Letters in Applied NanoBioScience
Open-Access Journal (ISSN: 2284-6808)

Article
Volume 14, Issue 1, 2025, 8
https://doi.org/10.33263/LIANBS141.008

The Theoretical Description for Bisphenol S and Bisphenol AF Cathodic Determination by Bivalent Chromium, Intercalated into Conducting Polymeric Material

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Received: 18.09.2023; Accepted: 12.05.2024; Published: 21.09.2024

Abstract: A theoretical description for bisphenol S and bisphenol AF electrochemical determination has been given. The electrochemical determination becomes possible via a cathodic process, in which both trifluoromethyl and sulfonic groups are reduced, as bivalent chromium is oxidized to trivalent. This process may be used for electroanalytical and environmental remediation as one of the stages, as it is an efficient diffusion- and kinetically-controlled system. As for the oscillatory behavior, it becomes more probable than for sucralose and chloropicrin removal in analogous conditions and more probable in relation to vanadium (III) oxyhydroxide use as a modifier.

Keywords: polycarbonate degradation; bisphenols; electrochemical determination; electrochemical remediation; conducting polymers; bivalent chromium; stable steady-state.

1. Introduction

Bisphenols (Figure 1) consist of a group of compounds used mainly in polyester plastics, including polycarbonates, fast-drying epoxy resin adhesives, and anticorrosion coatings [1–5]. They are also used in odontology for implant fixation.

