Electrocoagulation coupled adsorption for anaerobic wastewater post-treatment and reuse purposes

Janaína Terhorst Pizutti[a], Rita de Cassia dos Santos[b], Marcelo Hemkemeier[c], Jeferson Steffanello Piccin[a],*

[a]*Post-Graduate Program in Civil and Environmental Engineering, University of Passo Fundo, Campus I, BR 285,
Zip Code 99052–900, Passo Fundo, RS, Brazil, Phone +55(5)981212121, email: janapizutti@hotmail.com (J.T. Pizutti),
Phone +55(54)33168100, email: jefersonpiccin@upf.br (J.S. Piccin)*
[b]*Environmental Engineering, University of Passo Fundo, Campus I, BR 285, Zip Code 99052–900, Passo Fundo, RS, Brazil,
email: rita_de_cassia.85@hotmail.com (R.C. dos Santos)*
[c]*Chemical Engineering Department, University of Passo Fundo, Campus I, BR 285, Zip Code 99052–900, Passo Fundo, RS, Brazil*

ABSTRACT

Water reuse is an indispensable reality in the search for an equilibrium between water resources and water scarcity. The recovery and production of reclaimed water with a guarantee of safety of use requires advanced treatment technologies. This study proposes a conjugated system, composed of electrocoagulation followed by adsorption, for the post-treatment of UASB effluent for reuse purposes. For this purpose, initially the electrocoagulation process (EC) was optimized using response surface methodology. The optimum conditions were for a current density of 29.44 A/m² and pH 6.0, with 44.58% of COD removal and energy consumption of 31.1 kWh per kg of COD removed. The EC effluent was treated by adsorption on a fixed bed column of activated carbon. The conjugated system provided the complete removal of COD and BOD, and significant removals of turbidity (96.5%), phosphorus (97.5%) and total and thermotolerant coliforms. The quality of the final treated water reached parameters for reuse according to EPA.

Keywords: Activated carbon; Advanced treatment; Electrolysis; Fixed bed adsorption; Wastewater; Water reclaimed; Water reuse

1. Introduction

Conventional wastewater treatment processes, usually composed of physical processes including screening and sedimentation, physical–chemical coagulation and decantation, and biological processes, such as activated sludge and anaerobic reactors, are traditional and reliable systems. Although they are efficient for removing organic load and some micropollutants, they are not able to produce water for reuse purposes [1–3].

Wastewater post-treatment techniques, including adsorption [4–6], membrane separation [7–10], electrolysis [11–13], ozonization [14,15] and other processes, have been highlighted as advanced wastewater treatment systems for reuse. Electrocoagulation (EC) is an environmentally interesting and efficient technology to remove a variety of pollutants (organic and inorganic). Several mechanisms of removal and degradation may exist simultaneously in the electrochemical reactor [11–13,16]. It has been successfully applied in the treatment of water for reuse purposes [11–13], including post-treatment of UASB effluent [17,18], especially when there is the option of replacing energy matrices with renewable sources [19,20].

However, EC alone, as a post-treatment technique, is not able to achieve water quality levels for reuse, especially with regard to organic load, suspended solids and color. For this reason, EC has been coupled to different techniques, such as a membrane separation process [21], soni-

*Corresponding author.

cation [22], sand filtration [4], ultrasound [23] and anodic oxidation [24].

Adsorption represents a solution dedicated to the elimination of micro-pollutants and the polishing of wastewater [25,26] and has been applied as a complement to electrolytic systems [5,27–29]. The low energy requirement, absence of by-products with greater toxicity and the capacity of regeneration of adsorbents are considerable advantages of this process [25].

In this study, a conjugated system composed of electrocoagulation followed by fixed bed adsorption was proposed as a wastewater post-treatment technique for reuse purposes. Wastewater treated by a system composed of screening, sedimentation and a biological process with an UASB reactor was used as a case study. The operating conditions of electrocoagulation were optimized. Then, the effluent from the EC process was subjected to adsorption in fixed bed systems of activated carbon. Finally, the effluent characteristics were verified as to their quality for reuse.

2. Methodology

2.1. Wastewater collection

The wastewater used as a case study was collected at a treatment plant at a university campus located at the geographical coordinates 28°13′55.00″S and 52°22′42.74″W. The university has daily traffic of around 15 thousand people. In addition to academic activities, food services and a variety of technical service activities are provided that generate wastewater, with characteristics distinct from traditional domestic sewage. The station operates with an average flow of 280 m³/d. The treatment process consists of screening, sandbox, UASB (212 m³) and activated sludge (131 m³) biological reactors, and a secondary decanter (49 m³). The wastewater collection was performed after anaerobic UASB treatment.

2.2. Wastewater characterization

The UASB effluent before and after each treatment was characterized according the Standard Methods for the Examination of Water and Wastewater [30] to: pH (method 4500-PH), chemical oxygen demand (method 5220 D), biological oxygen demand (method 5210 B), total nitrogen (method 4500-N$_{org}$ C), total phosphorus (method 4500-P), suspended solids (method 2540 D), dissolved solids (method 2540 C), nitrite (method 4500-NO$_2^-$ B), nitrate (method 4500-NO$_3^-$ B), total and thermotolerant coliforms (method 9221), Surfactants (method 5440), aluminum (method 3111D) and temperature (method 2150). Turbidity is determined using nephelometric method (method 2130 B, DM-TU turbidimeter, Digimed) Color is measured in hazen scale by Spectrophotometry (EC 2000 Pt Co, Lovibond) according to 2120 method. Table 1 show the UASB effluent characterization.

