



Research article

Decolorization of reactive dyes in solar pond reactors: Perspectives and challenges for the textile industry



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ABSTRACT

In the past three decades, Fenton and photo-Fenton processes have been the subject of a large number of research studies aimed at developing a low-cost and robust alternative to treat complex wastewater. Aspects such as installation and operating costs and technical complexity of reactors have limited the commercial applications of Fenton processes. In this study, we evaluated the potential of solar pond reactors to carry out degradation of the dye reactive orange 16 (RO16). Decolorization ($D = 99 \pm 0.6\%$), chemical oxygen demand reduction ($COD = 55 \pm 2\%$), total organic carbon removal ($TOC = 28 \pm 0.5\%$), and biocompatibilization can be accomplished using 15% peroxide ($0.6 \text{ mg H}_2\text{O}_2/\text{mg RO16}$), which is theoretically required to mineralize the dye. Under dark conditions, decolorization and aromatic removal were scarcely affected (2%), whereas COD and TOC removal were reduced to 37% and 16%, respectively. The application of multivariable analysis and the use of low-cost reactors may lead to a reduction in annual treatment costs of colored effluents to $0.76 \text{ (US/m}^3\text{)}$. Furthermore, the treatment capacity can be increased from $0.6 \text{ m}^3 \text{ wastewater/m}^2 \text{ reactor surface}$ to $1.7 \text{ m}^3 \text{ wastewater/m}^2 \text{ reactor surface}$ without compromising process efficiency or the biodegradability (BOD_5/COD ratio) of the effluent. Dyeing auxiliaries, mainly NaCl, appreciably reduced the decolorization performance in Fenton ($13 \pm 0.4\%$) and photo-Fenton ($83 \pm 0.5\%$) processes due to the formation of iron-chloride complexes and less powerful oxidants. To reduce the impact of auxiliary agents on process performance and treatment capacity, the Fe^{2+} concentration should be increased from 5 mg/L to 15 mg/L. The results seem promising; however, additional studies at pilot and semi-industrial scales should be conducted to demonstrate the potential of low-cost reactors to carry out colored wastewater treatment.

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1. Introduction

Textile effluents are a complex mixture of organic (dyes, surfactants, biocides, leveling agents) and inorganic (salts, heavy metals, oxidants) compounds that are applied to make fibers resistant to physical, chemical and biological agents (IPPC, 2003). Textile wastewater is considered problematic due to large variations observed in flow rate, organic load, salt concentration, total solids, temperature, pH, and biodegradability (dos Santos et al., 2007; IPPC, 2003; Khatri et al., 2015). Reactive dyes represent 25% of the world dye market (~175 000 ton) and are applied to cotton, silk, wool, and cellulose over a wide range of temperatures and pH values (dos Santos et al., 2007; IPPC, 2003). The fixation efficiency

of a reactive dye is highly variable (40–95%) and depends on the type of fiber, the chemicals used in the dyeing process, the applied technology, the required shade, and the number of reactive groups present in the dye (IPPC, 2003; Khatri et al., 2015; Singh and Arora, 2011). This situation has created a major environmental problem because severe absorbance and organic load of textile wastewaters may lead to particular risks to aquatic organisms, robustness of conventional wastewater plants, and human health (Hai et al., 2007).

Advanced oxidation processes (AOPs) have emerged as a safe, easy-to-operate, and environmentally friendly technology for degrading complex substances at environmental temperature and pressure (Pouran et al., 2015; Singh and Arora, 2011; Spasiano et al., 2015). AOPs such as Fenton and photo-Fenton are applicable to both new and existing facilities; however, the scale-up, environmental impact, and cost analysis are scarce and highly variable (García-Segura et al., 2016; Hai et al., 2007; Silva et al., 2016). Fenton and

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photo-Fenton processes have been applied for the treatment of dyes and textile effluents at laboratory, pilot and semi-industrial scales (Alnuaimi et al., 2008; Hai et al., 2007; Hernández-Rodríguez et al., 2014; Hsueh et al., 2005; Karthikeyan et al., 2011; Papic et al., 2009). In such type of processes, pH, dye concentration, radiation source, temperature, dose of reagents, and reaction time affect directly the efficiency of the treatment, the volume of the required reactor, and costs (Malato et al., 2009; Monteagudo et al., 2009; Pouran et al., 2015; Rodrigues et al., 2013). Furthermore, Fenton and photo-Fenton processes have been successfully coupled to biological, electrochemical, and physicochemical processes for the treatment of dyes and industrial effluents (GilPavas et al., 2017; Hai et al., 2007; Kulik et al., 2007). Coupled processes can be overcome technical and economic disadvantages of single treatment, providing a more efficient and robust alternative for treatment of dyes and textile wastewaters. The technical, environmental and economic success of hybrid processes depends on wastewater characteristics, fixed and operating costs, synergism between processes, and presence of inhibitory or recalcitrant constituents (Malato et al., 2009; Oller et al., 2011).

Reactors required for Fenton applications are simple to operate; in addition, they can handle large flows and are less costly than those used for photo-Fenton processes. Compound parabolic concentrators (CPC) (Prato-García and Buitrón, 2012; Silva et al., 2016; Spasiano et al., 2015; Tanveer and Guyer, 2013), parabolic trough collectors (Spasiano et al., 2015; Tanveer and Guyer, 2013), circulating-bed reactors (Bello et al., 2017; Tisa et al., 2014), stirred tanks (Modirshahla et al., 2007; Wang and Bai, 2016), fixed-bed reactors (Duarte et al., 2013; Fida et al., 2017; Tisa et al., 2014), fluidized-bed reactors (Bello et al., 2017; García-Segura et al., 2016; Su et al., 2011), falling films (Bahnemann, 2004; Expósito et al., 2017), and flat-plate reactors (Wyness et al., 1994a, 1994b) have been successfully applied for the treatment of toxic and/or recalcitrant compounds. The CPC has been widely applied to photocatalytic processes for the degradation of pesticides, endocrine disruptors, disinfection, landfill leachates, antibiotics, dyes, and petrochemical effluents (Chacón et al., 2006; Malato et al., 2009; Silva et al., 2016; Spasiano et al., 2015).

