



# Optimization and toxicity assessment of a combined electrocoagulation, $\text{H}_2\text{O}_2/\text{Fe}^{2+}/\text{UV}$ and activated carbon adsorption for textile wastewater treatment

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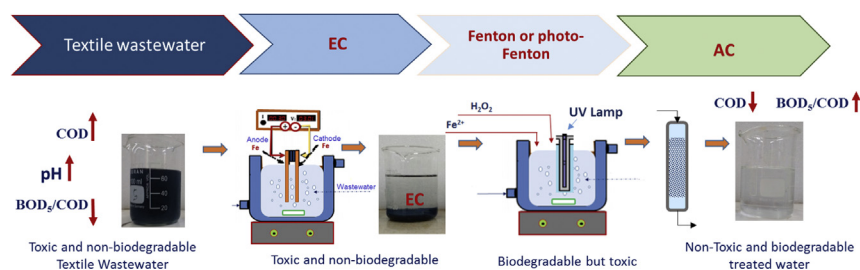
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## HIGHLIGHTS

- EC/ $\text{H}_2\text{O}_2/\text{Fe}^{2+}/\text{UV}$  process was evaluated for wastewater treatment.
- EC/ $\text{H}_2\text{O}_2/\text{Fe}^{2+}/\text{UV}$  performance was optimized by the Response Surface Methodology.
- Optimum conditions were found: pH = 4.3,  $[\text{Fe}^{2+}] = 1.1 \text{ mM}$  and  $[\text{H}_2\text{O}_2] = 9.7 \text{ mM}$ .
- EC/ $\text{H}_2\text{O}_2/\text{Fe}^{2+}/\text{UV}$  process yielded a colorless and highly mineralized effluent.

## GRAPHICAL ABSTRACT



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## ABSTRACT

In this study, the potential application of sequential Electrocoagulation + Fenton (F) or Photo-Fenton (PF) + Active carbon adsorption (EC + F/PF + AC) processes were analyzed as alternatives for the treatment of an industrial textile wastewater resulting from an industrial facility located in Medellín (Colombia). In order to maximize the organic matter degradation, each step of the treatment was optimized using the Response Surface Methodology. At first, the optimal performance of EC was achieved with Fe electrodes operating at pH = 7,  $j_{\text{EC}} = 10 \text{ mA/cm}^2$  and 60 rpm, during 10 min of electrolysis. At these conditions, EC let to remove 94% of the dye's color, 56% of the COD and 54% of the TOC. Next, sequentially applied Fenton or photo-Fenton process (i.e., EC + F/PF), operating at the optimized conditions (pH = 4.3,  $[\text{Fe}^{2+}] = 1.1 \text{ mM}$ ,  $[\text{H}_2\text{O}_2] = 9.7 \text{ mM}$ , stirring velocity = 100 rpm and reaction time = 60 min.), improved the quality of the treated effluent. The EC + F let to achieve total color reduction, as well as COD and TOC removals of 72 and 75%, respectively. The EC + PF reached 100% of color, 76% of COD and 78% of TOC reductions. The EC + F/PF processes were more efficient than EC in elimination of low molecular weight (<5 kDa) compounds from wastewater. Moreover, the BOD<sub>5</sub>/COD ratio increased from 0.21 to 0.42 and from 0.21 to 0.46 using EC + F and EC + PF processes, respectively. However, EC + F/PF were not fully effective for the removal of acute toxicity to *Artemia salina*: 20% and 60% of reduction in toxicity using EC + F and EC + PF, respectively, comparing to very toxic (100%) raw textile wastewater. Thus, activated carbon adsorption was applied as an additional step to complete the treatment. After AC adsorption, the acute toxicity decreased to 10% and 0% using EC + F and EC + PF, respectively. The total operational costs, including chemical reagents, electrodes, energy consumption and sludge disposal, were of 1.65 USD/m<sup>3</sup> and 2.3 USD/m<sup>3</sup> for EC + F and EC + PF, respectively.

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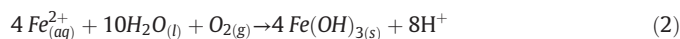
## 1. Introduction

Textile industry consumes large quantities of water and produces large volumes of wastewater, mostly during dyeing and finishing processes. Textile wastewater is often rich in color. Moreover, it is characterized with the presence of residues of reactive dyes and other hardly-degradable chemicals as well as high concentrations of Chemical Oxygen Demand (COD) and Biological Oxygen Demand (BOD<sub>5</sub>). Textile dyes are mainly aromatic and heterocyclic compounds. Their complicated and stable structure includes color-display and polar groups, both hardly-degradable. Indeed, the toxic effect of dyestuffs on both living species and the aqueous environment is widely recognized (Villanueva-Rodríguez et al., 2009; Shaolan Ding et al., 2010).

