



Two-step mineralization of Tartrazine solutions: Study of parameters and by-products during the coupling of electrocoagulation with electrochemical advanced oxidation processes



Abdoulaye Thiam^{a,b}, Minghua Zhou^{a,*}, Enric Brillas^b, Ignasi Sirés^{a,b,*}

^a College of Environmental Science and Engineering, Nankai University, Tianjin 300071, China

^b Laboratori d'Electroquímica dels Materials i del Medi Ambient, Departament de Química Física, Facultat de Química, Universitat de Barcelona, Martí i Franquès 1-11, 08028 Barcelona, Spain

ARTICLE INFO

Article history:

Received 7 October 2013

Received in revised form 3 December 2013

Accepted 6 December 2013

Available online 14 December 2013

Keywords:

Azo dyes

Electrocoagulation

Homogeneous catalysis

Photoelectro-Fenton

Tartrazine

ABSTRACT

A novel sequential electrochemical treatment consisting in an electrocoagulation (EC) pre-treatment and the subsequent advanced oxidation by photoelectro-Fenton (PEF) process with in-situ H_2O_2 electrogeneration and UVA light irradiation has been envisaged for the removal of organic pollutants from water, showing a high performance for the decolorization and mineralization of Tartrazine (Acid Yellow 23) solutions. EC has a dual purpose as a coagulation and catalyst source step since it allows the formation of $\text{Fe}(\text{OH})_n$ ($n = 2$ or 3) coagulant and $\text{Fe}^{3+}/\text{Fe}^{2+}$ ions. The influence of the anode material (Fe and Al), supporting electrolyte, pH and current during the individual EC on the abatement of color and total organic carbon (TOC) has been assessed. EC with Fe in 0.05 M NaCl yielded the best results. Next, various single electrochemical advanced oxidation processes (EAOPs) such as electro-oxidation (EO), electro-Fenton (EF) and PEF with a Pt or boron-doped diamond (BDD) anode and an air-diffusion cathode (ADE) have been tested. PEF with BDD/ADE yielded the quickest degradation among all the EAOPs in NaCl, due to the action of oxidants like active chlorine as well as $\cdot\text{OH}$ formed at the anode surface from H_2O oxidation and in the bulk from UV-enhanced Fenton's reaction between cathodic H_2O_2 and added Fe^{2+} . Therefore, the two-step EC (Fe/steel)/PEF (BDD/ADE) process was the best EC/EAOP combination. The EC treatment in 0.05 M NaCl at natural pH 6.3 and 200 mA, followed by PEF treatment of the supernatant at pH 3.0 and 200 mA, yielded the best conditions with a total decolorization in 15 min of EC and total mineralization in ca. 300 min of PEF. GC-MS analysis showed the formation of several benzenic by-products during the application of EC/PEF. Independent electrolyses revealed the ability of EC to accumulate soluble chlorobenzene derivatives, which can be completely destroyed in the PEF step under the action of the mixture of oxidants, particularly by successive hydroxylation via $\cdot\text{OH}$ largely promoted in the bulk by the Fe^{2+} ions generated in EC.

© 2013 Elsevier B.V. All rights reserved.

1. Introduction

Wastewaters generated by several industries pose some serious health and environmental risks worldwide owe to the significant concentrations and/or large variety of toxic aromatic pollutants. In this context, the advanced oxidation processes (AOPs) are very effective remediation technologies due to the action of highly reactive species (ROS) such as (mainly but not uniquely) hydroxyl radical ($\cdot\text{OH}$) in the reaction mechanism. The main catalyzed AOPs

can be divided into: (i) heterogeneous catalysis such as TiO_2 photocatalysis and electrocatalysis (EAOPs) with large O_2 -overvoltage anodes, and (ii) homogeneous catalysis such as catalyzed ozonation and chemical and electrochemical Fenton and photo-Fenton [1–3].

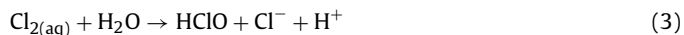
In recent years, the interest on new or enhanced electrochemical separation and degradation technologies has grown considerably. Most intensive research efforts are devoted to heterogeneous electrocatalysis, where electro-oxidation (EO) is predominant, electrocoagulation (EC) and EAOPs based on Fenton's reaction chemistry [4–6]. EO combines high simplicity and efficacy, favoring the oxidative decontamination of waters under the action of $\cdot\text{OH}$ formed at the anode surface by water oxidation via reaction (1). At present, boron-doped diamond (BDD) is the best electrocatalytic anode material to oxidize pollutants, which has led to the

* Corresponding authors. Tel.: +34 934021223/+86 2266229619;

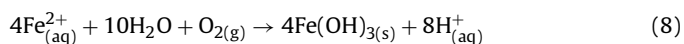
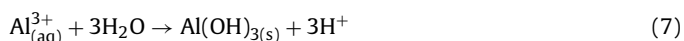
fax: +34 934021231/+86 2223501117.

E-mail addresses: zhoumh@nankai.edu.cn (M. Zhou), i.sires@ub.edu, siresa@gmail.com (I. Sirés).

development of successful flow reactors [7–10]. In chloride medium, the oxidation mediated by active chlorine species (Cl_2 , HClO and/or ClO^-) electrogenerated via reactions (2)–(4) competes with the $\cdot\text{OH}$ -mediated oxidation [6].



EC is a traditional physicochemical treatment for phase separation that is based on the release of $\text{Al}(\text{III})$ or $\text{Fe}(\text{II})$ cations in the bulk from dissolution of the corresponding sacrificial anode(s) via reactions (5) and (6). These are active coagulant precursors because the simultaneous production of OH^- anions by cathodic reduction of water leads to the formation of insoluble polymeric metallic hydroxides as shown in reactions (7) and (8). Such particles have large surface area and remain as a gelatinous suspension, then being able to remove pollutants by coagulation upon destabilization either by surface complexation or electrostatic attraction [11–13]. EC has been shown effective for the removal of colloidal and ionic pollutants from synthetic and real waters [14–22].