2.3. Electrocoagulation treatment

The electrolytic system was composed of a reactor with a useful volume of 2 L (ø = 165 mm and h = 190 mm) where 1.4 L of wastewater was added in each experiment. The rectangular electrodes, a carbon steel cathode and an

Table 1
Characterization of the effluent produced in the UASB reactor

Parameter	Value	Method
pH	7.02 ± 0.02	4500
COD (mg/L)	77.19 ± 0.75	5220
BOD (mg/L)	72.5 ± 7.1	5210
N-total (mg/L)	25.2 ± 0.9	4500
P-total (mg/L)	5.72 ± 0.59	4500
Suspended solids (mg/L)	84.0 ± 14.0	2540
Dissolved solids (mg/L)	240.0± 17.0	2540
Turbidity (NTU)	70.33 ± 0.58	2130
Color (hazen)	72.33 ± 1.53	2120
NO$_3^-$ (mg/L)	0.04 ± 0.01	4500
NO$_2^-$ (mg/L)	1.12 ± 0.02	4500
Thermotolerant coliforms (MPN/mL)	> 1100	9221
Total coliforms (MPN/mL)	> 1100	9221
Surfactants (mg/L)	0.44 ± 0.03	5540
Aluminum (mg/L)	<0.01	3111
Temperature (°C)	22.0 ± 0.1	2550

Mean ± standard deviation, n = 3

Table 2
Real and coded level of central composite design (CDD) for current density (J) and pH optimization on the electrocoagulation process

Variables	Levels				
	–1.414	–1	0	1	+1.414
X$_1$ (J, in A/m²)	14.4	20.0	30.0	40.0	43.2
X$_2$ (pH)	3.2	4.0	6.0	8.0	8.8

aluminum anode (with a projected area of 49.89 cm²), were arranged vertically and distally at 10 mm, according to the values reported in the literature [17,31,32]. The electrodes were supplied by electric current under galvanostatic conditions (continuous and constant current intensity) by an electrical source (New Dawer, FCC5002D). The experiments were conducted without forced stirring at 20°C for 60 min. Thereafter, the liquid phase of the effluent was filtered downward into a bed of calcined sand 10 cm in height, at an application rate of 2 cm/min [4] for the removal of suspended solids produced during EC [33].

The effect of current density (X$_1$) and the initial pH of the wastewater (X$_2$) during electrocoagulation was optimized using a central composite design based on previous work [21]. The pH of the wastewater was adjusted using HCl or NaOH (0.1 mol/L). Table 2 shows the actual and coded levels of the studied variables. The responses evaluated were the COD removal (COD rem) and specific energy consumption (w), according to Eqs. (1) and (2), respectively:

$$CODrem(\%) = \left(1 - \frac{C_f}{C_i}\right) \cdot 100 \qquad (1)$$

$$w = \frac{\int i \cdot V \cdot dt}{V_t \cdot \left(C_i - C_f\right)} \quad (2)$$

where C_i and C_f are the initial and final COD concentration (kg/m³), i is the current intensity (A), V is current tension (V) and V_t is the effluent treated volume (m³). The integral of Eq. (2) was graphically solved considering measurements of i and V at intervals of 5 min.

The effects of the studied variables (linear, quadratic and interactions) on the responses were evaluated through analysis of variance, and the response surfaces were obtained from the generated statistical model. Then, the global optimization of the variables studied was performed from Derringer's Desirability Test [34]. At the optimized condition the energy consumption ($W_{v'}$ in kWh/m³ of treated wastewater) was calculated according to Eq. (3).

$$W_v = \frac{\int i \cdot V \cdot dt}{V_i} \quad (3)$$

2.4. Adsorption experiments

After EC optimal conditions, were found adsorption experiments were carried out with the purpose of polishing the final effluent. The granular activated carbon (VETEC) was previously treated with 6 mol/L HCl at open reflux (at boiling temperature) with a volumetric ratio of 1.5:1 (HCl:activated carbon) for 60 min, washed with distilled water and dried at 60°C for 24 h. This process was carried out to remove the inorganic material present in the activated carbon, thus causing an increase in the surface area and activation of the adsorption sites [35,36]. The surface area of the activated carbon was determined at 856.4 m²/g through adsorption isotherms of N_2 (BET method). The pH of zero charge (pH_{PCZ}) was determined according Saucier et al. [35] using 20 mL of 0.050 mol/L NaCl and 50 mg of the adsorbent. The pH values of the solutions were adjusted from 1 to 10 with 0.1 mol/L of HCl or NaOH. The scanning electron microscopy (SEM - Fig. 1) is obtained using particle coating with gold, acceleration of 5 kV and magnification of 2000× (VEGA 3, Tescan). The activated carbon show pH_{PCZ} of 6.3 and a porous and uneven surface.

The adsorption system consisted of a fixed bed of 100 mm length and 25 mm diameter, fed towards the top by a peristaltic pump at a flow rate of 10 mL/min [4]. At predetermined intervals, samples of the effluent were collected, and the effluent color was monitored as an easy criterion for the observation of saturation of the adsorption bed. The adsorption effluent obtained between the color intervals of 0–5 Hazen, 5–10 Hazen and 10–15 Hazen were collected for final characterization and comparison with the original wastewater.

2.4. Options for reuse

The analysis of the possibility of reuse was performed by comparing the wastewater parameters with the reuse standards suggested by the USA Environmental Protection Agency [37], and with the literature applied for different situations [19,33,39,40].
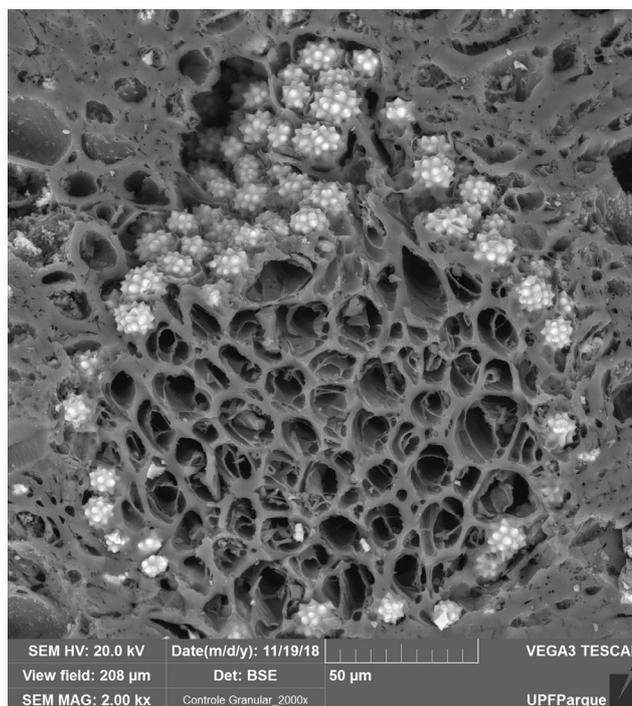

Fig. 1. SEM of activated carbon.

3. Results and discussion

3.1. Electrocoagulation process optimization

Table 3 presents the results obtained in the central composite design (CCD) experiments as a function of the studied variables. COD removals were observed between 21.7% and 48.28%, while energy consumption ranged from 11.23 kWh/kg to 55.37 kWh/kg. In general, there was an inverse relationship between these two factors, since the increase in removal causes a reduction in the specific energy consumption.

