













# Electrochemical Determination of Sudan Dyes and Two Manner to Realize it: a Theoretical Investigation

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**Abstract:** In this work, a general theoretical description of the electrochemical determination of Sudan dyes by cathodic and anodic route has been made. Two mathematical models have been developed for each case, being, afterward, analyzed by means of linear stability theory and bifurcation analysis. It has been shown that in both cases, the systems are efficient from the electroanalytical point of view. The possibility for the oscillatory and monotonic instability has also been verified.

**Keywords:** food safety; concentration measurements; Sudan dyes; electrochemical sensors; cathodic reduction; anodic oxidation; stable steady-state.

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## 1. Introduction

The use of the chemically modified electrodes has turned one of the most modern tools for electroanalysis of pharmaceutical compounds, pesticides, and other biologically active compounds [1–6]. Their main advantages are precision, rapidity, and affinity to the analyte.

On the other hand, the group of Sudan dyes [7–9] is the group of azo dyes, widely used in histological investigations. Moreover, being fat-soluble, they are used to color vax, oils, petrol derivatives, solvents, and polishes in bright colors—red, orange, and yellow [10–13].

In some countries, these dyes are used (mostly illegally) in food production [14–15]. For example, they are used in order to deepen the color of curry and chilly pepper [16–17]. Also, they are used to falsifythespecies[18-19].

The most dangerous is that these dyes may present a high toxicity level [20–23], provoking DNA mutations [24]. Moreover, they are classified as carcinogenic compounds [25], the reason why their use in food is banned in 32 countries [26]. Therefore, the development of a rapid and efficient method for their quantification is really actual [27–30].

Many electrochemical methods have been developed to quantify Sudan dyes [31 – 40], by either anodic or cathodic manner. Nevertheless, all of those works analyze each Sudan dye separately without analyzing the Sudan dyes as a family.

Also, the development of new electroanalytical methods and their application also depend on the resolution of some problems like:

- the indecision about the mechanism of electrooxidation or electrochemical reduction of Sudan dyes during the electroanalytical process;

- the possibility of the electrochemical instabilities, characteristic for either cathodic or anodic transformation of organic compounds (including the electropolymerization) [41–45];

The resolution of these problems is not possible without the development of a mathematical model capable of describing adequately the system's behavior. Moreover, the modeling lets us compare their behavior with that of the other systems.

Therefore, the general objective of this work is a mechanistic theoretical investigation of the Sudan dyes' behavior during their electrooxidation and electroreduction. Its realization is achieved by the realization of specific goals like:

- the suggestion of the mechanism of the electrooxidation and electroreduction of the Sudan dye family;

- development of the mathematical model, correspondent to the mechanism;

- the analysis of the model and its interpretation in terms of steady-state stability and in electroanalytical terms;

- the comparison of the behavior of this system with the behavior of similar ones [46-49].

## 2. Materials and Methods

### 2.1. System and its modeling.

The exact electrooxidation mechanism for Sudan dyes haven't been established yet. Nevertheless, considering the involvement of the equal number of protons and electrons, like also the conjugated nature of the compound, it's possible to suppose the most probable electrooxidation mechanism (with an example of Sudan I) as on the Fig. 1.

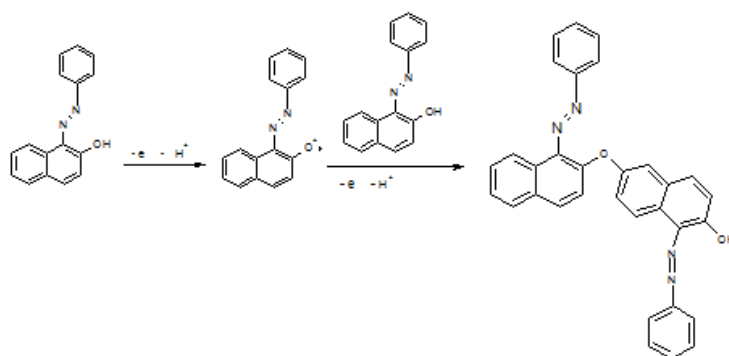
The possibility of electropolymerization isn't discarded, being electrooxidation in oligomers inter- and intramolecular. The electropolymerization of Sudan dyes may yield a semiconducting or conducting polydopamine-like polymer. Nevertheless, no Sudan I electropolymerization processes have been described yet.

