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Effect of bipolar electrode material on the reclamation of urban wastewater by an integrated electrodisinfection/electrocoagulation process

*Javier Llanos**, *Salvador Cotillas*, *Pablo Cañizares*, *Manuel A. Rodrigo*

Chemical Engineering Department, University of Castilla-La Mancha, Edificio Enrique Costa Novella, Campus Universitario s/n, 13005 Ciudad Real, Spain

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ABSTRACT

This work presents an integrated electrodisinfection/electrocoagulation (ED–EC) process for urban wastewater reuse that employs iron bipolar electrodes. Boron doped diamond (BDD) was used as the anode and stainless steel (SS) as the cathode. A perforated iron plate was introduced between the anode and cathode to function as a bipolar electrode. This ED–EC combined cell makes it possible to conduct the simultaneous removal of microbiological content and elimination of turbidity from urban wastewater. The results show that current densities greater than or equal to 6.70 A m^{-2} enable complete disinfection of the effluent and the removal of more than 90% of its initial turbidity. Hypochlorite and chloramines formed during the ED–EC process were found to be the main compounds responsible for the disinfection process. Furthermore, a cell configuration of cathode (inlet)-anode (outlet) improves the process performance by enhancing turbidity removal. Finally, the influence of the bipolar electrode material (iron or aluminium) was assessed. The results indicate that the efficiency of the electrodisinfection process depends mainly on the anodic material and is not influenced by the material of the bipolar electrode. In contrast, the removal of turbidity is more efficient when using iron as a bipolar electrode, especially at low current densities, due to the formation of a passive layer on the aluminium that hinders the dissolution of the bipolar electrode.

1. Introduction

In recent years, electrochemical engineering has experienced great advances due to the development of new electrode materials. For this reason, electrochemical technologies have been widely used in wastewater treatment processes such as the removal of nutrients (Bektas et al., 2004; Lacasa et al., 2011; Tran et al., 2011; Zaleschi et al., 2013), disinfection of wastewater effluents (Cano et al., 2011; Ghernaout et al., 2011; Cui et al., 2013) and degradation of organic compounds (Martínez-Huitle and Brillas, 2009; Alves et al., 2013).

The choice of an appropriate electrode material is a matter of major importance to ensure the success of an electrochemical process. In the case of coagulation, a technology applied in urban wastewater treatment and reclamation (Pouet and Grasmick, 1995; Rodrigo et al., 2010), aluminium and iron are the most commonly used electrode materials

* *Corresponding author*. Tel.: +34 926 29 53 00x3508; fax: +34 926 29 52 56.
E-mail address: javier.llanos@uclm.es (J. Llanos).

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(Farhadi et al., 2012; Mohora et al., 2012; Dubrawski et al., 2013). For the treatment of various target effluent types, the different behaviours of iron and aluminium electrodes have been assessed in the electrocoagulation of textile waste (Kobya et al., 2003; Zongo et al., 2009), baker's yeast wastewaters (Kobya and Delipinar, 2008) and leachate (Ilhan et al., 2008), among others. The results obtained have proven that the efficiency of the electrode material is directly dependent on the nature of the wastewater being treated. Specifically, iron electrodes have proved more efficient for Chemical Oxygen Demand (COD) removal during the treatment of textile wastewaters, whereas the use of aluminium electrodes had led to higher elimination percentages in the treatment of leachate and baker's yeast wastewaters.

Electrochemical disinfection has become a good alternative for the removal of microbiological content in wastewater (Martínez-Huitle and Brillas, 2008; Ghernaout and Ghernaout, 2010; Pérez et al., 2010). This process consists mainly of the elimination of faecal coliforms present in water through two potential mechanisms: the electroadsorption of *Escherichia coli* (*E. coli*) on the electrode surface (direct disinfection) (Golub et al., 1987; Ninomiya et al., 2013) and the formation of disinfectant species that attack and eliminate *E. coli* (indirect disinfection) (Schmalz et al., 2009; Bergmann, 2010). Again, the selection of the appropriate anode material for the electrodisinfection process can affect the nature of the oxidising species formed; the most common anode materials are boron doped diamond (BDD) and dimensionally stable anodes (DSA) (Jeong et al., 2009; Gusmão et al., 2010; Haaken et al., 2012).

