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The Stability Study for the Electrochemical Determination of Permethrin over Cobalt (III) Oxyhydroxide in Neutral Medium

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Abstract: The electrochemical determination of permethrin in neutral solutions using CoO(OH) has been investigated. A corresponding mathematical model was developed and examined through linear stability theory and bifurcation analysis. The results indicate that the analytical process may be more efficient on CoO(OH) in neutral media than under alkaline conditions. The electrochemical response should remain clear and readily interpretable. Furthermore, both oscillatory and monotonic instabilities were confirmed. These oscillations arise from DEL effects associated with the electrochemical step and the hydrolysis chemical stage.

Keywords: medication safety; permethrin; cobalt (III) oxyhydroxide; electrochemical sensors; steady-state stability.

1. Introduction

Demodicosis [1–4] is a disease caused by two arachnid species, Demodex folliculorum and Demodex brevis, which were first described in the middle of the 19th century [1].

Demodex folliculorum, at all developmental stages, inhabits small hair follicles and eyelash follicles. Both immature and adult forms feed on epithelial cells, causing follicular distention, hyperplasia, and increased keratinization. In eyelashes, this leads to cuffing, which is composed of keratin and lipid material. *Demodex brevis*, also at all stages, resides in the sebaceous glands of eyelashes, small hair follicles, and the lobules of the meibomian glands. Adults and immature forms consume glandular cells at these sites and, in cases of heavy infestation, can impair the formation of the superficial lipid layer of the tear film. Demodectic mites induce histologically observable tissue and inflammatory changes, epithelial hyperplasia, and follicular plugging. [2–4]. *Demodex* was considered a commensal but is now considered parasitic. However, in most cases, the mites go unnoticed and cause no adverse symptoms; in certain cases (usually related to a suppressed immune system caused by stress or illness), mite populations can dramatically increase. This results in a condition known as demodicosis.

Depending on the location, it may be small pustules (pimples or pustules) at the hair follicle opening, on inflamed, congested skin. Demodicosis is accompanied by itching, swelling, and erythema of the eyelid margins, as well as the appearance of scales at the base of the eyelashes. Typically, patients complain of eyestrain. Characteristic of the view of the affected century: plaque on the edge of the eyelids, eyelashes stuck together, surrounded by crusts as a clutch.

One of the substances used for demodicosis treatment is permethrin (Figure 1, right) [5–7], also sold under the name Nix, among others. It is a naturally derived insecticide (based on pyrethrin I – Figure 1 on the left) and arachnicide, capable of use as a drug in human and veterinary medicine for treating acaridosis and other mite-related diseases. It may be applied to human or animal bodies, or sprayed onto cloth (including military uniforms) and mosquito nets, killing mites and insects on contact. Although it is considered safe, its action is dose-related and may include side effects like nausea, headache, muscle weakness, excessive salivation, shortness of breath, and seizures. Worker exposure to the chemical can be monitored by measurement of the urinary metabolites, while severe overdose may be confirmed by measurement of permethrin in serum or blood plasma [8–12]. Thus, the development of a method capable of rapidly and efficiently detecting its concentration is urgent [13, 14].

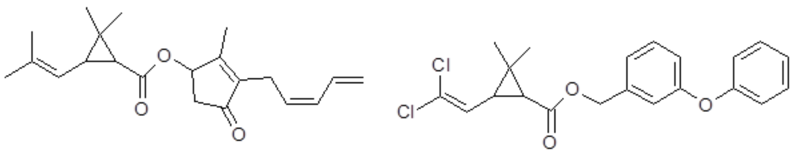

Figure 1. Pyrethrin I and permethrin.

Containing the easily oxidized and reduced groups, permethrin is electrochemically active for both anodic and cathodic processes. So, cobalt(III) oxyhydroxide, already used for similar compounds [15–18], could serve as an interesting modifier for permethrin electrochemical detection. Therefore, in this work, we theoretically describe the possibility of electrochemically determining permethrin in a neutral medium at a CoO(OH)-modified electrode. This is realized through the mechanism suggestion, its mathematical description via the development and analysis of the mathematical model from a stability perspective, and its comparison with similar processes [19–21].