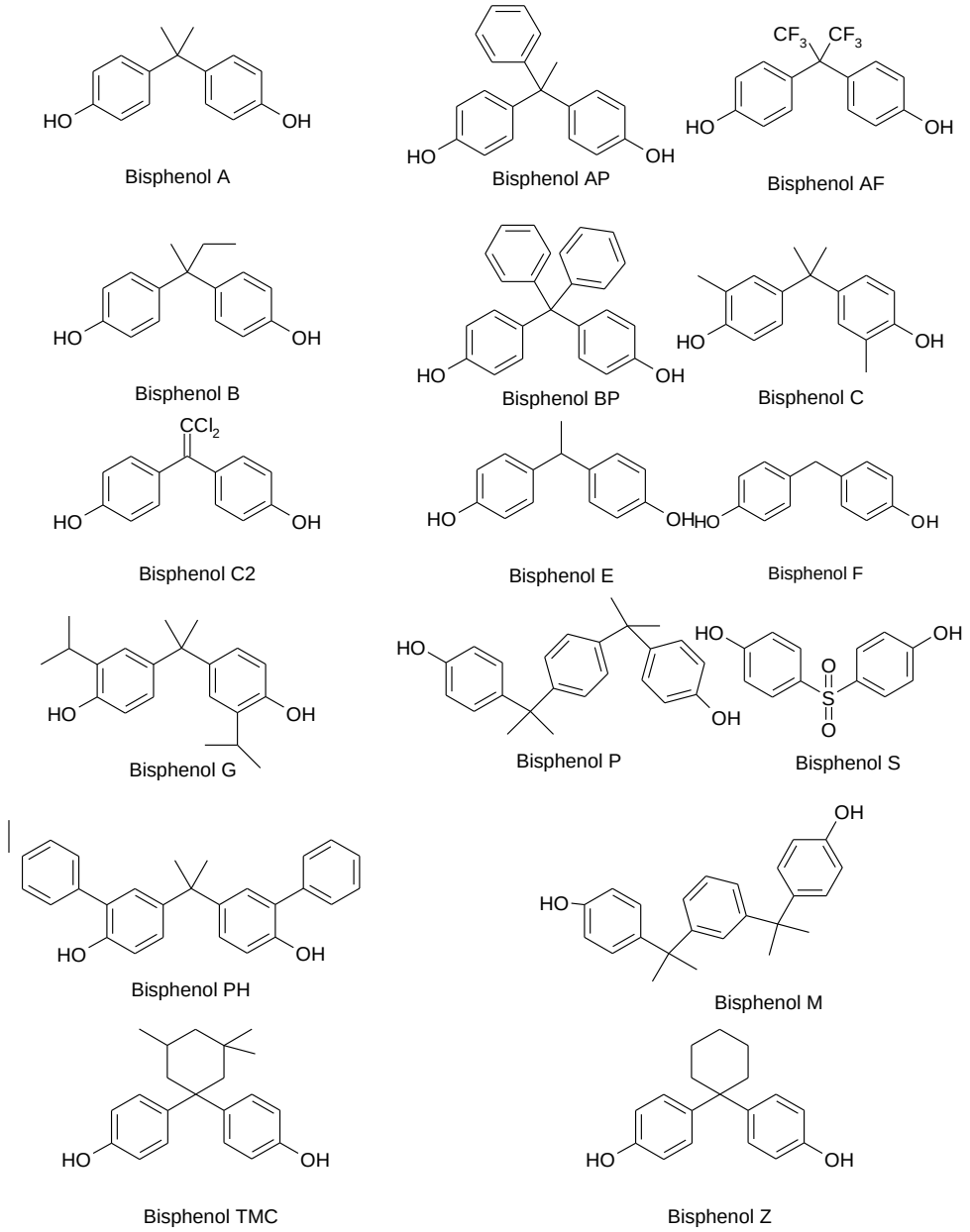

Figure 1. The most used bisphenols.

Bisphenol S is the only bisphenol possessing a π-conjugated system involving the entire molecule and the most acidic of all.

These compounds are used mostly in polycondensation polymers, which degrade while heated [6–10]. This degradation divides the polymer molecules into monomers, which may penetrate the food or water stored in the polyester ducts or vessels. Their main toxic action of bisphenols, principally A, F, and S, are endocrine disruptions, mainly xenoestrogenic, affecting behavior, fertility, and homeostasis. A recent EFSA investigation has shown high levels of bisphenol A intoxication in tested people in some EU members, including Portugal [10].

For this reason, the development of a method capable of detecting bisphenols in food and water and of removing them from it is actual [11–13], and the electrochemical methods, widely used for phenolic compound determination [14–23], could be an interesting solution for this task.

Generally, phenolic compounds, including bisphenols, are detected by anodic route, the main manifestation of phenols' electrochemical behavior. Nevertheless, the bisphenols C2, AF, and S may be detected cathodically in an acidic medium. The same process may also be applied to bisphenols substituted derivatives, like dinitro- or tetrabromobisphenol A.

In order to increase the working pH, the electrodes may be modified by materials capable of catalyzing the sulfonic group and reducing organic halide. One of these materials may be a bivalent chromium cation, generated *in situ* on conducting polymer matrix [24–26] by chrome nanoparticles reaction with acids in an inert atmosphere or mainly by trivalent chromium ion cathodic reduction:

$$Cr^{3+} + e^- \rightarrow Cr^{2+} \quad (1)$$

Even in mildly acidic mediums, bivalent chromium is a strong reducent that may even reduce water. Its reaction with sulfonic group and organic chlorine may be exposed as (2 – 3):

$$4Cr^{2+} + 4H^+ + R - SO_2 - R \rightarrow 4Cr^{3+} + R - S - R + 2H_2O \quad (2)$$

$$R - CF_3 + 6Cr^{2+} + 6H^+ \rightarrow 6Cr^{3+} + R - CH_3 + 3HF \quad (3)$$

The bivalent chrome will be regenerated in the electrochemical stage (1), providing an efficient electroanalytical process resembling the one described for sucralose [27,28]. Nevertheless, the changes in double electric layer (DEL) electrophysical properties may affect the electroanalytical process and analytical signal interpretation.

For this reason, the theoretical investigation of the electroanalytical process, capable of investigating the system from the mechanistic point of view, analyzing the steady-state stability and the most common instabilities, in order to foresee the possible difficulties in analytical signal interpretation is necessary, and it will be made in this work.

This analysis will include the development and analysis of the mathematical model, correspondence to the most probable reaction mechanism, detection of the steady-state stability and most frequent instabilities conditions, and comparison of the behavior of this system with that of similar ones [27,28].

2. Materials and Methods

The reduction of bisphenol S will yield a thiophenol eter moiety, and the bisphenol AF will be reduced to the proper bisphenol A. The schematic representation of the electroanalytical system is given in Figure 2.

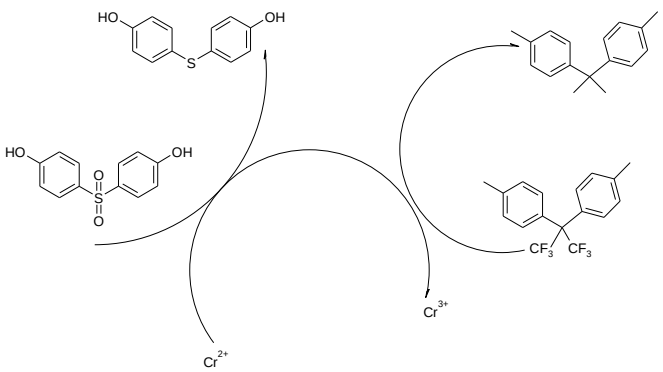

Figure 2. The schematic representation of bisphenols S and AF electrochemical determination on Cr^{2+}-modified polymer backbone.

