



Treatment of highly toxic cardboard plant wastewater by a combination of electrocoagulation and electrooxidation processes



Erhan Gengec

University of Kocaeli, Department of Environmental Protection, Kocaeli, Turkey

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ABSTRACT

The objective of this study was to investigate the removal efficiencies of the electrochemical treatment systems as an alternative for the treatment of cardboard plant wastewater (CPW). In accordance with this purpose, CPW was treated by electrocoagulation (EC) with Al electrodes and the effects of current density (CD), operating time (*t*), and initial pH (pH_i) were investigated. The results showed that EC at optimum treatment conditions (CD: 7.5 mA/cm², pH_i : 7.0 and *t*: 60 min) have limited removal efficiencies for total organic carbon (TOC; 17.1%) and chemical oxygen demand (COD, 14.2%), on the contrary of turbidity (98.7%). Due to the low TOC and COD removal efficiencies, a secondary treatment was needed and the electrocoagulated effluent was subjected to electrooxidation (EO) by using a boron doped diamond (BDD) electrode for investigating the effect of CD, *t*, pH_i and electrolyte concentration (C_e). Higher TOC (83.7%) and COD (82.9%) removal efficiencies were obtained by EO under the optimum treatment conditions (CD: 100 mA/cm², pH_i : 7.2, C_e : 5.0 g/L Na₂SO₄ and *t*: 180 min). In addition, a toxicity test was carried out to the raw and treated wastewater under the optimum operating conditions. This study demonstrated that the combination of EC and EO have a satisfactory potential for real industrial wastewater with a high organic content, suspended solids and toxicity.

1. Introduction

The pulp and paper industry use lignocellulose materials (wood and/or recycled paper etc.) as a raw material, and produce paper, cardboard and other cellulose-based products. During production of the paper and cardboard, wastewater that has high suspended solids, color, and organic substance in the range from 75 to 275 m³ per ton of product (Jaafarzadeh et al., 2016) is generated. Moreover, this industrial sector wastewater contains the chlorinated lignosulphonic acids, resin acids, phenols and hydrocarbons besides dibenzo-p-dioxin and dibenzofuran, highly toxic and recalcitrant compounds (Kumar et al., 2015). Due to these components, these effluents are extremely hazardous for the ecosystem because they exhibit a strong mutagenic effect, have acute toxicity, cause a decreasing of oxygen, and prevent light penetration. Therefore, various wastewater treatment systems and their combinations have been employed to minimize the negative influence of pulp and paper industry effluents on the environment until now (Birjandi et al., 2014). Inside these treatment systems, coagulation and flocculation processes in which polyaluminium chloride (PAC) and aluminum sulphate are used are very effective for the removal of suspended solids (SS), colloids and toxicity in wastewater. However, there are two problems involving the potential ecotoxicity of metallic coagulants and other toxic components in wastewater: (I) The production

of large volumes of toxic sludge, and (II) an increase in metal concentration in treated water are the main disadvantages of these systems. Another alternative treatment system in the paper industry is the biological method (mainly aerated lagoon), which is commonly used due to the low cost and minimal operation requirements. The biological methods are also very effective for the removal of soluble and biodegradable organic pollutants (Pokhrel and Viraraghavan, 2004). However, the existence of bio-refractory components in wastewater, the sensitivity of microorganisms to toxic material, production of high amounts of sludge and lengthy time requirements are the disadvantages of the biological methods (Renault et al., 2009). Due to the mentioned disadvantages, a physico-chemical method is usually needed before a biological method. Furthermore, adsorption, ozonation and membrane filtration of chlorinated phenolic compounds and adsorbable organic halides and chemical oxidation of colorful components were carried out effectively in paper industry wastewater (Pokhrel and Viraraghavan, 2004). However, these processes involve a great deal of disadvantages such as high cost, selective removal and difficulties in practice. On the other hand, due to the increasing fresh water cost and the limited discharge standards of wastewater, the focus of researchers has shifted to relatively new alternatives (Mansour and Kesentini, 2008) such as electrochemical treatment systems. Electrocoagulation (EC), electro-oxidation (EO), and electro-fenton (EF) are the most promising

E-mail address: erhan.gengec@kocaeli.edu.tr.

