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Electrochemical coagulation of treated wastewaters for reuse

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ABSTRACT

In this work, the disinfection capability of electrocoagulation with iron and aluminium electrodes is studied. To do this, two different effluents of conventional municipal wastewater treatment plants (WWTP) are treated. Results show that electrocoagulation is able to reduce satisfactorily chemical oxygen demand, turbidity and nutrients and it allows increasing significantly the water quality. Likewise, it does not affect conductivity (just the contrary conventional coagulation–flocculation treatment) and helps to buffer pH in a value around 8. Precipitation, floc enmeshment and adsorption seem to be the primary mechanisms in this process. Regarding the variation of faecal coliforms, this technique behaves as a disinfection technology and allows increasing the efficiencies of WWTP processes in a very cheaper way. Energy consumption lower than 0.2 kWh m^{-3} is required to attain standard disinfection levels.

Keywords: Electrocoagulation; Disinfection; Treated wastewater

1. Introduction

The search of new sources of water has become one of the major challenges for the near future. In this context, regeneration of urban wastewater is one of the better possibilities [1], and nowadays, countries like Spain (in which the lack of water resources is a serious problem) are making a great effort in the development of treated wastewaters regeneration plants (WRP) to provide water from the effluents of municipal wastewater treatment plants (WWTP). In these cases, depending on the further uses of the effluent (irrigation, repletion of aquifers, etc.), its quality should be further improved in a single tertiary treatment or in a more complex WRP. In many cases, membrane technologies complement this flow scheme, reducing the conductivity of the regenerated water [2]. Some significant problems for direct reuse of reclaimed wastewater are, according to technical literature, the high content of suspended solids, oils, phenols, etc. However, the use of these waters for certain purposes might be excluded or at least hampered because of the possible pathogenicity for human beings. Thus, *Escherichia coli* or other microbiological parameters including faecal coliform and total coliform becomes very important parameters for the assessment of regeneration technologies.

In general, a satisfactory reduction of pathogens is only possible by disinfection procedures such as different chlorination technologies (chlorine, hypochlorite, chloramines, etc.) or ultraviolet disinfection. However, some interesting improvements have been obtained recently by MBR systems [3,4].

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Wastewater disinfection levels are determined by standards and recommendations that are specific to each country and region. In this context, Spanish legislation related to regeneration of water [5] requires the disinfection of the regenerated wastewater (primarily in terms of removal of nematodes eggs and faecal coliforms) to allow its safe reuse or recycle. The disinfection targets stipulated vary according to application.

In this context, in literature [6–9] it has been recently proposed the use of electrochemical techniques to disinfect water. It is proved that electrolysis technology is able to remove efficiently *E. coli* and other bacteria by the action of the bactericide oxidants (mainly chlorine species) electrogenerated on the reaction media. Unfortunately, there is lack of information about the availability of electrocoagulation for the disinfection of treated wastewaters.

The electrochemical coagulation involves the *in situ* production of coagulants by electrolytic oxidation of an appropriate anode material (carbon steel or aluminium sheets) [10]. This technique differs from conventional coagulation in the way of dosing coagulant reagents in which the addition of hydrolyzing metal salts (of iron or aluminium) as coagulant reagents is carried out by salt solution dosing [11]. When compared with the conventional coagulation process, the electrocoagulation reports several advantages: the simplicity of the equipment required, versatility, safety and easy automation of the process. Likewise, this process results in high energy efficiency, selectivity and cost effectiveness, as well as a lower amount of precipitate or sludge.

With this background, the goal of this work is to check the possibility of use of electrocoagulation as an integrated treatment to get reuse water from the effluents of conventional municipal WWTP. Some conclusions were forwarded (for other municipal wastewater effluents) in a previous paper of our group [12], but here some very interesting results confirming the observations shown in this previous work, and other important advantages of the use of electrocoagulation are shown, in particular the significant effect of electrocoagulation on disinfection. To evaluate the effect of electrocoagulation on the treatment of the effluent of municipal WWTP, two different effluents were treated and both iron and aluminium were studied as electrode material.