Throughout the world, the discoloration and degradation of textile wastewater is mandatory before its final disposal. In fact, the latest environmental regulations have required the development of alternative treatment techniques to meet the standards. It has been proven that physico-chemical processes (v. g., coagulation/flocculation, precipitation, adsorption, ion exchange, membrane separation and oxidation) result in incomplete mineralization of pollutants (Zhou et al., 2009). On the other hand, in many cases, due to the toxicity of textile wastewater, biological methods are inefficient especially if the pollutants are present in high concentrations (Ghanbari and Moradi, 2015). Recently, electrochemical technologies (such as electrocoagulation, electroflotation, electrodecantation, electrooxidation, etc....) and Advanced Oxidation Processes (AOPs) have been considered as efficient alternatives for textile wastewater remediation (Brillas and Martínez-Huitle, 2015). The combination of these processes have been successfully applied for the treatment of several types of wastewaters. Sequential electrocoagulation/electrochemical Fenton (Flores et al., 2018) and coagulation/acid cracking/Fenton process (Yazdanbakhsh et al., 2015) were used for the treatment of olive oil mill wastewater. The sequential permanganate, electro-Fenton and Co<sub>3</sub>O<sub>4</sub>/UV/peroxymonosulfate (Jaafarzadeh et al., 2017) process was applied for pulp and paper resulting wastewater. An integrated electrocoagulation-electrooxidation (Ibarra Taquez et al., 2017) was used to treat wastewater resulting from a soluble coffee plant. Finally, combination of coagulation/photocatalytic methods (Jorfi et al., 2016) was also considered for dye and real textile wastewater treatment. In all these cases, the arrangement of different treatment processes improved global wastewater characteristics, minimized operational costs, slightly providing into no generation of secondary recalcitrant wastes and letting to obtain treated effluents with appropriate quality to be discharged into water bodies.

In this work, the potential application of a sequential EC + (Fenton (F) or photo-Fenton (PF)) + AC adsorption process is analyzed as an alternative for the treatment of an industrial textile wastewater (ITWW). First, electrocoagulation (EC) is used. It is a very versatile method for water and wastewater remediation. It has been successfully applied to remove a wide variety of pollutants, among them heavy metals, different anions and organic compounds including dyes (Koby et al., 2003; Garcia-Segura et al., 2017; Vepsäläinen et al., 2011). EC presents many advantages over other techniques, such as high efficiency at low capital and operational costs, equipment simplicity and easiness of process control. It relies on the electrochemical dissolution of a sacrificial metal electrode (usually made of iron or aluminum) into soluble or insoluble species that enhance the coagulation, the adsorption or the precipitation of soluble or colloidal pollutants (Mollah et al., 2004). Pollutant nature and concentration, electrode material, current density, solution pH and electrolyte type are known as factors influencing the effectiveness of EC treatment. The following reactions describe the processes involved in EC, if iron electrodes are used:

At the anode:



At the cathode:



It is important to notice that H<sub>2</sub> generation can provide an extra removal of organic material by flotation.

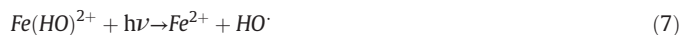
On the other hand, AOPs deal with the use of hydroxyl radicals (•OH) to attack the pollutant molecules present in wastewater. They are recognized as valuable methods to increase the biodegradability of textile wastewater due to its high oxidative efficiency. A variety of AOPs have been reported in the literature, among them UV/H<sub>2</sub>O<sub>2</sub>, UV/O<sub>3</sub> (Boczkaj and Fernandes, 2017), TiO<sub>2</sub>-assisted photocatalysis, Fenton process (Fe<sup>2+</sup>/H<sub>2</sub>O<sub>2</sub>) (De Lima et al., 2017; Rosa et al., 2015), electrochemical AOPs (Chatzisympson et al., 2006) and ultrasonication (Darvishi et al., 2016). The Fenton oxidation is an attractive method to mineralize dyes remaining in textile wastewater. Indeed, H<sub>2</sub>O<sub>2</sub> is an environmentally friendly oxidant, which is decomposed into water and oxygen, and iron is highly abundant and presents low toxicity (Ghanbari and Moradi, 2015). Fenton processes exhibit the following advantages: equipment simplicity, relatively low cost and easy operation and maintenance (Zazo et al., 2005). Moreover, due to its non-selectivity nature, it provides the removal of wide spectrum of contaminants together with a dramatic reduction in toxicity (Arzate-Salgado et al., 2016). The coupling of EC and Fenton process has been shown to be highly effective due to the generation of strongly oxidizing species. In this case, the electrocoagulated water (i.e., the resulting supernatant) can be further subjected to Fenton or photo-Fenton process. During Fenton reaction, H<sub>2</sub>O<sub>2</sub> decomposition is catalyzed by ferrous ion to generate hydroxyl radicals (•OH) according to Eq. (4) (El-Ghenymy et al., 2012):



The highly reactive •OH radicals can in turn react with dyes present in wastewater, resulting in their oxidation. The Fe<sup>3+</sup> ion can be recycled via Eq. (5), but this process is relatively slow (Rahim Pouran et al., 2015).



It has been established that photo-Fenton reaction can further improve the degradation of organic pollutants, either by direct photolysis or by increasing the production of •OH radicals, according to the following reactions (GilPavas et al., 2017):



Unfortunately, the partial oxidation of organic contaminants may also result in the formation of toxic intermediates (even more noxious than the parent compounds). In such a case, an additional treatment must be applied to the resulting effluent prior to its final disposal into the aqueous environment.

Adsorption processes are commonly used as a final step in the treatment of industrial wastewater. Activated carbon (AC) has been proven as an efficient adsorbent for the removal of a wide variety of environmental pollutants. Indeed, harm by-products (v.g., volatile, semi-volatile and non-volatile chlorinated organic pollutants) can be removed by AC adsorption even at very low concentration levels (Lemus et al., 2012; Pavoni et al., 2006). Among the unique properties of AC, the following can be mentioned: (i) high specific surface area; (ii) easy and wide availability; (iii) stability in acidic/basic environment; and (iv) structural stability at high temperatures. Thus, final adsorption

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