The analysis of variance (ANOVA) of the results presented in Table 3 (supplementary material) showed that for COD removal only the linear effect of pH (X_2) was not significant ($p > 0.05$). In addition, negative quadratic effects indicate a convex response surface, with the presence of optimal points. Regarding energy consumption (EC), only the linear effect of current density (X_1) and its interaction with pH (X_2) were significant. Thus, excluding non-significant effects on responses (Table S2), the statistical models of COD removal and specific energy consumption are represented by Eqs. (4) and (5), respectively. These models, as well as the response surfaces (Fig. 2), were generated for the real values of the studied variables.

$$COD\ rem(\%) = 44.58 + 5.75 \cdot X_1 - 6.84 \cdot X_1^2 - 8.19 \cdot X_2^2 + 4.69 \cdot X_1 \cdot X_2 \quad (4)$$

$$w(kWh/kg) = 32.21 + 11.20 \cdot X_1 - 8.94 \cdot X_1 \cdot X_2 \quad (5)$$

As shown in Fig. 2a, the surface concavity indicates that the COD removal has an optimized value in the study region, close to the central region. In relation to the spe-

cific energy consumption, the formation of a plateau in the region between the central point and the higher values of current density and pH is observed in Fig. 2b in the region of acid pH and low current density, where a reduction in energy consumption is observed.

In electrocoagulation with aluminum electrodes there are two principal removal mechanisms: coagulation, due to the adsorption of pollutants in the complexes of Al^{3+} formed, and the flotation of the Al^{3+} complexes with the contaminants removed, due to the hydrogen bubbles formed at the cathode.

During coagulation, aluminum ions produce hydroxide ions and hydroxides of neutral metals [17,20,26,41]. These aluminum hydroxides (coagulating agent) are responsible for increasing the pH during the removal process and for the neutralization of electrostatic charges of dispersed particles (reducing the repulsion between the particles and favoring the Van der Waals attractions), facilitating the agglomeration and separation of particles in the wastewater [16,42]. The low solubility of these hydroxides (mainly at pH 6.0–7.0) promotes the generation of sweep flakes inside the treated waste and the removal of pollutants by their fitting in these flakes [41].

Simultaneously, during electrocoagulation, the water dissociation generates H^+ ions, which reduce at the cathode to form H_2 gas. The release of this gas raises the concentration of OH^- ions and thus increases the pH of the solution. In addition, the increase in OH^- ions also enhances the generation of hydroxyl radicals (OH^*). These can cause the oxidation of organic matter to smaller intermediate compounds and, finally, to CO_2 and H_2O, causing degradation of pollutants [13]. In the case of the Al electrodes, when the wastewater pH is maintained in the range of 4 to 8, all the aluminum cations produced in the anode form the polymer species of $Al_{13}O_4(OH)_{24}^{7+}$, which precipitates as $Al(OH)_3$, leading to a more effective treatment. Moreover, when the medium is strongly alkaline, aluminate ions $Al(OH)_4^-$ are formed, thus inhibiting the treatment (a soluble species which does not contribute to flocks formation) [43,44].

The increase of 22% in the COD removal (experiments 8–5) with a greater current density of 14.4–30 A/m^2, for example, is due to the increase of the dissolution of aluminum in the anode, related to Faraday's Law. This therefore causes the potentiation of coagulation and the removal of contaminants. The greater current density also increases the coagulant surface area, due to the active sites and the H_2 bubble density generated at the cathode [22,45], implying the flotation of pollutants and greater removal of COD. Moreover, the increased current intensity results in a significant decrease in the diameter of the gas bubbles, which accelerates the efficiency of the electrocoagulation treatment [44].

From the statistical models generated [Eqs. (4) and (5)], multi-response optimization was performed using the Derringer desirability function, according to Fig. 3. This

Table 3
Design and results of the central composite design (CCD) from chemical oxygen demand removal (COD removal) e specific energy consumption (*w*) of EC treatment

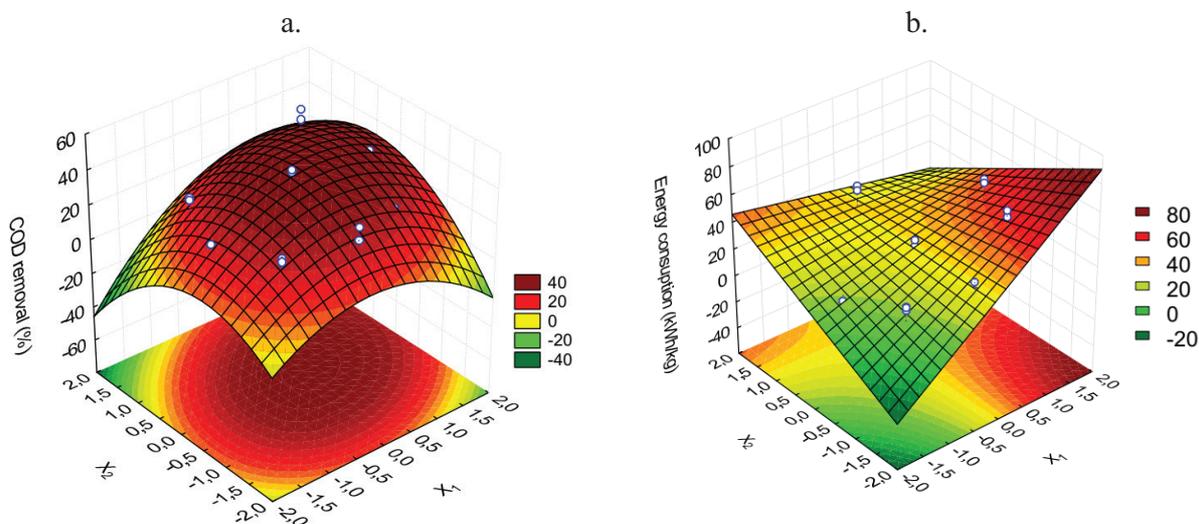| Exp.	J (A/m^2, X_1)	pH (X_2)	COD Removal (%)	EEC (kWh/kg)
1	20 (–1)	4.0 (–1)	24.38 ± 1.54	19.57 ± 1.20
2	40 (+1)	4.0 (–1)	26.11 ± 1.57	55.37 ± 3.37
3	20 (–1)	8.0 (+1)	26.11 ± 1.05	23.02 ± 0.54
4	40 (+1)	8.0 (+1)	48.28 ± 4.18	19.29 ± 0.77
5	30 (0)	6.0 (0)	44.58 ± 1.05	31.11 ± 2.69
6	30.0 (0)	3.2 (–α)	32.55 ± 5.12	27.05 ± 4.25
7	30.0 (0)	8.8 (+α)	21.70 ± 1.27	44.94 ± 2.65
8	14.4 (–α)	6.0 (0)	22.60 ± 0.01	11.23 ± 0.01
9	43.2 (+α)	6.0 (0)	37.08 ± 1.28	54.58 ± 1.88

