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The Mathematical Modeling for Salvarsane Electrochemical Determination over Cobalt Oxyhydroxide and Squaraine Dye

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Abstract: In this work, for the first time, the theoretical evaluation of the possibility of electroanalytical detection of salvarsan, an antisyphilitic drug, is carried out. Cobalt oxyhydroxide, in its compound with the squarainic dye, is used as an electrode modifier. Through stability analysis, its efficiency in the Salvasano detection processes has been confirmed. The linearity of the dependence between the electrochemical parameter and the drug concentration is readily achieved and maintained. On the other hand, oscillatory and monotonic instabilities are observed in the system and are caused by the electrochemical stages' influence on the capacitance of the electrical double layer (DEL).

Keywords: salvarsane; electrochemical sensor; cobalt oxyhydroxide; squarainic dye; electrochemical oscillations; stable steady state.

1. Introduction

Arsphenamine, that is, 3-3'diamino-4-4-dihydroxyarsenebenzene (Figure 1, left) (CAS: 139-93-5, M = 368 g / mol, marketed as Salvarsane and even as "compound 606", is one of the drugs widely used in the treatment of syphilis [1–5], being one of the first developed and used for such. Recent studies have shown that arsphenamine usually enters the cyclooligomerization reaction, forming itself, a. trimer, and a pentamer (Figure 1, center and right).

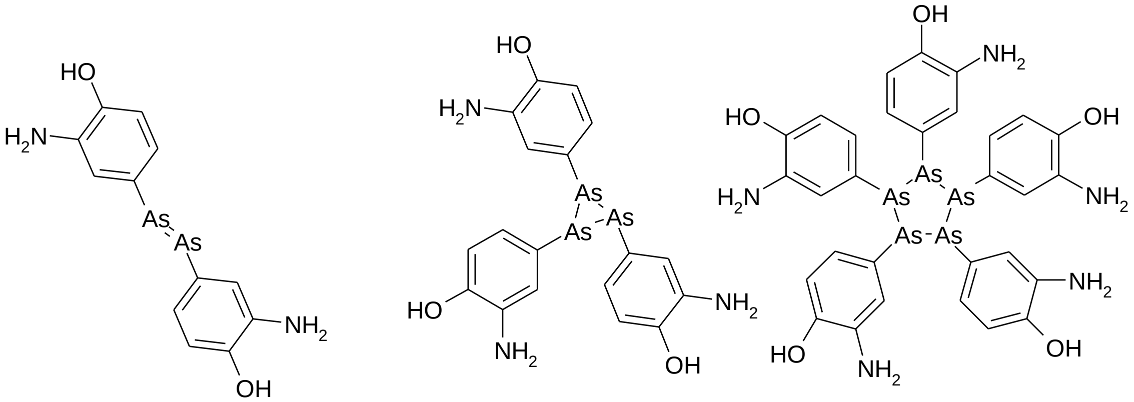

Figure 1. Salvasan and its oligomeric forms.

The compound was developed in 1910. Two years later, its derivative neosalvarsane, more soluble in water and easier to prepare, was developed. Both compounds have been used not only in the treatment of syphilis but also in the cure of trypanosomiasis [5, 6]. However, despite its effectiveness, salvasan and derivatives cannot be used in large quantities due to the presence of Arsenic, which is known for its toxicity [7–10]. Furthermore, there are difficulties in storing salvasano and its derivatives due to their high reactivity (nitrogen or noble gas atmospheres are used). That is why the use of salvasano had already decreased in the 1940s, with the discovery of penicillin. Today, salvasano is only used in some countries in Latin America, sub-Saharan Africa, and Asia. That is why the development of a method for its qualitative and quantitative detection remains current [11–14].

For now, there are no electrochemical methods of detecting salvasano. However, the other Arsenium compounds have been detected electrochemically [15,16]. For this, several modifiers have been used, including cobalt oxyhydroxide [17–20], a p-type semiconductor considered an alternative to titanium dioxide.

However, before being implemented in practice, new electroanalytical processes must be analyzed theoretically. This solves problems such as indecision about the most probable mechanism of the chemical and electrochemical steps that lead to the appearance of the electroanalytical signal, the electroanalytical process, and monotonic instabilities, characteristics of analogous systems [21–26].

The resolution of the three problems mentioned involves an important stage: an a priori theoretical investigation of the behavior of the electroanalytical system. Thus, in this work, the possibility of electrochemically determining salvasan in the presence of cobalt oxyhydroxide deposited on a squarainic dye is investigated from a mathematical perspective. Through the model development and analysis, the steady-state stability requirements are verified, as well as the conditions for oscillatory and monotonic instabilities. Furthermore, through theoretical analysis, the behavior of this system is compared (with high accuracy and without additional experimental tests) with that of similar systems [27, 28].

