar systems [27,28] and can be described by the positivity of these diagonal elements.  $ak_{h_1}h(1-p)\exp(-ah)$  and  $bk_{h_2}h_2(1-p)\exp(-bh_2) > 0$ , if  $a, b > 0$  and  $jk_0p \exp\left(\frac{zF\phi_0}{RT}\right) > 0$ , 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 depend on the background electrolyte composition, as demonstrated experimentally and theoretically [27,28].

The analogous model may be used to quantify two different metals and two valent states of the same metal. Nevertheless, some specific cases – for example, parallel chemical metal-oxidation or side reactions - will require special attention and will be evaluated in our next work.

#### 4. Conclusions

From the behavior investigation of the system with the mercury cations electrochemical detection and elimination, assisted by a conducting polymer, based on mushroom polyphenols, based on *C. orellanus*, *P. involutus*, and *A. xanthodermus*, it was possible to conclude that the polyphenolic conducting polymer, based on a natural source, may be an efficient anode modifier for mercury determination and removal.

#### Author Contributions

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

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## Informed Consent Statement

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Data supporting the findings of this study are available upon reasonable request from the corresponding author.

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## Conflicts of Interest

The authors declare no conflict of interest.

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