Mean ± standard deviation, *n* = 2

Fig. 2. Response surface graph for the effect of current density (X_1) and pH (X_2) on the COD removal (a) and specific energy consumption (b) of electrocoagulation treatment.
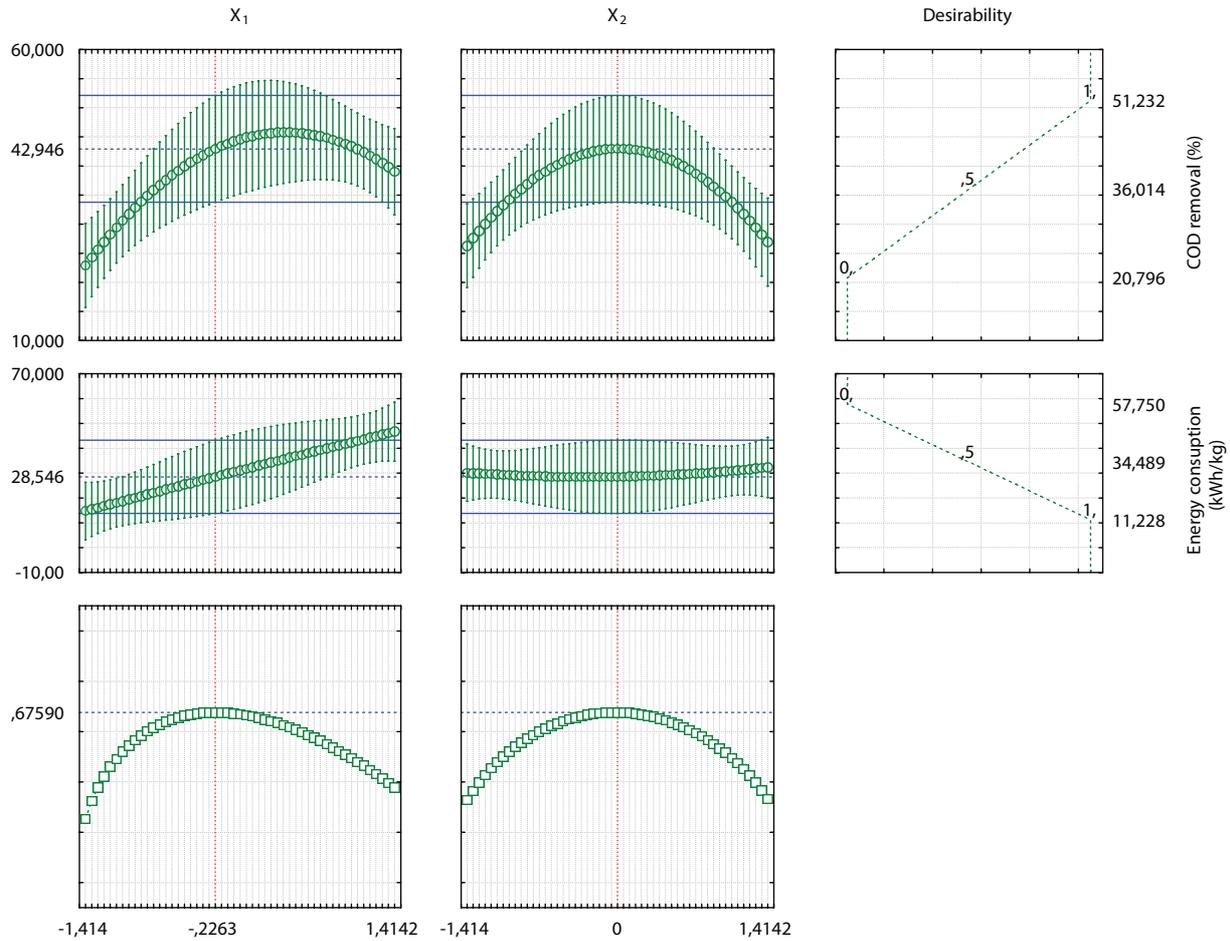
Fig. 3. Desirability profile for optimization of the process parameters.

function was applied in order to maximize the removal of COD and reduce the specific energy consumption. The conditions were optimized for a current density of 29.44 A/m^2 and pH 6. The predicted values for this condition were 42.9% of COD removal and a treatment energy consumption of 28.5 kWh/kg, corresponding to 1.3 kWh/m^3 of treated wastewater. Gerek et al. [46] observed a specific energy consumption of around 7 kWh/kg (8.33 kWh/m^3) for 82% of COD removal of tannery wastewater at 200 A/m^2 and pH 7. However, tannery wastewater has a COD concentration of around 1000 mg/L (four times higher than this study), which facilitates removal and explains the lower specific consumption (in kWh/kg). El-Ashtoukhy et al. [47] observed a specific energy consumption of 15.1 kWh/kg for the treatment of car-wash effluents with COD concentrations between 350 and 550 mg/L at 115 A/m^2 and pH 7.15. Asaithambi et al. [22] demonstrate that the lower the COD concentration, the higher the specific energy consumption, and the specific energy consumption value observed in this work is average compared to those commonly observed by other authors [13,17].

The electrocoagulation effluent in optimized conditions was completely characterized, according to Table 4. It is observed that COD reduced from 77.19 mg/L to 31.58 mg/L. This reduction is greater than expected from the statistical model. In addition, reductions of above 90% of phosphorus, turbidity, total and thermotolerant coliforms were observed. However, the presence of aluminum was observed in the EC effluent, due to the production of Al at the anode by electrolysis. In addition, the presence of color (up to 20 Hazen) and residual COD justify the use of an effective method for the removal of soluble substances present in the EC effluent.