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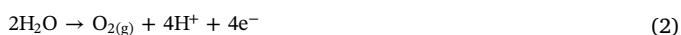
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electrochemical treatment systems.

Due to a great deal of competitive advantages such as no selectivity towards pollutants, low operating cost, easy operable equipment, short operation time, low requirement of chemicals, and low production amount of sludge, the EC process is frequently suggested in literature for the removal of chemical oxygen demand (COD), biological oxygen demand (BOD), total organic carbon (TOC), suspended solids (SS), turbidity, and toxicity (David et al., 2015; Makwana and Ahmed, 2016). Generally, a power supply (alternating current; AC or direct current; DC) and electrodes (aluminum or iron) are used during the EC process and anode electrodes are consumed under a potential. Thus, three successful stages during this process are realized: (i) formation of coagulants by electrolytic oxidation of the sacrificial anode electrode, (ii) destabilization of the pollutants, particulate suspension, and breaking of emulsions, (iii) aggregation of the destabilized phases to form flocs. The main reaction at the Al anode is shown as follows (Dastyar et al., 2015; Elabbas et al., 2015; Ezechi et al., 2014; Hamdan and El-Naas, 2014; Makwana and Ahmed, 2016; Mella et al., 2015; Vasudevan, 2012; Zaidi et al., 2016):



In addition to aluminum dissolution, oxygen evolution is realized via a reaction (Eqn. 2) at the anode:



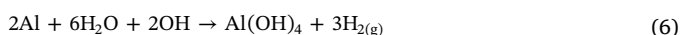
At cathode, hydrogen evolution is realized by the following reaction (Eq. (3)) and the formed hydrogen causes the floatation of pollutants. Moreover, the produced hydroxyl ions at cathode generally cause an increase of pH.



The hydrolysis constants for aluminum cover a very narrow range, and all of the aluminum deprotonations are 'squeezed' into an interval of three pH units. Therefore, except for a narrow pH region (approximately 5–8), the dominant soluble species are Al^{3+} at low pH and $\text{Al}(\text{OH})_4^-$ at high pH (Gengec et al., 2012; Kobya et al., 2014; Kobya and Gengec, 2012). The produced positively charged metal ions (Al^{3+}) in the acidic conditions neutralize the negatively charged pollutants. At about neutral pH (5–8) metal ions hydrolyze to monomeric-polymeric aluminum hydroxides and amorphous species, $\text{Al}(\text{OH})_3$ (sweep coagulation). Monomeric-polymeric species such as $\text{Al}(\text{OH})_2^+$, $\text{Al}(\text{OH})_3^0$, $\text{Al}(\text{OH})_4^-$, $\text{Al}_2(\text{OH})_6^{4+}$, $\text{Al}_6(\text{OH})_{15}^{3+}$, $\text{Al}_7(\text{OH})_{17}^{4+}$, $\text{Al}_8(\text{OH})_{20}^{4+}$, $\text{Al}_{13}\text{O}_4(\text{OH})_{24}^{7+}$, and $\text{Al}_{13}(\text{OH})_{34}^{5+}$ transform initially into $\text{Al}(\text{OH})_{3(\text{s})}$ and finally polymerize to $\text{Al}_n(\text{OH})_{3n}$ (Eqs. (4) and (5)) according to the pH of the wastewater and concentration of Al:



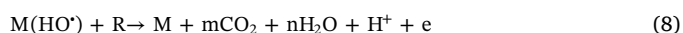
These hydroxides are excellent coagulants and cause adsorption of pollutants. On the other hand, the formed negatively charged hydroxides such as $\text{Al}(\text{OH})_4^-$ (Eq. (6)) usually reduce the removal efficiencies at high pH values (> 9);



Until now, EC technology has been reported as an effective treatment system for industrial wastewater such as electroplating (Kobya et al., 2016), distillery (David et al., 2015), tannery (Elabbas et al., 2015), baker's yeast production (Gengec et al., 2012) and textile dyeing (Kobya et al., 2014). Therefore, removal of turbidity and toxicity in cardboard plant wastewater (CPW) can be carried out successfully by EC. However, the mineralization of organic components in CPW up to the legal discharge standards of wastewater does not seem possible via EC.

