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The Mathematical Modeling for Permethrin and Fenvalerate Electrochemical Determination on VO(OH)

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Abstract: For the first time, the possibility of permethrin electrochemical determination in the presence of fenvalerate has been theoretically described. The electrochemical determination of both permethrin and fenvalerate occurs via the ester group. Nevertheless, fenvalerate may also be reduced by the nitrile group, providing an additional DEL-affecting chemical stage. Nevertheless, vanadium (III) oxyhydroxide may be used as an excellent electrode modifier for the electrochemical determination of both insecticides in mildly acidic up to neutral solutions.

Keywords: fenvalerate; permethrin; vanadium(III) oxyhydroxide; electrochemical sensors; electrochemical oscillations; stable steady-state.

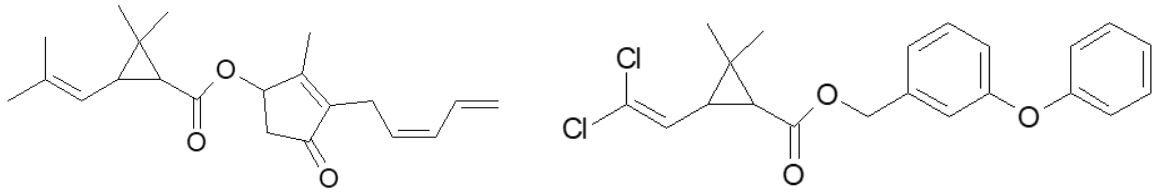
1. Introduction

Demodicosis [1–4] is a disease caused by two arachnid species, *Demodex folliculorum* and *Demodex brevis*, known from the middle of the XIX century [1].

Demodex folliculorum (all stages) is found in small hair follicles and eyelash hair follicles. In all forms, immature and adult, it consumes epithelial cells, produces follicular distention and hyperplasia, and increases keratinization, leading (in eyelashes) to cuffing, which consists of keratin and lipid moieties. *Demodex brevis* (all stages) is present in the eyelash sebaceous glands, small hair sebaceous glands, and lobules of the meibomian glands. Adults and immature forms consume the gland cells in all of these loci and, when infestations are heavy, can affect the formation of the superficial lipid layer of the tear film coacervate. Demodectic mites produce histologically observable tissue and inflammatory changes, epithelial hyperplasia, and follicular plugging [2–4]. Demodex was considered a commensal but is now considered a parasite. However, in most cases, the mites go unobserved without any adverse symptoms, though in certain cases (usually related to a suppressed immune system functionality 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 exit of hair, placed on inflamed, congested skin. Demodicosis is accompanied by itching, swelling, erythema of the eyelid margins, and the appearance of scales at the base of the eyelashes. Typically, patients complain of eyestrain. Characteristic of 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 1b) [5–7], also sold as Nix, among other names. It is a naturally inspired insecticide (based on pyrethrin I–Figure 1a), arachnicide, capable of being used as a human and veterinary medicine drug for treating acaridosis and other mite-related diseases. It may be applied to human or animal bodies or sprayed on 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 detecting its concentration rapidly and efficiently is really actual [13,14].



(a) **(b)**

Figure 1. (a) Pyrethrin I; **(b)** permethrin.

Fenvalerate (Figure 2) is another pyrethrin-based insecticide used in insecticidal fogs and aerosols [15–18]. Although it has moderate mammalian toxicity, it is highly toxic to bees and fishes [19–21]. Moreover, it is harmful to the skin and eyes on contact, as well as on swallowing. Many pesticide formulations combine the use of fenvalerate and permethrin, which is why the determination of fenvalerate alongside permethrin is really actual [22–24].

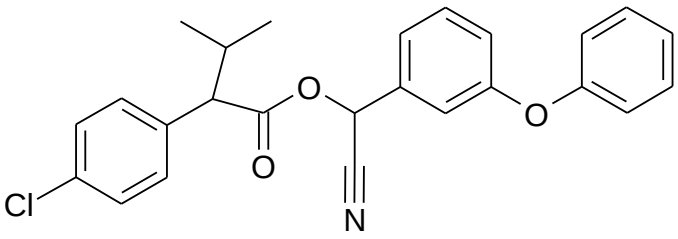


Figure 2. Fenvalerate.

Containing the easily reduced groups, both permethrin and fenvalerate may be electrochemically reduced for electroanalytical purposes. The electrochemical reduction will include proton and electron transfer towards the analyte. Thus, the vanadium (III) oxyhydroxide [25,26] could serve as an interesting modifier for the electrochemical detection of both of the analytes. Therefore, in this work, we describe theoretically the possibility of the electrochemical determination of permethrin and fenvalerate in neutral and mildly acidic medium over VO(OH)-modified electrode. This is realized by the mechanism suggestion, its mathematical description by development and analysis of the mathematical model from the stability point of view, and its comparison with similar processes [27,28].

