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Diazo dye Congo Red degradation using a Boron-doped diamond anode: An experimental study on the effect of supporting electrolytes

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HIGHLIGHTS

- Use of AOx/BDD to treatment CR diazo dye.
- Could be possible success in diazo CR degradation via AOx/BDD?
- Which can be the effect from the supporting electrolyte?

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ABSTRACT

Diazo dye Congo Red (CR) solutions at 100 mg/L, were degraded using different supporting electrolytes in an electrochemical advanced oxidation process (EAOPs), like the anodic oxidation (AOx/BDD). All experiments were carried out in a 3 L flow reactor with a Boron-doped diamond (BDD) anode and stainless steel cathode (AISI 304), at 7.5, 15, 30 and 50 mA/cm² current densities (j). Furthermore, each experiment was carried out under a flow rate of 7 L/min. Additionally, HClO₄, NaCl, Na₂SO₄, and H₂SO₄ were tested as supporting electrolytes at a 50 mM concentration. The degradation process was at all times considerably faster in NaCl medium. Solutions containing SO₄²⁻ or ClO₄⁻ ions were less prompted to degradation due to the low oxidation power of these species into the bulk. Dissolved organic carbon (DOC) analysis, was carried out to evaluate the mineralization of CR. The degradation of CR, was evaluated with the HPLC analysis of the treated solutions.

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1. Introduction

Recently, synthetic dyes produced and released by different economic activities such as textile, paper, pharmaceutical, cosmetics and food can affect the water bodies and diminish light penetration for photosynthetic organisms, resulting in lessened oxygen levels in aquatic ecosystems [1,2]. In this context, it is well known that some azo dyes characterized mainly by nitrogen to nitrogen double bond (–N=N–) are toxic, and in many cases their degradation products are carcinogenic and mutagenic to humans [3]. According to this, many physicochemical approaches have been reported for treating dye wastewater, nevertheless some of these processes have intrinsic disadvantages like: (i) high cost, (ii) sludge generation, (iii) concomitant secondary pollution and (iv) difficult treatment procedures [4].

Recently, comparing with physical and chemical methods, EAOPs are considered good techniques for treating dyed wastewater, since they can degrade dyes via free hydroxyl radicals (•OH) at room temperature and at specific pH conditions, with a low or minimum secondary impact on the environment and ecosystems and at a low cost [5–7].

Last decade, treatment of wastewaters containing dyes has been carried out applying mainly anodic oxidation (AOx), being the most popular electrochemical process for degradation of organic compounds in wastewaters [8,9].

The popularity of this electrochemical method is based on the fact that the organic compounds are oxidized by direct electron transfer to the anode and/or mediated oxidation with •OH radical, formed from water discharge on the anode surface at a high current [10–12]. On the other hand, the best anodes found to prompt this procedure are the non-active Boron-doped diamond (BDD) thin-film electrodes, because they interact very weakly with the physisorbed BDD(•OH) produced. As shown in Eq. (1), thin-film electrodes interact very weakly with physisorbed radicals [BDD-

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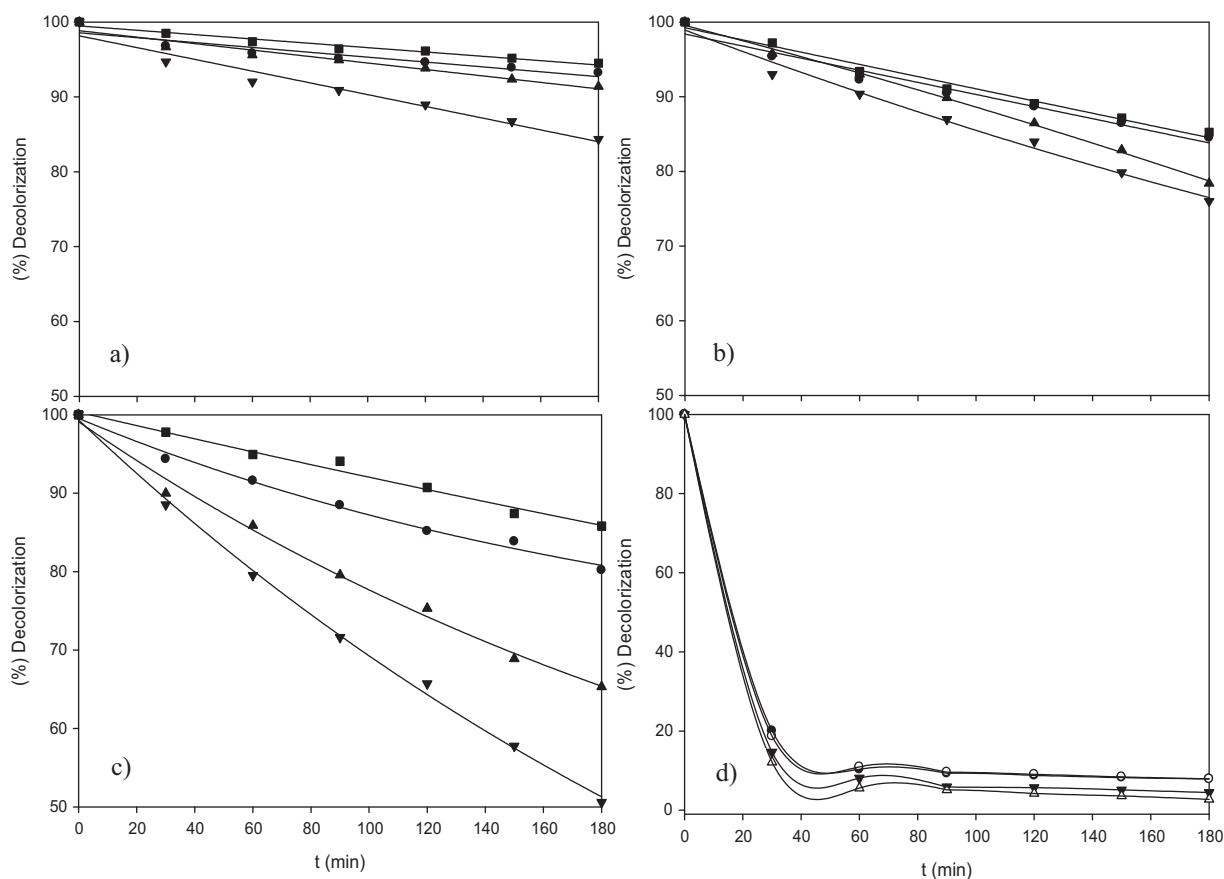
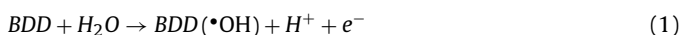