3.2. Fixed bed adsorption by activated carbon

After the electrolytic treatment, significant increases in the wastewater quality were observed. However, considering the need for a higher quality for reuse in more restrictive situations, the wastewater was polished in adsorption columns with activated carbon. Fig. 4 shows the characterization of color and residual turbidity as a function of treatment time.

It is observed at the beginning that the residual color concentrations were close to 2 Hazen, with a removal of approximately 90%. After a few minutes, the residual color began to increase, reaching the first break point (5 Hazen) at 240 min. The second break point (10 Hazen) was reached at

Table 4
Characterization of the effluent after each treatment step applied

| Parameter	Electrocoagulation	Adsorption		
		<5 hazen	5–10 hazen	>10 hazen
pH	7.07 ± 0.12	7.61 ± 0.04	7.68 ± 0.03	7.69 ± 0.11
COD (mg/L^{-1})	31.58 ± 1.41	<1	19.12 ± 0.46	66.11 ± 0.92
BOD (mg/L^{-1})	6.67 ± 0.94	<1	18.89 ± 0.18	<1
N-Total (mg/L^{-1})	22.4 ± 0.8	49.7 ± 1.9	55.1 ± 2.4	56.0 ± 2.2
P-total (mg/L^{-1})	0.16 ± 0.01	0.14 ± 0.03	<0.01	<0.01
Suspended solids (mg/L)	18.0 ± 0.1	17.0 ± 4.2	24.0 ± 0.1	16.0 ± 2.8
Dissolved solids (mg/L)	359.3 ± 5.0	355.0 ± 7.1	434.0 ± 42.1	770.0 ± 33.9
Turbidity (NTU)	0.6 ± 0.02	0.53 ± 0.03	0.61 ± 0.03	0.91 ± 0.25
Color (hazen)	22.02 ± 1.08	2.81 ± 0.28	7.11 ± 0.31	10.67 ± 0.28
NO$_3^-$ (mg/L^{-1})	<0.04	<0.04	<0.04	<0.04
NO$_2^-$ (mg/L^{-1})	1.08 ± 0.02	1.09 ± 0.03	1.08 ± 0.04	1.08 ± 0.02
Thermotolerant coliforms (MPN/mL)	<1	3.6	<1	<1
Total coliforms (MPN/mL)	<1	43	23	<1
Aluminum (mg/L^{-1})	0.43 ± 0.01	0.05 ± 0.01	<0.01	<0.01
Surfactants (mg/L)	0.23 ± 0.03	0.10 ± 0.01	0.11 ± 0.03	0.11 ± 0.02

Mean ± standard deviation, $n = 3$
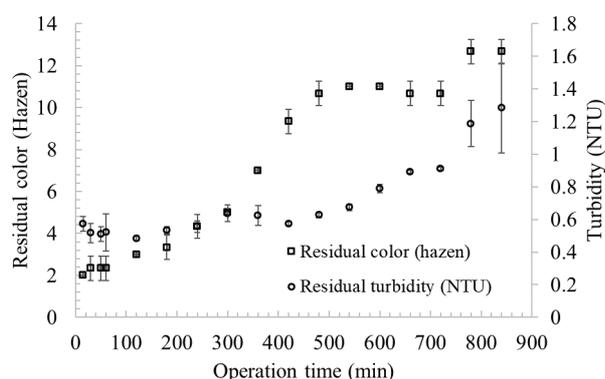
Fig. 4. Breakthrough curve analysis for fixed-bed removal of color of reclaimed water at $Q = 10$ mL/min (2 cm/min); L = 10 cm; T = 20°C.

420 min. This behaviour suggests that the adsorption bed is partially saturated throughout the experiment. At the end of the experiment (14 h), the residual color reached 16.67 Hazen, equivalent to a 24.22% removal. This shows that the adsorption system has not been fully saturated.

However, the turbidity of the wastewater treated by adsorption presents values approximately equal to the feed (0.6 NTU) up to 500 min. After this, the turbidity increases gradually, reaching values higher than 1 NTU after 780 min, due to effluent loading particles.

Table 4 shows the global characterization of the adsorption effluent for the three color ranges. It can be observed that the adsorption process was efficient to reduce most of the effluent parameters, especially in the first 240 min of operation. EC coupled with adsorption was able to remove the COD, BOD, aluminum and surfactants at trace concentrations close to the limit of detection of the methods. The

increase of microbiological load and nitrogen concentration was observed, due to their presence in the activated carbon. Similar results were observed by García et al. in 2018 [48], and the microbiological contamination can be reduced with the addition of Cl$_2$. The increase in COD and dissolved solids was observed after 420 min, suggesting that these substances were charged in the effluent, drastically reducing its quality. Adsorption is well known to remove pollutants efficiently, even at low concentrations such as those observed in this work.

3.3. Options for reuse

The characterization of the electrocoagulation coupled with adsorption effluent is shown in Table 4. Compared to the water quality parameters (Table 4) for reuse suggested by the *Guidelines for water reuse* of the USA Environmental Protection Agency [37], in Table 5, it is possible to observe that the physical–chemical and biological parameters were reached in the first adsorption break point (240 min or color < 5 hazen) for most reuse class. According to the guide, the reclaimed water obtained is amenable to residential urban use, irrigation of gardens, discharge of sanitary appliances, as well as non-potable applications with public access. In addition, reuse for irrigation of food crops for processed and non-food, including industrial (applications and facilities, energy production and extraction of fossil fuels), recreational (restricted and non-restricted), environmental and commercial (soil compacting, concretes, etc.) can also be carried out with the obtained effluent.

In addition, several authors have reported the successful reuse of wastewaters treated by conventional and advanced techniques with similar characteristics to the reclaimed water obtained in this work.