2. Experimental

2.1. Analytical procedure

The chemical oxygen demand (COD) was used to monitor the organic load of the wastes. This parameter and the total nitrogen and phosphorus were determined using a HACH DR200 analyzer. The zeta potentials and turbidity were measured to the clarified liquid using a Zetasizer Nano ZS (Malvern, UK) and a 115 VELP SCIENTIFICA Turbidimeter, respectively. Measurements of pH and conductivity were carried out with an InoLab WTW pH-meter and a GLP 31 Crison conductimeter, respectively. The aluminium and iron concentrations were determined by dilution 50:50 v/v of samples with HNO_3 4 N, and measured using an Inductively Coupled Plasma LIBERTY SEQUENTIAL VARIAN according to a standard method [19] (Plasma Emission Spectroscopy).

The faecal coliforms of the wastewaters were estimated using the most probable number technique [13]. The micro-organism counts were carried out by the multiple-tube-fermentation technique (24 h of incubation at 44°C) using five tubes in each dilution (1:10, 1:100, 1:1,000). The modified Bailenger method [14] was used to estimate nematodes eggs. The number of eggs per litre are calculated from the Eq. (1), where N is the number of eggs per litre of sample, A is the number of eggs counted in the McMaster slide, X is the volume of the final product (ml), P is the volume of the McMaster slide (0.3 ml) and V is the original sample volume (l).

$$N = AX/PV \tag{1}$$

2.2. Experimental devices

The electrocoagulation assays have been carried out in a bench-scale plant with a single compartment electrochemical flow cell working under a batch-operation mode (Fig. 1). Aluminium and iron electrodes were used as the anode and cathode. Both electrodes were square in shape (100 mm side), each with a geometric area of 100 cm^2 and with an electrode gap of 9 mm. The electrical current was applied using a DC power supply FA-376 Promax. The current flowing through the cell was measured with a 2000 digital multimeter Keithley. The wastewater (2 dm^3) was stored in a glass tank stirred by an overhead stainless steel rod stirrer Heidolph RZR 2041. The wastewater was circulated through the electrolytic cell by means of a peristaltic pump (50 $dm^3\,h^{-1}$). Before each experiment, the electrodes were treated with a solution of HCl 1.30 M in order to avoid any effect due to the different prehistory of the electrodes.

2.3. Wastewater characterization

In this work, actual effluents coming from the biological stage of two different WWTP were used. The

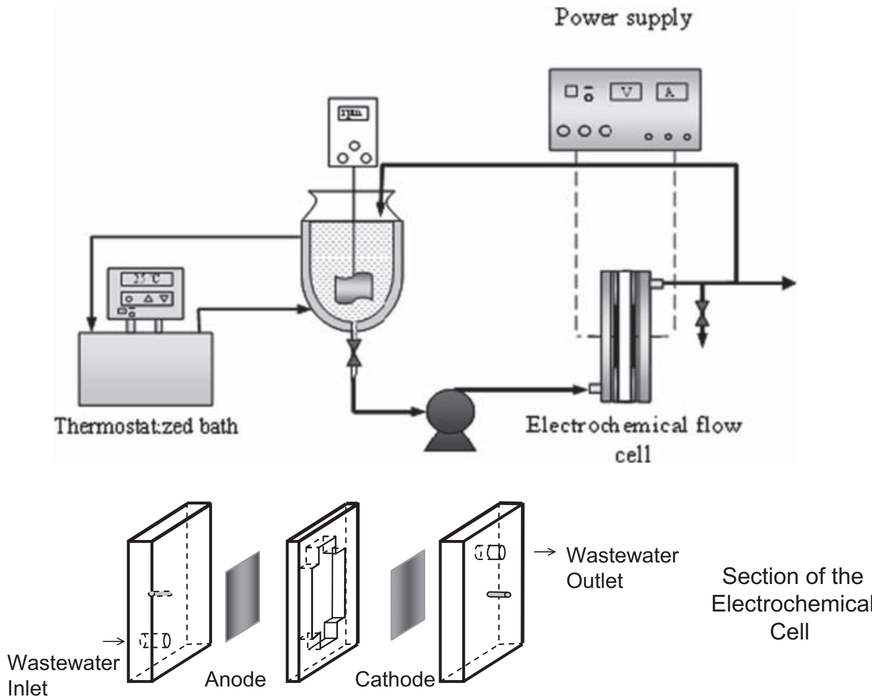

Fig. 1. Layout of the experimental set up and the electrochemical flow cell.

