Evaluation of electrocoagulation process for the removal of cypermethrin and chemical oxygen demand from livestock wastewater

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Abstract

The direct discharge of wastewater from livestock into freshwater courses, and surface runoff originating from processing areas, threatens aquatic ecosystems and the quality of drinking-water taken from streams. The present work aims to study the removal of chemical oxygen demand (COD) and cypermethrin from livestock wastewater generated after cattle bath by continuous electrocoagulation (EC) process. The effects of three major operating variables such as pH, retention time and voltage supply on COD and cypermethrin removals were examined. All these factors were found to have significant effects on COD and cypermethrin removals. At the optimal operating conditions (initial pH: 5, retention time: 30 min, and electric potential: 29.5 V), the EC effectively reduced the COD (93%) and cypermethrin (96%). In conclusion, continuous EC was successfully applied for the treatment of livestock wastewater (bathing cattle), with an operating cost of 2.2 US\$/m³.

Keywords: Cattle bath; pesticides; Chemical Oxygen Demand; Box–Behnken design; Response Surface Methodology

1. Introduction

The wastewater produced from rural agriculture and livestock production, is one of the leading causal factors in the loss of biodiversity and the leading source of water pollution in developed and emerging countries (FAO, 2006). In Colombia, cattle is one of the most representative sectors of the national economy; it contributes to 1.7% of the national gross domestic product (GDP), 20% of the agricultural GDP and 53% of the livestock sector GDP, and is considered as a socioeconomic line of great importance to the development of the field (Lafaurie, 2008). However, it has been and is challenged strongly by its productive performance and negative environmental impact (Mahecha et al., 2002; Steinfeld et al., 2009). It is well known that livestock wastewater contains high strength of chemical oxygen demand (COD), biological oxygen demand (BOD), color, nitrogen, phosphorus and suspended solids when mixed with cattle feces (Ferreira et al., 2003; Juteau et al., 2004; Lee and Shoda, 2008), and organic contaminants coming from livestock baths could not be disregarded. Control and elimination of ectoparasites (flies, ticks, mites) on cattle can be accomplished by either spraying or dipping the animal in a bath of pesticide. This frequent activity has a significant environmental impact on water bodies because a pesticide or a mixture of pesticides (e.g. chlorpyrifos, amitraz, ethion and cypermethrin) is discharged to aquatic ecosystems, causing resistance in ectoparasites because of its high residual and persistence effect (Benavides, 2001; Helena et al., 2003). Thus, studies of advanced and optimized treatment processes for livestock wastewater are necessary. In recent years, strict environmental regulations have called for new processes for efficient and adequate treatment of various wastewaters with relatively low operating costs. Electrocoagulation (EC) is an alternative wastewater treatment technology (Sahu et al., 2014; Moussa et al., 2017; Hakizimana et al., 2017). Currently, the interest of this technology has been renewed with a view to designing treatment plants with continuous operation especially for particular industrial applications; this, coupled with its proven ability to efficiently

remove various contaminants and their inherent simplicity in design and operation, has prompted the EC to be evaluated again as a suitable treatment technology for livestock wastewater (Holt et al., 2005). During the EC process, characterized by its simple equipment, easy operation and low levels of sludge, the coagulant is dissolved from the anode with a simultaneous formation of hydroxyl ions and hydrogen gas occurring at the cathode (Tezcan et al., 2014). The EC has been proved to be an efficient method to treat numerous varieties of wastewater. It has been successfully applied to treat different kinds (e.g. municipal, tanning, etc...) of wastewater (Kamaraj and Vasudevan, 2015; Sharma et al., 2014; Huang et al., 2017, Yetilmezsoy et al., 2009, Ilhan et al., 2008, Avsar et al., 2007, Asselin et al., 2008, El-Naas et al., 2008, Parga et al., 2005, Adhoum et al., 2004; Ahmad et al., 2005, Elabbas et al., 2016, Zhang et al., 2017). Moreover, the performance of EC technique for decolorization and COD reduction of livestock wastewater was successful investigated in a laboratory batch study (Tak et al., 2016) using Box–Behnken response surface design (BBD) coupled with desirability function methodology.

A similar approach has been undertaken at the present study to remove COD and cypermethrin from livestock wastewater after a common cattle bath operation. To achieve this goal, the individual and the interactive effect of process variables such as pH, the retention time and the applied voltage to get the maximum removal efficiency were investigated and optimized. The obtained results will be helpful to the implementation of EC treatment process in industrial level with lower environmental impact.

