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The Theoretical Description for Sulfite and Nitrite Anodic Detection and Removal from Wine over Poly(9-Triphenylphosphazo)Acridine-Modified Electrode

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Abstract: A novel electroanalytical process for nitrite and sulfite anodic electrochemical determination in wine, based on the 9-triphenylphosphazoacridine conducting polymer as an anode modifier, has been investigated. 9-triphenylphosphazoacridine monomer may be obtained from 9-chloroacridine via 9-azidoacridine by reacting it with triphenylphosphine in ether. As for the sulfite and nitrite electrooxidation to sulfate and nitrate correspondently, it is given in conducting polymer matrix by two doping types. The analysis of the model has confirmed the efficiency of poly(9-triphenylphosphazo)acridine as an electrode modifier for sulfite and nitrite determination in wine, although the oscillatory behavior is relatively probable.

Keywords: nitrite; sulfite; electrochemical sensor, acridine derivative; conducting polymer; electrochemical oscillations; stable steady-state

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1. Introduction

Both sulfites and nitrites are ions naturally occurring in food and beverages [1 – 4] from correspondent plant biosynthesis. Moreover, sulfites are widely used as conservant food additives in food preparation, storage, and even in natural violin and guitar catgut string production [4].

Nevertheless, sulfites and especially nitrite high concentrations in food and drink may be dangerous for human organisms [5 – 8]. Nitrite ion is thereby transformed into the highly toxic, mutagenic, and cancerogenic C- and N-nitrosocompounds. As for sulfite, it provokes hypersensitivity, allergic reaction, vitamin deficiency, and dysbiosis in gut microbiota (the same effect in violin/guitar string production). Moreover, both of the ions serve as markers for acid rain areas, the reason why their determination in food, beverages, and natural water is really actual [9 – 14].

Both ions are amphoteric from the electrochemical point of view, as they correspondingly possess trivalent nitrogen and tetravalent sulfur. Therefore, both cathodic and anodic processes are viable for these ions. In this aspect, the anodic process involving the neutral conducting polymer [15 – 22] would be an interesting option from both electroanalytical and removal points of view, as both ions enter the polymer matrix by doping, thereby oxidizing. If the monomer possesses basic groups or pyridinic nitrogen atoms, two types of doping will be realized.

The above-mentioned aspects are important for the mechanistic and dynamic behavior of the electroanalytical process. Moreover, the presence of the electrochemical instabilities, which is difficult to interpret the analytical signal, may be associated with the conducting polymer doping and electrooxidation [22 – 25]. Those instabilities are typical and may limit the electroanalytical and removal use of the electrochemical process. In order to foresee the possibility of the realization of the electrochemical instabilities, like the effect they may produce, it's necessary to investigate the process from the mechanical point of view and analyze its behavior theoretically.

So, the goal of this work is the mechanical evaluation for sulfite and nitrite determination and removal from wine on poly(9-triphenylphosphazo)acridine-modified anode, in which two doping mechanisms are realized. The correspondent mathematical model is developed and analyzed by means of linear stability theory and bifurcation analysis. The theoretical investigation includes the comparison of the behavior of this system with that of similar ones [26 – 28].

2. Materials and Methods

The 9-triphenylphosphazoacridine monomer may be synthesized from 9-chloroacridine via 9-azidoacridine (Fig. 1), as reported in [26]. The monomer is obtained as a yellow-green crystalline compound.

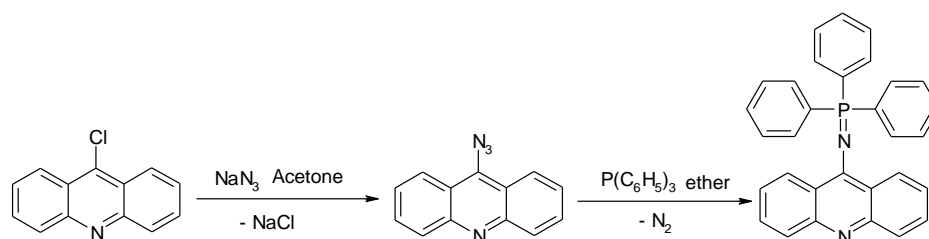


Figure 1. The synthesis of 9-phosphazoacridine monomer.

Taking into account that the triphenylphosphazenic moiety has highly donating properties (donation of 20 electrons towards the acridine ring), the color of the monomer (yellow-green) slightly differs from the other acridine derivatives, which are mostly red, orange and yellow. Moreover, its polymerization potential is slightly lower than the other acridine derivatives, including the yet polymerized acridine orange and acridine yellow (acriflavine) [18-21, 26]. The polymerization will be thereby realized via positions 2 and 7 of the acridine ring, yielding an alternant conducting polymer (Fig. 2).

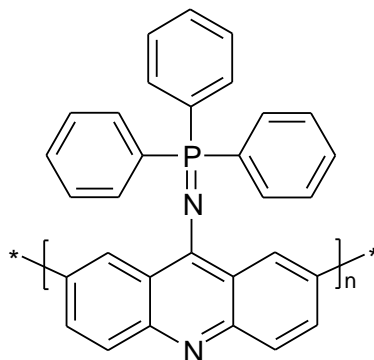


Figure 2. 9-phosphazoacridine polymerization scheme.