2. Materials and Methods

As the salvarsane molecule has several groups that tend to be oxidized (two o-aminophenol fragments and two arsenium atoms), cobalt oxyhydroxide may participate in its oxidation in different ways. In the case of the oxidation of *o*-aminophenol, CoO will be formed (Fig. 2):

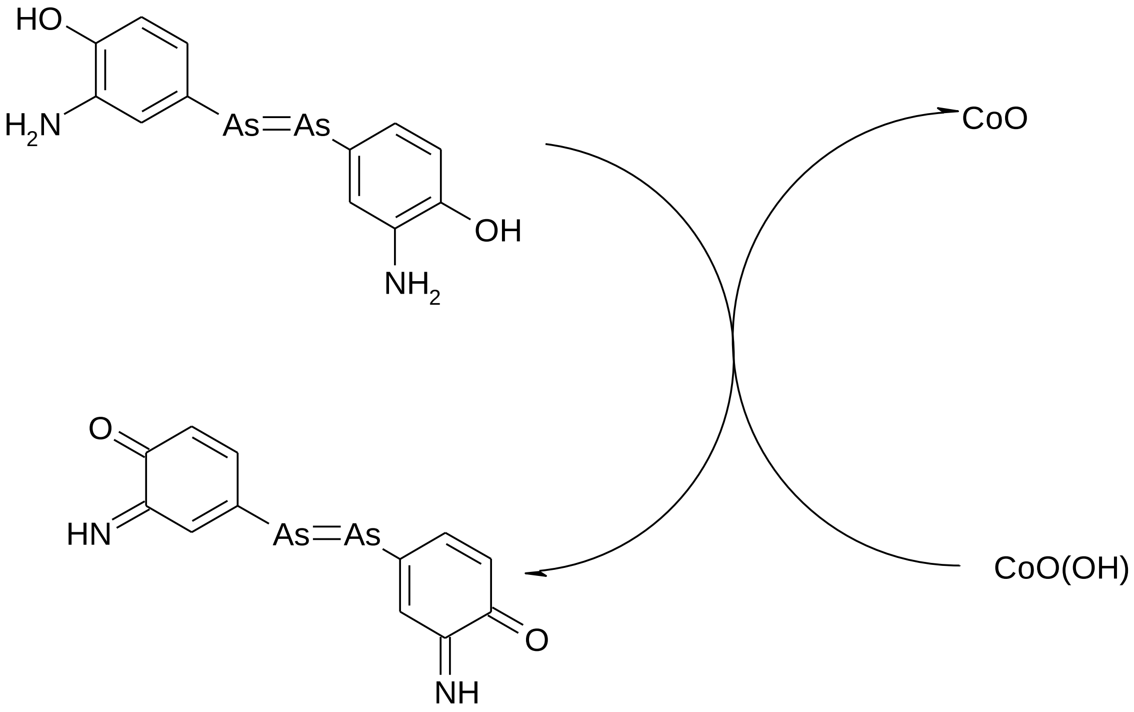

Figure 2. Oxidation of salvasano by 2-aminophenol.

When the Arsenium atom is oxidized, cobalt oxide (II–III) will be obtained (Figure 3) [16].