In the context of wastewater regeneration by electrochemical processes, our research group recently demonstrated the technical viability of a combined reactor for the simultaneous disinfection and removal of turbidity in actual effluents from municipal wastewater treatment facilities, WWTF (Cotillas et al., 2013). In that work, the combined electrodisinfection/electrocoagulation cell proposed was equipped with aluminium bipolar electrodes. This cell was able to completely remove the microbiological content in urban wastewaters at low applied electric charges (0.008 Ah dm^{-3}) and low current densities (6.65 A m^{-2}). In addition, this cell facilitated a decrease in the turbidity of wastewater by the electrodissolution of the aluminium bipolar electrode and the subsequent formation of coagulant species. However, the applied electric charge required to reduce the turbidity was found to be higher than that necessary to disinfect the urban wastewaters.

One of the key elements of the cell, which strongly affects the removal of turbidity, is the material of the bipolar electrode. For this reason, and with the aim of optimizing the efficiency of the proposed cell, it is necessary to confront a detailed study of the influence of the bipolar electrode material on the process performance. Likewise, the comparison of the behaviour of both materials can provide valuable information for the treatment of actual wastewater by combined electrochemical processes involving electrocoagulation.

Based on these results, the main aim of the present work was to study the influence of the bipolar electrode material (iron or aluminium) on the performance of an integrated electrodisinfection/electrocoagulation process for the regeneration of urban wastewater from the WWTF of Ciudad Real (Spain). The effects of the current density and cell flow path on the process performance were also studied using iron as a bipolar electrode material.

2. Material and methods

This section describes the analytical techniques, experimental setup and experimental procedure for the electrodisinfection/electrocoagulation process.

2.1. *Analytical techniques*

The total iron and aluminium concentration was measured off-line using an inductively coupled plasma spectrometer (Liberty Sequential, Varian) (detention limit <1.5 ppb) according to a previously published standard method (APHA-AWWA-WPCF, 1998) (plasma emission spectroscopy). To evaluate the total metal concentration, samples were diluted to 50:50 (v/v) using 4 N HNO_3 to ensure total solubility of the metal.

Turbidity was measured using a 115 Velp Scientifica turbidimeter (measuring accuracy: ±2%) according to a standard method described in the literature (APHA-AWWA-WPCF, 1998). This measurement was carried out after sedimentation (30 min without agitation, typical settle time in reclamation of wastewaters) and filtration (0.45 μm filter) of the samples.

Nitrogen and chloride inorganic anions (NO_3^-, NO_2^-, Cl^-, ClO^-, ClO_2^-, ClO_3^-, ClO_4^-) were measured by ion chromatography using a Shimadzu LC-20A equipped with a Shodex IC I-524A column; mobile phase, 2.5 mM phthalic acid at pH 4.0; flow rate, 1.0 ml min^{-1} (concentration accuracy: ±0.5%). The peak corresponding to hypochlorite interferes with that of chloride; therefore, the determination of hypochlorite was carried out by titration with 0.001 M As_2O_3 in 2.0 M NaOH (Wilpert, 1957; Freytag, 1959). This method consists of a redox determination to selectively quantify the hypochlorite concentration, specifically, it is based on the redox reaction between the hypochlorite and arsenite. Hypochlorite is reduced to chloride by the continuous addition of arsenite whereas this last one is oxidized to arsenate. The pretreatment of the samples consists of the addition of 2 ml of 2.0 M NaOH in order to increase the pH.

The same ion chromatography equipment (Shodex IC YK-421 column; mobile phase, 5.0 mM tartaric, 1.0 mM dipicolinic acid and 24.3 mM boric acid; flow rate, 1.0 ml min^{-1}) was used to measure the nitrogen inorganic cation (NH_4^+). Inorganic chloramines were measured following the DPD standard method described in the literature (APHA-AWWA-WPCF, 1998).

The faecal coliforms from wastewaters were estimated using the most probable number (MPN) technique (APHA-AWWA-WPCF, 1998) (confidence level: 95%). Microorganism counts were carried out by the multiple-tube-fermentation technique (24 h of incubation at 44 °C) using 5 tubes at each dilution (1:10, 1:100, and 1:1000).

The presence of trihalomethanes (THMs) was evaluated by gas chromatography (detention limit <0.2 ppb) using a SPB 10 column (30 m × 0.25 mm; macroporous particles with 0.25 μm diameter). Injection volume was set to 1 μL. Chloroform was