Loy et al. [38] demonstrated that municipal wastewater treated by conventional activated sludge and anaerobic

Table 5
Summarized guidelines for water reuse from EPA [37]

Parameter	More restrictive class	Less restrictive class
pH	6,5–8,5[10,11,12]	6–9[1,2,3,4,5,6,7,8,9]
BOD (mg/L)	≤ 10[1,3,5]	≤ 30[2,4,6,7,8,9]
Turbidity (NTU)	≤ 2[1,3,5,10,11,12]	NA[2,4,6,7,8,9]
Total Col. (MPN/100 mL)	ND[1,3,5,10,11,12]	< 200[2,4,6,7,8,9]
TSS (mg/L)	< 30[2,4,7,8,9]	NA[1,3,5,6,10,11,12]
TOC (mg/L)	< 2[10,11,12]	NA[1,2,3,4,5,6,7,8,9]
Cl_2 (mg/L)	> 1[1,2,3,4,5,6,7,8,9,10,11,12]	

Biological oxyge demand (BOD), Suspended solids (TSS), Total coliforms (Total Col.), Total organic carbon (TOC) residual chloro (Cl_2), not applicable (NA) and not detectable (ND). The class of EPA guidelines were:

[1]Unrestricted urban reuse

[2]Restricted urban reuse

[3]Agricultural reuse in food crops

[4]Agricultural reuse in processed food crops or non-food crops

[5]Unrestricted impoundments

[6]Restricted impoundments

[7]Environmental reuse

[8]Industrial reuse in once-through cooling

[9]Industrial reuse in recirculating cooling towers

[10]Groundwater recharge by spreading into potable aquifers

[11]Groundwater recharge by injection into potable aquifers

[12]Augmentation of surface water supply reservoirs

digesters causes less soil degradation than the use of brackish groundwater for the irrigation of farms. Intriago et al. [19] used reclaimed water with similar microbiological contamination for lettuce irrigation. Šrámková et al. [39] verified that wastewater with turbidity, color or organic load similar to this work can be used for irrigation or cooling of industrial systems. Sala and Gutiérrez-Bouzán [40] noted that reclaimed water with organic loads similar to this study did not cause changes when used in the tannery process, making it possible to reduce by up to 70% the water consumption in this type of industry with reuse techniques. Compared with the results observed by Moazzem et al. [33] the reclaimed water of this study can be utilized for car washing.

4. Conclusions

In this work we used an electrocoagulation process coupled with adsorption in fixed bed columns of activated carbon for the post-treatment of wastewater for reuse purposes. The electrocoagulation process was optimized using the response surface methodology. With the pH of the wastewater and current density applied at the optimum conditions (pH 6.0 and current density of 29.44 A/m^2) it was possible to remove 44.58% of the COD at an energy consumption of treatment of 31.1 kWh/kg (1.3 kWh/m^3). Under these conditions, we also observed significant reduc-

tions of phosphorus (70.27%), surfactants (63.33%), total coliforms (98.54%) and thermotolerant coliforms (96.1%). The electrocoagulation process coupled to fixed bed adsorption of activated carbon was able to remove almost 100% of the BOD and to qualify the wastewater for reuse according to international standards. Finally, comparing the quality of the effluent obtained with reuse standards and literature data, we observed that the electrocoagulation process conjugated to fixed bed adsorption of activated carbon is an effective treatment method for the post-treatment of wastewater for various reuse purposes.

References

[1] J. Löwenberg, A. Zenker, M. Baggenstos, G. Koch, C. Koch, T. Wintgens, Comparison of two PAC/UF processes for the removal of micropollutants from wastewater treatment plant effluent: process performance and removal efficiency, Water Res., 56 (2014) 26–36.

[2] Y. Luo, W. Guo, H.H. Ngo, L.D. Nghiem, F.I. Hay, J. Zhang, S. Liang, X.C. Wang, A review on the occurrence of micropollutants in the aquatic environment and their fate and removal during wastewater treatment, Sci. Total Environ., 473 (2014) 619–641.

[3] Y. Lorenzo-Toja, I. Vázquez-Rowe, S. Chenel, D. Marín-Navarro, M.T. Moreira, G. Feijoo, Eco-efficiency analysis of Spanish WWTPs using the LCA + DEA method, Water Res., 68 (2015) 651–666.

[4] L. Rizzo, A. Fiorentino, M. Grassi, D. Attanasio, M. Guida, Advanced treatment of urban wastewater by sand filtration and graphene adsorption for wastewater reuse: Effect on a mixture of pharmaceuticals and toxicity, J. Environ. Chem. Eng., 3 (2015) 122–128.

[5] A. Barhoumia, S. Ncib, W. Bouguerra, B. Hamrouni, E. Elaloui, Combining adsorption on activated carbon with electrocoagulation process for copper removal from used water, Desal. Water Treat., 83 (2017) 212–221.

[6] M. Mittersteiner, F. Schmitz, I.O. Barcellos, Reuse of dye-colored water post-treated with industrial waste: Its adsorption kinetics and evaluation of method efficiency in cotton fabric dyeing, J. Water Process. Eng., 17 (2017) 181–187.

[7] P. Bhattacharya, S. Sarkar, S. Ghosh, S. Majumdar, A. Mukhopadhyay, S. Bandyopadhyay, Potential of ceramic microfiltration and ultrafiltration membranes for the treatment of gray water for an effective reuse, Desal. Water Treat., 51 (2013) 4323–4332.

[8] M. Ebrahimi, S. Kerker, S. Daume, M. Geile, F. Ehlen, I. Unger, S. Schutz, P. Czermak, Innovative ceramic hollow fiber membranes for recycling/reuse of oilfield produced water, Desal. Water Treat., 55 (2015) 3554–3567.

[9] X. Song, W. Luo, J. McDonald, S.J. Khan, F.I. Hai, W.E. Price, L.D. Nghiem, An anaerobic membrane bioreactor–membrane distillation hybrid system for energy recovery and water reuse: Removal performance of organic carbon, nutrients, and trace organic contaminants, Sci. Total Environ., 628 (2018) 358–365.

[10] L. Wang, W. Liang, W. Chen, W. Zhang, J. Mo, K. Liang, B. Tang, Y. Zheng, F. Jiang, Integrated aerobic granular sludge and membrane process for enabling municipal wastewater treatment and reuse water production, Chem. Eng. J., 337 (2018) 300–311.

[11] M. Saleem, A.A. Bukhari, M.N. Akram, Electrocoagulation for the treatment of wastewater for reuse in irrigation and plantation, J. Basic Appl. Sci., 7 (2011) 11–20.

[12] J.S. Kim, T. Kim, I. Han, Phosphorus removal by a combined electrocoagulation and membrane filtration process for sewage reuse, Desal. Water Treat., 54 (2015) 956–965.

[13] S.B. Kausley, C.P. Malhotra, A.B. Pandit, Treatment and reuse of shale gas wastewater: Electrocoagulation system for enhanced removal of organic contamination and scale causing divalent cations, J. Water Process Eng., 16 (2017) 149–162.