2. Materials and Methods

Both of the pesticides contain an ester functional group, which is the most obvious object for electrochemical reduction in both acidic and neutral mediums. Moreover, fenvalerate also contains a nitrile group, which will be reduced to amino group in the same conditions (Figure 3).

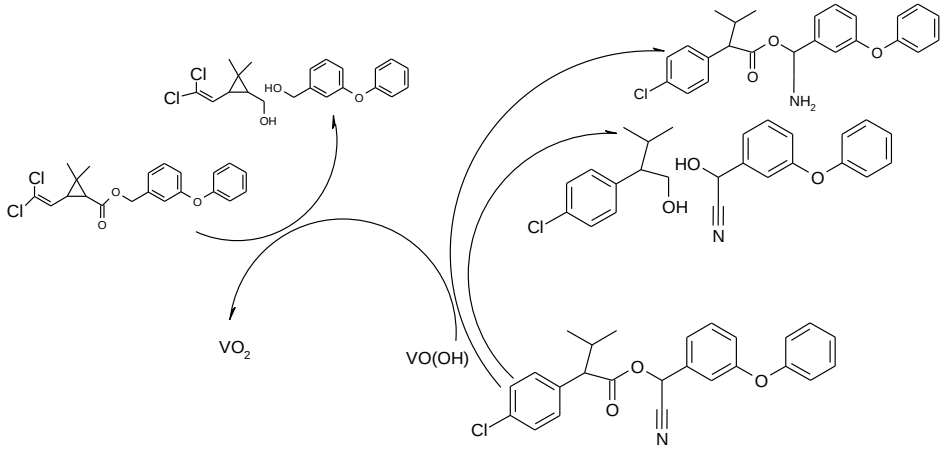


Figure 3. The schematic representation of the electroanalytical process.

The Nitrile group will be reduced to the amino group, which will be ionized in an acidic medium and will thereby affect the double electric layer (DEL) ionic force. The electrochemical dechlorination of both of the compounds may also occur, and it also affects the DEL. Nevertheless, it will require lower pH and cathode potential values.

For this reason, considering certain assumptions [27,28], we describe the behavior of this system by the trivariate equation-set (1):

$$\begin{cases} \frac{dp}{dt} = \frac{2}{\delta}\left(\frac{\Delta}{\delta}(p_0 - p) - r_1\right) \\ \frac{df}{dt} = \frac{2}{\delta}\left(\frac{\Phi}{\delta}(f_0 - f) - r_{21} - r_{22}\right) \\ \frac{dv}{dt} = \frac{1}{V}(r_1 + r_{21} + r_{22} - r_3) \end{cases} \quad (1)$$

Herein, p and f are the permethrin and fenvalerate pre-surface concentrations, p_0 and f_0 are their bulk concentrations, Δ and Φ stand for their diffusion coefficients, v is vanadium oxyhydroxide surface coverage degree, V is its maximal surface concentration, and the parameters r stand for the correspondent reaction rates, calculated as:

$$r_1 = k_1 p(1-v)^4 \quad (2)$$

$$r_{21} = k_{21} f(1-v)^4 \quad (3)$$

$$r_{22} = k_{22} f(1-v)^6 \exp(-af) \quad (4)$$

$$r_3 = k_3 v \exp\left(-\frac{F\varphi_0}{RT}\right) \quad (5)$$

The parameters k are the correspondent 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 terms, has to obey the classical model for VO(OH)-assisted ester electrochemical reduction to the correspondent alcohols, but with the additional details linked to ionic compounds formation during the hydrolysis, as described below.

3. Results and Discussion

To investigate the behavior of the system with the electrochemical detection of permethrin and fenvalerate on VO(OH), we investigate the equation set (1) alongside the algebraic relations (2–4) by means of linear stability theory. The steady-state Jacobian functional matrix elements for this equation set may be expressed 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:

$$a_{11} = \frac{2}{\delta}\left(-\frac{\Delta}{\delta} - k_1(1-v)^4\right) \quad (7)$$

$$a_{12} = 0 \quad (8)$$

$$a_{13} = \frac{2}{\delta}(4k_1(1-v)^3) \quad (9)$$

$$a_{21} = 0 \quad (10)$$

$$a_{22} = \frac{2}{\delta}\left(-\frac{\Phi}{\delta} - k_{21}(1-v)^4 - k_{22}(1-v)^6\exp(-af) + ak_{22}f(1-v)^6\exp(-af)\right) \quad (11)$$