Thus, in order to describe the electrooxidation of Sudan dyes, we introduce three variables:

$c$  – the Sudan dye concentration in the pre-surface layer;

$\theta$  – the anode Sudan dye surface coverage degree;

$\theta^*$  – the Sudan dye oligomer surface coverage degree.



**Figure 1.** The Sudan dyes electrooxidation.

To simplify the modeling, we suppose that the reactor is intensively stirred, so we can neglect the convection flow. Also, we assume that the background electrolyte is in excess, so we can neglect the migration flow. The diffusion layer is supposed to be of a constant thickness, equal to  $\delta$ , and the concentration profile in it is supposed to be linear.

It is possible to show that the electroanalytical process will be described by the three-dimensional equation set, exposed as:

$$\begin{cases} \frac{dc}{dt} = \frac{2}{\delta} \left( \frac{\Delta}{\delta} (c_0 - c) + r_{-1} - r_1 \right) \\ \frac{d\theta}{dt} = \frac{1}{G} (r_1 - r_{-1} - r_2) \\ \frac{d\theta^*}{dt} = \frac{1}{G^*} (r_2 - r_3) \end{cases} \quad (1)$$

Herein,  $\Delta$  is the diffusion coefficient,  $c_0$  is the bulk dye concentration,  $G$  is the colorant maximal surface concentration,  $G^*$  is the maximal surface concentration of the oligomeric oxidation product, and the parameters  $r$  stand for adsorption, desorption, and correspondent reaction rates, calculated as:

$$r_1 = k_1 c (1 - \theta - \theta^*) \exp(\alpha\theta) \quad (2)$$

$$r_{-1} = k_{-1} \theta \exp(-\alpha\theta) \quad (3)$$

$$r_2 = k_2 \theta^m \exp\left(\frac{nF\varphi}{RT}\right) \quad (4)$$

$$r_3 = k_2 \theta^* \exp\left(\frac{\mu F\varphi}{RT}\right) \quad (5)$$

In which  $k$  are respective reaction rate constants,  $n$  and  $\mu$  are the numbers of electrons transferred during the electrochemical processes,  $F$  is the Faraday number,  $R$  is the universal gas constant, and  $T$  is the absolute temperature.

In general terms, this is a system with two consecutive electrochemical reactions, the behavior of which will be described below.

### 3. Results and Discussion

In order to investigate the behavior of the system with the Sudan dyes electrooxidation, we analyze the differential reaction set (1), by means of linear stability theory. The steady-state Jacobian matrix members may be exposed as:

$$\begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{31} & a_{33} \end{pmatrix} \quad (6)$$

where:

$$a_{11} = \frac{2}{\delta} \left( -\frac{\Delta}{\delta} - k_1 (1 - \theta - \theta^*) \exp(\alpha\theta) \right) \quad (7)$$

$$a_{12} = \frac{2}{\delta} (k_1 c \exp(\alpha\theta) - \alpha k_1 c (1 - \theta - \theta^*) \exp(\alpha\theta) + k_{-1} \exp(-\alpha\theta) - \alpha k_{-1} \theta \exp(-\alpha\theta)) \quad (8)$$

$$a_{13} = \frac{2}{\delta} (k_1 c \exp(\alpha\theta)) \quad (9)$$

$$a_{21} = \frac{1}{G} (k_1 (1 - \theta - \theta^*) \exp(\alpha\theta)) \quad (10)$$