[14] X. Li, H. Shi, K. Li, L. Zhang, Combined process of biofiltration and ozone oxidation as an advanced treatment process for wastewater reuse, Front. Env. Sci Eng., 9 (2015) 1076–1083.

[15] G. Balcıoğlu, Z.B. Gönder, Baker's yeast wastewater advanced treatment using ozonation and membrane process for irrigation reuse, Process. Saf. Environ. Prot., 117 (2018) 43–50.

[16] C. Wang, W. Chou, Y. Kuo, Removal of COD from laundry wastewater by electrocoagulation/electroflotation, J. Hazard. Mater., 164 (2009) 81–86.

[17] A.R. Makwana, M.M Ahammed, Continuous electrocoagulation process for the post-treatment of anaerobically treated municipal wastewater, Process Saf. Environ., 102 (2016) 724–733.

[18] R. Mores, H. Treichel, C.A. Zakrzevski, A. Kunz, J. Steffens, R.M. Dallago, Remove of phosphorous and turbidity of swine wastewater using electrocoagulation under continuous flow, Sep. Purif. Technol., 171 (2016) 112–117.

[19] J.C. Intriago, F. López-Gálvez, A. Allende, G.A. Vivaldi, S. Camposeo, M.N. Nicolás, J.J. Alarcón, S.P. Salcedo, Agricultural reuse of municipal wastewater through an integral water reclamation management, J. Environ. Man., 213 (2018) 135–141.

[20] M. Millán, M.A. Rodrigo, C.M. Fernández-Marchante, S. Díaz-Abad, M.C. Peláez, P. Cañizares, J. Lobato, Towards the sustainable powering of the electrocoagulation of wastewater through the use of solar-vanadium redox flow battery: A first approach, Electrochim. Acta., 270 (2018) 14–21.

[21] V.B. Brião, M. Hemkemeier, J.S. Piccin, E.B. Brião, Micro and Ultrafiltration Coupled with Electrocoagulation for the Advanced Treatment of Sewage for Urban Water Reuse,In: Jenessa Terrell. (Org.), Water Filtration Systems: Processes, Uses and Importance, Nova Science Publishers, New York 2016, pp. 0–149.

[22] P. Asaithambi, A.R.A Aziz, B. Sajjadi, W.M.A.B.W. Daud, Sono assisted electrocoagulation process for the removal of pollutant from pulp and paper industry effluent, Environ. Sci. Pollut. Res., 24 (2017) 5168–5178.

[23] J.Y. Chu, Y.R. Li, Experimental study on the ultrasound enhanced electro-coagulation for treatment of car-washing wastewater, Environ. Pollut. Control, 34 (2012) 1–3.

[24] M. Panizza, C. Giacomo, Applicability of electrochemical methods to carwash wastewaters for reuse, Part 2: Electrocoagulation and anodic oxidation integrated process, J. Electroanal. Chem., 638 (2010) 236–240.

[25] H. Zhou, C. Wey, F. Zhang, J. Liao, Y. Hu, H. Wu, Energy-saving optimization of coking wastewater treated by aerobic bio-treatment integrating two-stage activated carbon adsorption, J. Clean. Prod., 175 (2018) 467–476.

[26] N.V. Narayanan, M. Ganesan, Use of adsorption using granular activated carbon (GAC) for the enhancement of removal of chromium from synthetic wastewater by electrocoagulation, J. Hazard. Mater., 16 (2009) 575–580.

[27] S. Bellebia, S. Kacha, A.Z. Bouyakoub, Z. Derriche, Experimental investigation of chemical oxygen demand and turbidity removal from cardboard paper mill effluents using combined electrocoagulation and adsorption processes, Environ. Prog. Sustain., 31 (2012) 361–370.

[28] E. Bazrafshan, M.R. Alipour, A.H. Mahvi, Textile wastewater treatment by application of combined chemical coagulation, electrocoagulation, and adsorption processes, Desal. Water Treat., 57 (2016) 9203–9215.

[29] A. Aouni, R. Lafi, A. Hafiane, Feasibility evaluation of combined electrocoagulation/adsorption process by optimizing operating parameters removal for textile wastewater treatment, Desal. Water Treat., 60 (2017) 78–87.

[30] APHA (2012) Standard methods for the examination of water and wastewater, 22th ed.Washington (DC): American Public Health Association.

[31] X. Zhu, J. Ni, J. Wi, X. Xing, H. Ling, Destination of organic pollutants during electrochemical oxidation of biologically-pretreated dye wastewater using boron-doped diamond anode, J. Hazard. Mater., 189 (2011) 127–133.

[32] S. Zaidi, T. Chaabane, V. Sivasankar, A. Darchen, R. Maachi, T.A.M. Msagati, M. Prabhakaran, Performance efficiency of electro-coagulation coupled electro-flotation process (EC-EF) versus adsorption process in doxycycline removal from aqueous solutions, Process. Saf. Environ., 102 (2016) 450–461.

[33] S. Moazzem, J. Wills, L. Fan, F. Roddick, V. Jegatheesan, Performance of ceramic ultrafiltration and reverse osmosis membranes in treating car wash wastewater for reuse, Environ. Sci. Pollut. Res., 25 (2018) 8654–8668.

[34] D.C. Montgomery, Design and analysis of experiments, John Wiley & Sons, New Jersey, 2017.

[35] C. Saucier, M. Adebayo, E. Lima, L. Prola, P. Thue, C. Umpierres, M. Puchana-Rosero, F. Machado, Comparison of a homemade bacuri shell activated carbon with carbon nanotubes for food dye removal, Clean - Soil Air Water, 43 (2017) 1389–1400.

[36] A.A.C. Leite, P. Thue, G. dos Reis, S. Dias, E. Lima, J. Vaghetti, F. Pavan, W. de Alencar, Activated carbon from avocado seeds for the removal of phenolic compounds from aqueous solutions, Desal. Water Treat., 71 (2017) 168–181.

[37] EPA (2012) Guidelines for water reuse. Washington (DC): United States Environmental Protection Agency.

[38] S. Loy, A.T. Assi, R.H. Mohtar, C. Morgan, A. Jantrania, The effect of municipal treated wastewater on the water holding properties of a clayey, calcareous soil, Sci. Total Environ., 643 (2018) 807–818.