$$a_{23} = \frac{2}{\delta}(k_{21}(1-v)^4 + k_{22}(1-v)^6\exp(-af)) \quad (12)$$

$$a_{31} = \frac{1}{V}(k_1(1-v)^4) \quad (13)$$

$$a_{32} = \frac{1}{V}(k_{21}(1-v)^4 + k_{22}(1-v)^6\exp(-af) - ak_{22}f(1-v)^6\exp(-af)) \quad (14)$$

$$a_{33} = \frac{1}{V}\left(-\frac{2}{\delta}(4k_1(1-v)^3) - k_{21}(1-v)^4 - k_{22}(1-v)^6\exp(-af) - k_3\exp\left(-\frac{F\varphi_0}{RT}\right) + jk_3 v\exp\left(-\frac{F\varphi_0}{RT}\right)\right) \quad (15)$$

Avoiding the cumbersome expression during the determinant analysis, we introduce new variables and rewrite the determinant as (16):

$$\text{Det J} = \frac{4}{\delta^2 V}\begin{vmatrix} -\kappa - \Xi & 0 & N \\ 0 & -\varphi - P & \Phi \\ \Xi & P & -N - \Phi - \Omega \end{vmatrix} \quad (16)$$

Considering that:

$$-Det\,J\begin{cases}>0, for\ steady-state\ stability\\ =0\ monotonic\ instability\end{cases} \quad (17)$$

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 (18):

$$-\kappa(\varphi N+\varphi\Phi+\varphi\Omega+PN+P\Omega)-\Xi(2\varphi N+\varphi\Phi+\varphi\Omega+2PN+P\Omega)\begin{cases}>0, curve\ linearity\\ =0, detection\ limit\end{cases} \quad (18)$$

If –Det J>0, the Routh-Hurwitz stability criterion is valid, and the steady-state is thereby stable, providing an efficient steady-state drug determination. Moreover, the wide stability region lets us use this system in pharmaceutical formulations and biological liquids, as well as in the environment.

This criterion is readily satisfied if the kinetic parameters P and Ω are positive. In the vast majority of the cases, they both have positive signs, and considering that the other variables in the determinant are positive, it indicates the vast steady-state stability topological region. The electroanalytical process is mostly kinetically controlled.

In the absence of the side reactions or other factors capable of compromising the analyte and (or) modifier stability, excluding the reactions foreseen by the mechanism, the linearity between the electrochemical parameter and concentration is observed, providing an efficient analytical signal interpretation, which is really important for drug concentration monitoring.

The condition Det J=0 corresponds to the detection limit, manifested by the *monotonic instability*. It may be seen as an N-shaped part of the steady-state voltammogram, depicts the margin between stable and unstable states, and corresponds to 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 addendums in main diagonal elements.

Observing the main diagonal elements (7), (11), and (15), 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 [27,28] and may be described by the positivity of the elements $ak_{22}f(1-v)^6\exp(-af)$, if a>0 and $jk_rv\exp\left(-\frac{F\varphi_0}{RT}\right)>0$, 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 [27,28].

At lower pH and/or lower cathodic potentials, both compounds' cathodic dechlorination is added. It becomes similar to that observed for sucralose. In this case, a new addendum is added to equations 1 and 2 of the equation set (1), transforming it into (19):

$$\begin{cases}\frac{dp}{dt}=\frac{2}{\delta}\left(\frac{\Delta}{\delta}(p_0-p)-r_1-r_{d1}\right)\\ \frac{df}{dt}=\frac{2}{\delta}\left(\frac{\Phi}{\delta}(f_0-f)-r_{21}-r_{22}-r_{d2}\right)\\ \frac{dv}{dt}=\frac{1}{V}(r_1+r_{21}+r_{22}-r_3)\end{cases} \quad (19)$$

The dechlorination scenario will affect the DEL ionic force, and consequently, the steady-state stability and the equation set (19) will describe a somewhat more dynamic behavior, which will be described in our next systems.

4. Conclusions

From the theoretical description of permethrin and fenvalerate electrochemical cathodic determination by VO(OH)-modified cathode, it has been possible to conclude it may be an excellent modifier for the quantification of both pesticides in different neutral and mildly acidic media. The electroanalytical process is mostly kinetically controlled. The oscillatory behavior in this system may be caused by DEL influence by electrochemical and fenvalerate-related chemical stages. The system may be used as an electroanalytical, providing efficient analytical signal interpretation.

Author Contributions

All authors have read and agreed to the published version of the manuscript.

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Informed Consent Statement

Not applicable.

Data Availability Statement

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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