2. Materials and Methods

Even in a neutral medium, the permethrin molecule undergoes hydrolysis at the geminal dichlorohalogen group (Figure 2).

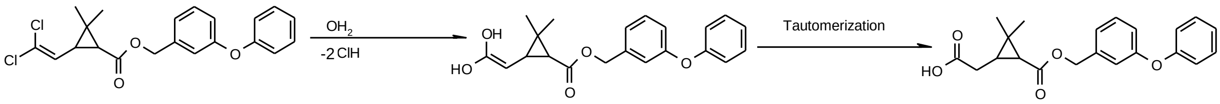

Figure 2. Permethrin hydrolysis.

Two electrooxidation scenarios, including cyclopropane ring cleavage and furan ring closure, may thereby be realized (Figure 3).

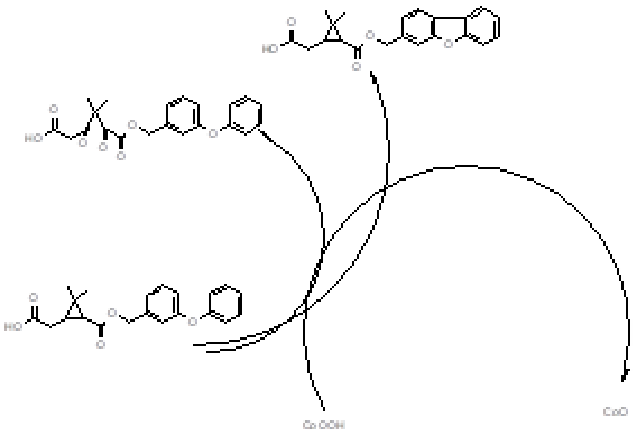

Figure 3. The scheme of the indirect electroanalytical process.

The last scenario may also give a path to electropolymerization. Given that hydrolysis may proceed rapidly, the electroanalytical process in this case is indirect.

Taking this into account and taking some assumptions [19–21], we describe the system´s behavior by equation set (1):

$$\begin{cases} \frac{dp}{dt} = \frac{2}{\delta}\left(\frac{\Delta}{\delta}(p_0 - p) - r_1\right) \\ \frac{dp*}{dt} = \frac{2}{\delta}(r_1 - r_{21} - r_{22}) \\ \frac{d\theta}{dt} = \frac{1}{C}(r_2 - r_3) \end{cases} \quad (1)$$

In which p and p* are permethrin and its hydrolysis product pre-surface layer concentrations, Δ is the diffusion coefficient, p_0 is the permethrin bulk concentration, C is the CoO maximal surface concentration, θ is the CoO surface coverage degree, and the parameters r are the corresponding reaction rates (2 – 5):

$$r_1 = k_1 p \exp(-ap) \quad (2)$$

$$r_2 = k_2 p * (1 - \theta) \quad (3)$$

$$r_{21} = k_2 p * (1 - \theta)^3 \quad (4)$$

$$r_{22} = k_2 p * (1 - \theta)^2 \quad (5)$$

$$r_3 = k_3 \theta \exp\frac{F\varphi_0}{RT} \quad (6)$$

being the parameters k, the corresponding reaction rate constants, a the DEL-related kinetic parameter, F the Faraday number, φ_0 the DEL potential slope, related to the zero-charge potential, R the universal gas constant, and T the absolute temperature.

The behavior of this system doesn´t tend to be autocatalytic and, in general, has to obey the classical model, also known as pyriproxyfen, but with the additional details related to ionic compound formation during hydrolysis, as described below.