2. Materials and methods

2.1 Wastewater

Livestock wastewater used in the experiments was obtained from a cattle estate Cordoba-Colombia, a day bathing cattle to control ectoparasites. The wastewater was

taken after completion of the activity once the barnyards were washed. Average production of wastewater effluent is 3000 L month⁻¹. The livestock wastewater had a Chemical Oxygen Demand (COD) of 2.500 ± 58 mg L⁻¹, cypermethrin (Cyp) of 30 ± 0.5 mg L⁻¹ and a pH of 6.2. The physicochemical characteristics are determined using APHA standard methods (2012). Cypermethrin analysis was performed by high-performance liquid chromatography (HPLC-DAD) using EPA method 1660 (EPA, 1993).

2.2. Experimental setup and procedure

The experimental set up used in this study is shown in Fig. 1, which mainly consisted of a cell of 5 L as a reactor to hold a sample of 4 L. An aeration system which increases the size of flocs and improves contact between precipitates and the target contaminant (Moussavi et al. 2011) was composed of two tank pump (1.5 mL min⁻¹) and a network of plastic tubes (diameter: 0.5 mm) distributed evenly in the cell. Iron (Fe) and Aluminum (Al: sacrificial electrode) plates of 0.25 cm thickness 2.5 cm width x 15 cm high dimensions were used as electrodes, with a distance between them of 1 cm. Because only 9 cm of electrode is submerged, the total effective surface area of both electrodes was 100.25 cm². The desired electrical potential was maintained constant by means of a direct current power supply DC - PHYWE (0–50 V). An hydraulic flocculator to generate more amounts of flocs avoiding their breakdown was established by an industrial transparent hose with three different diameters and lengths, adapted by an interconnection which allowed the flow directly enter the settler composed of a 50L plastic container and a bottom valve to expel sludge. All experiments were conducted in continuous mode of operation and pH of the wastewater was adjusted using 0.1 N HCl or NaOH. In each experimental run, a wastewater sample (40 L) was rigorously stirred by a mechanical stirrer. Livestock wastewater (bathing cattle) was fed into the reactor by a peristaltic pump (MICROGON

INC, Laguna Hills, Model No: 92653) with feeding rates of 267, 133.5 and 88.9 mL/min corresponding to the retention time of 15 min, 30 min and 45 min, respectively. After the experiments, the electrodes were polished, washed with dilute H₂SO₄ and then rinsed with distilled water before each run. After passing each current for each time period (i.e., after each batch experiment), the sample was transferred allowing to settle down. After a settling time of 20 min, the supernatant sample was collected to perform the analysis of COD and cypermethrin.

2.3. Experimental design

The experimental design and mathematical modelling were performed using Response surface methodology (RSM). Box–Behnken design (BBD) was used for optimization of operating parameters for the removal of pollutants by EC with three main factors at three levels: pH (A), retention time (B), and potential (C) under the RSM technique to optimize experimental variables (Table 1). Responses included in the model were removal efficiency of COD and cypermethrin. The aim of the experimental design and analysis was to define the effective factors, and to select the levels which give the maximum COD and cypermethrin removal efficiency. All the statistical analyses were done with the help of Statgraphics Centurion XV.II software.

2.4. Electrical energy consumption

The electrical energy consumption of present EC treatment process was calculated using the following equation (Thirugnanasambandham et al., 2015).

$$E = \frac{V \cdot I \cdot t}{V_s} \quad (1)$$

Where E is the electrical energy (kWh/m³), V is the cell voltage (in volt, V), I is the electrical current (in ampere, A), V_s is the volume of the treated wastewater (L), and t is the time of electrocoagulation process (h).

3. Results and discussion

Table 2 shows the predicted and experimental values of the response variables COD removal and cypermethrin removal, based on the BBD experiments and factors pH, retention time and voltage supply. The multiple regression analysis method was used to analyze the results obtained from the BBD experiments, and a second order polynomial equation was used to express the empirical relationship between the response and independent variables. The final empirical models developed to predict the performance of the process are presented as follows:

$$\text{COD removal, \%} = 44.7555 - 1.20982A - 2.05877B + 1.77684C - 0.226562A^2 + 0.0212795AB + 0.0075AC - 0.0371938B^2 + 0.00243812BC - 0.0301375C^2$$

(2)

$$\text{Cypermethrin removal, \%} = 72.496 - 6.92752A + 2.16959B + 1.04299C - 0.139375A^2 + 0.0149279AB + 0.172875AC - 0.0324756B^2 - 0.00918878BC - 0.02415C^2$$

(3)

where A is the solution initial pH, B is retention time, and C is the potential. Positive terms indicate a positive effect on the COD or the cypermethrin removal, whereas negative terms indicate that the parameter has negative effect.