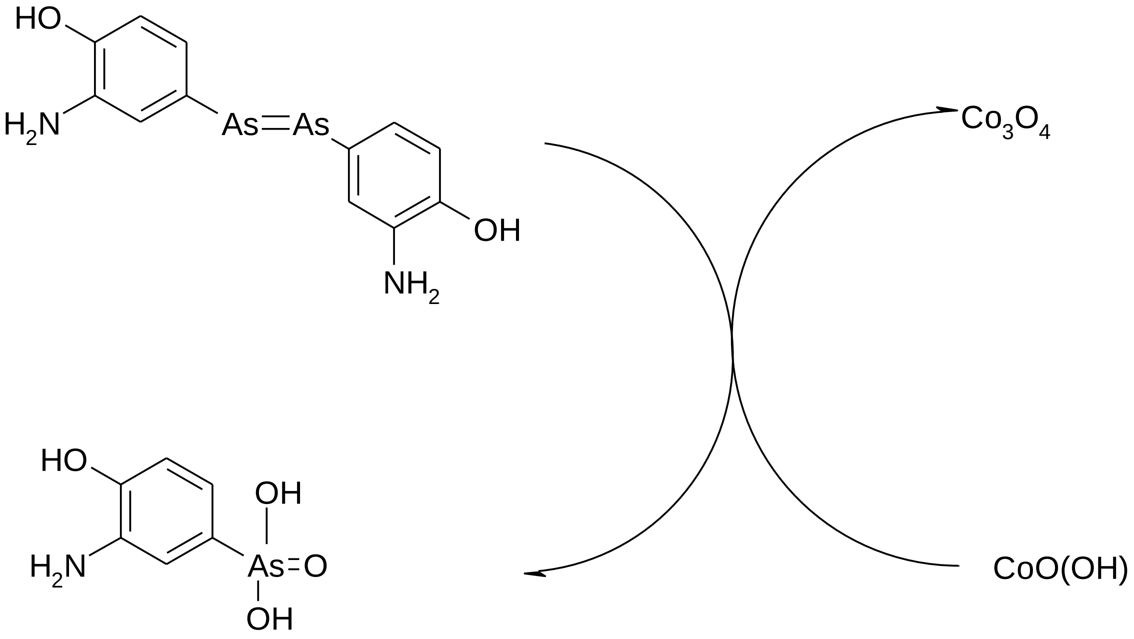

Figure 3. Oxidation of Salvasano by Arsenic.

The product of oxidation by Arsenium will be oxidized by the hydroquinonic system more readily, due to the electron-accepting influence of arsenate (V).

Thus, to describe the behavior of the system, a set of differential balance equations with three variables is introduced: s – the concentration of the salvasano in the presurface layer; c_1 – the degree of surface coating by free cobalt (II) oxide; c_2 – the degree of surface coating by cobalt oxide (II-III).

To simplify the model, we assume that the reactor is stirred intensely, which allows us to neglect the convection flow. Furthermore, we assume that the supporting electrolyte is present in excess, allowing us to underestimate the migration flow. We also assume that the concentration profile of the substances in the pre-surface layer is linear, and its thickness is constant and equal to δ.

It can be verified that the behavior of the system is described by the set of differential equations (1)

$$\begin{cases} \frac{ds}{dt} = \frac{2}{\delta}\left(\frac{S}{\delta}(s_0 - s) - r_{C1} - r_{C2}\right) \\ \frac{dc_1}{dt} = \frac{1}{\mathrm{C}_1}(r_{C1} - r_{O1}) \\ \frac{dc_2}{dt} = \frac{1}{\mathrm{C}_2}(r_{C2} - r_{O2}) \end{cases} \quad (1)$$

In which S is the diffusion coefficient of salvasan, s_0, its concentration inside the solution, C_1 and C_2 are, correspondingly, the maximum surface concentrations of CoO (OH) and Co_3O_4, and the parameters r are the corresponding reaction rates, which can be calculated according to:

$$r_{C1} = k_{C1}s(1 - C_1 - C_2)^2 \quad (2)$$

$$r_{C2} = k_{C2}s(1 - C_1 - C_2)^6 \quad (3)$$

$$r_{O1} = k_{O1}C_1 \exp\left(-\frac{F\varphi_0}{RT}\right) \quad (4)$$

$$r_{O2} = k_{O2}C_2 \exp\left(-\frac{F\varphi_0}{RT}\right) \quad (5)$$

In which the parameters k are rate constants of the respective reactions, F is the Faraday number, φ_0 is the potential jump in the electric double layer (DCE), related to the potential of zero charge, R is the universal constant of gases, and T is the absolute temperature.

In general, the behavior of this process is similar to that observed in [27,28]. However, the presence of electroanalytical cycles is felt in the appearance of two reduced forms of cobalt and, consequently, two electrochemical stages, each responsible for the appearance of instabilities, as will be described below.

3. Results and Discussion

To investigate the behavior of the system with the electroanalytical detection of salvarsane, we analyzed the set of differential equations (1) using linear stability theory. The stationary elements of the Jacobian matrix are 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 what

$$a_{11} = \frac{2}{\delta}\left(-\frac{S}{\delta} - k_{C1}(1 - C_1 - C_2)^2 - k_{C2}(1 - C_1 - C_2)^6\right) \quad (7)$$

$$a_{12} = \frac{2}{\delta}\left(2k_{C1}s(1 - C_1 - C_2)^{\square} + 6k_{C2}s(1 - C_1 - C_2)^5\right) \quad (8)$$

$$a_{13} = \frac{2}{\delta}\left(2k_{C1}s(1 - C_1 - C_2)^{\square} + 6k_{C2}s(1 - C_1 - C_2)^5\right) \quad (9)$$

$$a_{21} = \frac{1}{\mathrm{C}_1}\left(k_{C1}(1 - C_1 - C_2)^2\right) \quad (8)$$

$$a_{22} = \frac{1}{C_1}\left(-2k_{C1}(1 - C_1 - C_2)^{\square} - k_{O1}\exp\left(-\frac{F\varphi_0}{RT}\right) - pk_{O1}C_1\exp\left(-\frac{F\varphi_0}{RT}\right)\right) \quad (10)$$

$$a_{23} = \frac{1}{C_1}\left(-2k_{C1}(1 - C_1 - C_2)^{\square} \square -qk_{O1}C_1\exp\left(-\frac{F\varphi_0}{RT}\right)\right) \quad (11)$$

$$a_{31} = \frac{1}{C_2}(k_{C2}(1 - C_1 - C_2)^6) \quad (12)$$

$$a_{32} = \frac{1}{C_2}\left(-6k_{C2}s(1 - C_1 - C_2)^5 \square -pk_{O2}C_2\exp\left(-\frac{F\varphi_0}{RT}\right)\right) \quad (13)$$

$$a_{33} = \frac{1}{C_2}\left(-6k_{C2}s(1 - C_1 - C_2)^5 -k_{O2}\exp\left(-\frac{F\varphi_0}{RT}\right) -qk_{O2}C_2\exp\left(-\frac{F\varphi_0}{RT}\right)\right) \quad (14)$$

