Electrocoagulation as a key technique in the integrated urban water cycle – A case study in the centre of Spain

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Abstract

Electrocoagulation (EC) presents operational advantages over chemical coagulation, including no requirement for the addition of chemicals. This work compares the performance of electrocoagulation in two stages of the urban water cycle: drinking water production and wastewater regeneration. A case study focussed on the treatment of actual effluents from different locations in the centre of Spain is performed. It was observed that electrocoagulation with Al anodes is an efficient technique to remove turbidity and total organic carbon (TOC) from surface water and to deplete turbidity and *Escherichia coli* (*E. coli*) from urban treated wastewater (taken from the outlet of an urban wastewater treatment facility, WWTF). Although electrocoagulation can be applied to the treatment of both effluents, the technique is more efficient in the case of the production of drinking water from surface sources. This behaviour is related to the nature of the natural organic matters (NOM) present in both effluents.

Keywords

Wastewater treatment, drinking water, electrocoagulation

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1. Introduction

The use of electrochemical technologies for water treatment has attracted increasing interest from the scientific community within the last decades (Brillas and Martínez-Huitle 2015). In the urban water cycle, electrochemical technology has been applied to the production of drinking water (Mezule *et al.* 2015) and, more recently, to the reclamation of previously treated urban wastewaters (Cotillas *et al.* 2015, Liu *et al.* 2015). Regarding secondary treatment of urban wastewater, biological processes are the most common treatment technologies due to the high biodegradability of these effluents.

Among the electrochemical technologies, electrocoagulation stands out due to its high efficiency, low operation cost and advantages with respect to conventional coagulation (e.g. the buffering effect of electrode reactions) (Sahu *et al.* 2014). For the case of urban water treatment by electrocoagulation (either drinking or wastewater), this technique has been recently applied (among other applications) to the removal of metal ions (Mouedhen *et al.* 2008), phosphates (Attour *et al.* 2014), greywater treatment for reuse (Church *et al.* 2015), *E. coli* depletion (Ndjomgoue-Yossa *et al.* 2015) and emerging organic pollutants (Govindaraj and Pattabhi 2015). This technique is based on the electrodisolution of a sacrificial anode (aluminium or iron) by the application of an electric field. The result is the generation of coagulant species that contribute to the removal of turbidity or organic matter (Cotillas *et al.* 2014). In addition, electrocoagulation does not require the addition of chemicals in contrast to the conventional coagulation process and, therefore, different physical-chemical parameters are not modified (e.g., pH and conductivity)

This work presents a case study that compares the performance of this emerging electrochemical technology in two different stages of the urban water cycle: drinking

water production (from surface water) and wastewater regeneration. In both cases, target effluents are actual water sources obtained from different locations in the centre of Spain. In the first stage, the performance of EC as a technique for the removal of organic matter and turbidity from surface water was assessed. These parameters should be near-zero according to the Spanish regulations for drinking water (R.D. 140/2003). Next, the treatment of wastewater taken from the outlet of a wastewater treatment facility (WWTF) by EC was performed, and the results were compared to those observed for the production of drinking water. In this case, the concentration of *E. coli* was monitored because it is an indicative of faecal pollution and it is included in the Spanish Legislation for wastewater reuse (R.D. 1620/2007).

2. Experimental

2.1 Analytical techniques

A 115 Velp Scientific turbidimeter was used to determine the turbidity by means of a standard method (APHA-AWWA-WPCF 1998). The most probable number (MPN) technique was used to estimate the population of faecal coliforms (APHA-AWWA-WPCF 1998). Microorganism counts were conducted 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).

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, was used to determine the inorganic anions (Cl⁻, ClO⁻, ClO₃⁻ and ClO₄⁻) concentrations. For the determination of hypochlorite interference with chloride hypochlorite was measured by titration with

As₂O₃ in 2.0 M NaOH. The DPD colorimetric standard method (APHA-AWWA-WPCF 1998) was used to determine the concentration of chloramines in the wastewater.