Two types of doping are possible in this case. Besides the classical conducting polymer doping, in which the dopant-ion enters the matrix by cationic site, the N-doping, in which the pyridinic nitrogen atom is protonized or quaternized is also realized. Moreover, if the classical doping is purely electrochemical, the second is purely chemical. Both influence the double electric layer (DEL) electrophysical and electrochemical properties, as described below.

Taking this into account and accepting some assumptions [26 – 28], we describe the behavior of this system by the balance differential equation-set (1):

$$\begin{cases} \frac{dn}{dt} = \frac{2}{\delta} \left(\frac{\Delta}{\delta} (n_0 - n) - r_{c1} - r_{n1} \right) \\ \frac{ds}{dt} = \frac{2}{\delta} \left(\frac{D}{\delta} (s_0 - s) - r_{c2} - r_{n2} \right) \\ \frac{dp}{dt} = \frac{1}{P} (r_{c1} + r_{n1} + r_{c2} + r_{n2} - r_o) \end{cases} \quad (1)$$

in which n and s are nitrite and sulfite concentrations in the pre-surface layer, n_0 and s_0 are the bulk concentrations of the correspondent ions, δ is the pre-surface layer thickness, Δ , and D are the diffusion coefficients, p is the doped polymer surface coverage degree, P is the doped polymer maximal surface concentration, and the parameters r stand for the correspondent reaction rates:

$$r_{c1} = k_{c1}n(1 - p) \exp\left(\frac{x_1 F \phi_0}{RT}\right) \quad (2)$$

$$r_{n1} = k_{n1}n(1 - p) \exp(-an) \quad (3)$$

$$r_{c2} = k_{c2}s(1 - p) \exp\left(\frac{x_2 F \phi_0}{RT}\right) \quad (4)$$

$$r_{n2} = k_{n2}s(1 - p) \exp(-as) \quad (5)$$

$$r_o = k_o p \exp\left(\frac{y F \phi_0}{RT}\right) \quad (6)$$

Herein, the parameters k are the correspondent reaction rate constants, a is the variable relating the DEL electrochemical and electrophysical properties with nitrite and sulfite concentrations, x_1 , x_2 and y are the numbers of electrons transferred during the electrochemical stages, F is the Faraday number, φ_0 is the zero-charge related potential slope in DEL, R is the universal gas constant, and T is the absolute temperature.

In this system, both types of doping and polymer electrooxidation are responsible for the appearance of the oscillatory and monotonic instabilities. Nevertheless, the system will be efficient from both electroanalytical and electrosynthetical points of view, as shown below.

3. Results and Discussion

We describe the behavior of this system by analyzing the equation-set (1), alongside the algebraic relations (2 – 6) by means of linear stability theory and expose the steady-state Jacobian matrix members as (7):

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

In which:

$$a_{11} = \frac{2}{\delta} \left(-\frac{D}{\delta} - k_{c1}(1-p) \exp\left(\frac{x_1 F \varphi_0}{RT}\right) - k_{n1}(1-p) \exp(-an) + ak_{n1}n(1-p) \exp(-an) \right) \quad (8)$$

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

$$a_{13} = \frac{2}{\delta} \left(k_{c1}n(1-p) \exp\left(\frac{x_1 F \varphi_0}{RT}\right) - jk_{c1}n(1-p) \exp\left(\frac{x_1 F \varphi_0}{RT}\right) + k_{n1}n(1-p) \exp(-an) \right) \quad (10)$$

$$a_{21} = 0 \quad (11)$$

$$a_{22} = \frac{2}{\delta} \left(-\frac{D}{\delta} - k_{c2}s(1-p) \exp\left(\frac{x_2 F \varphi_0}{RT}\right) - k_{n2}(1-p) \exp(-as) + ak_{n2}s(1-p) \exp(-as) \right) \quad (12)$$

$$a_{23} = \frac{2}{\delta} \left(k_{c2}s \exp\left(\frac{x_2 F \varphi_0}{RT}\right) - jk_{c2}s(1-p) \exp\left(\frac{x_2 F \varphi_0}{RT}\right) + k_{n2}s \exp(-as) \right) \quad (13)$$

$$a_{31} = \frac{2}{\delta} \left(k_{c1}(1-p) \exp\left(\frac{x_1 F \varphi_0}{RT}\right) + k_{n1}(1-p) \exp(-an) - ak_{n1}n(1-p) \exp(-an) \right) \quad (14)$$

$$a_{32} = \frac{2}{\delta} \left(k_{c2}s(1-p) \exp\left(\frac{x_2 F \varphi_0}{RT}\right) + k_{n2}(1-p) \exp(-as) - ak_{n2}s(1-p) \exp(-as) \right) \quad (15)$$

$$a_{33} = \frac{2}{\delta} \left(-k_{c1}n(1-p) \exp\left(\frac{x_1 F \varphi_0}{RT}\right) + jk_{c1}n(1-p) \exp\left(\frac{x_1 F \varphi_0}{RT}\right) - k_{n1}n(1-p) \exp(-an) - k_{c2}s \exp\left(\frac{x_2 F \varphi_0}{RT}\right) + jk_{c2}s(1-p) \exp\left(\frac{x_2 F \varphi_0}{RT}\right) - k_{n2}s \exp(-as) - k_o \exp\left(\frac{y F \varphi_0}{RT}\right) + jk_o p \exp\left(\frac{y F \varphi_0}{RT}\right) \right) \quad (16)$$