WWTPs treat the wastewater of two small towns (20,000 and 30,000 p.e.) located in the centre of Spain. In both cases, the influent is domestic wastewater without a significant industrial contribution. Their main characteristics of the samples used in this study are shown in Table 1. The two WWTP were selected because of the different characteristic of the effluents expressed in terms of COD, turbidity, nutrients and micro-organisms.

3. Results and discussion

3.1. Electrochemical dosing of reagents

Fig. 2 shows the variation with the electrical charge applied to the aluminium and iron concentrations electrogenerated during the electrocoagulation process of a treated wastewater. It is important to point that unlike other environmental remediation electrochemical processes, no salts are added in this case to increase ionic conductivity of water, but just the raw water is treated as it is. This means that cell voltage can be higher than expected in other works about electrocoagulation [15,16] (in which salts are added to wastewaters in order to increase ionic conductivity), for the same current density applied. However, it has been done for practical reasons, as the dosing of salts to improve the performance of the electrochemical process would make this process less attractive to be used in the treatment of the effluent of municipal WWTP, because of the associated decrease in the quality due to the increase in the salinity. Anyway, conductivity of the effluents of WWTP was used to be enough to achieve good results in electrocoagulation processes as it is going to be discussed during this work. For the current density applied ($10\,\mathrm{A\,m^{-2}}$), the cell voltage was maintained constant during the experiments in around 3.0 V for both electrode material. No significant changes in the cell potential were observed during the treatments.

Concerning Fig. 2, as it can be seen, the electrodissolved metal concentration increases linearly with the electrical charge for both aluminium and iron, and the experimental values are greater than the values calculated if the process is considered to be purely

Table 1
Characterization of the wastewaters used in this work

Type of the raw wastewater (before treatment)	$WWTP_1$	$WWTP_2$
COD/$\mathrm{mg\,O_2\,dm^{-3}}$	43	22
Total nitrogen/$\mathrm{mg\,N_T\,dm^{-3}}$	6.0	11.9
Total phosphorus/$\mathrm{mg\,P_T\,dm^{-3}}$	1.2	0.8
Conductivity/$\mathrm{\mu S\,cm^{-1}}$	291	195
pH	7.4	7.7
Turbidity/NTU	1.3	2.1
E. coli/CFU/100 $\mathrm{dm^{-3}}$	9,550	18,000

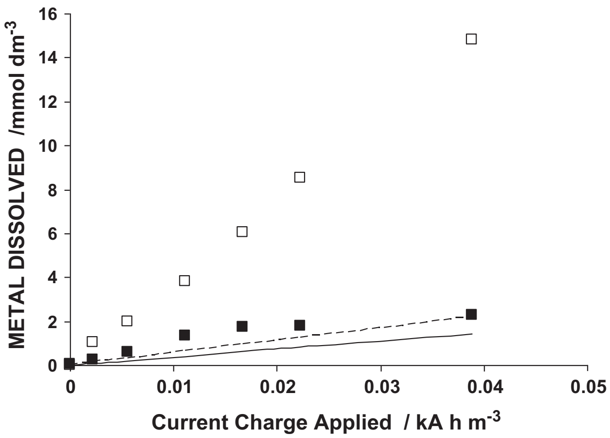

Fig. 2. Variation of aluminium (□) and iron (■) concentration electrogenerated in the electrochemical processes with the electrical charge passed in batch mode operation: Operation conditions: T: 25℃; flowrate: $50\,dm^3\,h^{-1}$; current density: $10\,A\,m^{-2}$; cell voltage: 3 V. Straight lines: Theoretical values predicted by Faraday's Law.

electrochemical (according to Faraday's Law). In this point, it can be observed that the rate of generation of aluminium ($1.15\,g\,min^{-1}\,m^{-2}$) is higher than iron generation rate ($0.37\,g\,min^{-1}\,m^{-2}$). This fact is related to the chemical dissolution of the electrode surfaces that in the case of aluminium sheets is significantly favoured at the alkaline pH of the cathode surface [16].