To consider the relationship between the predicted values from the model calculated by Eqs. (2) and (3) and the observed values, the data obtained was very close to linear (significant correlations with the results obtained experimentally COD: $R = 0.989$; Cyp: $R = 0.972$) indicating that both values were accurate and reliable, as presented in Fig. 2. Table 3 shows the analysis of variance (ANOVA) tests at 95% confidence level. Response function estimates agreed well with the experimental data, i.e. $R^2 = 98\%$ for COD and $R^2 = 94\%$ for cypermethrin. It can be seen that linear term (A), i.e. pH, is effective factor on the COD and cypermethrin removal. As can be seen in Table 3, the square of terms, such as B^2 and C^2 are statistically significant ($p < 0.05$) for COD

removal. Unlike the B^2 and interaction AC are effective factors on the cypermethrin removal because of their p values ($p < 0.05$).

3.1. Effect of operational parameters

The individual and interactive effects of the process variables on the responses were studied through a three dimensional (3D) response surface plots (Figure 3). The response surface plots were constructed from the developed models, and they were also used to locate the optimal condition of each factor in order to find out the maximum removal efficiency of COD and cypermethrin. In general, the removal of both variables increases at acidic pH values, possibly due to the higher rate of floc formation (Prieto, 2014). Other studies reported that acid affects the mechanism of coagulation leading to the formation of different chemical species in wastewater (Mahesh et al., 2006, Yetilmezsoy et al., 2009, Tak et al., 2015). At basic pH values, the removal efficiency possibly decreases due to the formation of hydroxide radical species that do not react with the aluminum cation (Chen, 2004, Bayramoglu, 2004, Daneshvar et al, 2007).

On the other hand, the effect of the retention time on treatment efficiency results is shown in Fig. 3. The COD and cypermethrin removal efficiency increase with increasing retention time which can be explained by the high mass transport due to the forced convection in the bulk, the reaction and the hydrogen production at the cathode as a function of the electrolysis time, build-up of more ions by the sacrificial electrode in the early stages of the electrolysis, and consequently increasing the removal efficiency by causing the precipitation of the organic matter (Mouedhen et al., 2008; Zaroual et al., 2009; Tezcan et al., 2014; Tak et al., 2015).

The potential is one of the most important parameters that affect the electrochemical treatment method to treat livestock wastewater significantly. The applied voltage is the major driving force in electrochemical reactors, because increased voltage promotes the availability of new reactions by enabling higher half cell voltage and current density increase for oxidation reactions (Card et al., 1990; Tezcan et al., 2014). As can be seen in Fig.3, the initial elimination rate of COD and cypermethrin increase with increasing applied voltage, proving that fastest electrochemical reactions are enabled also with increased voltage. Generally, the mechanisms thought to be responsible for organic matter degradation can be stated not only accomplished by the oxidants that are formed electrochemically, but also by direct anodic oxidation where the pollutants are adsorbed onto the anode surface and destroyed by the anodic electron transfer reaction (Güvena et al., 2009).

3.2 Optimization of treatment conditions

Using the application simultaneous optimization of multiple responses and desired function methodology on the results of BBD, the optimal operating conditions provided by the model were: initial pH (5), retention time (30min) and potential (29.5 V), with removal efficiencies for COD and cypermethrin in 94 and 100%, respectively, with electrical current 0.05 A. No significant differences were observed when the predicted response values were compared with the corresponding experimental value (COD: 92.8%, cypermethrin: 96.2%, $p>0.05$), which confirms that RSM can be used to optimize the process parameters (Raghu and Basha, 2007). The consistency between the predicted and experimental results verified the validation of the model and the existence of an optimal point. In addition, the energy supplied is 18.4 kWh/m³ considering the optimal operating conditions. Therefore, taking into account the price of energy in Colombia is on average around \$340 COP/KWh (0.12 US\$/KWh), the cost is 2.2 US\$/m³. In comparison with other studies, the cost is lower than that for the