Fig. 1. Effect of different supporting electrolyte under decolorization of diazo dye Congo Red at 498 nm, of 3 L of 100 mg/L in: 50 mM of (a) HClO_4 , (b) H_2SO_4 , (c) Na_2SO_4 , (d) NaCl , using a flow plant with a AOx/BDD stainless steel reactor at (■) 7.5 mA/cm^2 , (●) 15 mA/cm^2 , (▲) 30 mA/cm^2 and (▼) 50 mA/cm^2 .

(•OH)], and promote a much greater O_2 -overpotential than other conventional anodes like Pt, DSA (Dimensional Stable Anodes) and PbO_2 [13,14], enhancing a mineralization process of azo dyes due to the efficient removal of final generated carboxylic acids [15,16].



Electrochemical oxidation with BDD, presents advantages in terms of efficiency, stability, and cost compared to other electrodes and methods [9,17–19].

Therefore, BDD thin-films have been defined as non-active anode material since they do not provide any catalytically active site for the adsorption of reactants and/or products in aqueous media [2,20–22].

The most important characteristic of BDD anodes, is the physisorbed $\text{BDD}(\bullet\text{OH})$, generated from water discharge on its surface, expressed in Eq. (1), which is considered the responsible specie for the electrochemical combustion of organic compounds [11,12,23]. Moreover, slower reactions with other ROS (Reactive Oxygen Species) like H_2O_2 and O_3 molecules were found. Additionally, oxidants generation, like active chlorine species, peroxodisulfate, peroxodicarbonate or peroxodiphosphate, is also feasible [24–27].

This is based on the important technological properties of this material like: (i) inert surface with low adsorption, (ii) remarkable corrosion stability even in strongly acidic media and (iii) extremely high O_2 -evolution overvoltage. The higher efficiency of BDD in relation to other anodes ensures a greater cost performance for electrochemical oxidation. The study of the degradation of diazo dyes under electrochemical advanced oxidation methods is little known and only a limited number of researches have reported on

using an AOx [28–30]. Could it be possible to succeed in the diazo dye Congo Red (CR) degradation via AOx/BDD? Which could be the effect from the supporting electrolyte? Here in, we wish to report on a comparative decolorization, dye removal and mineralization of diazo CR solutions, at 100 mg/L, under anodic oxidation conditions using a constant flow BDD/stainless steel reactor.

It is well known that CR dye has a strong affinity, apparently non-covalently, with fibers; this is why it is widely used in the textile industry. However, the use of CR in this industry has declined, particularly because of its high or moderate toxicity. The role of the different supporting electrolytes was explained by the influence of the applied current density (j) and flow rate on EAOPs. The study was further extended to the AOx process using a flow reactor equipped with a BDD/stainless steel filter-press reactor. The effect of (j) the supporting electrolyte and the flow rate on the AOx treatment, was analyzed in order to determine its optimum operating conditions. In all the assays the dye decay was evaluated by UV–vis determinations, the mineralization was determined by DOC techniques, and the dye concentration abatement was evaluated by liquid chromatography (HPLC).

2. Experimental section

2.1. Chemicals

Commercial Congo Red dye (85% purity, with the rest of inorganic ions for stabilization) was purchased from Sigma–Aldrich and used as received. Analytical grade sodium sulfate, perchloric acid, sodium chloride and sulfuric acid used as background electrolytes were purchased from Fluka and Sigma–Aldrich,

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