3.2. Effect on colloids and COD

Fig. 3 shows the changes in the COD and turbidity during the electrocoagulation of the two actual effluents of municipal WWTP using both iron and aluminium as electrodic material. As it can be observed, both, COD and turbidity decrease significantly for both wastewaters studied during the treatment. The decrease is not the same for the two wastewaters, although for the same wastewater, both coagulant reagents got the same results for large doses added. This is important because it states that electrocoagulation is a robust and proper technology to improve the quality of the effluent of a municipal WWTP, but results depend strongly on the particular influent treated, and although the effluents of WWTP have the similar characteristics, it is difficult to estimate the particular efficiency of these technologies.

Two different stages can be discerned during the process. For small current charges applied, removal of COD and turbidity seems to be very efficient, with a high removal rate (Q is directly related to time). Then, the efficiency decreases down to a constant value

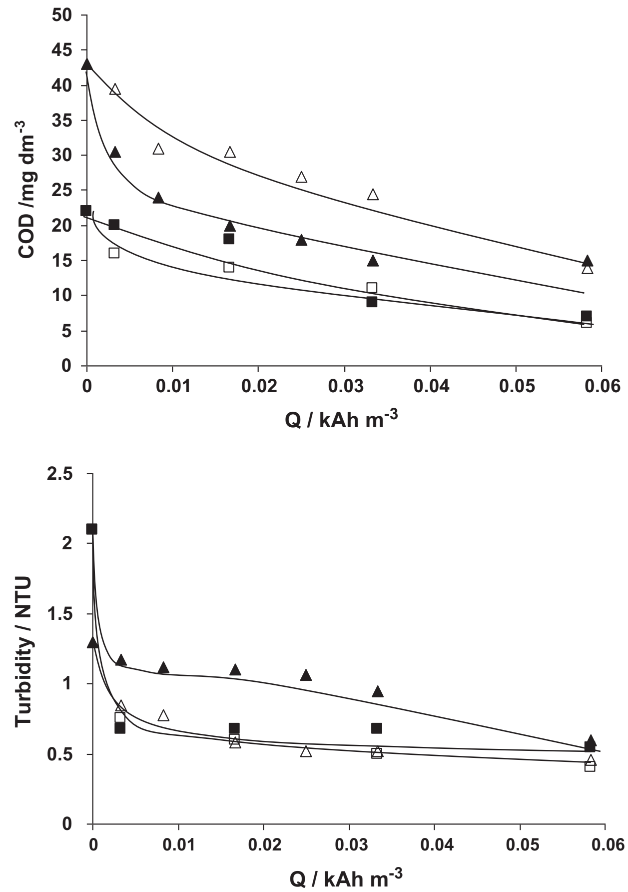

Fig. 3. Influence of the current applied on the removal of COD and turbidity (after 40 μm filtration) $WWTP_1$ ▲ iron Δ aluminium; $WWTP_2$ ■ iron □ aluminium. Operation conditions: T: 25℃; flowrate: $50\,dm^3\,h^{-1}$; Current density: $10\,A\,m^{-2}$.

(linear decrease of both COD and turbidity with charge). This may be explained in terms of the removal of particulate species during the first stage, followed by a less efficient, but still effective, floc enmeshment and adsorption mechanisms, during a second stage, in which the concentration of the coagulant dosed becomes very significant [17,18]. During the different processes, not a clear influence of the anode material can be observed suggesting that for the same wastewater it is more efficient with iron in terms of dosing as it gets the same results for a smaller dose (as state in Fig. 1 iron is produced in smaller amounts for the same current applied).