Although this system's behavior is similar to that observed for sucralose and chloropicrin [27,28], it will be slightly different due to the ionic form transformation in the polymer backbone, changing the cathode material conductivity and ionic force. This will affect the behavior of the electroanalytical system, as shown below.

Taking this into account and accepting some assumptions [27,28], we describe the behavior of this system by the trivariate equation-set (4):

$$\begin{cases} \frac{ds}{dt} = \frac{2}{\delta}\left(\frac{D}{\delta}(s_0 - s) - r_s\right) \\ \frac{df}{dt} = \frac{2}{\delta}\left(\frac{\Phi}{\delta}(f_0 - f) - r_f\right) \\ \frac{dc}{dt} = \frac{1}{C}(r_r - r_s - r_p) \end{cases} \quad (4)$$

Herein, δ is the diffusion layer thickness, s, and f are the pre-surface concentrations of S- and AF-bisphenol, D, and Φ stand for the correspondent diffusion coefficients, s_0 and f_0 stand for their bulk concentrations, c is the bivalent chromium polymer matrix coverage degree, C stands for its matrix concentration, and the parameters r correspond to the correspondent reaction rates, calculated as:

$$r_s = k_s s c^4 \exp(-\alpha c) \quad (5)$$

$$r_f = k_f c^{12} \exp(-\alpha c) \exp(-\beta f) \quad (6)$$

$$r_r = k_r (1 - c) \exp\left(-\frac{F\varphi_0}{RT}\right) \quad (7)$$

in which the parameters k are the correspondent reaction rate constants, α and β are parameters describing the impact of the chemical stages on the polymer matrix and DEL electrophysical properties correspondently, F is the Faraday number, φ_0 is the zero-charge-related potential slope, R is the absolute gas constant, and T is the absolute temperature.

Contrarily to sucralose and chloropicrin removal and determination by vanadium oxyhydroxide [27,28], only one of two analytes influences the DEL during the chemical stage. Nevertheless, in this system, the DEL and surface electrophysical properties are affected, and this influence may cause oscillatory behavior. Despite this, this system is an efficient electrochemical process, as shown below.

3. Results and Discussion

We investigate the electroanalytical process of bisphenols AF and S electrochemical determination on bivalent chrome intercalated in conducting polymer backbone by analyzing the equation-set (4) and the algebraic relations (5 – 7) using linear stability theory. The steady-state Jacobian matrix members may be calculated as (8):

$$\begin{pmatrix} a_{11} & a_{12} & a_{13} \\ a_{21} & a_{22} & a_{23} \\ a_{31} & a_{32} & a_{33} \end{pmatrix} \quad (8)$$

In which:

$$a_{11} = \frac{2}{\delta}\left(-\frac{D}{\delta} - k_s c^4 \exp(-\alpha c)\right) \quad (9)$$

$$a_{12} = 0 \quad (10)$$

$$a_{13} = \frac{2}{\delta}(-4k_s s c^3 \exp(-\alpha c)_{\square} + \alpha k_s s c^4 \exp(-\alpha c)) \quad (11)$$

$$a_{21} = 0 \quad (12)$$

$$a_{22} = \frac{2}{\delta}\left(-12k_f c^{11} \exp(-\alpha c) \exp(-\beta f) + \beta k_f c^{12} \exp(-\alpha c) \exp(-\beta f)\right) \quad (13)$$

$$a_{23} = \frac{2}{\delta}\left(-12k_f c^{11} \exp(-\alpha c) \exp(-\beta f) + \alpha k_f c^{12} \exp(-\alpha c) \exp(-\beta f)\right) \quad (14)$$

$$a_{31} = \frac{1}{C}(-k_s c^4 \exp(-\alpha c)) \quad (15)$$

$$a_{32} = \frac{1}{C}\left(-12k_f c^{11} \exp(-\alpha c)\exp(-\beta f) + \beta k_f c^{12} \exp(-\alpha c)\exp(-\beta f)\right) \quad (16)$$

$$a_{33} = \frac{1}{C}\left(-k_r \exp\left(-\frac{F\varphi_0}{RT}\right) + \xi k_r(1-c)\exp\left(-\frac{F\varphi_0}{RT}\right) - 4k_s c^3 \exp(-\alpha c) + \alpha k_s c^4 \exp(-\alpha c) - 12k_f c^{11} \exp(-\alpha c)\exp(-\beta f) + \alpha k_f c^{12} \exp(-\alpha c)\exp(-\beta f)\right) \quad (17)$$