EO is an effective method for the mineralization of organic substances in wastewater, though not for turbidity. Thus, the combination

of EC with EO can be a good alternative for treatment of the CPW (Jaafarzadeh et al., 2016; Zaidi et al., 2016). In addition, EO presents a large number of advantages: (I) the oxidation of pollutants up to the final product or to less toxic by-products in contrast with coagulation, membrane, filtration, and adsorption processes, (II) short oxidation time requirements, no sludge problem, non-selective to toxic and non-biodegradable material in contrast with biological oxidation processes, and (III) addition of chemical reagents not required (except for electrolytes or catalyst addition at low amounts) in contrast with chemical oxidation. EO can be categorized as direct and indirect oxidation. In the indirect oxidation process, oxidizing reagents such as chlorine, hypochlorous acid, hypochlorite, hydrogen peroxide and ozone are formed at the electrodes surfaces and then, destroy pollutants in wastewater. The direct oxidation process involves (I) electron transfer to the pollutants and (II) the forming of hydroxyl radicals on anode's surface (Särkkä et al., 2015; Wu et al., 2014). When direct and indirect oxidations are compared, direct oxidation has some advantages such as requiring a lower amount of chemical substance and producing no secondary contaminants (Särkkä et al., 2015). The electrocatalytic activity and electrochemical stability of the electrode is a key point for direct oxidation. Although several anode materials have been used in the direct oxidation, the complete mineralization of the pollutants has been possible by use of high oxygen overvoltage anodes such as SnO_2 , PbO_2 and boron doped diamond (BDD) (Panizza and Cerisola, 2001). The BDD electrode has been identified as an excellent non-active anode for the removal of pollutants in wastewater (Özcan et al., 2008) and the formation of hydroxyl radical ($\cdot\text{OH}$) on the BDD anode surface has been reported as the main removal mechanism. Due to having an inert surface and low adsorption capacity, the BDD surface interacts weakly with the $\cdot\text{OH}$. During EO with BDD, the complete mineralization of pollutants in wastewater is possible due to the physisorbed $\cdot\text{OH}$ by the following reactions (Eqs. (7) and (8), R: organic pollutants in wastewater) (Gao et al., 2004; Marco Antonio Quiroz et al., 2006; Martínez-Huitle and Ferro, 2006; Scialdone, 2009; Sires et al., 2014)



Although scientists recent attention has focused on EO, there is a lack of information in literature regarding EO: (I) removal efficiencies of EO in different real wastewater (only 20% EO paper in the last 3 years have dealt with treatment of the real wastewater), (II) comparison of the electrodes performance, (III) contribution level on removal efficiencies of EO at a combination of different treatment systems, (IV) removal mechanism, (V) electrical consumption (Sires et al., 2014) and (VI) possible formation of toxic by-products. To the best of the author's knowledge, the combination of the electrocoagulation (Al electrode) and electrooxidation processes (BDD electrode) has yet to be studied for the treatment of CPW. Moreover, the influence of the EC and EO combination on toxicity of real wastewater has not been investigated sufficiently.

Therefore, the purposes of this study were: (I) present the combination of EC and EO as a competitive alternative for the treatment of industrial wastewater with high suspended solids, organic content and toxicity, (II) investigate the effect of process conditions on removal efficiencies, and (III) calculate the electrical and electrode consumption during these processes.

2. Material and methods

2.1. Wastewater

The wastewater was taken from the starting point of a cardboard plant treatment system. The coated cardboard is made out of recycled paper and the annual cardboard production capacity of the plant has reached 240,000 t (Table 1).

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