Fig. 4 shows the influence of the current applied on the z-potential. It can be observed that in every case z-potential increased with the applied current but it is always in negative values. This means that not charge neutralization but floc enmeshment and adsorption of soluble COD are the primary mechanisms in this process.

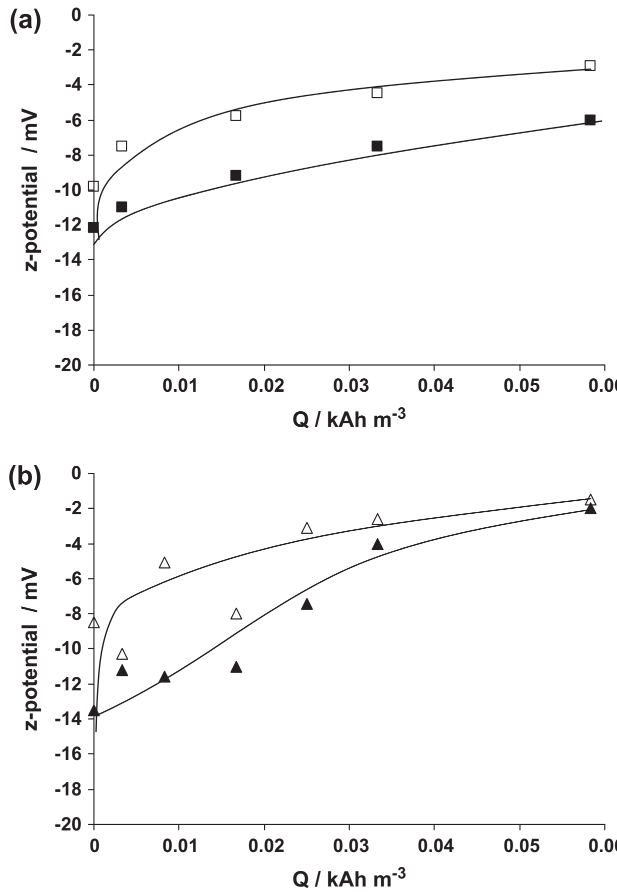

Fig. 4. Changes in the z-potential during the eletrocoagulation treatment. (a) $WWTP_1$ ▲ iron Δ aluminium; (b) $WWTP_2$ ■ iron □ aluminium. Operation conditions: T: 25℃; flowrate: 50 $dm^3 h^{-1}$; current density: 10 $A m^{-2}$.

3.3. Effect on nutrient removal

Fig. 5 shows the changes in the nutrient concentration during metal dosage electrochemically. As it can be observed phosphorus is removed very efficiently. In contrast, the removal of nitrogen is smaller, especially in the treatment of the effluent namely $WWTP_2$, but it is important to state that it is very significant in every case. In the case of phosphorus, and according to literature [19–23], the combined effect of precipitation and of coagulation (by both adsorption and enmeshment) allows explaining the good results observed. In the case of nitrogen, although most nitrogen is present in the form of nitrates (effluent of a secondary treatment) some recent studies demonstrate the very effective role of electrocoagulation in the removal of nitrates by adsorption of these species into the growing metal hydroxide precipitate formed during the electrochemical dosing [24–26], although in several studies, the nature of the salts contained in the treated water was found to influence strongly on the results. This may help to explain the results better found for the wastewater named $WWTP_1$ [27,28] as compared to the other wastewater treatment. When the same dosage of iron and aluminium was provided to the treated wastewater same results were obtained.