A CPC-type reactor is a tubular reactor manufactured in borosilicate glass with low iron content that uses two involutes covered with anodized and electro-polished aluminum as reflecting surfaces (Blanco and Malato, 2003; Spasiano et al., 2015). In CPC-type reactors a higher area to reactor volume is required to increase degradation rate and degree of mineralization of pollutants; this operational condition may be considered adverse for large-scale applications (Spasiano et al., 2015).

A CPC presents high efficiencies, but aspects such as fixed costs (200–400 US/m²), pressure drops, dirt on the reflecting surfaces, fragility of reactor materials, and technical complexity increase treatment costs and limit its commercial viability (Blanco and Malato, 2003; Silva et al., 2016; Spasiano et al., 2015). Several studies provide evidence raceway reactors, waste stabilization ponds, solar lagoons, and flat-plate reactors are efficient for degrading micro-pollutants and phenolic compounds (Carra et al., 2014, 2015; Rivas et al., 2015; Wyness et al., 1994b). Solar lagoons (a type of shallow pond reactor) have been extensively used for nutrient removal, disinfection, algae cultivation, disinfection, and wastewater treatment (Lam and Lee, 2013; Pawar, 2016; Polprasert and Kittipongvises, 2011; Wyness et al., 1994b). Solar pond reactors present low optical efficiencies but are advantageous because of their high treatment capacity and low cost (US\$15/m²). Additionally, they are simpler to construct and operate, have low energetic consumption (1.5–9 W/m³), and are more robust than CPC-type reactors (Carra et al., 2014, 2015; Lam and Lee, 2013).

The textile industry is a complex and heterogeneous sector dominated mainly by small and medium facilities (IPPC, 2003). In developing countries, sustainable alternatives for wastewater treatment have been applied rarely due to technical and economic barriers (IPPC, 2003; Ozturk et al., 2016; Vajnhandl and Volmajer, 2014). Therefore, the application of operative strategies that will allow reduction in reagent consumption and the use of low-cost reactors are fundamental to extending the application of photo-assisted processes to an industrial scale. In this work, we evaluate the potential of solar pond reactors to carry out decolorization of the azo compound reactive orange 16 (RO16). The first two stages of this study include optimized selection of reagents and analysis and modeling of the decolorization process. In the third stage, the role of operational factors such as solar irradiation, reactor depth, and the presence of chemical auxiliaries in process performance are evaluated.

2. Materials and methods

2.1. Reagents

Reactive orange 16 (RO16, C₂₀H₁₇N₃Na₂O₁₁S₃, λ_{max} = 496 nm, purity = 70%W, M.W. = 617, Sigma-Aldrich), NH₄VO₃ (Sigma-Aldrich, ACS, 99.9%), Na₂S₂O₃ (Sigma-Aldrich, ACS, 99.9%), CH₃COONa (Sigma-Aldrich, ACS, 99.9%), Na₂CO₃ (Fisher Scientific, ACS, 99%), CH₃OH (Sigma-Aldrich, ACS, 99.9%), NaCl (Fisher Scientific, 99%), FeSO₄ × 7H₂O (Fisher Scientific, ACS, 99%), H₂SO₄ (Sigma-Aldrich, 96%), NaOH (Sigma-Aldrich, ACS, 97%). H₂O₂ (concentration 30% w/w) was purchased from Mollabs.

2.2. Analytical determinations

The percentage of decolorization (PD) was assessed on the basis of absorbance changes at 496 nm (the strongest tautomeric form of dye). In Eq. (1), A₀ and A_t are the absorbance of the sample at reaction time 0 and t, respectively. Additionally, a spectral scan between 250 nm and 650 nm was performed to study absorbance reduction at 388 nm and 296 nm. The peaks at 388 nm and 296 nm are attributed to the presence of the azo form of RO16 and naphthalene-type rings, respectively (Thomas and Burgess, 2007). Analyses were carried out in a UV–visible spectrophotometer (Spectroquant Pharo-300, Merck, Germany, 10 mm quartz cuvettes) at least in triplicate. Samples (1 mL) were quickly injected into a vial containing a methanol-water solution (4 mL, 250 mM) to reduce their optical density and to stop the Fenton reaction.

$$PD = \left(\frac{A_0 - A_t}{A_0} \right) \quad (1)$$

The percentage of carbon removal was determined using a total organic carbon (TOC) analyzer (Shimadzu-5050A, Japan), using the standard method 5310B as a reference (APHA et al., 2005). Chemical oxygen demand (COD) assays were performed in low range vials (0–150 mg O₂/L) as indicated in the standard method 5220D (APHA et al., 2005). The biochemical oxygen demand (BOD₅) was performed in triplicate at constant temperature (20 ± 1 °C) using microorganisms extracted from an urban wastewater treatment plant (Cali, Colombia). BOD₅ tests were conducted according to the standard procedures (5210 B). The biodegradability (B) of the effluent was determined from the BOD₅/COD ratio. To reduce interferences caused by H₂O₂ in the COD and BOD assays, samples were treated with a NaOH-Na₂S₂O₃ solution. The concentration of H₂O₂ was determined using the spectrophotometric method developed by Nogueira et al. (2005). All analyses were performed at least in triplicate. Assays to determine inhibition of the oxygen

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