Electroanalytical process cycles are here. In this system, there are two electrochemical reactions (each with its own). Thus, the DCE will be influenced by both reactions, and *oscillatory instability* will be more likely than in similar systems involving cobalt (III) oxyhydroxide [27,28]. It will be caused by the influences mentioned above.

For the Hopf bifurcation corresponding to oscillatory instability to be realized, positive elements on the main diagonal of the matrix must be present. Looking at elements (7), (10), and (14), it can be seen that elements $-qk_{O2}C_2\exp\left(-\frac{F\varphi_0}{RT}\right)$ and$-pk_{O1}C_1\exp\left(-\frac{F\varphi_0}{RT}\right)$ They are positive if p<0 and q<0. This positivity is only obtained with the strong influences of the electrochemical reactions on the DCE capacitance, which will lead to periodic changes in the capacitance and conductivity of the layer. Oscillations are expected to be frequent and of low amplitude.

To investigate the steady-state stability of this system, we apply the Routh-Hurwitz criterion to the set of differential equations (1). Simplifying the analysis of the matrix, we introduce new variables, so that their determinant is described according to:

$$\frac{2}{\delta C_1 C_2}\begin{vmatrix} -\sigma - \Sigma - \varsigma & \alpha + \beta & \alpha + \beta \\ \Sigma & -\alpha - \xi - \omega & -\alpha - \psi \\ \varsigma & -\beta - \omega & -\beta - \lambda - \psi \end{vmatrix} \quad (15)$$

Applying the Det requirement J <0, leaving the criterion, we obtain the stability condition, described as:

$$-\sigma(\xi\beta + \omega\beta + \alpha\lambda + \xi\lambda + \omega\lambda + \alpha\psi + \xi\psi - \beta\psi - \alpha\omega) - \Sigma(\xi\beta + 2\omega\beta + \xi\lambda + \omega\lambda + \xi\psi - 2\beta\psi - \beta\lambda) - \varsigma(\alpha\lambda + \xi\lambda + \omega\lambda + \alpha\psi + \xi\psi - 2\alpha\omega - \xi\alpha) < 0 \quad (16)$$

This corresponds to an efficient electroanalytical system, although the steady state stability zone is narrower than in similar systems [27,28]. The electroanalytical process will be controlled both by the diffusion of the analyte and by the kinetics of the process.

Requirement (16) is easily satisfied if p>0 and q>0, describing the weakness of the influences of the electrochemical stages of the electroanalytical process. Since there are no side reactions in the system, the steady state stability corresponds to the linear dependence between the electrochemical parameter and the concentration of the analyte.

When the steady state is in effect, the determinant has negative values. When the determinant is canceled, the detection limit is obtained, described by monotonic instability. His spot serves as a margin between stable and unstable, and, for that matter, it is described as:

$$-\sigma(\xi\beta + \omega\beta + \alpha\lambda + \xi\lambda + \omega\lambda + \alpha\psi + \xi\psi - \beta\psi - \alpha\omega) - \Sigma(\xi\beta + 2\omega\beta + \xi\lambda + \omega\lambda + \xi\psi - 2\beta\psi - \beta\lambda) - \varsigma(\alpha\lambda + \xi\lambda + \omega\lambda + \alpha\psi + \xi\psi - 2\alpha\omega - \xi\alpha) = 0 \quad (17)$$

The presence of trimer and pentamer will also lead to the appearance of additional peaks, referring to the formation of cyclooligomers. Therefore, the model will be modified as it will be described in one of our next works.

4. Conclusions

The theoretical analysis of the electrochemical detection of salvasan on cobalt oxyhydroxide, in combination with both divalent cobalt oxide and cobalt oxide (II, III), concluded that the analyte-detection mechanism is hybrid, and that it forms an efficient electroanalytical system with a stable complex. The electroanalytical process is controlled in the same way by diffusion or by reaction. Oscillatory behavior in this system is more likely than in similar systems, since two stages influence the capacitance, conductivity, and ionic strength of the electrical double layer. The presence of salvarsane cyclooligomers has a significant impact on the system's behavior. This impact contributes to the appearance of an additional peak.

Author Contributions

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

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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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