[39] M.V. Šrámková, V. Diaz-Sosa, J. Wanner, Experimental verification of tertiary treatment process in achieving effluent quality required by wastewater reuse standards, J. Water Process Eng., 22 (2018) 41–45.

[40] M. Sala, M.C. Gutiérrez-Bouzán, Electrochemical treatment of industrial wastewater and effluent reuse at laboratory and semi-industrial scale, J. Clean. Prod., 65 (2014) 458–464.

[41] M. Kobya, H. Hiz, E. Senturk, C. Aidyner, E. Demirbas, Treatment of potato chips manufacturing wastewater by electrocoagulation, Desalination, 190 (2006) 201–211.

[42] M.Y.A. Mollah, R. Schennach, J.R. Parga, D.L. Cocke, Electrocoagulation (EC) – science and applications, J. Hazard. Mater., 84 (2001) 29–41.

[43] P.T. Srinivasan, T. Viraraghavan, K.S. Subramanian, Aluminium in drinking water: An overview, Water SA, 25 (1999) 47–55.

[44] G. Chen, Electrochemical technologies in wastewater treatment, Sep. Purif. Technol., 38 (2004) 11–41.

[45] G.K. Akkaya, E. Sekman, S. Top, E. Sagir, M.S. Bilgili, S.Y. Guvenc, Enhancing filterability of activated sludge from landfill leachate treatment plant by applying electrical field ineffective on bacterial life, Environ. Sci. Pollut. Res., 24 (2017) 10364–10372.

[46] E.E. Gerek, S. Yılmaz, A.S. Koparala, Ö.N. Gerek, Combined energy and removal efficiency of electrochemical wastewater treatment for leather industry, J. Water Process Eng., (2017) in press.

[47] E.S.Z. El-Ashtoukhy, N.K. Amin, Y.O. Fouad, Treatment of real wastewater produced from Mobil car wash station using electrocoagulation technique, Environ. Monit. Assess., 187 (2015) 628.

[48] E.A. García, M. Agulló-Barceló, P. Bond, J. Keller, W. Gernjak, J. Radjenovic, Hybrid electrochemical-granular activated carbon system for the treatment of greywater, Chem. Eng. J., 352 (2018) 405–411.
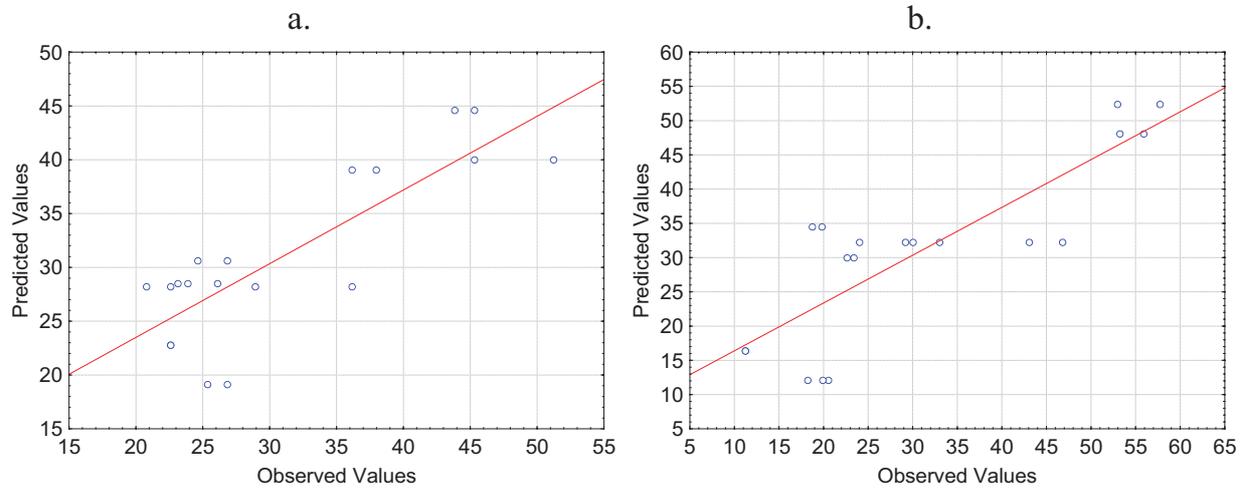
Supplementary material

Fig. S1. Residual plots of observed and predict values of COD removal (a) and energy consumption (b).

Table S1

Analysis of variance of current density (X_1) and pH (X_2) on the removal and energy consumption of electrocoagulation treatment

Effect	SS	DF	MS	F	p
ANOVA – COD removal					
X_1 (L)	543.6	1	543.6	14.4	0.002
X_1^2 (Q)	270.7	1	270.7	7.2	0.020
X_2 (L)	28.6	1	28.6	0.8	0.399
X_2^2 (Q)	389.3	1	389.3	10.3	0.007
$X_1 * X_2$	205.5	1	205.5	5.4	0.036
Error	491.0	13	37.8		
Total	1649.0	1649.0			
ANOVA – Specific energy consumption (w)					
X_1 (L)	2135.330	1	2135.330	21.99186	<0.001
X_1^2 (Q)	0.206	1	0.206	0.00212	0.964
X_2 (L)	28.884	1	28.884	0.29747	0.595
X_2^2 (Q)	10.789	1	10.789	0.11112	0.744
$X_1 * X_2$	730.386	1	97.096	7.52229	0.016
Error	1262.253	13	37.7708		
Total SS	4344.979	18			

Table S2

Regression coefficients of statistical model on the COD removal and energy consumption of electrocoagulation treatment (significant variables at p < 0.05)

Parameter	Reg. Coeff.	Std. error	t	p	+95%	–95%
COD removal						
Mean	44.58	4.31	10.3	<0.001	35.34	53.82
X_1 (L)	5.75	1.48	3.9	0.002	2.57	8.94
X_{12} (Q)	−6.84	2.52	−2.7	0.017	−12.24	−1.43
X_{22} (Q)	−8.19	2.52	−3.3	0.006	−13.60	−2.79
$X_1 * X_2$	4.69	2.04	2.3	0.038	0.31	9.07
Energy consumption (w)						
Mean	32.21	2.09	15.4	<0.001	27.79	36.64
X_1 (L)	11.20	2.21	5.1	<0.001	6.52	15.88
$X_1 * X_2$	−8.94	3.04	−2.9	0.010	−15.38	−2.50