3.4. Effect on pH and Conductivity

Fig. 6 shows the changes in the conductivity and pH during the electrocoagulation or the two WWTP effluents with iron and aluminium. It is important remark that electrocoagulation process does not affect conductivity (just the contrary that it can be found in a conventional coagulation–flocculation treatment due to the addition of large amount of salts) and that it helps to buffer pH in a value around 8.0, which is clearly inside the recommended limits fixed by the environmental authorities for water reuse. Thus, there

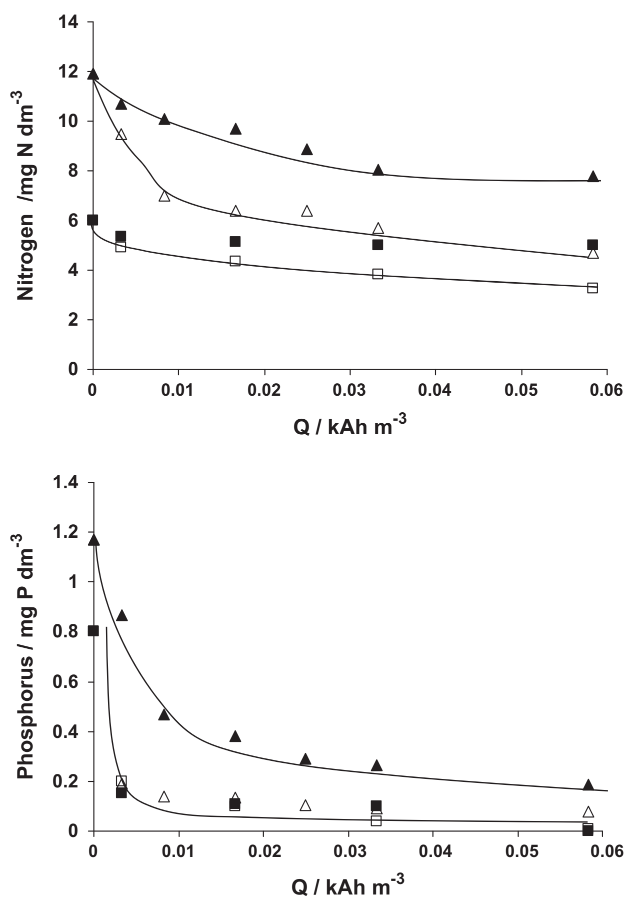

Fig. 5. Influence of the reagent dosed on the removal of nutrients (after 40 μm filtration) by electrocoagulation. $WWTP_1$ ▲ iron Δ aluminium; $WWTP_2$ ■ iron □ aluminium. Operation conditions: T: 25℃; flowrate: 50 $dm^3 h^{-1}$; current density: 10 $A m^{-2}$.

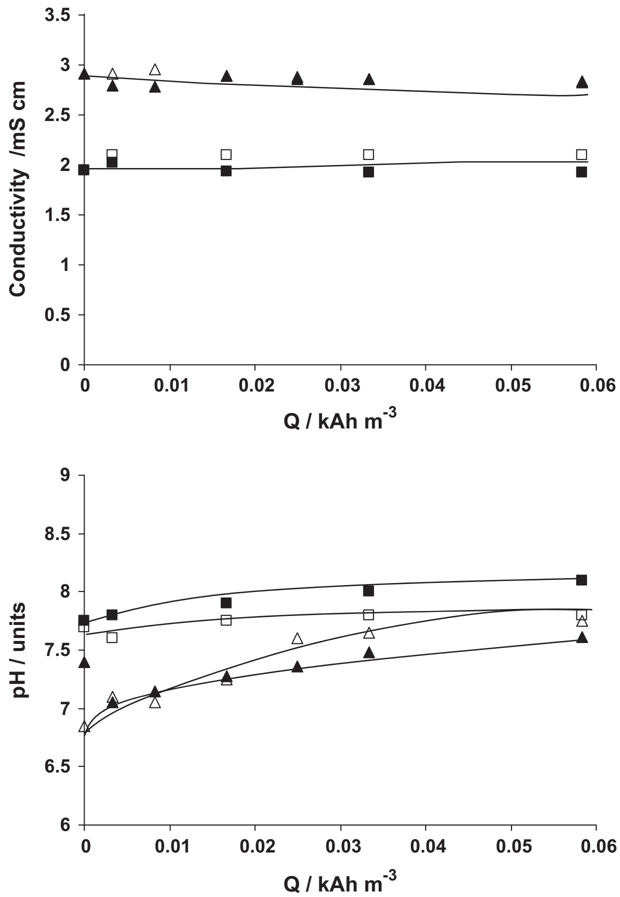

Fig. 6. Changes in the pH and conductivity (after 40 μm filtration) during the electrocoagulation process. $WWTP_1$ ▲ iron Δ aluminium; $WWTP_2$ ■ iron □ aluminium Operation conditions: *T*: 25℃; flowrate: $50\,dm^3\,h^{-1}$; current density: $10\,A\,m^{-2}$.