EAOPs such as electro-Fenton (EF) and photoelectro-Fenton (PEF) processes are even more efficient than EO and EC for the destruction of aqueous pollutants [23–36]. They are based on the in-situ electrogeneration of H_2O_2 into acidic polluted solutions from the two-electron O_2 reduction at a carbonaceous cathode. The addition of a catalytic amount (0.1–1.0 mM) of Fe^{2+} to the medium allows a high production of $\cdot\text{OH}$ in the solution bulk from the well-known Fenton' reaction [5]:



PEF has the greatest performance owing to the concomitant action of both the generated $\cdot\text{OH}$ and the UV radiation on the organic molecules and their $\text{Fe}(\text{III})$ -complexes [30,33,35]. Although EF and PEF are promising technologies, their potential exploitation is still limited due to the relatively high operation costs that are inherent to the long treatment time required. Aiming to shorten the reaction time, the so-called peroxi-coagulation (PC) process with a sacrificial iron anode and a cathode able to generate H_2O_2 has been further devised [37–39]. This one-pot method allows the simultaneous coagulation and oxidation of organic pollutants thanks to Fe^{2+} catalyst generated on site via reaction (6), but it presents some major drawbacks: (i) The excess of iron cations causes the destruction of $\cdot\text{OH}$, (ii) the cathode lifetime and its ability for producing H_2O_2 are drastically reduced due to both, adsorption of coagulated particles that block the diffusion pores and the active sites, and surface abrasion caused by precipitated hydroxides and coagulated matter, and (iii) the effect of UV light in photoperoxi-coagulation is almost negligible due to the barrier effect of the iron precipitate.

Synthetic dyes are being largely produced due to their unique properties that allow limitless industrial applications. Azo dyes are particularly pre-eminent since they represent 60–70% of the world production [18]. However, their occurrence in wastewaters entails

major concerns due to the proven toxicity and carcinogenicity and the production of toxic by-products like aromatic amines [31,40]. Among them, food azo-colors have received much less attention than their textile counterparts, despite being considered dangerous due to their possible connection to hyperactivity in children in cases of intakes of potent mixes of colorings. Recent issues over the toxicity of food additives have led to a stricter legislation. For example, the UK Foods Standards Agency has recommended that they should be phased out over a period from food and drink in the EU [41]. Tartrazine (Acid Yellow 23, trisodium 1-(4-sulfonatophenyl)-4-(4-sulfonatophenylazo)-5-pyrazolone-3-carboxylate) is primarily used as a common synthetic food coloring agent, also known as additive E102, to provide a vivid yellow color to processed commercial foods like sweets, beverages and snacks. It is also used in personal care and cosmetic products, as well as in medications. Sensitivity to Tartrazine may be relevant since it is suspected to be responsible for most of the allergic and intolerance reactions among all azo dyes, even being temporarily banned in some European countries. Its adverse reactions in human beings have lately been under consideration by the European Food Safety Agency [42]. A large range of dyestuffs has been removed from water electrochemically by EO [8,9,17,43,44], EC [14,17,19,21] and EAOPs like EF and PEF [23–26,31,33,45,46]. However, there are no studies assessing the performance of BDD or PEF for Tartrazine, and just a couple of studies report its treatment by EC [47,48].

As far as we know, this work reports for the first time the sequential combination of EC and EAOPs based on Fenton's reaction chemistry, aiming at: (i) inducing the coagulation of a large amount of organic matter via the EC pre-treatment, which also acts as a source of iron catalyst, and (ii) controlling the electrogeneration of oxidizing agents on site to reach the complete mineralization of dye solutions during the EAOP post-treatment. This intends to overcome the drawbacks exposed for PC, therefore achieving a higher degradation performance for dye solutions. Previous EC/EAOPs couplings reported in literature for decontamination have dealt with the EC/EO system [49–51]. The present study focuses on the treatment of solutions of 130 ml of Tartrazine as a model food azo dye in different electrolytes at constant current. First, the individual EC with Fe or Al anodes, and EO, EF and PEF with Pt or BDD anodes were tested to assess the effect of the experimental parameters on the decolorization and TOC removal rates. Then, the best combined EC/EAOP was optimized to achieve the quickest decontamination. The degradation by-products formed during this combined treatment were identified by gas chromatography coupled to mass spectrometry (GC–MS).

2. Experimental

2.1. Chemicals

Tartrazine ($\text{C}_{16}\text{H}_9\text{N}_4\text{Na}_3\text{O}_9\text{S}_2$, C.I. 19140, dye content $\geq 85\%$) was purchased from Sigma-Aldrich and was used as received. Anhydrous sodium sulfate, sodium chloride and sodium nitrate used as background electrolytes, as well as iron(II) sulfate heptahydrate, used as catalyst in EF and PEF, were of analytical grade from Merck and Fluka. Solutions were prepared with ultra-pure water from a Millipore Milli-Q system with resistivity $>18\text{ M}\Omega\text{ cm}$ and their pH was adjusted with analytical grade sodium hydroxide or sulfuric, hydrochloric or nitric acid from Merck. Other chemicals for performing analyses were obtained from Merck and Sigma-Aldrich.

2.2. Electrochemical cells

All the electrolytic trials were conducted in an open and undivided cylindrical glass cell of 150 ml capacity with a double jacket

Download English Version:

<https://daneshyari.com/en/article/45916>

Download Persian Version:

<https://daneshyari.com/article/45916>

[Daneshyari.com](https://daneshyari.com)