$$a_{22} = \frac{1}{G} \left( -k_1 c \exp(\alpha\theta) + \alpha k_1 c (1 - \theta - \theta^*) \exp(\alpha\theta) - k_{-1} \exp(-\alpha\theta) + \alpha k_{-1} \theta \exp(-\alpha\theta) - m k_2 \theta^{m-1} \exp\left(\frac{nF\varphi}{RT}\right) + j k_2 \theta^n \exp\left(\frac{nF\varphi}{RT}\right) \right) \quad (11)$$

$$a_{23} = \frac{1}{G} \left( k_1 c \exp(\alpha\theta) + p k_2 \theta^n \exp\left(\frac{nF\varphi}{RT}\right) \right) \quad (12)$$

$$a_{31} = 0 \quad (13)$$

$$a_{32} = \frac{1}{G^*} \left( m k_2 \theta^{m-1} \exp\left(\frac{nF\varphi}{RT}\right) - j k_2 \theta^n \exp\left(\frac{nF\varphi}{RT}\right) - j k_3 \theta^* \exp\left(\frac{\mu F\varphi}{RT}\right) \right) \quad (14)$$

$$a_{33} = \frac{1}{G} \left( k_2 \exp\left(\frac{\mu F\varphi}{RT}\right) + p k_3 \theta^* \exp\left(\frac{\mu F\varphi}{RT}\right) - k_2 \exp\left(\frac{\mu F\varphi}{RT}\right) + p k_3 \theta^* \exp\left(\frac{\mu F\varphi}{RT}\right) \right) \quad (15)$$

Observing the expressions (7), (11) and (15), it is possible to see that the oscillatory behavior, in this case, is possible, as the matrix main diagonal contains the positive elements, describing the positive callback.

These are the elements  $\alpha k_1 c (1 - \theta - \theta^*) \exp(\alpha\theta)$  and  $\alpha k_{-1} \theta \exp(-\alpha\theta)$ , describing the attraction between the colorant adsorbed particles, like also the elements  $j k_2 \theta^n \exp\left(\frac{nF\varphi}{RT}\right)$ ,  $-p k_2 \theta^n \exp\left(\frac{nF\varphi}{RT}\right)$  and  $p k_2 \theta^* \exp\left(\frac{\mu F\varphi}{RT}\right)$ , describing the DEL influences on the electrochemical stages. Both of the influences are characteristic for similar systems [46–49].

In order to investigate the steady-state stability, we apply the Routh-Hurwitz criterion to the differential equation set (1). Avoiding the cumbersome expressions, we introduce the new variables, rewriting the matrix determinant as:

$$\frac{2}{\delta G G^*} \begin{vmatrix} -\kappa_1 - \varepsilon & \Lambda & P \\ \varepsilon & -\Lambda - X & -P + \Sigma \\ 0 & -X - \Omega & -\Sigma - \Psi \end{vmatrix} \quad (16)$$

Opening the parenthesis and applying the Det J < 0 requisites, salient from the criterion, we obtain the stability condition as:

$$(-\kappa_1 - \varepsilon)(\Lambda \Sigma + \Lambda \Psi + X \Psi - X P - \Omega P - \Omega \Sigma) + \varepsilon(-X P - \Omega P + \Lambda \Sigma + \Lambda \Psi) < 0 \quad (17)$$

Rearranging the equation (17), expelling the elements with equal modules and opposed signs, we obtain the stability requisite, expressed as:

$$(-\kappa_1)(\Lambda \Sigma + \Lambda \Psi + X \Psi - X P - \Omega P - \Omega \Sigma) - \varepsilon(X \Psi - \Omega \Sigma) < 0 \quad (18)$$

Easily satisfied, if X, Ω, Σ and Ψ have values correspondent to the DEL influences fragility, and the parameter Λ, the values, correspondent to the adsorbed particle repulsion. Therefore, the equation on the left side of the inequity (18) will have more negative values, and, consequently, will be satisfied. It is also characteristic for similar systems [46 – 49].