Avoiding the cumbersome expression during the determinant analysis, we introduce new variables and rewrite the determinant as (18):

$$\text{Det J} = \frac{4}{\delta^2 C}\begin{vmatrix} -\kappa - \Xi & 0 & -\Lambda \\ 0 & -\varphi - P & -\Phi \\ -\Xi & -P & -\Omega - \Lambda - \Phi \end{vmatrix} \quad (18)$$

Considering that:

$$-Det\,J \begin{cases} > 0, for\ steady - state\ stability \\ = 0\ monotonic\ instability \end{cases} \quad (19)$$

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 (20):

$$\Omega(\kappa\varphi + \kappa P + \Xi\varphi + \Xi P) + \Lambda(\kappa\varphi + \kappa P) + \Phi(\kappa\varphi + \Xi\varphi) \begin{cases} > 0, curve\ linearity \\ = 0, detection\ limit \end{cases} \quad (20)$$

If –Det J>0, the Routh-Hurwitz stability criterion is valid, and the steady-state is thereby stable, providing an efficient bisphenols electrochemical determination. Moreover, the wide stability region allows us to use this system as an electroanalytical for sensing purposes.

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

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 important for bisphenol 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.

Really, if we analyze the main diagonal elements (9), (13), and (17), we may observe four elements capable of being possible:

$\alpha k_f c^{12} \exp(-\alpha c)\exp(-\beta f) > 0$ and $\alpha k_s c^4 \exp(-\alpha c) > 0$, if $\alpha > 0$, describing the impact of the changes in ionic configuration (and conducting polymer matrix doping degree and, thereby, conductivity) on conducting polymer configuration, resulting in cyclic changes in cell current, manifested in the oscillatory behavior;

$\beta k_f c^{12} \exp(-\alpha c)\exp(-\beta f) > 0$, if $\beta > 0$, describing the impact of the ionic forms cyclic transformations in DEL, manifested in cyclic changes in its ionic force and conductivity and, thereby, in current oscillatory behavior.

$\xi k_r(1-c)\exp\left(-\frac{F\varphi_0}{RT}\right) > 0$, if $\xi > 0$, describing the cyclic changes in DEL and the surface during the electrochemical stages, resulting in the oscillatory behavior. These factors tend to be manifested far beyond the detection limit, such as in similar systems [27,28].

The same process may also be used as one of the stages of bisphenol transformation into more biodegradable compounds. It may also be applied for bisphenol C2 and substituted bisphenol derivatives with halide and acceptor substituents. Suppose the bisphenol C2 is used in bisphenol AF or tetrabrombisphenol A. In that case, the system's behavior becomes similar to [27,28]. Yet, if bisphenol C2 is used alongside bisphenol S or dinitro bisphenol A, the behavior of this system will be described by the model, which is exposed in the present paper.

For safety reasons, membrane electrolysis [27], capable of removing the halide ion without permitting halogen evolution on the anode, may be used for electroanalytical or environmental remediation.

4. Conclusions

From the theoretical description of bisphenols AF and S electrochemical cathodic determination on bivalent chromium, cation intercalated into the polymer matrix, and it has been possible to conclude it may be an excellent modifier for both removal and quantification of phenolic pollutants. The electrochemical removal becomes diffusion-controlled or kinetically controlled with the prevalence of the kinetic factor. The oscillatory behavior in this system may be caused by both DEL and matrix influence by electrochemical and chemical stages. The system may be efficiently used for monitoring and remediation purposes, as well as adapted to other bisphenols containing halide and accepting fragments.

Funding

This research received no external funding.

Acknowledgments

Volodymyr V. Tkach acknowledges the support of the Engineering Faculty of the University of Porto and the University of Trás-os-Montes and Alto Douro during these difficult times for Ukraine and its research.

Conflicts of Interest

The authors declare no conflict of interest.

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