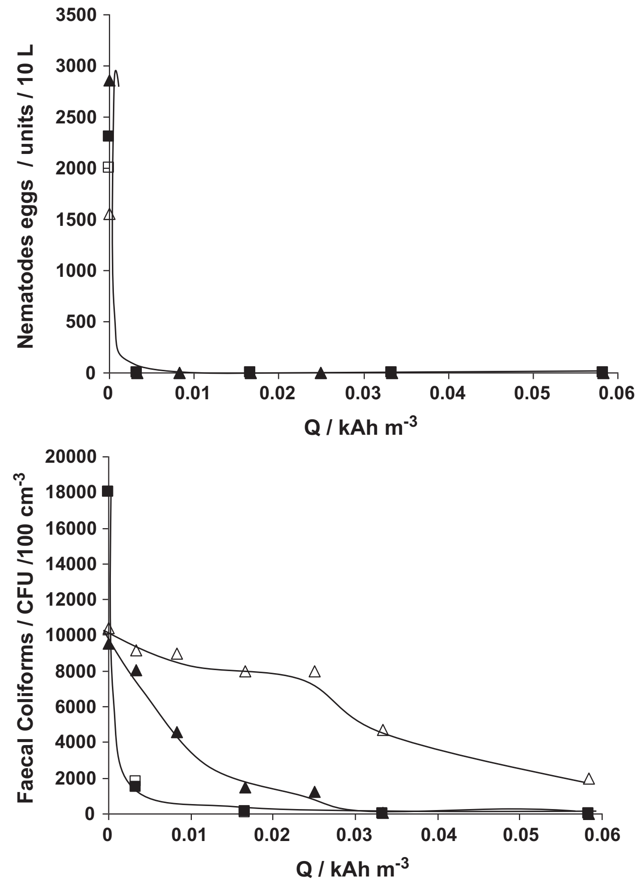

Fig. 7. Variation of the concentration of nematodes eggs and faecal coliforms during the electrocoagulation process of two secondarily treated wastewaters using iron and aluminium electrodes. ▲ $WWTP_1$, iron; Δ $WWTP_1$, aluminium; ■ $WWTP_2$, iron; □ $WWTP_2$, aluminium. Operation conditions: *T*: 25℃; flowrate: $50\,dm^3\,h^{-1}$; current density: $10\,A\,m^{-2}$.

is no necessity of neutralizing after treatment. This fact is a clear advantage on comparison to conventional coagulation processes in which the addition of metal salts leads to a decrease in the pH of the reaction media [29].

Therefore, this technique shows a good effect in the removal of COD and nutrients and it allows increasing significantly the water quality and the efficiencies of WWTP processes in a very cheaper way avoiding some of the drawbacks associated with coagulation (need of neutralization and increase of salinity).

3.5. Effect on the population of micro-organisms

In order to check the feasibility of this treatment as disinfection technique, the concentration of faecal coliforms and nematodes eggs haven been monitored during the electrocoagulation process (Fig. 7). In this point it is important to point out that to be effective, electrocoagulation should always be followed by a filtration system. Separation of solids in an electrocoagulation cell is carried out by a very effective combination of sedimentation and flotation processes. This later process is assisted by bubbles formed during electrolysis of water (main side reactions of the electrocoagulation process). Because of this combined outcome, effluents of an electrocoagulation process use to have very low concentrations of solids. However, the efficiency is not 100% and a filtration unit is required when a very high-quality effluent is required. This is the case of removal of nematode eggs. In previous experiments (not shown in this manuscript) it was obtained that the raw effluent of electrocoagulation contained random concentration of nematode eggs (ranging from 0 to very low different values) and that in every case a filtration of the samples simulating a filtration unit in a treatment facility prevented this concentration. For this, aqueous samples taken during the electrocoagulation were filtered (using filters of 40 μm). This fact can justify the rapid decrease observed in the nematodes eggs.