2.2 Experimental setup

The electrocoagulation process was conducted in a bench-scale, single-compartment, electrochemical reactor equipped with an aluminium (Al) plate as the anode and a stainless steel (SS) plate as the cathode (included as supplementary material, Sup. 1). Both electrodes were circular, with a diameter of 94 mm (anode area of 69.4 cm²). The electrode gap between the anode and cathode was 6 mm, and the internal volume of the electrocoagulation cell was 60 ml. A power supply (0-30V, 0-10A, Delta Electronika ES030-10) was used to apply current in the experiments.

Drinking water and wastewater were stored in a glass tank. Both systems work in total recirculation mode, with a peristaltic pump (JP Selecta Percom N-M328) continuously recycling the target water.

2.3. Experimental procedure

Surface water was collected at the entrance of the drinking water treatment plant (DWTP) of Granátula de Calatrava (Ciudad Real, Spain). The influent of this DWTP comes from a natural reservoir (Vega del Jabalón) located at the province of Ciudad Real (38°46'21.4"N 3°46'59.0"W, centre of Spain). Fresh actual wastewater was collected daily at the outlet of the secondary clarifiers of the WWTF of Ciudad Real (38°57'57.8"N 4°00'37.6"W). Bench-scale electrocoagulation tests of 4 dm³ of drinking water and wastewater were conducted under galvanostatic conditions. The evolution of the tests is expressed with respect to the applied electric charge (Q, Ah dm⁻³) and not vs. time.

1

2 **3. Results and discussion**

3 In the first part of the work, the treatment of actual surface waters was performed.

4 Figure 1 shows the evolution of the turbidity and TOC with applied electric charge for
5 electrocoagulation with Al anodes at a current density (j) of 50 A m^{-2} .

6 It is possible to completely remove the initial turbidity of the effluent by
7 electrocoagulation with applied electrical charges lower than 0.2 A h dm^{-3} . In addition,
8 TOC is also partially removed throughout the process, whereas the pH is maintained
9 around the initial value of 8 (data not shown). The removal of turbidity (Llanos *et al.*
10 2014a) and dissolved organic matter (Ben-Sasson *et al.* 2013) has been previously
11 reported, so the results presented here confirm the potential of electrocoagulation in the
12 treatment of actual effluents and the depuration of drinking water. Furthermore, the
13 removal of TOC is a matter of great importance, mainly to prevent the subsequent
14 formation of trihalomethanes (THMs) in the latter disinfection processes (Ulu *et al.*
15 2015). The invariability of the pH (when the initial pH is slightly basic) has been
16 observed by other authors (Mouedhen *et al.* 2008) and can be explained by the effect of
17 alkalizing phenomena (e.g., hydrogen evolution) offsetting acidifying phenomena (e.g.,
18 precipitation of Al(OH)_3).

19 Figure 2 shows the influence of the current density in the removal of turbidity. The
20 efficiency of turbidity removal increases with the applied current density for values
21 between 10 A m^{-2} and 50 A m^{-2} . At lower current densities (10 A m^{-2}) it is not possible
22 to remove turbidity due to the probable formation of a passive layer on the Al electrode
23 (Mouedhen *et al.* 2008). For current densities higher than 10 A m^{-2} , evolution of TOC
24 and pH (data not shown) followed the same trend as described for a current density of
25 50 A m^{-2} .

Next, the treatment of treated urban wastewater by electrocoagulation was studied. In this case, both the evolution of turbidity and *E. coli* was monitored. Figure 3 shows the evolution of these two parameters with the applied electric charge for the electrocoagulation of actual wastewaters with Al electrodes, working at the same current density as for the treatment of surface water (50 A m^{-2}).

The turbidity decreased with applied electric charge, being completely removed at a value of 0.125 Ah dm^{-3} . As was the case for the drinking water treatment, the removal of turbidity is directly linked to the production of soluble and non-soluble aluminium species that destabilize colloids or trap them into the growing flocs. The removal of *E. coli* can be achieved either by trapping microorganisms in the formed flocs or by the attack of *E. coli* by the formed disinfectant species, that is, oxidants formed from the oxidation of ions naturally present in the target effluent (Ricordel *et al.* 2013). To evaluate the role of these oxidants, the evolution of free and combined chlorine species with respect to the applied electric charge is presented as supplementary material (Figure sup. 2). Hypochlorite is the main chlorine derivative formed by the electrooxidation of chloride (Eqs. 1-3).