3. Results and Discussion

To investigate the system's behavior in electrochemical detection of permethrin via its alkaline-assisted hydrolysis, we use linear stability theory to analyze equation set (1). The steady-state Jacobian functional matrix elements for this equation set may be calculated as:

$$\begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix} \quad (7)$$

In which:

$$a_{11} = \frac{2}{\delta}\left(-\frac{\Delta}{\delta} - k_1 \exp(-ap) + ak_1 p \exp(-ap)\right) \quad (8)$$

$$a_{12} = 0 \quad (9)$$

$$a_{13} = 0 \quad (10)$$

$$a_{21} = \frac{2}{\delta}(k_1 \exp(-ap) - ak_1 p \exp(-ap)) \quad (11)$$

$$a_{22} = \frac{2}{\delta}(-k_2(1-\theta)^3 - (1-\theta)^2) \quad (12)$$

$$a_{23} = \frac{2}{\delta}(3k_2 p * (1-\theta)^2 + 2k_2 p * (1-\theta)) \quad (13)$$

$$a_{31} = 0 \quad (14)$$

$$a_{32} = \frac{1}{C}(k_2(1-\theta)) \quad (15)$$

$$a_{33} = \frac{1}{C}\left(-3k_2 p * (1-\theta)^2 - 2k_2 p * (1-\theta) + jk_3\theta \exp\left(\frac{F\varphi_0}{RT}\right)\right) \quad (16)$$

In which the parameter j describes the influence of the electrochemical oxidation on the double electric layer (DEL).

Similarly to [19–21], the oscillatory behavior for this system is possible, and it will be caused by influences of electrochemical oxidation on DEL capacitances of both permethrin hydrolysis and CoO electrooxidation, regenerating CoO(OH), described by the positivity of main-diagonal elements $jk_4\theta \exp\left(\frac{2F\varphi_0}{RT}\right) > 0$, if j>0 and $ak_1 p \exp(-ap) > 0$, if a>0 (positive main-diagonal elements describe the positive callback).

These influences (and also the oscillations' amplitude and frequency) will depend not only on the solution content and ionic strength (which are "stronger" in the presence of phenoxide ions), but also on the modifying material and its doping level, as observed in analogous systems. In this case, it may depend on the composition of the matrix over which cobalt (III) oxyhydroxide is deposited.

The steady-state stability analysis, made using the Routh-Hurwitz criterion, is applied to this system. Applying this criterion and the Det J<0 condition, surging of it, it is possible to obtain the steady-state stability requirement, expressed as:

$$\frac{4}{\Gamma\delta^2} K_1(k_2 W(1-\theta) < 0 \quad (17)$$

In which $K_1 = -\frac{\Delta}{\delta} - k_1 \exp(-ap) + ak_1 p \exp(-ap)$ and $W = -k_3 \exp\left(\frac{F\varphi_0}{RT}\right) + jk_3\theta \exp\left(\frac{F\varphi_0}{RT}\right)$ may be positive or negative, depending on the value of the parameter j. If it is positive, as in the absence or fragility of oxidation effects in DEL capacitances, the left-hand side of the inequation is always negative, and steady-state stability is warranted. This means that CoO(OH) may be compatible as a modifier for permethrin.

Suppose the kinetic effect of an electrochemical reaction is equal to its capacitance effect. In that case, the parameter W will be equal to zero, and the Jacobian determinant will be equal to nil. So, the *monotonic instability* will be realized, and the detection limit will be identified. In this case, its condition will be described as:

$$\frac{4}{\Gamma\delta^2}K_1(k_2W(1-\theta)=0 \qquad (18)$$

If hydrolysis is relatively slow under the analysis conditions, the model will be transformed to describe semidirect or direct analysis. Those cases will be described in the next work.

Also, it is important to keep in mind that, to reduce the influence of pH decrease during hydrolysis, CoO(OH) may be deposited on a polymer with basic properties (e.g., chitosan or basic conducting polymers). By this, the protons formed during hydrolysis interact with the basic polymer, neutralizing the pH in the electroanalytical process, reducing the hydrolysis DEL influences, and stabilizing the system.

