



An overview on the removal of synthetic dyes from water by electrochemical advanced oxidation processes

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HIGHLIGHTS

- In-depth analysis of various electrochemical advanced oxidation processes (EAOPs) for dye removal.
- Recent trends in the dye removal by EAOPs.
- Comparative dye removal efficiency by different EAOPs.
- Dye removal mechanisms during different EAOPs.
- Discussion of scale-up and associated problems of the dye removal processes.

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ABSTRACT

Wastewater containing dyes are one of the major threats to our environment. Conventional methods are insufficient for the removal of these persistent organic pollutants. Recently much attention has been received for the oxidative removal of various organic pollutants by electrochemically generated hydroxyl radical. This review article aims to provide the recent trends in the field of various Electrochemical Advanced Oxidation Processes (EAOPs) used for removing dyes from water medium. The characteristics, fundamentals and recent advances in each processes namely anodic oxidation, electro-Fenton, peroxo-coagulation, ferred Fenton, anodic Fenton, photoelectro-Fenton, sonoelectro-Fenton, bioelectro-Fenton etc. have been examined in detail. These processes have great potential to destroy persistent organic pollutants in aqueous medium and most of the studies reported complete removal of dyes from water. The great capacity of these processes indicates that EAOPs constitute a promising technology for the treatment of the dye contaminated effluents.

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1. Introduction

Water pollution due to various industrial effluents is a global environmental problem. Due to the rapid industrialization, the use of coloring chemicals like dyes also increases day by day. Overall, 40,000 dyes and pigments with more than 7000 different chemical structures have been reported recently (Demirbas, 2009). Production of dyestuff and pigments annually across the world is more than 700,000 tonnes and in India itself it is close to 80,000 tonnes

(Mathur et al., 2005; Gong et al., 2007; Mane et al., 2007). Among this 10,000 different types of dyes and pigments are being manufactured annually across the world (Ponnusami et al., 2008). These dyes are chemically, photolytically and biologically highly stable and are highly persistent in nature (Suteu and Bilba, 2005). Various industries like textile, leather, food, cosmetic, paper, pharmaceutical etc. are using variety of synthetic dyes. Among these industries, textile industries are the largest consumer of dyeing stuffs and pigments and produces large amount effluents after dyeing process. For example, 1.5 million liters per day of effluent are discharging into natural water bodies from an average mill producing 60×10^4 m of fabric (COINDS, 2000). This wastewater contains various kinds of pollutants apart from dyes and most of them are

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hazardous. Depending on the textile processes, effluents generated from the industry contains various types of solvents, salts, detergents etc. apart from many types of dyes (Barredo-Damas et al., 2006). Typical characteristics of the effluent generated from textile industries are given in Table 1. Among these contaminants, removal of dyes from the wastewater needs to be a special attention.

The effluents containing dyes are one of the major threats to environment. Even in low concentrations, the dyes are highly visible (esthetic pollution) and affect the aquatic life and food chain (chemical pollution) (Namasivayam and Kavitha, 2002; Malik, 2003). However, the average concentration of dyes in a textile wastewater is around 300 mg L⁻¹ (Couto, 2009). Disposal of this highly colored wastewater into natural water bodies hamper light penetration, distress biological process in the water medium and provide an aesthetically displeasing appearance (Arivoli et al., 2009). Reduced light penetration with the introduction of colored wastewater reduces the photosynthetic activity within the water body and this also affect the symbiotic process (Ju et al., 2008).

The hazardous, toxic and carcinogenic natures of dyes are also well known. Physiological disorders in aquatic organisms happen by the consumption of dyes in textile effluents via food chain (Karthikeyan et al., 2006). Some of the azo dyes are responsible for causing bladder cancer in humans and chromosomal aberration in mammalian cells (Medvedev et al., 1988; Percy et al., 1989). Cytotoxicity of reactive dyes (three monochlorotriazinyl dyes: yellow, red and blue with different concentrations) using human keratinocyte HaCaT cells in vitro was investigated by Klemola et al. (2007) and the mean inhibitory concentration values after 72 h of exposure was measured as 237 µg mL⁻¹ for yellow dye, 155 µg mL⁻¹ for red dye and 278 µg mL⁻¹ for blue dye. Similarly, spermatozoa motility inhibition test of these dyes showed the mean inhibitory concentration values after 24 h of exposure as 135 µg mL⁻¹ for yellow dye, 124 µg mL⁻¹ for red dye and 127 µg mL⁻¹ for blue dye (Klemola et al., 2006). Frame-shift mutation and base pair substitution in Salmonella in the presence of CI disperse blue was observed by Umbuzeiro et al. (2005). Experimental investigations of Bae and Freeman (2007) reported that C.I. Direct Blue 218 is very toxic to daphnids. The 50% of mortality of daphnids was observed for concentrations of 1–10 mg L⁻¹ of C.I. Direct Blue 218 after 48 h. Impact of textile effluents on a proteinous edible fresh water fish *Mastacembelus Armatus* was examined by Karthikeyan et al. (2006) and they observed a decrease in Na⁺ and Cl⁻ concentrations and increase in K⁺, Mg²⁺ and Ca²⁺ concentrations after 35 d of Acid Blue 92 exposure. The toxicity of the textile dye industry effluent on freshwater female crab, *Spiralothelphusa hydrodroma* was investigated by Sekar et al. (2009). The authors observed a decrease in protein, carbohydrate and lipid contents in ovary, spermatheca, muscle, hepatopancreas, gill, brain, thoracic ganglia and eyestalks at 30 d of sublethal (69.66 mg L⁻¹) concentration of textile dye industry effluent exposure. Mathur et al. (2005) tested