The evolution of the hypochlorite concentration follows the typical profile of a reaction intermediate, with an initial increase and a further decrease after an applied electric charge of 0.1 A h dm^{-3} . The concentration of chloramines increases when the hypochlorite concentration decreases, which confirms that the formation of chloramines is one of the main reactions causing the decrease in the concentration of hypochlorite (Eqs. 4-6).

4 The formation of chloramines is a desired side reaction due to the lower oxidative
5 power of combined chlorine compared to other disinfectants, preventing the formation
6 of organochlorinated species, which are harmful to human health (Lyon *et al.* 2012).

7 Figure 4 shows the influence of the current density on the removal of turbidity. The
8 initial efficiency of the process is similar, regardless of the value of the current density
9 applied. Nevertheless, there is a recalcitrant value of turbidity that cannot be removed if
10 the current density is lower than 50 A m⁻².

11 If these results are compared to those obtained in the treatment of drinking water,
12 several differences can be noted. It is necessary to apply a minimum current density to
13 begin the process of turbidity removal for drinking water and not for the regeneration of
14 treated wastewaters. Once the process is initialized, it is much more efficient (lower
15 required electric charge to obtain the same removal of turbidity) when the
16 electrocoagulation process is used to treat drinking water.

17 To explain these tendencies, it is necessary to consider the composition of both
18 effluents. The dissolution of Al electrodes can be hindered by the formation of passive
19 layers that are formed on the Al surface. The presence of small ions, such as chloride,
20 enhances the dissolution of this passive layer and, thus, increases the efficiency of the
21 dissolution of the anode. The concentration of chloride in drinking water (100 mg dm⁻³
22 in this study) was less than half the concentration of chloride in the treated water taken
23 from the WWTF (220 mg dm⁻³). This lower chloride concentration may explain why it

1 is necessary to apply a minimum current density to achieve efficient dissolution of the
2 anode and to initialize the removal of turbidity.

3 Second, the removal of NOM (natural organic matter) and turbidity by coagulation with
4 Al is more efficient for the removal of the hydrophobic and high molecular weight
5 fraction of NOM (Soh *et al.* 2008). In a recent work, it was shown that the organic
6 matter of treated wastewater is mainly composed of low molecular weight hydrophilic
7 organic matter (Llanos *et al.* 2014b). In contrast, the NOM present in natural surface
8 waters is generally high molecular weight and hydrophobic (hydrophobic humic acids
9 and fulvic acids) (Cho *et al.* 2000). This dissimilar composition explains the higher
10 efficiency in the removal of turbidity from drinking water, in which the NOM is highly
11 hydrophobic and, thus, easily removed by coagulation.

12 Finally, the results of this research open the door to the scale-up of the
13 electrocoagulation process for the treatment of drinking and waste water with higher
14 values of TOC, turbidity and microorganisms. Prior to process industrialization, it is
15 necessary to evaluate the energy consumption and costs of electrocoagulation. In this
16 context, several studies have evaluated the costs of this process for the treatment of
17 different types of wastewater, obtaining an energy cost of approximately 0.10 € m^{-3}
18 (Lacasa *et al.* 2013, Cotillas *et al.* 2014).

Conclusions

From the results presented in this work, the following conclusions can be drawn:

- Electrocoagulation with Al anodes is an efficient technique to remove turbidity and TOC from surface water and to decrease the turbidity and *E. coli* of treated urban wastewaters.
- It is necessary to apply a minimum current density to perform the process of turbidity removal for drinking water but not for the regeneration of treated wastewaters. Nevertheless, the process is more efficient when the electrocoagulation process is used to treat drinking water.
- The dissimilar behaviour of electrocoagulation in the treatment of different water sources in the urban water cycle (surface water and treated wastewater) is related to the dissimilar composition of the effluents, mainly the chloride concentration and the nature of the NOM.

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