4. Conclusions

Analyzing the system using CoO(OH)-assisted electrochemical determination of permethrin indicates that the electroanalytical process is efficient. Although the oscillatory behavior is more probable than in the simplest case, it does not destabilize the system much in a neutral medium. As for the electroanalytical process, it is either diffusion or kinetically controlled, and the electroanalytical signal is easy to interpret.

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.; M.V.K.; S.C.O.; V.M.O.; M. P.K.; I.M.B.; A.I.B.; O.V.L.; V.V.K.; V.I.D.; Z.O.K.; Y.G.I.; O.P.M.; O.O.M.; M.V.M.; O.V.A.; P.I.Y.; Z.Z.M.; M.V.K.; O.O.A.; O.P.A.; J.R.G.; J.I.F.P.M.; V.P.M.; O.A.B.; O.V.K. and O.O.V.. formal analysis, V.V.T.; T.V.M.; M.V.K.; S.C.O.; V.M.O.; M. P.K.; O.V.A.; P.I.Y.; Z.Z.M.; M.V.K.; O.O.A.; O.P.A.; J.R.G.; J.I.F.P.M.; V.P.M.; O.A.B.; O.V.K. and O.O.V..investigation, V.V.T.; T.V.M.; M.V.K.; S.C.O.; V.M.O.; M. P.K.; I.M.B.; A.I.B.; O.V.L.; V.V.K.; V.I.D.; Z.O.K.; Y.G.I.; O.P.M.; O.O.M.; M.V.M.; O.V.A.; P.I.Y.; Z.Z.M.; M.V.K.; O.O.A.; O.P.A.; J.R.G.; J.I.F.P.M.; V.P.M.; O.A.B.; O.V.K. and O.O.V..; resources, V.V.T.; T.V.M.; M.V.K.; S.C.O.; V.M.O.; M. P.K.; I.M.B.; A.I.B.; O.V.L.; V.V.K.; V.I.D.; Z.O.K.; Y.G.I.; O.P.M.; O.O.M.; M.V.M.; O.V.A.; P.I.Y.; Z.Z.M.; M.V.K.; O.O.A.; O.P.A.; J.R.G.; J.I.F.P.M.; V.P.M.; O.A.B.; O.V.K. and O.O.V..data curation, V.V.T.; T.V.M.; M.V.K.; S.C.O.; V.M.O.; M. P.K.; I.M.B.; A.I.B.; O.V.L.; V.V.K.; V.I.D.; Z.O.K.; Y.G.I.; O.P.M.; O.O.M.; M.V.M.; O.V.A.; P.I.Y.; Z.Z.M.; M.V.K.; O.O.A.; O.P.A.; J.R.G.; J.I.F.P.M.; V.P.M.; O.A.B.; O.V.K. and O.O.V.. ; writing—original draft preparation, V.V.T.; T.V.M.; M.V.K.; Z.O.K.; Y.G.I.; O.P.M.; O.O.M.; M.V.M.; O.V.A.; P.I.Y.; Z.Z.M.; M.V.K.; O.O.A.; O.P.A.; J.R.G.; J.I.F.P.M.; V.P.M.; O.A.B.; O.V.K. and O.O.V.. ; writing—review and editing, V.V.T.; T.V.M.; M.V.K.; S.C.O.; V.M.O.; M. P.K.; I.M.B.; A.I.B.; O.V.L.; V.V.K.; V.I.D.; Z.O.K.; Y.G.I.; O.P.M.; O.O.M.; M.V.M.; O.V.A.; P.I.Y.; Z.Z.M.; M.V.K.; O.O.A.; O.P.A.; J.R.G.; J.I.F.P.M.; V.P.M.; O.A.B.; O.V.K. and O.O.V.. visualization, V.V.T.; T.V.M.; M.V.K.; S.C.O.; V.M.O.; M. P.K.; I.M.B.; A.I.B.; O.V.L.; V.V.K.; V.I.D.; Z.O.K.; Y.G.I.; O.P.M.; O.O.M.; M.V.M.; O.V.A.; P.I.Y.; Z.Z.M.;

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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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