Taking into account the main diagonal elements (8), (12), and (16), we may observe the positive elements, describing the positive callback. The presence of these elements depicts the possibility of the Hopf bifurcation, which is correspondent to the *oscillatory behavior*. As in similar systems [26 – 28], the oscillatory behavior will be caused by DEL influences of both chemical and electrochemical reactions, and the oscillation pattern will be dependent on the background electrolyte ionic composition, including pH.

Besides the positivity of the elements $jk_{c1}n(1-p) \exp\left(\frac{x_1 F \varphi_0}{RT}\right) > 0$, $jk_{c2}s(1-p) \exp\left(\frac{x_2 F \varphi_0}{RT}\right) > 0$ and $jk_o p \exp\left(\frac{y F \varphi_0}{RT}\right) > 0$ if $j > 0$, describing the oscillatory behavior caused by the DEL conductivity influences of the electrochemical doping and oxidation, the positive elements are $ak_{n1}n(1-p) \exp(-an) > 0$ and $ak_{n2}s(1-p) \exp(-as) > 0$ if $a > 0$, describing the same influences of chemical doping. In this system, the positive elements, related to the chemical stages and electrochemical stages, belong to different matrix elements.

As for the *steady-state stability*, we investigate it by applying the Routh-Hurwitz criterion to the equation set (1). In order to facilitate the analysis, we rewrite the Jacobian determinant as (17):

$$\frac{4}{\delta^2 C} \begin{vmatrix} -\kappa - \Xi & 0 & T \\ 0 & -\lambda - \Sigma & P \\ \Xi & \Sigma & -T - P - \Omega \end{vmatrix} \quad (17)$$

avoiding thereby the cumbersome expressions.

Opening the brackets, applying the $\text{Det } J < 0$ requisite, salient from the criterion, and changing the signs to the opposite, we obtain the steady-state stability requisite, exposed as (18):

$$\kappa(\lambda T + \lambda P + \lambda \Omega + \Sigma T + \Sigma \Omega) + \Xi(\lambda P + \lambda \Omega + \Sigma \Omega) > 0 \quad (18)$$

And this requisite will be readily satisfied if the kinetic parameters T, P, Ω , Ξ , and Σ are positive (their negativity may lead to the instabilities, including the oscillatory instability described above and the monotonic instability, described below). Really, considering that the diffusion parameters κ and λ are always positive, the left-side expression of the inequation (18) will be driven to more positive values, stabilizing the system, which will be thereby kinetically controlled, with the possibility of transition to the diffusion-controlled mode.

From the electroanalytical point of view, it corresponds to the linear dependence between the electrochemical parameter and concentration, which provides efficient analytical signal interpretation. From the removal point of view, it corresponds to efficient sulfite and nitrite removal from wine in an easily maintained steady-state mode. The topological region correspondent to this behavior, is relatively wide, confirming the efficiency of poly(9-triphenylphosphazo)acridine-modified electrode for nitrite and sulfite determination and removal.

As for the *detection limit*, it is described by the monotonic instability, which occurs in the point at which the stable steady states are separated from the unstable states. Its condition for this system is $\text{Det } J = 0$ or (19):

$$\kappa(\lambda T + \lambda P + \lambda \Omega + \Sigma T + \Sigma \Omega) + \Xi(\lambda P + \lambda \Omega + \Sigma \Omega) > 0 \quad (19)$$

The oscillatory instability will require more negative values of the parameters T, P, Ω , Ξ and Σ , being therefore realized beyond the detection limit (19).

The same type of behavior will be observed if the squaraine dyes containing pyridinic nitrogen atoms are used. If not, the elements r_{n1} and r_{n2} will be annihilated, and the model (1) will be simplified, according to [27 – 28].

4. Conclusions

From the analysis of the system with poly(9-triphenylphosphazo)acridine-assisted nitrite and sulfite electrochemical determination and removal from wine, it was possible to conclude that this process is a highly efficient, mostly kinetically controlled process, in which the linear dependence between the electrochemical parameter and concentration is easily obtained and maintained. Although the oscillatory behavior is highly probable in this system, it is realized beyond the detection limit, and its pattern depends on the background electrolyte

composition. Either way, poly(9-triphenylphosphazo)acridine is an efficient electrode modifier for nitrite and sulfite determination and elimination from wine.

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Conflicts of Interest

The authors declare no conflict of interest.

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