the mutagenic activity of seven dyes by Ames assay, using strain TA 100 of Salmonella typhimurium and reported that except violet dye, all are mutagenic. Among these mutagenic dyes, Congo red and royal blue dyes are moderately mutagenic while bordeaux is highly mutagenic or extremely mutagenic. Similarly, effect of malachite green on immune and reproductive systems was observed by Srivastava et al. (2004). The authors also observed the potential genotoxic and carcinogenic nature of the mentioned basic dye.

These non-exhaustive examples highlight how much the presence of the dyes in water is harmful to the aquatic environment. Therefore, removal of dyes from aqueous medium is an important environmental issue in the environmental safety point of view.

2. Electrochemical advanced oxidation processes (EAOPs) for dye removal

Various treatment techniques such as adsorption, coagulation, filtration, electrocoagulation, photolysis, sonolysis, biodegradation, wet land treatment, ozonation, photocatalysis, membrane filtration etc. have been used for removing synthetic dyes from aqueous solution. Advantages and disadvantages of dye removal methods have been discussed in detail (Crini, 2006; Martínez-Huitle and Brillas, 2009; Nidheesh et al., 2013). Based on the principle mechanism behind the removal of dyes, these processes can be divided into two broad classes: Separative (physical, and physicochemical) methods, and degradative (chemical and biological) methods. The schematic diagram of the various methods used for the dye removal is shown in Fig. 1. Most of the methods used for dye removal are separation process and the main disadvantage of these processes is the disposal of dye containing sludge as in coagulation process, dye sorbed adsorbents and concentrated dye solution as in membrane processes. In contrary to this, complex dye compounds undergo a series of degradation in chemical degradation methods. In the case of use of advanced oxidation processes, degradation procedure advance until ultimate oxidation degree, i.e. mineralization of organic pollutants. These methods produce carbon dioxide, water and various inorganic ions (following heteroatoms presents at the starting organic pollutants) as the final products.

Among various degradation techniques, advanced oxidation processes (AOPs) received great attention for the efficient degradation of dyes, during recent years (Martínez-Huitle and Brillas, 2009; Oturan and Aaron, 2014). These processes are based on the production of highly reactive oxidants, mainly hydroxyl radical ([•]OH). This radical is the second most powerful oxidizing agent (after fluorine) with a redox potential of E° ([•]OH/H₂O) = 2.8 V/SHE. Once these radicals are produced *in situ*, they attack organic pollutants with high reaction rates through following three different ways (Oturan, 2000; Brillas et al., 2009; Sirés et al., 2014): electron transfer (redox reaction) (Eq. (1)), H atom abstraction (dehydrogenation) (Eq. (2)), and electrophilic addition to π systems (hydroxylation) (Eq. (3)). These reactions produce organic radicals and start a radical chain including reactions with oxygen (formation of peroxy radicals) and formed reaction intermediates undergo further oxidation reactions with generated oxidizing agents ([•]OH, HO₂, H₂O₂ ...) until the complete mineralization of organic pollutants.



where RX and PhX represent aliphatic and aromatic halogens, respectively.

Table 1
Characteristics of textile wastewater. Reprinted with permission from Sandhya et al. (2008), Copyright 2007, Elsevier.

Parameters	Concentration
Solution pH	9.5–12.5
Total Suspended Solids (mg L ⁻¹)	60–416
Total Dissolved Solids (mg L ⁻¹)	4500–12800
Total Organic Carbon (mg L ⁻¹)	26,390–73190
Biochemical Oxygen Demand (mg L ⁻¹)	25–433
Chemical Oxygen Demand (mg L ⁻¹)	1835–3828
Aromatic Amines (mg L ⁻¹)	20–75
Ammonia (mg L ⁻¹)	2–3
Chloride (mg L ⁻¹)	1200–1375
Sulphate (mg L ⁻¹)	700–2400

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