From the electroanalytical point of view, the steady-state stability is correspondent to the linear dependence between the concentration and the electrochemical parameter. The electroanalytical process will be diffusion-controlled.

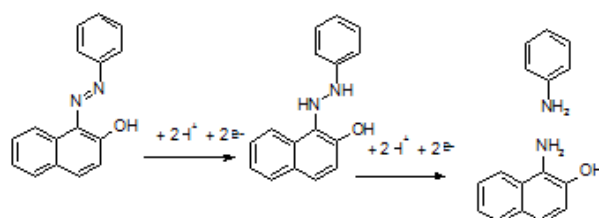
The monotonic instability, correspondent to the detection limit from the electroanalytical point of view, is also possible for this system, and its condition will be described as:

$$(-\kappa_1)(\Lambda \Sigma + \Lambda \Psi + X \Psi - X P - \Omega P - \Omega \Sigma) - \varepsilon(X \Psi - \Omega \Sigma) = 0 \quad (19)$$

In the case of the presence of autocatalytic processes (for example, the autocatalytic chain growth), the expressions, describing the respective process rates, will be modified. Moreover, the autocatalytic influences will be one more factor, responsible for the oscillatory behavior.

### 3.1. Cathodic Sudan Dye determination.

In the case of the cathodic detection, the electroanalytical process will be realized in acidic media and, depending on the solution acidity, will lead to the appearance of a hydrazocompound and, furtherly, of correspondence aromatic amines, as on the Fig. 2.



**Figure 2.** The cathodic Sudan dyes determination.

In this case, the system's behavior will be similar to that observed for benzodiazepines, described in [49]. Therefore, it will be described briefly in this system.

As in [49], the system will include the so-called “hydrogen evolution” and will be described by a tridimensional differential equation system, as:

$$\begin{cases} \frac{d\varepsilon}{dt} = \frac{2}{\delta} \left( \frac{\Delta}{\delta} (\varepsilon_0 - \varepsilon) + \rho_{-1} - \rho_1 \right) \\ \frac{d\eta}{dt} = \frac{1}{\psi} (\rho_1 - \rho_{-1} - \rho_2) \\ \frac{dh}{dt} = \frac{2}{\delta} \left( \frac{d}{\delta} (h_0 - h) - \rho_2 - \rho_3 \right) \end{cases} \quad (20)$$

Analyzing the differential equation set (20), it will be a possible conclusion that:

- The steady-state stability in this system is maintained in the case of the repulsion of the adsorbed particles and of the fragility of the electrochemical influences in DEL. The reaction process will be controlled by the diffusion of both dye and protons.

- The topological zone of the electroanalytical efficiency of the steady-state stability will be less vast than of the general stability conditions, due to the “hydrogen evolution”. Nevertheless, in order to augment the electroanalytical efficiency, moderately acidic solutions have to be used. Another alternative is the use of the electrode materials, impeding the hydrogen evolution.

- The oscillatory behavior in this system is caused by surface and electrochemical factors, but it is less probable than in the case of anodic oxidation. The augment o pH will augment the wavelength but will decrease the oscillation frequency.

Therefore, among the anodic and cathodic reactions, it is preferable to use the cathodic reduction, as it is more sensitive and exact for the determination of Sudan dyes.

## 4. Conclusions

The behavior analysis of the system with Sudan dyes electrochemical oxidation and reduction, realized for electroanalytical purposes, let us conclude that in both of the cases, we deal with efficient diffusion-controlled electroanalytical systems. The stability of the steady-state, corresponding to the linear dependence between the current and dye concentration, is safeguarded by the adsorbed particle repulsion, and DEL influences fragility. The opposite factors are responsible for the oscillatory and monotonic instability. Also, the cathodic process,

if realized on the materials impeding the hydrogen evolution, is realized by a more efficient manner than the anodic electrochemical determination.

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## Conflicts of Interest

The authors declare no conflict of interest.

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