removal of COD in the treatment of meat industry wastewater (3.99 US\$/m³) (Pinedo et al, 2015). Moreover, the cost is quite similar to that reported by Varank et al. (2014) for tannery wastewater treatment (2.92 US\$/m³), and for the removal of color and COD in the treatment of meat industry wastewater (1.6 US\$/m³) (Thirugnanasambandham et al., 2015). Results indicated that this electrochemical treatment process in continuous mode reduces the cost of traditional start-stop-start electrocoagulation process with a reasonable operating cost. Moreover, it must be taken into account that the cost could be reduced when implementing photovoltaic systems.

4. Conclusions

Treatment of livestock wastewater by continuous electrocoagulation has been proven as an effective process for the removal of COD and cypermethrin, being a viable alternative for its application in the cattle sector. Under optimal operating conditions, such as pH: 5, electrolysis time: 30 min and voltage supplied: 29.5 V, the removal efficiency of COD and cypermethrin were found to be 92.8% and 96.2%, respectively, with a reasonable operating cost of 2.2 US\$/m³.

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Figure 1. Schematic diagram of the experimental set-up.

Figure. 2. Regression plots of the actual data against the predicted values from the response surface models describing (a) COD removal and (b) Cypermethrin removal efficiencies by the EC process.

Figure 3. 3D Response surface plots for COD removal (a) and Cypermethrin removal (b)

a)

b)

Table 1. The level and range of factors chosen for the design of experiments for the EC optimization.

Variable (unit)	Factor	Coded levels of variables		
		-1	0	+1
pH	A	3	5	7
retention time (min)	B	15	30	45
potential (V)	C	20	30	40

Table 2. Factors and responses from the BBD

Run	Independent variables			Removal efficiency (%)			
				Experimental		Predicted	
	pH	Retention time (min)	Potential (V)	COD	Cyp	COD	Cyp
1	1	0	1	83.33	96.56	82.85	94.88
2	0	1	-1	81.97	89.91	82.67	90.64
3	1	0	-1	81.53	82.53	81.35	82.84
4	-1	0	-1	92.31	99.31	92.78	100.98
5	-1	0	1	93.51	99.51	93.68	99.19
6	-1	1	0	91.55	99.55	90.54	98.00
7	0	-1	1	83.58	92.58	82.96	92.87
8	0	-1	-1	83.19	89.19	82.11	86.46
9	1	1	0	77.45	83.45	78.78	85.70
10	1	1	0	81.98	88.98	81.29	88.08
11	0	1	1	84.03	90.03	84.94	91.72
12	-1	-1	0	90.16	97.16	90.51	97.34
13	0	0	0	90.82	97.82	91.59	98.08
14	0	0	0	91.76	97.47	91.59	98.08
15	0	0	0	92.19	98.19	91.59	98.08

Table 3. ANOVA for responses COD and Cypermethrin.

Source	Sum of squares		DF		Mean square		F		P	
	COD	Cyp	COD	Cyp	COD	Cyp	COD	Cyp	COD	Cyp
A:pH	196.884	207.834	1	1	196.884	207.834	125.5	37.85	0.0001*	0.0016*
B:Retention time	3.23851	4.63601	1	1	3.23851	4.63601	2.04	0.84	0.2103	0.4003
C:Potential	4.35555	25.1011	1	1	4.35555	25.1011	2.78	4.57	0.1565	0.0855
A ²	3.03245	1.14759	1	1	3.03245	1.14759	1.93	0.21	0.2232	0.6667
A B	1.72704	0.84992	1	1	1.72704	0.84992	1.10	0.15	0.3421	0.7102
A C	0.09	47.8172	1	1	0.09	47.8172	0.06	8.71	0.8202	0.0318*
B ²	107.824	82.2034	1	1	107.824	82.2034	68.73	14.97	0.0004*	0.0118*
B C	0.56680	8.05075	1	1	0.56680	8.05075	0.36	1.47	0.5740	0.2801
C ²	33.5361	21.5344	1	1	33.5361	21.5344	21.38	3.92	0.0057*	0.1045
Error total	7.8442	27.4543	5	5	1.56884	5.49086				
Total (corr.)	384.019	464.423	14	14						
R ²	0.979	0.940								
adjust R ²	0.942	0.834								

* $P < 0.05$