Regarding the variation of faecal coliforms, it can be observed that electrocoagulation behaves as a disinfection technology and that applied current charges lower than 0.06 kAh m^{-3} are required to attain the complete removal of faecal coliforms from water. The trend and energy requirement depends on both types of treated wastewater and of electrode material. Thus, the disinfection of the effluent $WWTP_2$ is attained rapidly and very efficiently (taking into account the cell voltage, specific energy lower than 0.05 kWh m^{-3}). On the other hand, the disinfection of the effluent $WWTP_1$ requires the addition of higher amount of coagulant and thus the energy consumption is higher (0.1 kWh m^{-3} in the case of using iron and 0.2 kWh m^{-3} in the case of using aluminium).

The disinfection observed is very interesting although not expected. In order to clarify the formation of potential disinfection reagents, synthetic water consisting of a solution of 1.6 g dm^{-3} of sodium chloride was electrolyzed in the same electrochemical cell, using iron or aluminium electrodes. Concentration of hypochlorite obtained is shown in Fig. 8. As it can be observed, a large concentration of this disinfection reagent is produced in the system. It is also important to state that no chlorates or perchlorates were found in the reaction media. This means that disinfection of micro-organisms is not due to the enmeshment of them in the growing flocs of metal hydroxide, but to the action of electrochemically produced hypochlorite.

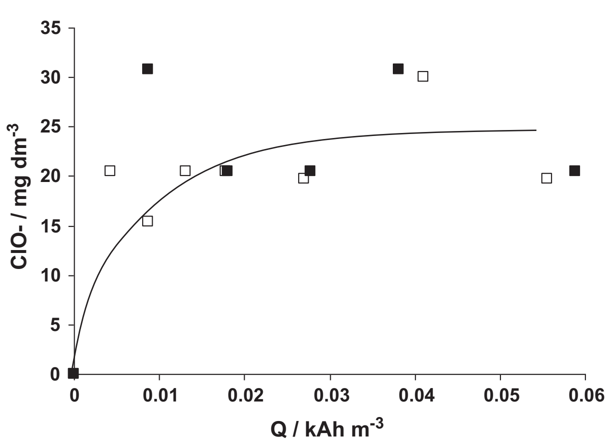

Fig. 8. Production of hypoclorite during the electrolyses of a solution 1.6 mg dm^{-3} NaCl in the electrocoagulation cells. ■ iron; □ aluminium. Operation conditions: *T*: 25°C; flowrate: 50 dm^3 h^{-1}; current density: 10 A m^{-2}.

By comparing turbidity and removal of faecal coliforms, it is interesting to see that removal of faecal coliforms does not fit the turbidity decreases. This means that this process is not only related to the removal of micro-organisms by enmeshment, but it has to be promoted by the formation of disinfectant species from the salts contained in the reaction media [12].

On base of these results and taking into account the present unitary electricity cost for industrial use in Spain (0.10 € kWh^{-1}), energy cost of electrocoagulation as disinfection technique is around 0.02 € m^{-3}. This value is significantly lower than expected and is within a good range to study scale-up of the process.

4. Conclusions

Electrocoagulation with iron or aluminium shows good effect in the removal of COD, turbidity and nutrients and it allows increasing significantly the water quality and the efficiencies of WWTP processes in a very cheaper way avoiding some of the drawbacks associated to coagulation (need of neutralization and increase of salinity). The combined effect of precipitation and of coagulation (by both adsorption and enmeshment) allows explaining the good results observed. Regarding the variation of faecal coliforms, this technique behaves as a disinfection technology. Results seem to indicate disinfection which is not only related to the removal of micro-organisms by enmeshment, but it has to be promoted by the formation of disinfectant species from the salts contained in the reaction media.

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